

# BASIC RADIATION PHYSICS AND SOURCES OF RADIATION

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## 1. INTRODUCTION

Treatment of materials and products with radiation in order to modify their physical, chemical and biological properties is defined as radiation processing of materials. Radiation processing can be controlled and used for the development of the novel materials and products with desirable properties.

The knowledge of the basic radiation physics, including the structure of matter, elements of nuclear physics, the nature of electromagnetic radiation, and radiation interaction with matter is required to understand irradiation processing and its potential in material sciences.

### 1.1. CLASSIFICATION OF RADIATION

Radiation-induced changes in materials depend on the origin and type of radiation and the deposited energy (Fig.1).

Energy deposition processes in turn depend on the origin of the radiation:

- particulate radiation, including electrons, positrons, protons, neutrons, ions;
- electromagnetic radiation, which covers a broad wavelength range, including infrared, visible and ultraviolet radiation and X-rays and gamma rays.

The following Table 1 gives approximate wavelengths, frequencies, and energies for selected regions of the electromagnetic spectrum.

Deposition of kinetic energy of accelerated particles in a target is considered when discussing the interaction of particulates with matter. In the case of electromagnetic radiation, interaction with matter, in general, energy transferred by the individual quanta, known as photons, is taken into account. The energy of photon (quantum energy),  $E$ , is given by:

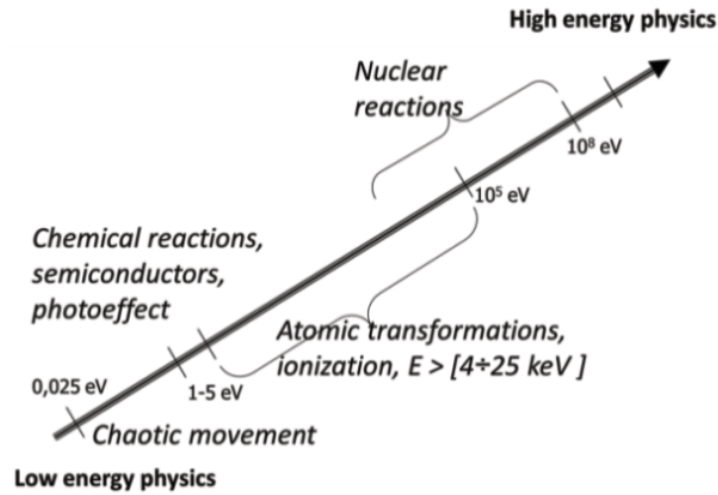


Fig.1. Comparison of energies.

Table 1. Spectrum of the electromagnetic radiation.

Wave region	Wavelength [m]	Frequency [Hz]	Energy [eV]
Radio	$> 0.1$	$< 3 \times 10^9$	$< 10^{-5}$
Microwave	$0.1-10^{-4}$	$3 \times 10^9-3 \times 10^{12}$	$10^{-5}-0.01$
Infrared	$10^{-4}-7 \times 10^{-7}$	$3 \times 10^{12}-4.3 \times 10^{14}$	$0.01-2$
Visible	$7 \times 10^{-7}-4 \times 10^{-7}$	$4.3 \times 10^{14}-7.5 \times 10^{14}$	$2-3$
Ultraviolet	$4 \times 10^{-7}-10^{-9}$	$7.5 \times 10^{14}-3 \times 10^{17}$	$3-10^3$
X-rays	$10^{-9}-10^{-13}$	$3 \times 10^{17}-3 \times 10^{21}$	$10^3-10^7$
Gamma rays	$< 10^{-11}$	$> 3 \times 10^{19}$	$> 10^5$

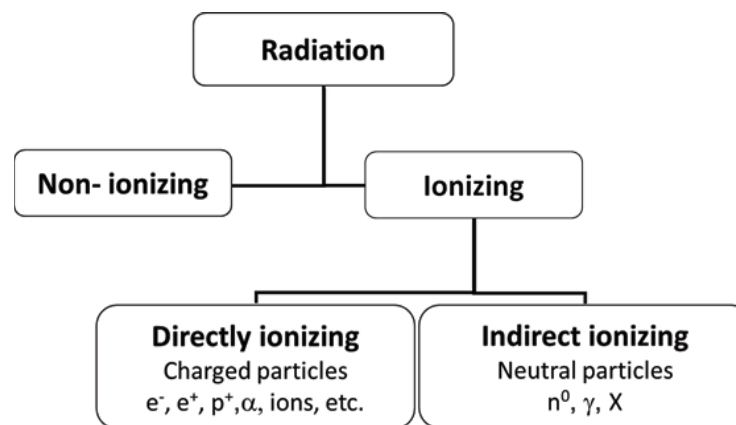


Fig.2. Classification of radiation.

$$E = h\nu = h \frac{c}{\lambda} \quad (1)$$

where: the constant  $h$  is known as Planck's constant,  $\nu$  – a frequency,  $c$  – a speed of light in vacuum,  $\lambda$  – wavelength.

Radiation is classified in two main categories, non-ionizing and ionizing radiation depending on its ability to ionize matter (Fig.2):

- Direct ionizing radiation corresponds to the energy deposition in the material by energetic charged particle which have Coulomb interaction with an orbital electron of a target atom.
- Indirect ionizing radiation is realized in two steps. First, fast charged particles (electrons and positrons) are released in the material due to the photon energy deposition or due to kinetic energy deposition by neutrons, protons or heavier ions. Second, the released charged particles deposit their energy directly in the material through Coulomb interactions between these particles and orbital electrons of an atom.

## 2. ATOMIC AND NUCLEAR STRUCTURE

### 2.1. BASIC DEFINITIONS FOR ATOMIC STRUCTURE

The atom is composed of a central nucleus surrounded by a cloud of negatively charged electrons. An atomic nucleus consists of  $Z$  protons, and  $N$  neutrons. The main characteristics of the constituents of an atom are shown in Table 2.

Table 2. Main characteristics of the atom constituents.

Particle	Symbol	Mass [kg]	Energy [MeV]	Charge
Proton	p	$1.672 \times 10^{-27}$	938.2	+
Neutron	n	$1.675 \times 10^{-27}$	939.2	0
Electron	e	$0.911 \times 10^{-30}$	0.511	-

The radius of an atom is  $\sim 10^{-10}$  m, the radius of the nucleus is about  $10^{-14}$  m. Protons and neutrons are commonly referred to as nucleons.

The number of protons in atom is known as the atomic number,  $Z$ . It equals the number of electrons in a non-ionized atom, thus making an atom neutral.

Atomic mass number,  $A$ , equals to the number of protons plus neutrons in the nucleus.

Atomic mass,  $M$ , might be expressed in mass units – g or in atomic mass units – u, where u is equal to 1/12 of the mass of the  $^{12}\text{C}$  atom or  $931.5 \text{ MeV}/c^2$ .

Some binding energy is required to keep the nucleons within the nucleus. Thus the atomic mass of particular nuclide is smaller than the sum of the individual masses of constituent particles.

Number of atoms,  $N_a$ , per mass of an element is:

$$\frac{N_a}{m} = \frac{N_A}{A} \quad (2)$$

where  $N_A$  is Avogadro's number ( $N_A = 6.022 \times 10^{23}$  atoms/g-atom).

Number of electrons,  $N_e$ , per mass of element is:

$$\frac{N_e}{m} = Z \frac{N_a}{m} = \frac{Z}{A} N_A \quad (3)$$

Number of electrons,  $N_e$ , per volume,  $V$ , of an element is:

$$\frac{N_e}{V} = \rho Z \frac{N_a}{m} = \rho Z \frac{N_A}{A} \quad (4)$$

In nuclear physics, a nucleus  $X$  with atomic mass number  $A$  and atomic number  $Z$  is denoted as  ${}^A_Z X$ , for example  ${}^{60}_{27} \text{Co}$  and  ${}^{137}_{55} \text{Cs}$ .

An atomic nucleus identified by its atomic element and its mass number is defined as nuclide.

Atoms having an identical atomic number,  $Z$ , but different atomic mass numbers,  $A$ , related to different numbers of neutrons in the nucleus are called isotopes of a given element, for example:  ${}^{11}_6 \text{C}$ ,  ${}^{12}_6 \text{C}$ ,  ${}^{13}_6 \text{C}$ ,  ${}^{14}_6 \text{C}$ .

In ion physics it is usual to provide ions with superscripts  $+/-$ . For example,  ${}^4_2 \text{He}^{2+}$  stands for a doubly ionized He atom which is the alpha particle [1].

## 2.2. ATOMIC STRUCTURE

The currently accepted simplified atom model relies on the 1913 Bohr theory [2, 3] and his famous postulates that combine classical non-relativistic mechanics with quantum mechanics adding the concept of angular momentum quantization. A variety of postulated formulations (physical content being the same) are provided in the literature [4-6]. Here is a summary of Bohr's postulates, as noted in Ref. [7]:

- Postulate 1: Electrons revolve about the Rutherford nucleus in well-defined, allowed orbits (planetary-like motion):

$$F_{\text{coul}} = \frac{1}{4\pi\epsilon_0} \frac{Ze^2}{r_e} \equiv F_{\text{cent}} = \frac{m_e v_e^2}{r_e} \quad (5)$$

- Postulate 2: While in orbit, the electron does not lose any energy despite being constantly accelerated (no energy loss while electron is in an allowed orbit).

- Postulate 3: The angular momentum  $L = m_e v r$  of the electron in an allowed orbit is quantized and given as  $L = n\hbar$ , where  $n$  is an integer referred to as a principal quantum number,  $\hbar = h/2\pi$  and  $h$  is Planck's constant.
- Postulate 4: An atom emits radiation only when an electron makes a transition from the initial orbit with a quantum number  $n_i$  to final orbit with a quantum number  $n_f$  (energy emission during orbital transitions).

$$h\nu = E_i - E_f \quad (6)$$

Electron transitions result in the emission of photons. The wavenumber  $k$  of the emitted photon is given by:

$$k = \frac{1}{\lambda} R_\infty \left( \frac{1}{n_f^2} - \frac{1}{n_i^2} \right) \equiv 109737 \text{ cm}^{-1} Z^2 \left( \frac{1}{n_f^2} - \frac{1}{n_i^2} \right) \quad (7)$$

where  $R_\infty$  is the Rydberg constant.

The radius  $r_n$  of one-electron Bohr atom is given by:

$$r_n = a_0 \left( \frac{n^2}{Z} \right) = 0.529 \text{ \AA} \left( \frac{n^2}{Z} \right) \quad (8)$$

where  $a_0 = 0.529 \text{ \AA}$  is the Bohr's radius.

The energy levels for orbital electron shells in one-electron atomic structure are given by:

$$E_n = -E_R \left( \frac{Z}{n} \right)^2 = -13.6 \text{ eV} \left( \frac{Z}{n} \right)^2 \quad (9)$$

where:  $E_R$  – the Rydberg energy,  $n$  – the principal quantum number ( $n = 1$ , ground state,  $n > 1$ , excited state),  $Z$  – atomic number of one-electron atomic structure.

An energy level diagram for the hydrogen (H) atom is shown in Fig.3A.

Bohr's theory works well for one-electron structures (hydrogen atom, singly ionized helium atom and doubly ionized lithium atom, *etc.*), but it does not apply directly to multi-electron atoms because of the repulsive Coulomb interactions among the atomic electrons. Development of the theory of quantum mechanics by Heisenberg, Schrödinger, Dirac, Pauli and others, contributed significantly to the explanation of possible energy levels (states) that might be occupied by electrons in a multi-electron atom. In this theory, individual energy states are defined by four quantum numbers as follows [8]:

- the principal quantum number,  $n$ , which specifies the ground (main) energy shell and can take integer values;
- the azimuthal quantum number,  $l$ , which specifies the total rotational angular momentum for the electron and can take integer values between 0 and  $n - 1$ ;
- the magnetic quantum number,  $m$ , which specifies a component of the angular momentum and can take integer values between  $-l$  and  $+l$ ;
- the spin quantum number,  $s$ , which specifies a component of the spin angular momentum of the electron and takes values  $-1/2$  or  $+1/2$ .

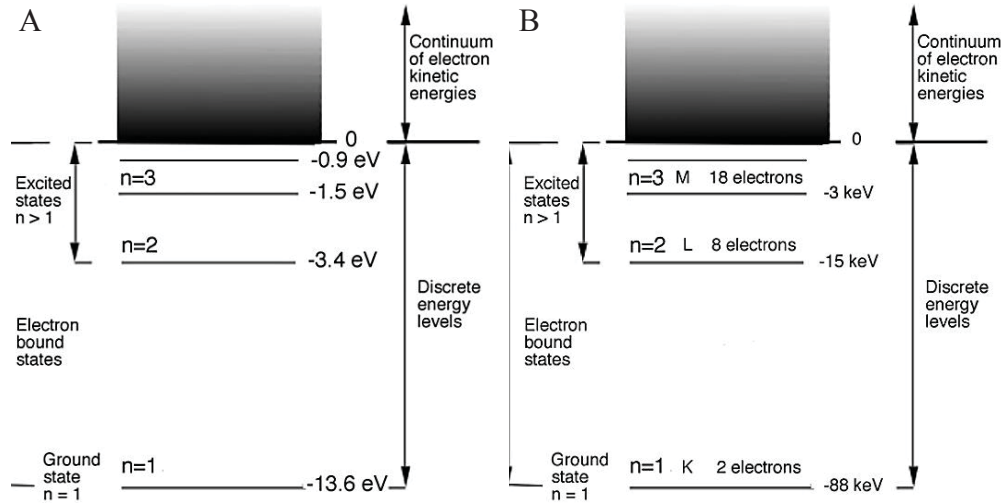


Fig.3. Energy level diagrams: A – for a hydrogen atom, B – for a lead atom. (Adapted from Ref. [7]).

Electrons occupy allowed shells; however, the number of electrons per shell according to the Pauli exclusion principle is limited to  $2n^2$ .

The distribution of energy levels in a multi-electron atom (Pb) is shown in Fig.3B.

The energy levels associated with the various electron orbits (not drawn to scale) increase with  $Z$  and decrease with quantum number  $n$  and the average distance from the nucleus. The outer electronic shell (the valence shell) determines the chemical properties of the element. The energy bands associated with  $n = 1, 2, 3, \text{etc.}$ , are known as the K, L, M, *etc.*, bands. The structure of each band arises from small differences in energy associated with both the  $l$  and  $s$  quantum numbers.

In a multi-electron atom, inner shell electrons are bound with much larger energies  $E_n$  than  $E_R$  in single-electron model:  $E_n = -E_R(Z_{\text{eff}}^2/n^2)$ , and the corresponding atomic radius is:  $r_n = a_0(n^2/Z_{\text{eff}})$ , where  $Z_{\text{eff}}$  is the effective atomic number, given by  $Z_{\text{eff}} = Z - s_c$ , with  $s_c$  as the screening constant, which equals to 2 for K-shell electrons.

There are two main processes when an electron is removed from a given shell in the atom: excitation and ionization. Both of them occur within the atom through various possible interactions (energy transition) which will be discussed in Chapter 2.

Excitation of an atom is present when an electron is moved from a given shell to a higher  $n$  shell which is empty or is not filled by corresponding number of electrons. Excitation energy (excitation potential) is a minimum energy required to excite an atom from its ground state to a higher state (Fig.4A).

Ionization of an atom occurs when an electron is removed from the atom (a certain amount of energy is transferred to the electron which is sufficient to

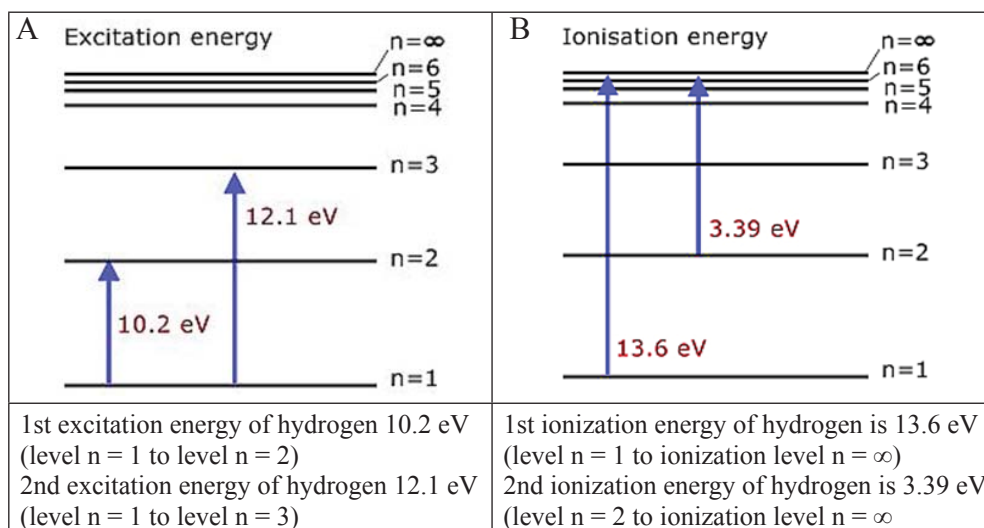


Fig.4. Excitation scheme (A) and ionization scheme (B) of hydrogen atom.

overcome its binding energy in the shell). Ionization energy (ionization potential) is a minimum energy required to release electron from atom or ion (Fig.4B).

An orbital electron from a higher  $n$  shell will fill the electron vacancy in a lower  $n$  atomic shell. The energy difference between two shells will be either emitted as a (fluorescent) photon or it will be transferred to the higher  $n$ -shell electron, which will be ejected from the atom as an Auger electron.

The minimum energy required to ionize the atom (ionization potential) ranges from a few (alkali elements) to 24.5 eV (helium) [9].

### 2.3. NUCLEAR STRUCTURE

Most of the mass of an atom is concentrated in the atomic nucleus, consisting of  $Z$  protons and  $N = (A - Z)$  neutrons and having radius:

$$r = r_0 \sqrt[3]{A} \quad (10)$$

where  $r_0$  is a constant ( $\sim 1.4$  fm) assumed equal to  $\frac{1}{2}$  of  $r_e$ , the classical electron radius.

The constituents of the nucleus, protons and neutrons (nucleons), are bound in the nucleus with a strong force. This short-range ( $10^{-15}$  m) force exceeds not only the long-range electromagnetic force between charged nucleons (protons), which is repulsive and tends to disrupt the nucleus, but also other known natural forces (gravitational, weak interaction) by several orders of magnitude and holds different nucleons (protons-neutrons) together in the nucleus. The energy associated with the strong force is called binding energy.

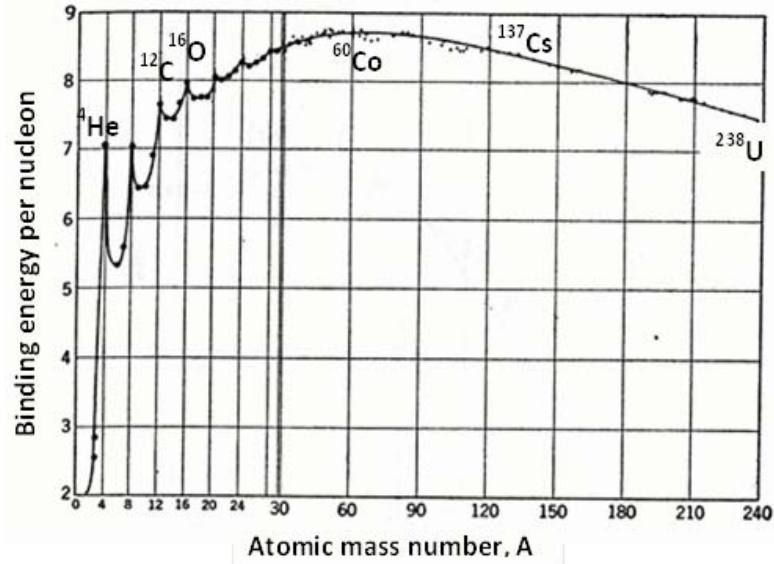


Fig.5. Binding energy per nucleon for different elements. (Adapted from Ref. [10]).

The binding energy per nucleon,  $E_B/A$ , in a nucleus varies gradually with the number of nucleons,  $A$  (Fig.5). It may be calculated from the energy equivalent of the mass deficit (defect),  $\Delta_m$ , of a given nucleus as:

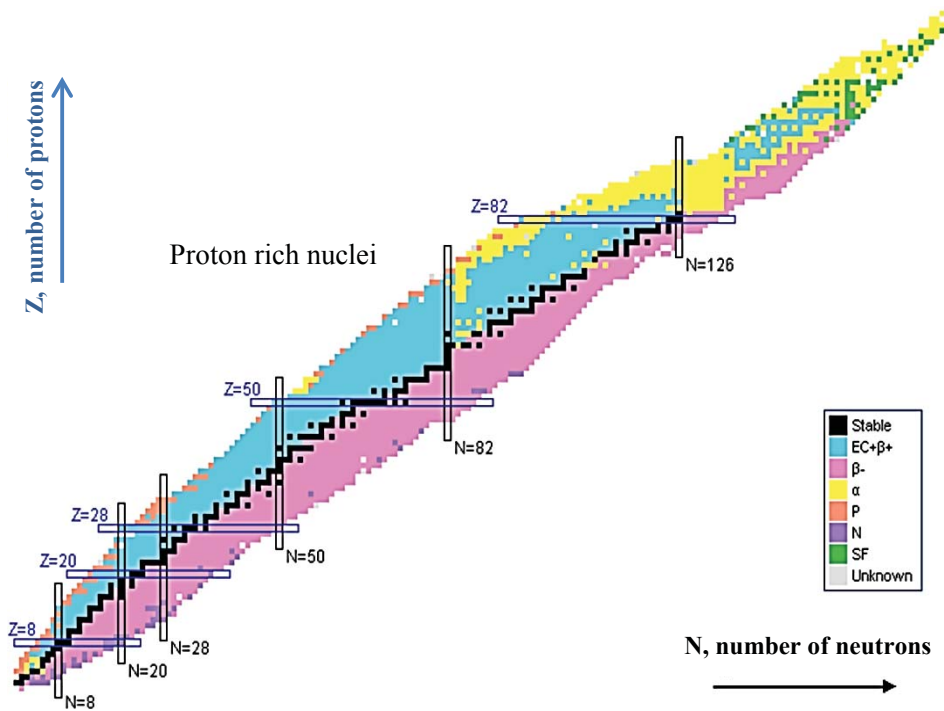


Fig.6. Nuclear stability chart. The stability line is indicated in black. (Adapted from Ref. [11]).



$$\frac{E_B}{A} = \frac{\Delta mc^2}{A} = \frac{Zm_p c^2 + (A - Z)m_n c^2 - Mc^2}{A} \quad (11)$$

where:  $M$  – the nuclear mass, expressed in atomic mass units –  $u$  ( $Mc^2 = 931.5$  MeV);  $m_p c^2$  – the proton rest energy;  $m_n c^2$  – the neutron rest energy.

A stable nucleus has enough binding energy to hold the nucleons together permanently. There is no basic relation between the atomic mass number  $A$  and atomic number  $Z$  of a nucleus, but an empirical relationship:

$$Z = \frac{A}{1.98 + 0.0155A^{2/3}} \quad (12)$$

gives a good approximation for a stable nucleus.

Strong nuclear forces and the associated binding energy in the nucleus determine the stability of the nucleus (balance of protons and neutrons). Too many neutrons or too many protons upset this balance disrupting the binding energy of the strong nuclear forces making the nucleus unstable (Fig.6).

### 3. NUCLEAR TRANSFORMATIONS

The nuclear transformations (transmutations) play a significant role in the development of new materials. Materials in which nuclear transformation processes take place are known as natural radiation sources. They represent a powerful tool for radiation-induced modification of materials (especially in nuclear energy and in the biomedical field), since the result of every transmutation process is an energy release [12]. Nuclear transmutation energy is released as:

- Kinetic energy of the product particles.
- Almost immediate emission of very high energy photons, *i.e.* prompt gamma rays, or it is a postponed energy release through gamma decay to the ground state of the nucleus, which is present, when nucleus is firstly transformed to a metastable state.
- A small amount of energy may also emerge in the form of X-rays. (Generally, the product nucleus has a modified atomic number, so the configuration of its electron shells is destroyed. As the electrons rearrange themselves and drop to lower energy levels, X-rays, due to internal transitions, may be emitted).

There are four major types of nuclear transmutation [4]:

- Radioactive decay, in which nuclei spontaneously eject one or more particles and lose energy to become the nuclei of lighter atoms. Examples are alpha, beta and gamma decays.
- Fission, which is the splitting of a nucleus into two “daughter” nuclei, *e.g.*  
 $n + {}_{92}^{235}\text{U} \rightarrow {}_{92}^{236}\text{U} \rightarrow {}_{56}^{141}\text{Ba} + {}_{36}^{92}\text{Kr} + 3n.$

- Fusion of two parent nuclei into one daughter nucleus, e.g.  ${}^1_1\text{N} + {}^1_1\text{N} \rightarrow {}^2_1\text{N} + e^+ + \nu_e$ , where  $\nu_e$  stands for (electron) neutrino – which is an elementary particle holding no electrical charge, travelling at nearly the speed of light, and passing through ordinary matter with virtually no interaction.
- Neutron capture, in which the nuclear charge ( $Z$ , the atomic number) is unchanged, the nuclear mass ( $A = \text{number of protons} + \text{neutrons}$ , the atomic mass) increases by one, and the number of neutrons ( $N$ ) increases by one.

The simplified overview of nuclear transmutations is provided in Fig.7.

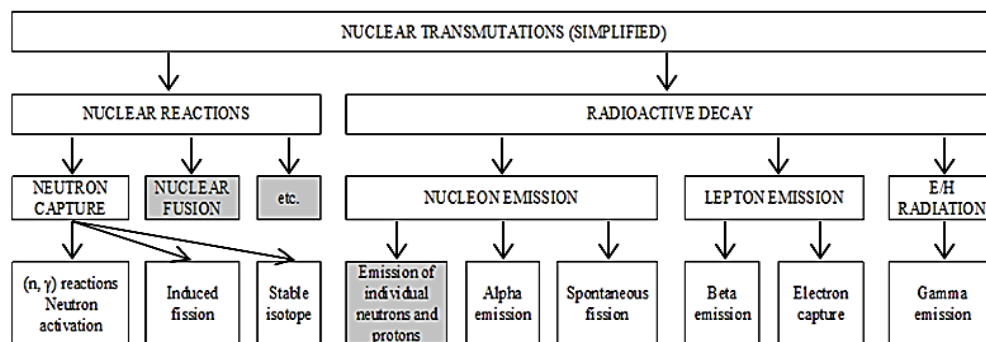


Fig.7. Nuclear transmutation processes. Processes indicated in gray boxes are not considered for discussion in this book.

### 3.1. RADIOACTIVITY

Radioactivity is a process characterized by a transformation of an unstable nucleus into more stable state that may also be unstable and will decay further through a chain of decays until a stable nuclear configuration is reached. The energy difference between the two quantum states is called the decay energy,  $Q$ , and is emitted from the nucleus in the form of electromagnetic radiation (gamma rays) or in the form of kinetic energy of the reaction products.

All radioactive processes are governed by the same formalism based on:

- substance related characteristic parameter called the decay constant  $\lambda$ ;
- activity,  $A(t)$ , which represents the total number of disintegrations (decays) of nuclei per unit time and is defined as:

$$A(t) = \lambda N(t) \quad (13)$$

where  $N(t)$  is the number of radioactive nuclei at time  $t$ .

The SI unit of activity is the becquerel, Bq ( $1 \text{ Bq} = 1 \text{ s}^{-1}$ ), but the older unit of activity, the curie, Ci ( $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$ ), originally defined as the activity of 1 g of  ${}^{226}\text{Ra}$ , is also used.

The simplest radioactive decay involves a transition with a decay constant  $\lambda_p$  from a quantum state of the unstable parent nucleus, P, to the quantum state of the stable daughter nucleus, D:  $P \xrightarrow{\lambda_p} D$ .

The rate of depletion of the number of radioactive parent nuclei,  $N_p$ , is equal to the activity of radioactive parent,  $A_p(t)$ , at time  $t$ :

$$\frac{dN_p(t)}{dt} = -A_p(t) = -\lambda_p N_p(t), \quad \int_{N_p(0)}^{N_p(t)} \frac{dN_p}{N_p} = -\int_0^t \lambda_p dt \quad (14)$$

where  $N_p(0)$  is the initial number of parent nuclei at time  $t = 0$ .

The number of radioactive parent nuclei  $N_p(t)$  in radioactive substance and the activity of radioactive parent  $A_p(t)$  as a function of time (Fig.8) may be defined as:

$$\begin{aligned} N_p(t) &= N_p(0)e^{-\lambda_p t} \\ A_p(t) &= \lambda_p N_p(t) = \lambda_p N_p(0)e^{-\lambda_p t} = A_p(0)e^{-\lambda_p t} \end{aligned} \quad (15)$$

where  $A_p(0)$  is the initial activity at time  $t = 0$ .

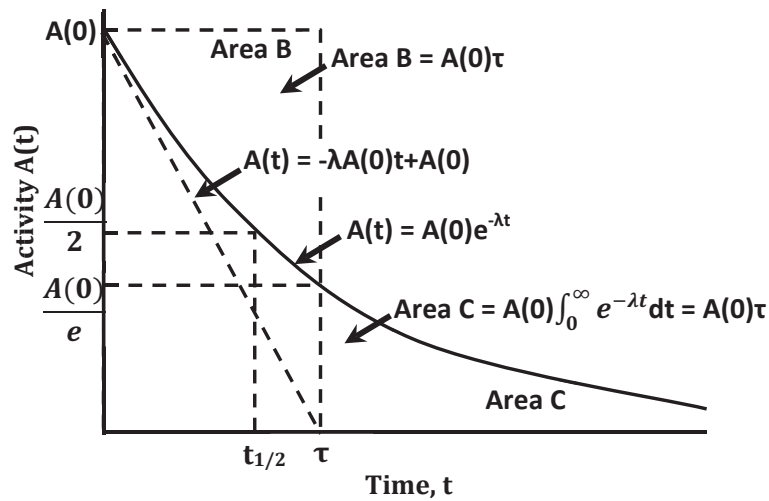


Fig.8. Plot of activity as a function of time.

The half-life,  $(t_{1/2})_p$  of radioactive parent, P, is the time during which the number of radioactive parent nuclei decay from the initial value  $N_p(0)$  at time  $t = 0$  to half of the initial value:

$$N_p(t = t_{1/2}) = \frac{N_p(0)}{2} = N_p(0)e^{-\lambda_p(t_{1/2})_p} \quad (16)$$

The same relationship is valid for the activity.

The average (mean) lifetime,  $\tau_p$  of a radioactive substance is the average life expectancy of all parent radioactive nuclei in the substance at time  $t = 0$ :

$$A_p(0)\tau = \int_0^{\infty} A_p(0)e^{-\lambda_p t} dt = \frac{A_p(0)}{\lambda_p} \quad (17)$$

The decay constant  $\lambda_p$ , the half-life  $(t_{1/2})_p$  and average lifetime  $\tau_p$  of a radioactive substance are related to each other as follows:

$$\lambda_p = \frac{\ln 2}{(t_{1/2})_p} = \frac{1}{\tau_p}, \quad (t_{1/2})_p = \tau_p \ln 2 \quad (18)$$

Specific activity  $a$  is the activity per unit mass:

$$a = \frac{A(t)}{M} = \frac{\lambda N(t)}{M} = \frac{\lambda N_A}{A} = \frac{N_A \ln 2}{A(\tau_{1/2})_p} \quad (19)$$

where  $N_A$  stands for Avogadro's number and  $A$  is the atomic mass number.

A more complicated radioactive decay occurs when a radioactive parent nucleus,  $P$ , decays with its decay constant  $\lambda_p$  into unstable daughter nucleus,  $D$ , which in turn decays with a decay constant  $\lambda_D$  into a stable granddaughter,  $G$ :  $P \xrightarrow{\lambda_p} D \xrightarrow{\lambda_D} G$ .

Time-dependent parent and daughter activities are shown in Fig.9.

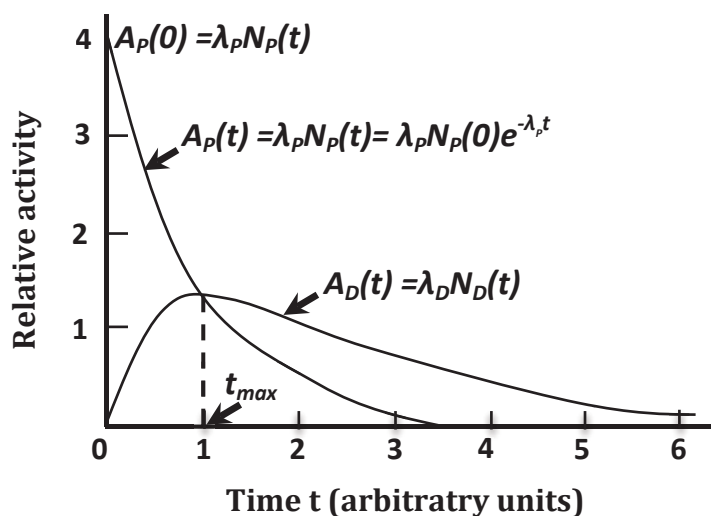


Fig.9. Parent and daughter activities plotted as a functions of time.

The activity of the daughter nuclei is expressed as:

$$A_D = \frac{\lambda_D}{\lambda_D - \lambda_p} A(0)(e^{-\lambda_p t} - e^{-\lambda_D t}) \quad (20)$$

The maximum activity of daughter nuclei occurs at time  $t_{max}$ :

$$t_{max} = \frac{\ln(\lambda_D / \lambda_p)}{\lambda_D - \lambda_p} \quad (21)$$

under condition, that  $N_D = 0$  at time  $t = 0$ .

There are some special considerations in the parent-daughter-granddaughter relationship:

- for non-equilibrium:

$$\lambda_D < \lambda_P \quad \text{or} \quad (t_{1/2})_D > (t_{1/2})_P, \quad \frac{A_D}{A_P} = \frac{\lambda_D}{\lambda_D - \lambda_P} \left[ 1 - e^{-(\lambda_D - \lambda_P)t} \right] \quad (22)$$

- for transient equilibrium:

$$\lambda_D > \lambda_P \quad \text{or} \quad (t_{1/2})_D < (t_{1/2})_P, \quad \frac{A_D}{A_P} = \frac{\lambda_D}{\lambda_D - \lambda_P}, \quad \text{for } t \gg t_{\max} \quad (23)$$

- for secular equilibrium:

$$\lambda_D \gg \lambda_P \quad \text{or} \quad (t_{1/2})_D \ll (t_{1/2})_P, \quad A_D/A_P \approx 1 \quad (24)$$

### 3.2. ACTIVATION OF NUCLIDES

Activation of nuclides is possible when a parent nuclide, P, interacts with thermal neutrons in a nuclear reactor. This interaction is followed by occurrence of a radioactive daughter nuclide, D, that decays into a granddaughter nuclide, G:  $P \xrightarrow{\sigma\dot{\phi}} D \xrightarrow{\lambda_D} G$ , where  $\dot{\phi}$  (in  $\text{cm}^{-2}\cdot\text{s}^{-1}$ ) indicates neutron fluence rate.

The probability for the parent nuclei activation is determined by nuclear reaction cross section,  $\sigma$ , expressed in barn/atom (1 barn =  $10^{-24}$   $\text{cm}^2$ ). In the case of parent nuclei activation, the expression for daughter activity is given in Eq. (21) where  $\lambda_p$  is replaced by the  $\sigma\dot{\phi}$ :

$$A_D(t) = \frac{\sigma\dot{\phi}\lambda_D}{\lambda_D - \sigma\dot{\phi}} N_p(0)(e^{-\sigma\dot{\phi}t} - e^{-\lambda_D t}) \quad (25)$$

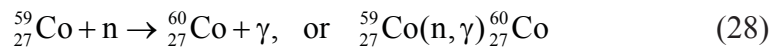
In the activation process the maximum activity of daughter nuclei occurs at time:

$$t_{\max} = \frac{\ln \lambda_D / \sigma\dot{\phi}}{\lambda_D - \sigma\dot{\phi}} \quad (26)$$

When  $\sigma\dot{\phi} \ll \lambda_D$ , the daughter activity expression transforms into:

$$A_D(t) = \sigma\dot{\phi}\lambda_D N_p(0)(1 - e^{-\lambda_D t}) \quad (27)$$

$^{60}_{27}\text{Co}$  being one of the most important radionuclides for industrial and medical irradiators is also produced artificially by bombarding stable  $^{59}_{27}\text{Co}$  with thermal neutrons in a nuclear reactor:



Estimated cross section for this process is  $\sigma = 37$  barn/atom. The typical neutron fluence rate  $\dot{\phi}$  is of the order of  $10^{14}$   $\text{cm}^{-2}\cdot\text{s}^{-1}$  [13, 14].

### 3.3. RADIOACTIVE DECAY MODES. NATURAL RADIATION SOURCES

Radioactive decay is a process in which an unstable parent nucleus, P, reaches a more stable daughter nucleus, D, through possible decay modes. The most important radioactive decay modes are: alpha ( $\alpha$ ) decay,  $\beta^+$  decay,  $\beta^-$  decay, electron capture ( $\epsilon$ ), and isomeric transition (IT) which is also called internal conversion (IC), spontaneous fission (sf), proton (p) decay, neutron (n) decay and special mixed beta-decay processes. Most of the decay processes are followed by gamma emissions.

Total energy of particles released from the decaying nucleus equals the decrease in the rest energy:

$$Q = [M_p - (M_D + m)]c^2 \quad (29)$$

where  $M_p$ ,  $M_D$  and  $m$  are the nuclear rest masses of the parent, daughter and emitted particle, respectively.

Radioactive decay processes are described in detail in Ref. [4]. Information on radionuclide decay data are found in special databases [11, 13, 14].

**Alpha decay** is a nuclear transformation in which an energetic alpha particle ( ${}^4_2\text{He}$  nucleus) is emitted:  ${}^A_Z\text{P} \rightarrow {}^{A-4}_{Z-2}\text{D} + {}^4_2\text{He}$ .

The process of alpha decay is found mainly in proton rich, high atom number ( $Z > 83$ ) nuclides (see Fig.6). The reason for alpha decay is an unbalance between two competing forces in the nucleus: electrostatic repulsion (Coulomb) force between protons and the strong interaction force between protons and neutrons.

The binding energy of alpha particle (potential barrier in the nucleus) is very high ( $EB = 28.3$  MeV).

The kinetic energy of alpha particles released by naturally occurring radionuclides is between 4 and 9 MeV.

An example of alpha decay for  ${}^{226}_{88}\text{Ra}$  is provided in Fig.10.

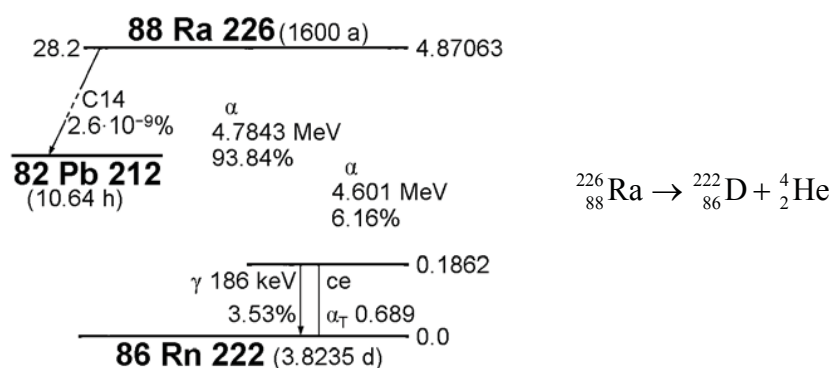


Fig.10. Alpha decay of  ${}^{226}_{88}\text{Ra}$ . Most important processes are:  $\alpha_1$  decay (4.7843 MeV, 93.84%) and  $\alpha_2$  decay (4.601 MeV, 6.16%) followed by gamma emission (186 keV, 3.53%). (Adapted from database [15], 2012).

**Beta decay** is a nuclear transformation in which an electron or positron is emitted. There are no electrons in nucleus. The reason for beta decay is quite different as compared to alpha decay. Weak interaction forces are responsible for beta decay, since the weak interaction causes transmutation of quarks, that are constituents of nucleons [16]. Six types of quarks are known, but nucleons are constructed from *up* and *down* quarks: a neutron consists of one *up* (+2/3e) and two *down* (-1/3e) quarks; a proton – two *up* (+2/3e) and one *down* (-1/3e) quarks. Weak interaction is the only process in which a quark can change to another quark.

In **neutron decay**, one *down* quark is changed to an *up* quark, transforming the neutron into a proton:

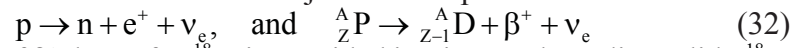


In **proton decay**, one *up* quark is changed to the *down* quark, transforming the proton into the neutron:



The number of nucleons and total charge are conserved in the beta decay process and the daughter, D, can be referred to as an isobar of the parent, P. This transformation is only possible, if  $M_p > M_D + m_e$ .

- $\beta^+$  decay occurs when, in a proton rich radioactive parent nucleus, a proton is converted into a neutron and a positron and neutrino. Sharing the available energy, positrons and neutrinos are ejected from parent nucleus:



An example of  $\beta^+$  decay for  ${}^{18}_9\text{F}$  is provided in Fig.11. The radionuclide  ${}^{18}_9\text{F}$  is a widely used tracer in nuclear medicine.

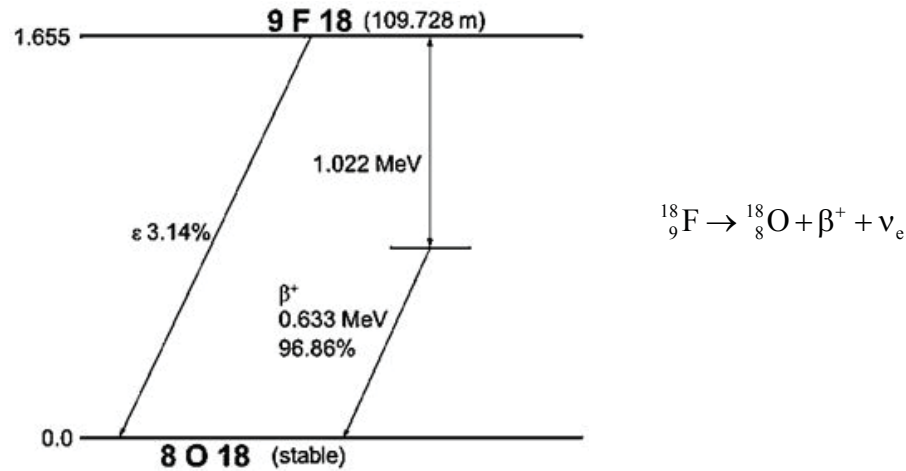


Fig.11.  $\beta^+$  decay of  ${}^{18}_9\text{F}$ . The main decay mode is positron emission (0.633 MeV, branching ratio 96.86%). Branching ratio for electron capture is 3.14%. No gamma emissions are observed. (Adapted from database [15], 2013).

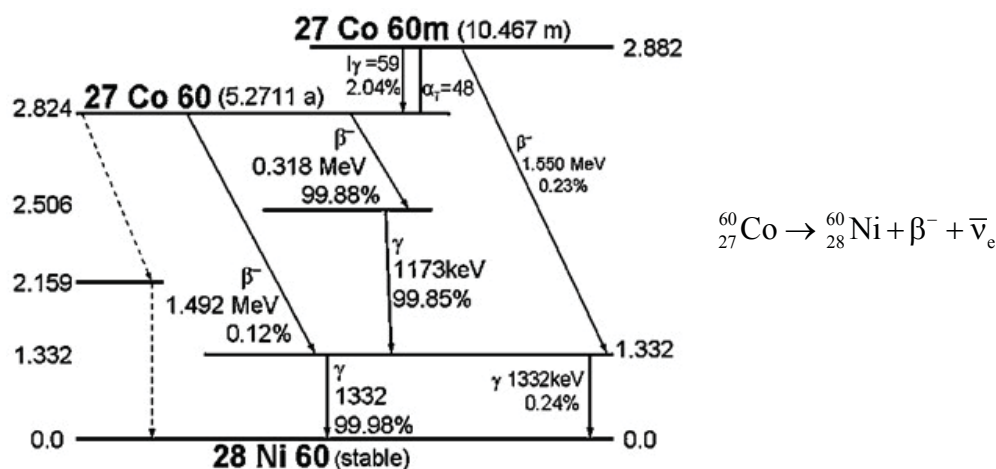
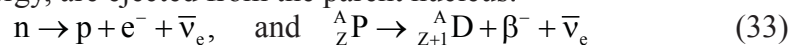


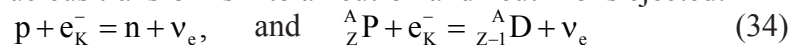
Fig.12.  $\beta^-$  decay of  $^{60}\text{Co}$ . In 99.88% cases electrons with a maximum energy of 0.318 MeV are emitted, the rest – 0.12% electrons – with a maximum energy of 1.492 MeV.  $\beta^-$  decay is followed by gamma emissions:  $E_{\gamma_1} = 1.173$  MeV, 99.85% and  $E_{\gamma_2} = 1.332$  MeV, 99.98%. A metastable state  $^{60\text{m}}\text{Co}$  is also indicated. The transition from metastable state to the ground state of  $^{60}\text{Co}$  occurs primarily through electronic capture. (Adapted from database [15], 2012).

- $\beta^-$  decay occurs when in a neutron-rich radioactive parent nucleus a neutron is converted into a proton and an electron and anti-neutrino, that share the available energy, are ejected from the parent nucleus:



An example of  $\beta^-$  decay for  $^{60}\text{Co}$  is provided in Fig.12.  $^{60}\text{Co}$  source is widely used in different industrial and medical applications. One gram of  $^{60}\text{Co}$  has an activity of *ca.* 43 TBq.

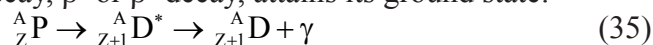
**Electron capture** is a process in which a neutron deficient nucleus captures an atomic electron from the inner K or L shells of the atomic orbits. As a result, a proton in the nucleus transforms into a neutron and neutrino is ejected:



This transformation is only possible, if  $M_p + m_e > M_D$ .

An example of electron capture process is provided in Fig.13.

**Gamma emission** is present when excited daughter nucleus, D, generally produced through alpha decay,  $\beta^-$  or  $\beta^+$  decay, attains its ground state:



If the daughter nucleus de-excites with a time delay, the excited state of the daughter is referred to as a metastable, \*, state. The nucleus in metastable state is called an isomer and the process of de-excitation is called an isomeric transition, with the emission of gamma photons. Radioactive decay related spontaneously emitted photons are from the range 5 keV-1.5 MeV, but in some cases also higher photon energies (6-7 MeV) are possible.

An example of gamma decay scheme of  $^{137}\text{Cs}$  source, is provided in Fig.14.



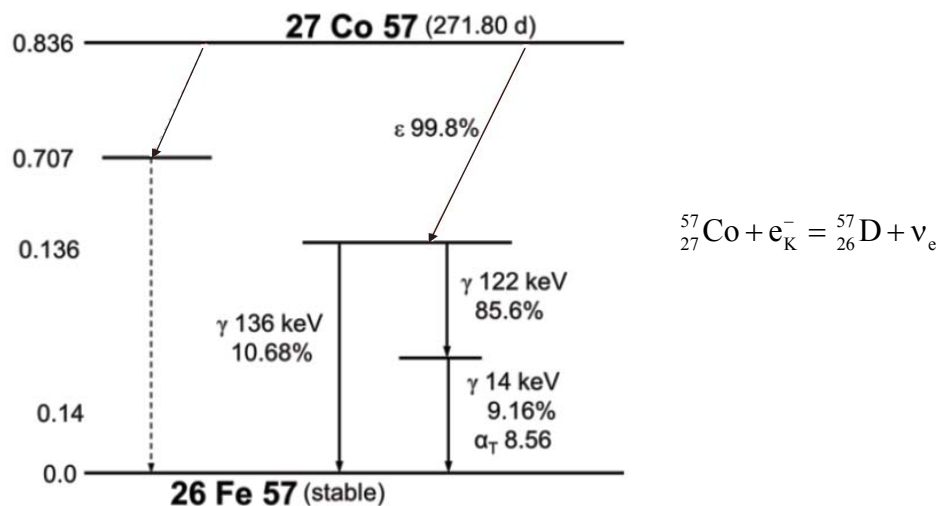


Fig.13.  ${}_{27}^{57}\text{Co}$  decay by electron capture. Electron capture (99.8%) is prevailing process when  ${}_{27}^{57}\text{Co}$  nuclei (0.836 MeV) transforms into the  ${}_{26}^{57\text{m}}\text{Fe}$  in the excited state (0.136 MeV). This transformation is caused by the fact that the Q value (0.836 MeV) is not sufficient for  $\beta^+$  decay (threshold energy for positron emission is 1.022 MeV). The process is followed by gamma emissions. (Adapted from database [15], 2012).

${}_{55}^{137}\text{Cs}$  is used as irradiation/calibration source in medical and industrial applications. One gram of  ${}_{55}^{137}\text{Cs}$  has an activity of *ca.* 3.215 TBq.

**Internal conversion** is a process in which the nuclear excitation energy may be transferred to a K- or L-shell orbital electron that is ejected instead of gamma photon emission:

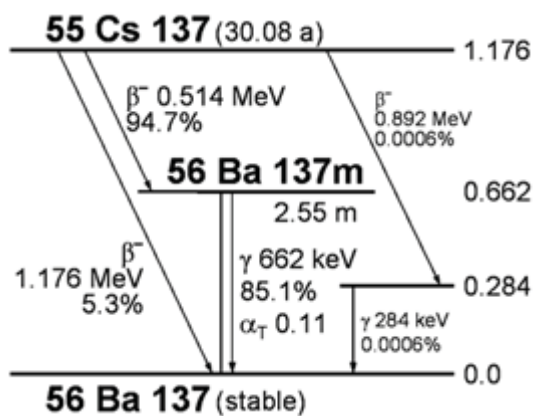


Fig.14. Metastable transition  ${}_{55}^{137}\text{Cs} \rightarrow {}_{56}^{137}\text{Ba}$ . 94.7% of  ${}_{55}^{137}\text{Cs}$  nuclei possess  $\beta^-$  decay to a metastable short-lived nuclear isomer of barium,  ${}_{56}^{137\text{m}}\text{Ba}$  ( $t_{1/2} = 153$  s); the 5.3% of  ${}_{55}^{137}\text{Cs}$  nuclei through  $\beta^-$  decay are transformed directly to the ground state of  ${}_{56}^{137}\text{Ba}$ , which is stable.  ${}_{56}^{137\text{m}}\text{Ba}$  is responsible for the emissions of gamma rays with the energy of 0.662 MeV (85.1%). The emission of conversion electrons is also possible: 7.8% – K shell, 1.8% – L and M shells. (Adapted from database [15], 2012).

$${}^A_Z\text{D}^* = {}^A_Z\text{D} + e^-_{\text{K}} \quad (36)$$

The kinetic energy of ejected electron equals to the difference between the excitation energy and the orbital electron binding energy. The resulting K-shell vacancy is filled with a higher level orbital electron and the transition energy is emitted in the form of characteristic photons or Auger electrons.

An example of internal conversion is the decay of metastable  ${}^{137\text{m}}_{56}\text{Ba}$  (Fig.14), which results from  $\beta^-$  decay of  ${}^{137}_{55}\text{Cs}$ , into stable  ${}^{137}_{56}\text{Ba}$  through emission of gamma rays and internal conversion electrons.

## 4. ELECTRICAL (ARTIFICIAL) RADIATION SOURCES

Fundamentals of radiation physics have been discussed in previous sections with the aim of introducing a basic knowledge of atomic and nuclear structures and their radioactive transformation processes. This knowledge is necessary to understand the origin and behaviour of natural radiation sources, which might be used for materials modification and in the radiation processing of materials. However radioactive decay of naturally occurring radionuclides has a probabilistic character. Even if the energies of the released particles might be relative high: up to 5 MeV for alpha particles and up to 3 MeV for beta particles, their scientific and especially industrial applications are limited by the difficulties in maintaining radioactive sources, their lack of purity for chemical processing, low intensity, poor geometry and sometimes uncontrolled broad range of energies. Radioactive sources can be expensive and require replenishment. This makes electrical (artificial) sources, beams of accelerated particles, produced using different physical phenomena and techniques, very attractive. These particles may additionally produce beams of secondary particles. Photons (X-rays, gamma rays, visible light) might be generated by the interaction of accelerated electrons with matter, while neutrons are generated by accelerated proton beams in neutron spallation sources. Primary and secondary beams are used for radiation processing of materials and in the analysis of material properties, or for the treatment and diagnosis of patients.

### 4.1. PRODUCTION OF X-RAYS

Particle interaction processes with matter are discussed in detail in Chapter 2. In order to have a full scope of particles that are produced within an atom, a physical background of X-ray production must be introduced.

Inelastic interactions between incident electrons and orbital electrons or nuclei result in production of X-rays of two types: characteristic X-rays, and

“bremsstrahlung” X-rays (braking radiation). X-rays may be of natural origin, but intensive X-ray beams that are used as radiation sources in medical, industrial and research applications, are generated electricity: low energy (kilovoltage range) X-rays are produced in conventional X-ray tubes, and megavoltage X-rays are produced by particle accelerators.

**Bremsstrahlung X-rays** are produced when a high energy electron beam strikes a target, letting the incident electron interact with a nucleus of the absorbing material (Fig. 15A). Due to deceleration in the E/H field of the nucleus, the electron loses its energy. This results in the appearance of the continuous spectrum of X-ray photons, that are emitted from the target material, with the energies ranging from zero to the kinetic energy of the incident electron.

**Characteristic X-rays** are produced when the high energy incident electron interacts with an orbital electron and ejects it from the absorber atom (ionization). This action results in appearance of a vacancy, which is filled by an orbital electron (secondary electron) from a higher level shell. The energy difference between the two shells,  $h\nu = W_2 - W_1$ , is well defined and is specific for each element in the periodic table. This energy may be either emitted in the form of characteristic X-rays or transferred to another orbital electron that is ejected from the atom as an Auger electron (Fig. 15B).

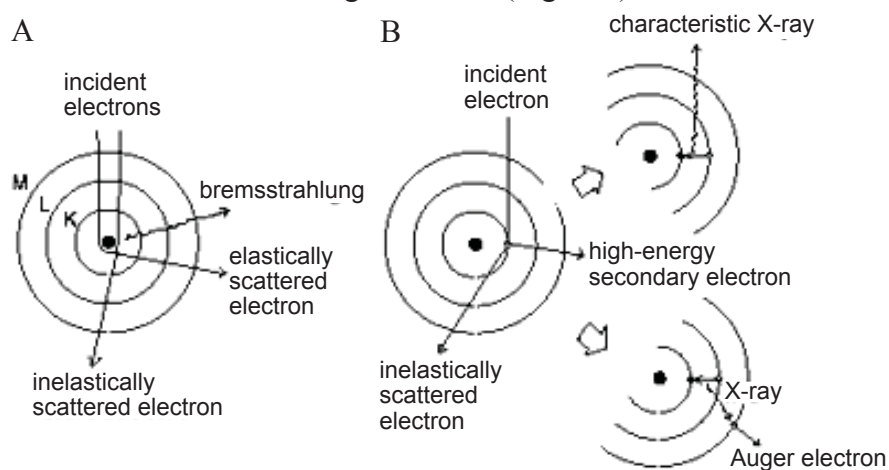


Fig. 15. X-ray production schemes and related scattering: A – bremsstrahlung X-rays, B – characteristic X-rays.

The bremsstrahlung spectrum produced by a given X-ray target depends upon the kinetic energy of the incident electron, the atomic number of the target and the thickness of the target. A typical X-ray spectrum, consisting of continuous bremsstrahlung spectrum and superimposed onto it linear characteristic X-ray spectrum, is shown in Fig. 16.

The relative proportion of the number of characteristic photons to bremsstrahlung photons in an X-ray beam spectrum varies with the kinetic energy of the electron beam striking the X-ray target and the atomic number of the target.

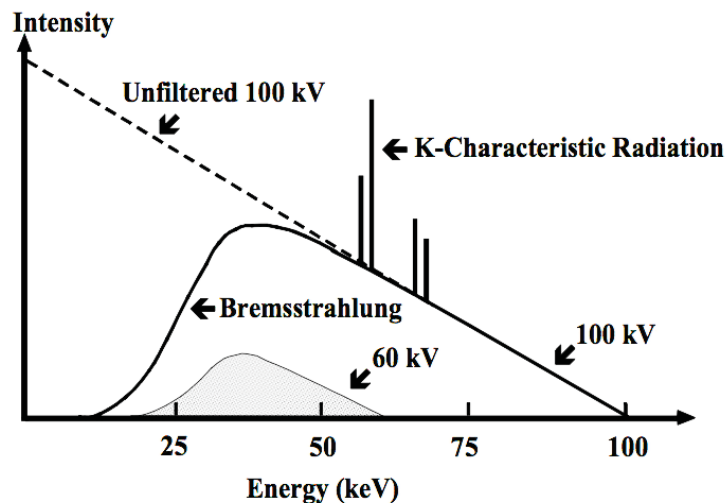


Fig.16. Typical X-ray spectrum.

For example, X-ray beams produced by a tungsten target by 100 keV electrons contain *ca.* 20% in characteristic photons and *ca.* 80% in bremsstrahlung photons. In the megavoltage range the contribution of characteristic photons to the total spectrum is negligible.

## 5. APPLICATION OF RADIATION SOURCES IN MATERIALS PROCESSING

It was estimated in 2013 [17] that more than 20 000 particle accelerators producing charged particle beams were used in the industrial processes. This number does not include the more than 11 000 particle accelerators that have been produced exclusively for medical therapy with electrons, ions, neutrons, or X-rays, and accelerators for physics research. An overview on industrial application of accelerated particles in different fields is shown in Fig.17.

Production of accelerated particle beams for different purposes is defined by the physical process of particle generation and operational parameters of generating equipment (Fig.18).

Accelerators for industrial applications, including radiation processing of materials are classified according to the particle generation processes:

- Direct voltage accelerators (accelerate either electrons or ions):
  - Dynamitrons™ and Cockcroft Walton accelerators (energies to 5 MeV and currents up to 100 mA),
  - Van de Graaff accelerators (energies from 1 to 15 MeV at currents of a few nA to a few mA),

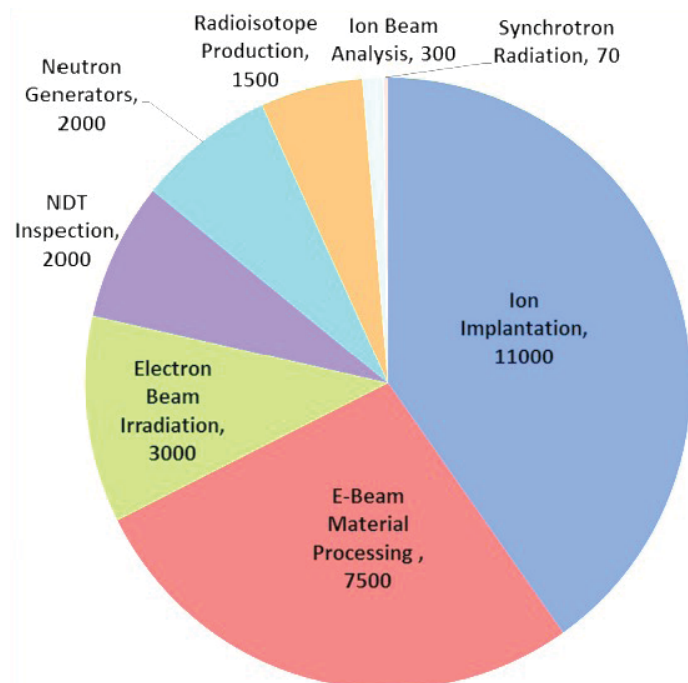


Fig.17. Number of particle accelerators in operation and fields of their industrial applications in 2013. (Adapted from Ref. [17]).

- inductive core transformers (ICT) (energies to 2.5 MeV at currents to 50 mA);
- RF linacs (within a wide range of operating RF frequencies for charged particles):
  - electron linacs (standing wave and travelling wave cavities from 0.8 to 9.0 GHz, energies from 1 to 16 MeV at beam powers to 50 kW),
  - ion linacs (RF from 100 to 600 MHz, energies from 1 to 70 MeV at beam currents to > 1 mA);
- circular accelerators:
  - betatrons (electron energies to 15 MeV at a few kW beam power),
  - cyclotrons (ion energies from 10 to 70 MeV at beam currents to several mA),
  - Rhodotrons™ (electron energies from 5 to 10 MeV at beam powers up to 700 kW),
  - synchrotrons (electron energies to 3 GeV and ion energies to 300 MeV/amu).

Useful and detailed information on industrial irradiators can be found in the textbooks edited by R.W. Hamm and M.E. Hamm [18], A. Chao *et al.* [19], and A. Sessler and E. Wilson [20].

In this book we will concentrate on electron, gamma (high activity  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  sources) and X-ray applications for materials processing (Table 3).

Electron beam processing applications fall into two broad categories: modifications of materials (welding, melting, cutting, drilling and hardening)

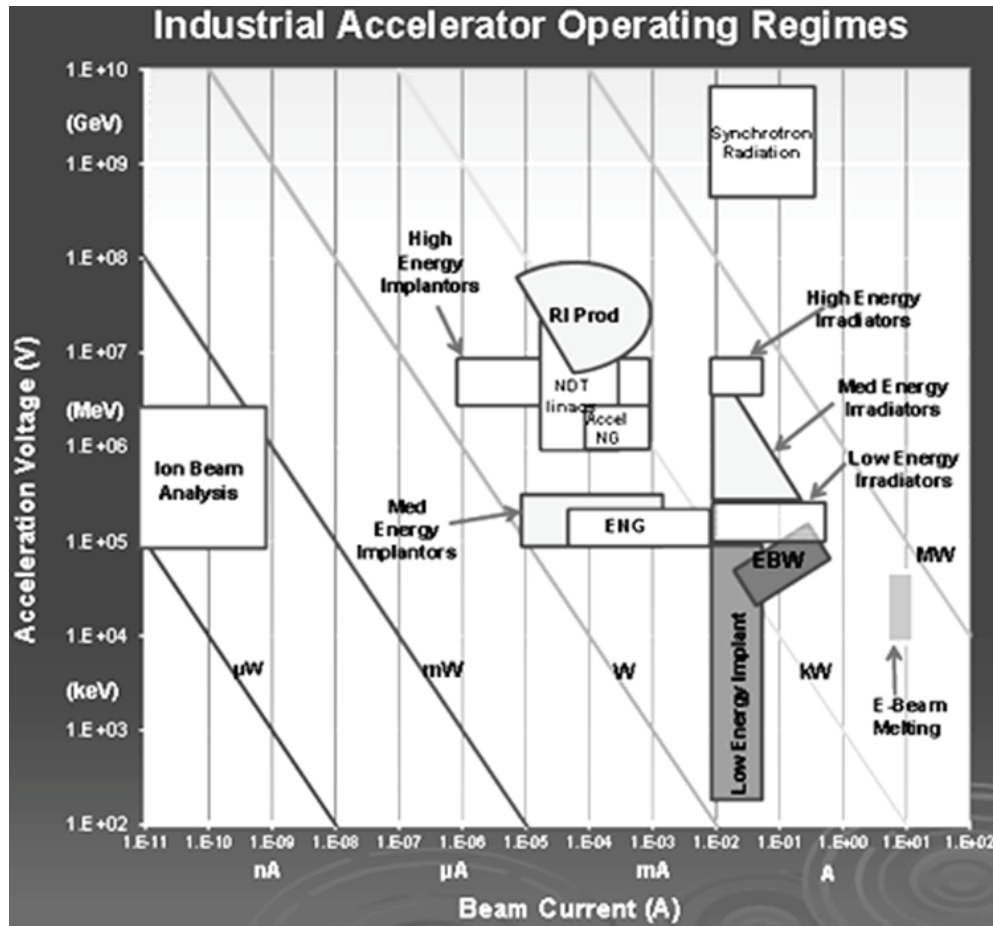


Fig.18. Operational regimes of industrial accelerators. (Adapted from Ref. [17]).

Table 3. Comparison of ionizing radiation sources. (Adapted from Ref. [21]).

	Electron beam	X-rays	Gamma rays
Power source	Electricity	Electricity	Radioactive isotope (mainly $^{60}\text{Co}$ )
Power activity	Electrical on-off	Electrical on-off	Half-life: 5.27 year
Properties	Electrons	Photons	Photons
Charge	$1.16 \times 10^{-19} \text{ C}$	$\lambda = 4.1 \times 10^{-3} \text{ nm}$	$\lambda = 1.0 \times 10^{-3} \text{ nm}$
Emission	Unidirectional (can be scanned and bent by magnets)	Forwards peaked	Isotropic
Penetration	Finite range	Exponential attenuation	Exponential attenuation
Dose rate	100 kGy/s	$2.7 \times 10^{-2} \text{ kGy/s}$	$2.8 \times 10^{-3} \text{ kGy/s}$ (fresh source)

and irradiation, which includes radiation processing of polymers, monomers and oligomers used in inks, coatings and adhesives or for composite matrices, food preservation, product sterilization and waste treatment.

Bombarding a material with energetic electrons can promote chemical processes by generating ions and slow electrons which modify the material's atomic bonds through interactions with the free radicals. This can alter both its chemical and physical properties. Applications of such radiation processing can be divided into four main categories: polymerization-crosslinking of inks, coatings and adhesives; production of crosslinked, scissioned and grafted polymers; sterilization and food preservation; and wastewater and gas treatment. There are other applications such as the curing composite matrices, viscose production, and thermo-mechanical pulp production that have been demonstrated but are not yet in wide-spread use [21].

The electron beam energy needed in these applications is determined by the thickness and density of the material being processed. The accelerators used cover a wide range of beam energies and can be classified as low (80 to 300 keV), medium (300 to 1000 keV) and high (1 to 10 MeV).

Low energy electron beam accelerators are used for the polymerization-crosslinking of thin film coatings on sheets of material, for the crosslinking of plastic laminates and for thin gauged, single strand wire. Medium energy systems are mainly used for crosslinking, as for wire insulation and heat shrinkable products and in the partial crosslinking of components used in tire manufacture.

High energy accelerators are used for the crosslinking or treatment of thicker materials and for the sterilization of medical products. High energy, high current accelerators are used with water cooled tantalum targets for medical device sterilization, as an alternative to the use of radioactive isotopes, and for bio-hazard elimination. High energy accelerators with a water-cooled tungsten target generate high energy X-rays that can be used for food irradiation, wastewater remediation, and gemstone colour enhancement, particularly topaz and diamonds. The energy limit of 10 MeV is set to avoid activation in the processed material of any metal into a radioactive isotope. For food irradiation, the regulatory approved upper energy limit is 7.5 MeV.

Radioactive isotope emitted gamma rays and electrically sourced X-rays are mainly used for food preservation and for the sterilization of packaged medical devices. Gamma rays and X-rays have the same penetration depth, but X-rays are one order higher in dose rate. High through-put industrial processes use high beam current accelerators. Low dose rate, small isotope sources have historically been used for research purposes, with medical device sterilization being the only major industrial market. Recent developments in modular beam tube technology have enabled small, self-shielded laboratory units to become available. The decision to use either electron beam or X-ray or gamma-ray treatment is made depending the size of the object being treated and industry

demands for product through-put. Many of the industrial applications using electron beam and X-ray processing have been described in Ref. [21].

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