3 Physical fundamentals

This chapter introduces the basic properties of atomic nuclei, their structure characteristics and transformations by radioactive decay and nuclear reactions. The particle and electromagnetic radiation associated with nuclear transformation is classified and their interaction with matter briefly described. Special emphasis is put on fission products leading to the radioactive inventory of nuclear reactors.

3.1 Natural radioactivity

Radioactivity was observed for the first time in 1896, when Henri Becquerel [96Bec] in Paris found that photographic plates were blackened even in the absence of light, if they were in contact with uranium containing minerals. In 1898 the same observation was made for thorium by Marie Curie [98Cur] in France and by G. C. Schmidt [98Sch] in Germany. Marie Curie found differences in the radioactivity of uranium and thorium and concluded that these elements must contain unknown radioactive elements. Together with her husband, Pierre Curie, she discovered polonium in 1898, and radium in the same year. More historical details can be found e.g. in [95AMC].

Since radioactive radiation cannot be detected by human senses, it is necessary to use for its detection and the identification of radioactive substances solid, liquid, or gaseous detectors, which indicate the radiation via its interaction with the respective materials.

Radioactive elements are widely distributed in the earth's crust in more or less small concentrations. They originate on one hand from extremely long-lived primordial radionuclides formed together with the other stable elements and from their longer or shorter lived decay products like the important minerals of uranium and thorium and their radioactive decay chains. Uranium and thorium are common elements in nature. Their concentrations in the crust of the earth are about 4 and 13 mg/kg, respectively, and the concentration of uranium in seawater is about 3 µg/l. The most important uranium mineral is pitchblende ($U_3 0_8$). The most important thorium mineral is monazite, which contains between about 0.1 and 15 % Th. Natural radioactivity is mainly observed with heavier primordial elements and seldom with light ones (e.g. ^{40}K and ^{87}Rb).

On the other hand, radioactive elements can be formed - especially in case of light shorter lived elements - by interactions of cosmic radiation with the atmosphere. This radiation entering the atmosphere from outside originates from the sun as well as from material in the deep interstellar space. It produces a variety of elementary particles (protons, neutrons, electrons, positrons, mesons, photons) and of radioactive atoms via cascades of interactions with the gas molecules in the atmosphere.

Among others, ^{14}C, ^{10}Be , ^{7}Be, and ^{3}H (tritium) are produced in the atmosphere by cosmic radiation. Examples for the production rate are: about 2.2×10^4 atoms of ^{14}C per second per m^2 of the earth's surface and about 2.5×10^3 atoms of tritium per second and m^2. Taking into account the radioactive decay and the dwell times in the atmosphere, these data result in a global equilibrium inventory of about 6.3×10^4 kg of ^{14}C and of about 3.5 kg of ^{3}H.

The measurement of local concentrations of natural radionuclides like ^{14}C, ^{40}K, ^{87}Rb is very useful for the determination of the age of the respective material. However, other sources have to be taken into account like the production of these nuclides in nuclear power plants and by explosions of nuclear bombs.

3.2 Elements, isotopes and radionuclides

3.2.1 Atoms, electrons and the Periodic Table of Elements

3.2.1.1 The atom

The atom is the smallest particle of an element, which can no more be subdivided by chemical methods. It consists of a (heavy) nucleus surrounded by a number of (light) electrons. The elements differ from each other by the structure of their atoms i.e. the composition of the nucleus and the number and distribution of the electrons. The diameter of an atom is approximately 10^{-8} cm. The nucleus contains positively charged protons and electrically neutral neutrons and has, therefore, a positive charge. Its diameter is in the order of 10^{-13} cm, which is five orders of magnitude smaller than the atomic diameter. Since atoms in their normal state are electrically neutral, the number of (negative) electrons in the shell must balance the number of (positive) protons in the atomic nucleus.

3.2.1.2 The electron shells

The electron distribution around the atomic nucleus is described and denoted according to Bohr's shell model. Electrons move in discrete shells with energy states (binding energies) decreasing with increasing distance from the nucleus. They are numerated beginning with n = 1 and denominated by capital letters K, L, M, ... The maximum number of electrons in one shell is 2n.

By energy transfer electrons can be excited into higher lying weaker bound states. The electrons are released into an unbound state by an energy transfer of at least the binding energy of the respective shell leaving the residual atom in an ionised state. The remaining hole in the electron shell is filled by one of the outer electrons releasing the energy difference in form of characteristic photon radiation (fluorescence) or by radiationless energy transfer to another bound electron, which in turn is emitted (Auger electron).

3.2.1.3 The Periodic Table of Elements

When ordering all elements according to their atomic weight, one finds a periodical repetition of special properties leading to several groups of elements with a remarkable similarity of their chemical and physical properties. Within these groups very similar differences of the atomic weights of neighbouring group members are found. The most outstanding element group is formed by the noble gases, similar observations are made for the halogens fluorine, chlorine, bromine, and iodine, as well as for the chalcogens sulphur, selenium, and tellurium. First indications of this periodical behaviour were already described in the early 19th century. In 1869 L. Meyer and D. Mendeleev succeeded in placing all known elements satisfactory in the periodical system.

In this periodical system all elements, except of the rare earth elements (lanthanides) and the transuranium elements (actinides) are sorted into 6 periods. This ordering shows two short periods with 8 elements each and three long periods comprising 18 elements, taking into account some not naturally occurring elements, like technetium. The three long periods are subdivided into two parts. Between these one finds three triples of very similar elements: Fe-Co-Ni, Ru-Rh-Pd, Os-Ir-Pt, respectively. In this arrangement chemically similar elements form columns combined in 8 groups, subdivided into main groups and auxiliary groups of transition elements. The 8th main group containing the noble gases by convention is called group 0.

This ordering was explained later on by Bohr's atomic model. The periods correspond to the filling of the different electronic shells, which can take up 2n electrons each. The noble gases in group 0 correspond to a completely filled shell in each period.

3.2.2 Atomic nuclei, nuclides and the Chart of Nuclides

3.2.2.1 Nuclei and nuclides

Atomic nuclei are composed of protons and neutrons having the common denotation nucleons. The corresponding number of protons Z is called the atomic number. It characterizes the element and its chemical behaviour, while the physical properties are defined by Z as well as by the neutron number N. Atoms with a defined nucleus and the corresponding number of electrons are called nuclides. Each nuclide is identified by its mass number $A = Z + N$ and is marked by the following denotation for the element symbol El:

${}_Z\mathrm{El}_N^A$ or in shorter form ${}^A\mathrm{El}$ (or El-A), e.g.: ${}^4\mathrm{He} = \mathrm{He}\text{-}4 = {}_2\mathrm{He}_2^4$, ${}^{235}\mathrm{U} = \mathrm{U}\text{-}235 = {}_{92}\mathrm{U}_{143}^{235}$

since Z defines uniquely the element and N is given as well by $N = A - Z$.

The following denotation is used for neighbouring nuclides (cf. Fig. 3.1):

- nuclides with equal atomic number Z are called *isotopes*
  mass number A are called *isobars*
  neutron number N are called *isotones*
- nuclides in long-lived excited states are called *isomers*.

Different isotopes of an element exhibit almost the same chemical behaviour (very small differences are observable in special experiments only: "isotopic effect"). In general, the elements represent a mixture of all of their stable isotopes.

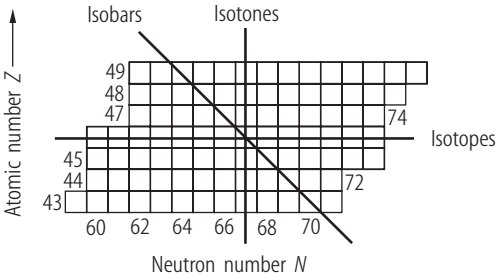

Fig. 3.1. Representation of nuclides in a chart of nuclides as proposed by E. Segrè and definition of isotopes, isotones, and isobars.

Today nearly 3000 nuclides are known most of which are extremely short-lived and can be produced in complicated experiments only. More than 300 nuclides are found in nature and can be assigned to one of four categories:

1. 258 stable nuclides
2. 25 nuclides with $Z < 80$ having extremely long half-lives (radioactive decay not proved uniquely in all cases)
3. about 15 *quasistable* nuclides with half-lives $>10^5$ years (including ^{238}U, ^{235}U, ^{232}Th and ^{244}Pu) existing since the genesis of the elements and called *primordial radionuclides*. Among these ^{238}U, ^{235}U and ^{232}Th and their radioactive decay products form the main sources of natural radioactivity
4. radionuclides continuously produced by the impact of cosmic radiation: e.g. ^{14}C, ^{10}Be, ^{7}Be, ^{3}H.

3.2.2.2 The Chart of Nuclides

In order to get a synopsis of this vast amount of nuclides several types of schematic representations have been introduced in the past. The most widespread scheme has been proposed by E. Segrè and has been realized among others in the Karlsruhe Chart of Nuclides [98PKS]. In this scheme the nuclides are arranged in such a way (cf. Fig. 3.1) that the proton number Z is given on the ordinate and the neutron number $N = A - Z$ on the abscissa, respectively. Each experimentally observed nuclide is represented by a square containing the symbol of the element and the number of nucleons (mass number) and in addition

various decay or structure data. For example, in the Karlsruhe Chart of Nuclides [98PKS] besides the half-lives, the decay modes, the types of emitted radiation and the energies of the most abundant radiation types are given as observable in respective experiments. Other charts, nowadays often distributed in electronic form (e.g. [99Mag]), are based on (evaluated) nuclear data files enabling access to all known decay data of all known radionuclides.

In these charts the stable nuclides lie around a central branch with approximately $N = Z$ for the lighter nuclides. With increasing mass this branch flattens due to the increasing neutron excess necessary for the stability of heavier nuclei. This branch *(valley of beta-stability)* separates the neutron rich nuclides undergoing β^--decay from the proton rich nuclides undergoing β^+-decay. Furthermore, with increasing distance from the valley of stability, i.e. with increasing neutron- or proton excess, the stability and consequently the half-lives of the nuclides decrease drastically. Thus, radionuclides with half-lives in the range of hours or longer are only found in narrow bands along the valley of stability.

The so called strong interaction, by which the nucleons are bound in the atomic nucleus, is a very short ranged pairing force binding the nucleons very close together. This leads to a very high density of the nuclear matter. With increasing mass number stable nuclei exhibit an increasing neutron excess (up to 50 % for the heaviest nuclei) necessary to balance the increasing repulsive force of the positively charged protons.

Nuclei – like the electron shells – can be excited by energy transfer into higher energetic states. The de-excitation of such states occurs under emission of characteristic nuclear radiation, which is in general γ-radiation. At sufficiently high excitation energies (above the separation energy of nucleons) the emission of nucleons or nucleon clusters becomes possible.

3.3 The structure of the atomic nucleus

3.3.1 Elementary particles

Though the modern high energy and particle physics deals with elementary particles far below the level of those forming the atomic nuclei, only the latter shall be discussed here and are – according to classical nuclear physics – denominated as elementary particles of the nucleus, the nucleons.

3.3.1.1 Charge, mass and stability of the nucleons

The proton is stable and has a positive electric charge equal and opposite to the elementary charge. The neutron is electrically neutral and its mass is slightly larger than that of the proton. The free unbound neutron is unstable and decays with a half-life of 10.25 min into a proton and an electron and an anti-neutrino.

The following units are used for atomic and nuclear quantities (see also Table 3.1), making use of the energy-mass equivalence in Einstein's equation $E = mc^2$, which leads to the correspondence 1 kg = 5.6095892×10^{29} MeV:

elementary charge: $e = 1.602176462 \times 10^{-19}$ C

mass: atomic mass unit: $u = 1.66053873 \times 10^{-27}$ kg = 9.31494013×10^2 MeV/c^2, where the basis of this unit is 1/12 of the mass of the neutral ^{12}C atom (including the masses of its 6 electrons).

Table 3.1. Frequently used general constants (main values taken from [99MoT]) and conversion factors. The standard deviations are given in parentheses.

Symbol	value	quantity
c	299792458 m s^{-1}	speed of light
h	6.62606876 (52)×10^{-34} J s	Planck constant
$\hbar = h/2\pi$	1.054571596 (82)×10^{-34} J s	
u	1.66053873 (13)×10^{-27} kg	atomic mass unit
	9.31494013 (37)×10^{2} MeV/c^2	
m_n	1.00866491578 (55) u	neutron rest mass
	1.67492716 (13)×10^{-27} kg	
	9.39565330 (38)×10^{2} MeV/c^2	
m_p	1.00727646688 (13) u	proton rest mass
	1.67262158 (13)×10^{-27} kg	
	9.38271998 (38)×10^{2} MeV/c^2	
m_e	9.10938188 (72)×10^{-31} kg	electron rest mass
	0.510998902 (21) MeV/c^2	
e	1.602176462 (63)×10^{-19} C	elementary charge
	4.803204197 (19)×10^{-10} esu	
e/m_e	1.758820174 (71)×10^{11} C kg^{-1}	specific electron charge
	5.27281023 (21)×10^{17} esu g^{-1}	
$r_e = e^2/m_e c^2$	2.817940285 (31)×10^{-15} m	electron radius
a_0	5.291772083 (19)×10^{-11} m	Bohr radius
α	7.297352533 (27)×10^{-3}	fine structure constant
R_∞	1.0973731568549 (83)×10^{7} m^{-1}	Rydberg constant
N_A	6.02214199 (47)×10^{23} mol^{-1}	Avogadro constant
V_m	2.2413996 (39)×10^{-2} m^3 mol^{-1}	molar volume of an ideal gas at s. t. p.
R	8.314472 (15) J mol^{-1} K^{-1}	molar gas constant
	8.314472 (15)×10^{7} erg mol^{-1} K^{-1}	
$k = R/N_A$	1.3806503 (24)×10^{-23} J K^{-1}	Boltzmann constant
	8.617342 (15)×10^{-5} eV K^{-1}	
$F = N_A \cdot e$	9.64853415 (39)×10^{4} C mol^{-1}	Faraday constant
	2.892557769 (12)×10^{14} esu mol^{-1}	

Some useful conversions:

1 Ci = 3.7×10^{10} Bq (= disintegrations s^{-1})
1 W ≙ 3.1×10^{10} fissions s^{-1}
1 MWd ≙ 2.7×10^{21} fissions ≙ 1g fissionable material
1 μA = 6.241509745×10^{12} e s^{-1}
1 eV/Atom ≙ 23 kcal mol^{-1}
1 MeV Ci ≙ 5.93×10^{-3} W

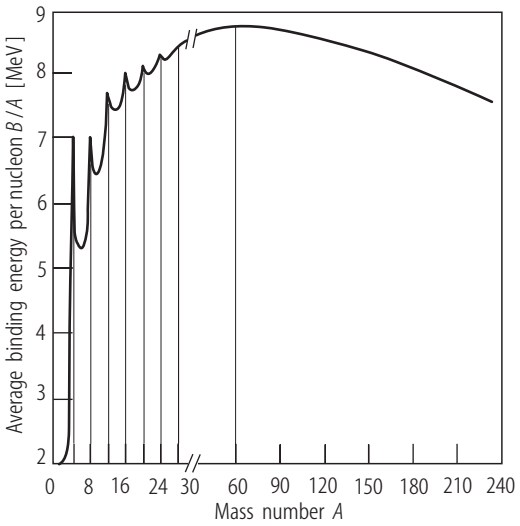

Fig. 3.2. Average binding energy per nucleon in dependence on the mass number.

3.3.1.2 Binding energy

The experimentally measured atomic weights M of stable pure isotopes are not integer multiples A of the atomic mass unit u. This deviation is called the mass defect $\Delta M = M - A \cdot u$. In addition, also the mass $M(Z, A)$ of a nucleus differs from the mass sum of its nucleons and is always smaller: $M(Z, A) < Z \cdot M_p + N \cdot M_n$.

According to Einstein's equation, the mass difference $\Delta M = Z \cdot M_p + N \cdot M_n - M(Z, A)$ corresponds to an energy $B = \Delta M \cdot c^2 = [Z \cdot M_p + N \cdot M_n - M(Z, A)] \cdot c^2$, which is necessary to separate all nucleons of the nucleus. B is called the total binding energy of the nucleus and is always positive for stable nuclides.

B/A is the average binding energy per nucleon, which has a mean value of about 8 MeV/nucleon, except for the very light and heavier elements (cf. Fig. 3.2). This behaviour has the most important consequence that energy can be gained by fission of heavy as well as by fusion of light nuclei, both leading to a higher energy for the product(s).

3.3.2 Nuclear transformations

3.3.2.1 Nuclear reactions

The vast amount of radioactive nuclides as shown in charts of nuclides do not occur in nature but can only be produced artificially by means of nuclear reactions. In a nuclear reaction a normally stable nuclide is bombarded with a beam of one of many types of radiation (charged particle or neutron radiation, electromagnetic radiation, electrons).

The simplest form of a nuclear transformation is the radioactive decay caused by internal excess energy: $A \rightarrow B + b + \Delta E$. This is a mononuclear reaction. Binuclear reactions, denoted generally as "nuclear reactions", are induced by bombarding target nuclei with a beam of specific projectiles. They are described by: $A + a \rightarrow B + b + \Delta E$, where A is the target nuclide, a the projectile, B the product nuclide, and b the "ejectile" (particle or photon emitted). The energy ΔE is also called the *Q value* of the reaction. The first nuclear reaction was observed in a cloud chamber in 1919 by Rutherford:

$^{14}N + {}^{4}He \rightarrow {}^{17}O + {}^{1}H$, in shorter form
$^{14}N(\alpha,p)^{17}O$ or generally
A(a,b)B.

When comparing chemical and nuclear reactions one finds the following main differences:

- In chemical reactions macroscopic amounts of matter as a whole are altered, in nuclear reactions individual atoms.
- In chemical reactions the total involved mass is conserved, in nuclear reactions the sum $\Sigma(E + mc^2)$ of the energies and the mass-energy equivalents.
- The energies of chemical reactions are relatively small and comparable with the energies of chemical bonds (of the order of eV), whereas the energies involved in nuclear reactions are about 6 orders of magnitude higher (of the order of MeV).

Many nuclear reactions pass over a transition state, similarly to chemical reactions:

$$A + a \rightarrow (C)^* \rightarrow B + b + \Delta E$$

The transition state $(C)^*$, which is excited to an elevated energy, is also called a compound nucleus; its lifetime is very short ($<10^{-13}$ s).

While the probability for a mononuclear reaction (i.e. for radioactive transmutation) is given by the decay constant λ, in the compound nucleus model the reaction probability is determined by both the probability that the projectile a will react with the nuclide A and by the probability that the nuclide B is produced.

The time duration of a nuclear reaction is in between about 10^{-23} and 10^{-13} s. The lower limit is given by the crossing time of a particle passing the nucleus with the velocity of light, and the upper limit holds for slow reactions, e.g. with thermal neutrons.

3.3.2.2 Projectiles for nuclear reactions

Charged particles

Positively charged particles inducing nuclear reactions, such as protons, deuterons or ions with higher atomic numbers Z, need a minimum energy to surmount the repulsive Coulomb barrier formed by the protons of the nuclei. The Coulomb barrier U can be calculated approximately from the equation

$$U \approx \frac{Z_A Z_a e^2}{4\pi\varepsilon_0 r} \approx \frac{Z_A Z_a}{A_A^{1/3} + A_a^{1/3}} \tag{3.3.1}$$

where Z_A and Z_a are the charge numbers of the nuclide A and the projectile a, respectively, e is the unit charge, ε_0 the electric field constant, and r the distance, which in this approximation is set to be

$$r \approx r_0(A_A^{1/3} + A_a^{1/3}) \tag{3.3.2}$$

A_A and A_a are the mass numbers of the nuclide A and the projectile a, respectively. For the Coulomb barrier U the following approximate values can be calculated:

- $U \approx 1.8$ MeV for the reaction of a proton with ^{12}C,
- $U \approx 13$ MeV for the reaction of a proton with ^{238}U,
- $U \approx 24$ MeV for the reaction of an α-particle with ^{238}U,
- $U \approx 130$ MeV for the reaction of ^{12}C with ^{238}U and
- $U \approx 700$ MeV for the reaction of ^{238}U with ^{238}U.

With increasing atomic numbers the approximate formula (3.3.1) becomes incorrect. For the reaction of ^{238}U with ^{238}U, e.g., a more exact value for the Coulomb barrier is $U \approx 1500$ MeV.

Charged particles for nuclear reactions originate from particle accelerators. They are produced in ion sources by bombarding a gas with energetic electrons. The positive ions are extracted by means of an electrode. For the further acceleration various set-ups are used. The two main groups are linear accelerators (linacs) with single (e.g. Cockroft-Walton type) and multiple (e.g. tandem Van de Graaff accelerators) stages and circular accelerators. With single-stage linear accelerators proton or deuteron beams of up to about 10 mA with energies of up to about 4 MeV are obtained. These accelerators are often applied as injectors in larger machines for the production of high-energy particles. In two-stage tandem Van de Graaff accelerators proton beams of about 20 MeV and α-particles of about 30 MeV are obtained. In addition, lighter heavy ions can be efficiently accelerated, too. The intensity of the beam current varies between about 10 and 100 μA.

In cyclotrons the ions move on spiral paths with increasing radius guided by a suitable magnetic field, while in synchrotrons the circular orbit of the particles remains constant and the guiding magnetic field increases with the momentum of the ions.

In modern machines, protons, deuterons and α-particles can be accelerated to energies of several 100 MeV up to about 1 GeV. For the production of radionuclides, relatively small accelerators are used producing particle energies of 10 to 30 MeV and ion currents of the order of 100 μA.

With light charged particles mainly nuclides on the left hand side of the valley of β-stability are produced (β^+- and electron capture activities). Increasing projectile energies lead then to an increasing neutron deficit of the produced radionuclides.

Heavier ions are produced in special types of linear or circular accelerators. The term “heavy ions” is used in this context for all ions heavier than α-particles and includes “light ions” e.g. of lithium, carbon or oxygen as well as “heavy ions” of elements up to uranium. These projectiles are widely used in basic as well as in applied research and techniques. Due to the great number of nucleons, which they can transfer to the target nuclide, the main application fields of heavy ions are: synthesis of new (including the so-called super-heavy) elements, production of nuclides far off the line of β-stability, investigation of nuclear matter at high densities, production of small holes with defined diameters in thin foils and irradiation of tumors in medicine.

Neutrons

Neutrons are frequently used projectiles for nuclear reactions. Since they do not carry a positive charge, they do not experience Coulomb repulsion, and even low-energy (thermal and slow) neutrons can easily enter the target nuclei. Neutrons with energies of the order of 1 to 10 eV (resonance neutrons) exhibit relatively high absorption maxima. Furthermore, neutrons are available in large quantities in nuclear reactors with fluxes of the order of about 10^{10} to 10^{16} cm^{-2} s^{-1}.

Neutrons may also be produced by nuclear reactions, such as $^{9}Be(\alpha,n)^{12}C$, $^{9}Be(d,n)^{10}B$, $^{9}Be(\gamma,n)2\alpha$, $d(\gamma,n)p$, $d(d,n)^{3}He$ or $t(d,n)\alpha$. Alpha particles are available from radionuclides like ^{226}Ra or ^{210}Po, and Ra-Be neutron sources have formerly often been applied in experimental neutron physics. Gamma-rays of sufficiently high energy for (γ,n) reactions may be supplied by radioactive nuclides, such as ^{124}Sb. Neutrons are also available from spontaneously fissioning nuclides like ^{252}Cf (1 μg of ^{252}Cf emits 2.3×10^6 neutrons per second). In neutron generators deuterons with energies between 0.1 and 10 MeV are produced in small accelerators and impinge on a suitable target, e.g. a tritium target. The neutron fluxes available from neutron sources and neutron generators vary between about 10^5-10^8 and 10^8-10^{11} $cm^{-2}s^{-1}$, respectively. Neutrons produced by the reaction $t(d,n)\alpha$ have energies of about 14 MeV.

Reactions with neutrons are used to produce nuclides on the right-hand side of the line of β-stability, i.e. nuclides undergoing β^--decay.

Electrons and γ-rays

For the acceleration of electrons to high velocities approaching the velocity of light, much less energy is needed than for the acceleration of ions. They are accelerated in linear or circular (betatrons, synchrotrons) accelerators. Depending on the size of the installations, energies of the order of 10 to 300 MeV are obtained in linear accelerators and betatrons, whereas in electron synchrotrons electrons with energies up to the order of 10 GeV are available.

Electrons are not directly used for the production of radionuclides. However, the high-energetic bremsstrahlung, which is emitted, when the electrons are slowed down in a target of high atomic number, is applied for the induction of nuclear reactions (photo-nuclear reactions). The maximum energy of this bremsstrahlung corresponds to the energy of the incident electrons and is in the range of high-energy γ-rays. Gamma rays are also available from γ-emitters, such as ^{60}Co or ^{124}Sb, but with comparably lower energies and intensities.

The monoenergetic synchrotron radiation emitted by electrons moving in the magnetic field of a synchrotron has found many applications because of the high intensities, broad energy ranges, and small beam diameters available. The energy of the synchrotron radiation is in the range of the energy of X-rays.

3.3.2.3 Artificial radionuclides for medical and technical applications

Numerous radionuclides are especially produced at accelerators or nuclear reactors for dedicated applications in medicine and technology. In particular, in nuclear medicine new diagnostic methods have been established and are applied with increasing frequency such as single photon emission computed tomography (SPECT) and positron emission tomography (PET). The main advantage of these methods as compared to other imaging processes is their capability not only to provide an image of inner organs but also to give information about their function (metabolism). These methods use short living accelerator isotopes like ^{11}C, ^{13}N, ^{18}F, ^{123}I or ^{153}Gd. The diagnostical goals are detection of malfunction or cancer stroke of thyroid gland, kidneys, liver, heart and brain.

For so-called targeted radiotherapy of cancer various β-emitters e.g. ^{90}Y, ^{125}I and ^{107}Pd are being used. Quite recently, α-emitters like ^{211}At, and ^{213}Bi/^{225}Ac are under development for such purposes which promise to have thousands fold better biological effectiveness.

Technical application of primordial (^{241}Am) and artificial radionuclides (e.g. ^{55}Fe, $^{57, 60}$Co, ^{137}Cs) range from smoke detection, measurement of moisture content in materials via thickness measurements for various materials to wear diagnostics of machine parts (^{57}Co). The latter method is industrially applied by many car factories.

3.3.2.4 Excited states, level- and decay schemes

Like the electrons, which can occupy only discrete states in the shell with defined energies, also the nucleons in the atomic nucleus are in discrete states with well-defined energies. The state with the lowest energy is called the groundstate, its energy is per definition set to zero and all higher lying states (excited levels) are referred to this groundstate.

In many cases of radioactive decay and even more often after nuclear reactions the product of the transformation is not in the groundstate but rather in an excited state. These and other excited states correspond to a series of defined energy levels, which are specific for each nuclide. They release their excitation energy by emission of one or several γ-rays, mostly within about 10^{-13} s after their formation by a nuclear reaction or by a preceding α- or β-decay. In some special cases, e.g. for very low transition energies, γ-transitions can be “hindered” (forbidden transitions) resulting in longer half-lives of these metastable “isomeric states”.

In order to visualize the level structure of a nuclide, the levels are ordered in a scheme according to their energy above the ground state and are denoted – besides the energy – by the specific quantum numbers nuclear spin and parity, resulting from the nucleons contributing to the excitation. In addition,

the assigned γ-transitions and for isomeric states the half-lives are indicated. In the decay schemes the nuclides with higher atomic number are ordered to the right hand side and the decay types and transition energies to the levels in the daughter nuclide are specified.

The construction of level schemes is performed either directly by sophisticated nuclear reaction experiments and/or by a comprehensive investigation of the radioactive decay of the mother nuclide feeding several states with subsequent γ-decay. While for light radionuclides the decay schemes are mostly relatively simple due to the fact that only few levels can be fed by the decay of the mother nuclide, heavier nuclides exhibit mostly very complicated schemes with many levels and γ-transitions.

3.4 Radioactive decay

3.4.1 Basic properties

3.4.1.1 Decay law for a single radionuclide

From the observation that the number of nuclei decaying per unit time is proportional to the actual number N of radioactive nuclei the following time law results:

$$dN/dt = -\lambda N \tag{3.4.1}$$

The constant of proportionality λ is characteristic for each nuclide and is called "decay constant". It corresponds to the fraction of the actually present nuclei, which decays per unit time.
The integration of equation (3.4.1) results in the general decay law:

$$N(t) = N(0)\cdot e^{-\lambda t} \tag{3.4.2}$$

The number of atoms of a nuclide decaying per unit time is called its decay rate (λN). It is measured in Bequerel (1Bq = 1/sec). The formerly used unit of radioactivity was the Curie (1 Ci corresponding to 3.7×10^{10} Bq). A specified activity refers to the respective nuclide only, independent of possible subsequent radioactive decays of daughter nuclides.

The decay rate λN is usually set equal to the activity A. In general, however, an experimentally *measured* activity A is lower but always proportional to the decay rate of a sample $A = F\lambda N$. This is due to the fact that the measured rate depends on the experimental setup, the sensitivity of the detector used and the thickness, backing and area of the sample. In the following, this factor F will be set to 1.
The general decay equation becomes therefore:

$$\lambda N(t) = \lambda N(0)\cdot e^{-\lambda t} \tag{3.4.3}$$

or

$$A(t) = A(0)\cdot e^{-\lambda t} \tag{3.4.4}$$

and

$$\ln A(t) = \ln A(0) - \lambda t \tag{3.4.4a}$$

where $A(t)$ means the activity at time t and $A(0)$ the activity at time $t = 0$.

If the original number of atoms $N(0)$ has decreased to one half, $N(t) = N(0)/2$ and $t = T$ (= $T_{½}$: half-life), equ. (3.4.2) gives $\lambda = \ln 2/T \approx 0.693/T$. With this expression for λ it follows from equ. (3.4.4a):

$$\ln A(t) = \ln A(0) - \ln(2)\cdot t/T \tag{3.4.4b}$$

The use of the half-life is often more convenient than that of the decay constant λ. Hence, in final expressions λ is replaced by ln 2/T. Half-lives are usually given in units of seconds (s), minutes (m), hours (h), days (d) and years (y).
Examples of radioactive decays into a stable final state are: ^{60}Co, ^{128}I, ...

3.4.1.2 Branching radioactive decay

Several nuclides can decay via more than one decay branch, e.g. by electron capture as well as by β^- or β^+-decay. The decay constant λ being characteristic for the nuclide's decay is in this case composed of the partial decay constants of the individual decay branches:

$$\lambda = \lambda_1 + \lambda_2 + ... + \lambda_n \tag{3.4.5}$$

Since the activity for each decay branch is $A_i = \lambda_i N$, the total activity of the nuclide is given by:

$$A = A_1 + A_2 + ... + A_n = \lambda_1 N + \lambda_2 N + ... + \lambda_n N \tag{3.4.6}$$

or

$$A(t) = \lambda_1 N(0)\, e^{-\lambda t} + \lambda_2 N(0)\, e^{-\lambda t} + ... + \lambda_n N(0)\, e^{-\lambda t} \tag{3.4.7}$$

The partial activities are proportional to the total number N of the respective nuclide decreasing with its corresponding half-life.
Example: ^{64}Cu $\rightarrow$ ^{64}Ni + ^{64}Zn (T=12.7 h; β^+=19 %, β^-=39 %, EC = 42 %)

3.4.1.3 Mixture of several nuclides without genetic relations

The total activity of a mixture of radionuclides at any time is proportional to the sum of the individual activities at the respective time:

$$A = A_1 + A_2 + ... + A_n = A_1(0)\cdot e^{-\lambda_1 t} + A_2(0)\cdot e^{-\lambda_2 t} + ... + A_n(0)\cdot e^{-\lambda_n t} \tag{3.4.8}$$

3.4.1.4 Activity of nuclides with genetic relations

For activity chains with generic mother-daughter relations the activity of the mother nuclide (index 1) is given by equ. (3.4.4). For the determination of the daughter activity (index 2) one has to consider both the formation via decay of the mother nuclide with the decay constant λ_1 and the decay of the daughter with its own decay constant λ_2:

$$dN_2/dt = \lambda_1 N_1 - \lambda_2 N_2 \tag{3.4.9}$$

The number of daughter nuclei at time t is:

$$N_2(t) = N_1(0)\frac{\lambda_1}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2(0)e^{-\lambda_2 t} \tag{3.4.10}$$

$$A_2(t) = A_1(0)\frac{\lambda_2}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + A_2(0)e^{-\lambda_2 t} \text{ or} \tag{3.4.10a}$$

$$A_2(t) = A_1(0)\frac{T_1}{T_1 - T_2}\left(e^{-\frac{t\ln 2}{T_1}} - e^{-\frac{t\ln 2}{T_2}}\right) + A_2(0)e^{-\frac{t\ln 2}{T_2}} \tag{3.4.10b}$$

If at t = 0 the daughter activity is zero, equ. (3.4.10b) becomes

$$A_2(t) = A_1(0)\frac{T_1}{T_1 - T_2}\left(e^{-\frac{t\ln 2}{T_1}} - e^{-\frac{t\ln 2}{T_2}} \right) \tag{3.4.11}$$

3.4.1.5 Secular radioactive equilibrium ($T_1 \gg T_2$)

If the half-life T_1 of the mother nuclide is much larger than that of the daughter nuclide T_2 the latter can be discarded in the quotient of equ. (3.4.11):

$$A_2(t) = A_1(0)\left(e^{-\frac{t\ln 2}{T_1}} - e^{-\frac{t\ln 2}{T_2}} \right) \tag{3.4.12}$$

If in addition $t \ll T_1$, equ. (3.4.12) becomes:

$$A_2(t) = A_1(0)\left(1 - e^{-\frac{t\ln 2}{T_2}} \right) \tag{3.4.13}$$

what means that the increase of the daughter activity depends only on T_2.

After a period of about 6 - 7 T_2 the daughter activity equals approximately that of the mother nuclide. The total activity for periods small when compared to T_1 is constant and equals $2A_1$.
Example: $^{90}\mathrm{Sr} \rightarrow {}^{90}\mathrm{Y} \rightarrow {}^{90}\mathrm{Zr}$ (T_1=28.64 y, T_2=64.1 h)

3.4.1.6 Transient equilibrium ($T_1 > T_2$)

If $T_1 > T_2$ but both are of the same order of magnitude equ. (3.4.11) is valid. It can be simplified for periods t larger than about 6-7 T_2, since the expression

$$e^{-\frac{t\ln 2}{T_2}}$$

decreases much faster with time than

$$e^{-\frac{t\ln 2}{T_1}}$$

$$A_2(t) = A_1(0)\frac{T_1}{T_1 - T_2} e^{-\frac{t\ln 2}{T_1}} \tag{3.4.14}$$

The system reaches an equilibrium state, where the daughter activity is

$$A_2 = \frac{T_1}{T_1 - T_2} A_1$$

and decreases with the half-life of the mother T_1.

The daughter activity A_2 for $t = 0$ equals zero and approaches zero again for very large times t. The time t_m, where A_2 has a maximum, is found from the first derivative of equ. (3.4.11):

$$t_m = \frac{T_1 T_2}{(T_2 - T_1)\ln 2} \ln \frac{T_2}{T_1} \tag{3.4.15}$$

For the total mother- and daughter activity

$$A_{total} = A_1 + A_2 = A_1(0) e^{-\frac{t \ln 2}{T_1}} + A_1(0) \frac{T_1}{T_1 - T_2} \left(e^{-\frac{t \ln 2}{T_1}} - e^{-\frac{t \ln 2}{T_2}} \right) \tag{3.4.16}$$

the time for the maximum is found correspondingly:

$$t_m = \frac{T_1 T_2}{(T_2 - T_1)\ln 2} \ln \frac{{T_2}^2}{2T_1 T_2 - {T_2}^2} \tag{3.4.17}$$

Example: $^{132}\mathrm{Te} \rightarrow {}^{132}\mathrm{I} \rightarrow {}^{132}\mathrm{Xe}$ (T_1=76.3 h, T_2=2.3 h)

3.4.1.7 Similar half-lives ($T_1 \approx T_2$)

If the half-lives of mother and daughter nuclide are very similar, equ. (3.4.11) can no more be used. By means of series development one obtains:

$$A_2(t) = A_1(0) \frac{t \ln 2}{T_2} e^{-\frac{t \ln 2}{T_2}} \left(1 + \frac{t(T_1 - T_2)\ln 2}{2T_1 T_2} \right) \tag{3.4.18}$$

The term

$$\frac{t(T_1 - T_2)\ln 2}{2T_1 T_2}$$

can be neglected if it adopts values «1. Equation (3.4.18) has then the following form:

$$A_2(t) = A_1(0) \frac{t \ln 2}{T_2} e^{-\frac{t \ln 2}{T_2}} \tag{3.4.19}$$

For

$$\frac{t(T_1 - T_2)\ln 2}{2T_1 T_2} \cong 1$$

the bracket in equ. (3.4.18) cannot be neglected. For

$$\frac{t(T_1 - T_2)\ln 2}{2T_1 T_2} > 1$$

instead of equ. (3.4.18) equ. (3.4.11) has to be used again. For the special case $T_1 = T_2$ one obtains formally again equation (3.4.19).

From the first derivative of equ. (3.4.19) one gets the time t_m of the maximum activity:

$$t_m = \frac{T}{\ln 2} = \frac{1}{\lambda} \tag{3.4.20}$$

If the mother nuclide at time $t = 0$ did not contain any daughter activity, t_m corresponds to the time where mother and daughter activities are equal.

Example: $^{101}Mo \rightarrow {}^{101}Tc \rightarrow {}^{101}Ru$ (T_1=14.6 m, T_2=14.2 m)

3.4.1.8 Half-life of mother nuclide shorter than half-life of daughter ($T_1 < T_2$)

In case of $T_1 < T_2$ equ. (3.4.11) can be rewritten as:

$$A_2(t) = A_1(0)\frac{T_1}{T_2 - T_1}\left(e^{-\frac{t\ln 2}{T_2}} - e^{-\frac{t\ln 2}{T_1}}\right) \tag{3.4.21}$$

In this equation $T_1/(T_2-T_1) <1$, if $2T_1 < T_2$. For times $t \ll T_2$ the increase of the daughter activity A_2 depends only on the half-life T_1 of the mother nuclide in contrast to the cases of secular and transient equilibrium:

$$A_2(t) = A_1(0)\frac{T_1}{T_2 - T_1}\left(1 - e^{-\frac{t\ln 2}{T_1}}\right) \tag{3.4.22}$$

For times $t > T_1$ the daughter activity decreases with its individual half-life:

$$A_2(t) = A_1(0)\frac{T_1}{T_2 - T_1}e^{-\frac{t\ln 2}{T_2}} \tag{3.4.23}$$

In this case no equilibrium is reached, since the ratio of daughter and mother activities increases permanently as function of time:

$$\frac{A_2(t)}{A_2(t)} = \frac{T_1}{T_2 - T_1}\left(e^{\frac{T_2 - T_1}{T_1 T_2}t\ln 2} - 1\right) \tag{3.4.24}$$

The daughter activity passes a maximum, like in the cases 3.4.1.6 and 3.4.1.7. The corresponding time t_m can be calculated from equ. (3.4.15), too.

Example: $^{135}I \rightarrow {}^{135}Xe \rightarrow {}^{135}Cs$ (T_1=6.61 h, T_2=9.1 h)

3.4.1.8 Successive activities in longer decay chains (radioactive families)

In case of further decay of a daughter nuclide (decay chains, radioactive families) the activity A_3 at time t of the next member in the chain is given by equ. (3.4.25) provided that at time $t = 0$ only the mother activity existed:

$$A_3(t) = A_1(0)T_1\left(\frac{T_1}{(T_1-T_2)(T_1-T_3)}e^{-\frac{t\ln 2}{T_1}} + \frac{T_2}{(T_2-T_1)(T_2-T_3)}e^{-\frac{t\ln 2}{T_2}} + \frac{T_3}{(T_3-T_1)(T_3-T_2)}e^{-\frac{t\ln 2}{T_3}}\right) \quad (3.4.25)$$

Example: $^{211}Pb \rightarrow {}^{211}Bi \rightarrow {}^{207}Tl \rightarrow {}^{207}Pb$ (T_1=36.1 m, T_2=2.17 m; T_3=4.77 m)
For $T_1 \gg T_2$ and $T_1 \gg T_3$ and for times $t \ll T_1$ equ. (3.4.25) is simplified to:

$$A_3(t) = A_1(0)\left(1 - \frac{T_2}{T_2-T_3}e^{-\frac{t\ln 2}{T_2}} + \frac{T_3}{T_2-T_3}e^{-\frac{t\ln 2}{T_3}}\right) \quad (3.4.26)$$

For $T_2 > T_1$ and $T_2 > T_3$ and for times $t \gg T_1$ and $t \gg T_3$ equ. (3.4.25) is further simplified to:

$$A_3(t) = A_1(0)\frac{T_1T_2}{(T_2-T_1)(T_2-T_3)}e^{-\frac{t\ln 2}{T_2}} \quad (3.4.27)$$

and for $T_1 < T_2$ and $T_3 \ll T_2$ to:

$$A_3(t) = A_1(0)\frac{T_1}{T_2-T_1}e^{-\frac{t\ln 2}{T_2}} = A_2(t) \quad (3.4.28)$$

The general equation for the activity of the nth member of a radioactive decay chain was given by Bateman [10Bat] for the case that at $t = 0$ there are no decay products of the mother nuclide:

$$A_n(t) = A_1(0)T_1\left(C_1e^{-\frac{t\ln 2}{T_1}} + C_2e^{-\frac{t\ln 2}{T_2}} + ... + C_ne^{-\frac{t\ln 2}{T_n}}\right) \quad (3.4.29)$$

with

$$C_n = \frac{T_n^{(n-2)}}{(T_n-T_1)(T_n-T_2)...(T_n-T_{n-1})},\ C_2 = \frac{T_2^{(n-2)}}{(T_2-T_1)(T_2-T_3)...(T_2-T_n)},\ C_1 = \frac{T_1^{(n-2)}}{(T_1-T_2)(T_1-T_3)...(T_1-T_n)}$$

3.4.2 Decay modes

In order to minimise their total energy, many nuclides undergo radioactive decay. Depending on the lowest reachable energy state various decay types occur either caused by the strong interacting nuclear force (non-isobaric decay with change of the mass number) or by the weak interaction (isobaric decay with constant mass number).

The most common radioactive decay modes observed for naturally occurring nuclides are α-decay (non-isobaric), β-decay (isobaric) in the 3 types β^--, β^+-decay, and electron capture, and γ-decay (electromagnetic transitions between nuclear states). Much less abundant decay modes are the spontaneous fission and the emission of nucleons (protons, neutrons) and nucleon clusters. The following scheme (Fig. 3.3) shows the nuclear transmutations (changes of atomic and mass number) originating from the main decay modes. A more complete compilation is given in Table 3.3.

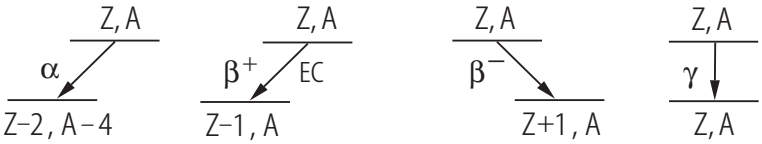

Fig. 3.3. Transmutations of atomic and mass numbers during the main nuclear decay modes.

During the *α-decay* $^4He^{2+}$ ions are emitted by the unstable radionuclide, whose mass and atomic numbers decrease consequently by 4 and 2 units, respectively. The transitions take place between well defined states in the mother and daughter nuclides leading to one or several monoenergetic α-lines for each α-decaying nuclide. The energy available for the α-decay can be obtained from the Einstein relation and the average binding energy per nucleon (Fig. 3.2) resulting in the fact that all nuclides with $A > 140$ should be unstable against α-decay. However, further effects have to be considered. In order to leave the nucleus, the α-particle has to surmount a high potential barrier, which can be passed at high energies only. Nevertheless, α-particles are observed with energies well below this barrier. This is a consequence of the quantum mechanical tunnel effect according to which a certain probability exists to pass this barrier at lower energies. Since the width of this barrier decreases with increasing potential, one could expect that α-particles with higher energy tunnelling at a narrower barrier width would pass the barrier with higher probability. This was observed phenomenologically already in 1911 by Geiger and Nutall who found a clear correlation between half-life and range of α-particles, the latter being a direct measure for the α-energy: $\log \lambda = a \cdot \log E_\alpha + b$ (Geiger-Nutall rule).

Table 3.2. Compilation of nuclear decay modes

Decay mode	Symbol	Radiation emitted	Decay process
α-decay	α	helium nuclei He^{2+}	$(Z, A) \rightarrow (Z-2, A-4) + {}^4He^{2+}$
β-decay	β^-	electrons e^-, antineutrinos	$(Z, A) \rightarrow (Z+1, A) + e^- + \bar{\nu}_e$
	β^+	positrons e^+, neutrinos	$(Z, A) \rightarrow (Z-1, A) + e^+ + \nu_e$
electron capture (EC)	ε	characteristic X-rays/ Auger electrons of the daughter nuclide, neutrinos	$(Z, A) \rightarrow (Z-1, A) + \nu_e$
γ-transition	γ	photons ($h\nu$)	release of nuclear excitation energy after particle emission
isomeric transition (IT)	I_γ	photons ($h\nu$)	delayed release of nuclear excitation energy
internal conversion (IC)	e^-	conversion electrons and characteristic X-rays	transfer of nuclear excitation energy to an inner shell (K-,L-,...) electron
proton decay	p	protons	$(Z, A) \rightarrow (Z-1, A-1) + p$
spontaneous fission	sf	fission products, neutrons	$(Z, A) \rightarrow (Z', A') + (Z-Z', A-A'-x) + x\,n$

The denotation *β-decay* combines all nuclear decay modes, by which the atomic number Z is changed by one unit, while the mass number A remains unchanged (cf. Fig. 3.3). During β^--decay a neutron in the nucleus is transformed into a proton, a negative electron together with an (anti-)neutrino is emitted and the atomic number changes from Z to $Z+1$. In contrast, during β^+-decay a proton in the nucleus is transformed into a neutron, a positive positron together with a neutrino is emitted and the atomic number changes from Z to $Z-1$. From the mass balance of this process it follows that it can take place only, if the atomic masses of mother and daughter nuclide differ by at least two electron rest mass units. The neutrinos emitted together with the β-particles are uncharged nearly mass-less particles which give rise to the continuous form of the β-spectra, since the available transition energy is shared between the $\beta^\pm$-particle and the usually not observed neutrino. The third possibility competing to the β^+-decay, the *electron capture*, results from the fact that the electron orbits of the inner shells - especially the K-shell -

can pass the nucleus and the electron can be captured by an excess proton forming a neutron. The hole in the electron shell is filled by an electron from outer shells giving rise to the emission of characteristic X-rays of the daughter nuclide, which are the only external radiation of this process.

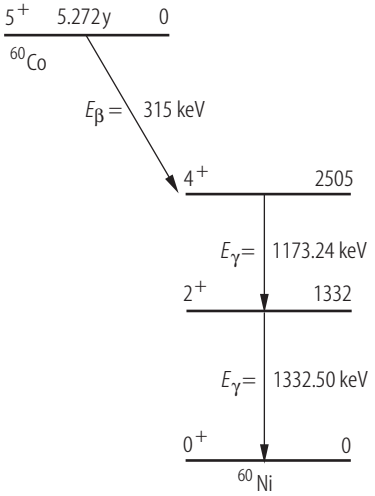

Fig. 3.4. Main part of the decay scheme of ^{60}Co decaying by β^--decay to ^{60}Ni. On the right hand side of the states their energy referred to the ground state is given (in keV), on the left hand side the nuclear spin and parity characterising the respective level.

Quite often the radioactive decay modes as discussed above do not populate the ground states but rather excited states of the daughter nuclides (cf. Fig. 3.4). These are de-excited mostly immediately by one or more *γ-transitions*, until the ground state is reached. The energy of the γ-rays corresponds to the energy difference of the nuclear levels involved in the respective transition.

Besides of γ-ray emission an excited nucleus can interact directly with a bound shell electron by transfer of its energy leading to emission of the electron. This process - called *internal conversion* - is a one-step process without the production of an intermediate γ-ray. It competes with the normal γ-emission, preferentially for heavy nuclei, and obeys the same selection rules like γ-decay. The emission of an electron of the K-shell is called K-conversion and analogous for the other shells L-, M-, N-conversion.

Spontaneous fission is a decay mode by which heavy nuclides with high neutron excess separate under energy release into two lighter fragments and several neutrons. Among natural radioactive elements it occurs only in very few cases for primordial or very long-lived nuclides heavier than uranium. Its probability compared to the competing α- or β-decay is very small for uranium (^{238}U: ~1:10^6), but increases markedly with increasing atomic number Z and neutron excess. In addition, several short-lived isomers decaying exclusively by spontaneous fission (fission isomers) have been observed for the transuranium elements Np to Bk. Spontaneous fission is described by the transformation:

$$(Z, A) \rightarrow (Z', A') + (Z-Z', A-A'-x) + xn + \Delta E,$$

where (Z', A') and $(Z-Z', A-A')$ denote the two fission fragments, x is the number of emitted neutrons and ΔE the energy released mainly in form of kinetic energy of the excited fragments and γ-radiation from their de-excitation. More details on fission products and further physical background of fission will be given in Chapter 3.6.

A further decay mode occurring, however, very rarely and mostly for nuclides far off the valley of β-stability is the emission of protons, neutrons or heavier nucleon clusters ($^{5\text{-}7}$Li, $^{7\text{-}9}$Be, $^{11\text{-}14}$C, $^{14\text{-}16}$N, $^{19\text{-}22}$F and $^{20\text{-}25}$Ne).

With increasing proton excess on the left-hand side of the valley of β-stability, the binding energy of the last proton decreases markedly, and proton emission from the ground state *(p-decay)* becomes energetically possible by tunnelling through the energy barrier of the nuclear potential as in the case of α-decay. The emission of monoenergetic protons of 1.06 and 1.23 MeV, respectively, by transmutation of the ground state of the mother nuclide into the ground state of the daughter nuclide was first observed for ^{147}Tm→^{146}Er + p (E_p = 1.051 MeV, $t_{1/2}$ = 0.56 s) and ^{151}Lu→^{150}Yb + p (E_p = 1.233 MeV, $t_{1/2}$ = 85 ms). Due to the competition with β^+-decay, which is favoured in most cases, p-decay from the ground state

occurs very rarely. Much more frequently, p-emission occurs after β^+-decay feeding an excited state in the daughter nuclide, from which the proton can easier surmount the energy barrier *(β^+-delayed proton emission)*. β^+-delayed emission of protons, in some cases even of two protons or an α-particle, was found for the lightest known isotopes of most of the elements between B and Zr.

The binding energy of additional neutrons is higher than that of additional protons and approaches zero only at large distances from the line of β-stability. Therefore, on the right-hand (neutron-rich) side of the line of β-stability all known nuclides are energetically stable to neutron emission from the ground state, which has not been observed up to now. In contrast, neutron emission immediately following β^--decay *(β^--delayed neutron emission)* is observed for many neutron-rich nuclides and many fission products.

In very recent years spontaneous emission of particles heavier than α-particles *(cluster-emission)* has been observed in several cases. Spontaneous fragmentation of nuclei with atomic numbers $Z > 40$ by emission of cluster nuclei is energetically possible with extremely large partial half-lives. Consequently, cluster radioactivity is a very rare event when compared to other decay modes.

3.4.3 The natural radioactive decay families

After the first detection of radioactivity in 1896 many other radioactive substances were identified in the investigated uranium and thorium minerals. Among these, there are three primordial isotopes: ^{232}Th, ^{238}U and ^{235}U, which initiate three naturally occurring decay series. They were called the thorium, uranium-radium and actinium families, the latter two according to their most important members ^{226}Ra and ^{227}Ac, respectively.

In these three decay series, only α- and β^--decay occurs. By the emission of an α-particle the mass number decreases by 4 and the atomic number by 2 units, whereas by the emission of a β^--particle the mass number remains unchanged and the atomic number increases by 1 unit. From this fact it follows that all members of such a decay series have mass numbers differing from each other only by multiples of 4 units. This means that all mass numbers occurring in a series have mass numbers $A = 4n + b$, where n varies within the series and b depends only on the mass number of the starting primordial isotope. Consequently, the thorium family originating from ^{232}Th ($A = 232 = 4n$ with $n = 58$) is characterised by the common label $A = 4n$; by variation of n, all possible mass numbers of the members of the decay series are obtained. For the uranium-radium family starting with ^{238}U, the respective label is $A = 4n + 2$, and for the actinium family starting with ^{235}U, $A = 4n + 3$. One radioactive decay series with $A = 4n + 1$ is obviously missing in nature. Members of this family can be produced, however, artificially via nuclear reactions. The longest half-life in this decay series belongs to ^{237}Np; therefore it is called the neptunium family. It was probably present in natural matter after the genesis of the elements before about 5×10^9 y but decayed due to the relatively short half-life of ^{237}Np.

The final members of all these decay series are of course stable nuclides: ^{208}Pb at the end of the thorium family, ^{206}Pb for the uranium-radium family, ^{207}Pb for the actinium family, and ^{209}Bi for the neptunium family. Furthermore, in all four decay series one or more branchings occur due to the capability of several nuclides to decay by α-decay as well as by β^--decay. In nearly all cases, however, one branch preponderates strongly and the weak branch ends up at the same common stable end product of the family.

In the early years of radioactivity only few radioactive elements and no concept of isotopy had been established. Consequently, the newly detected activities, mainly distinguished from their differing half-lives, were given historical names, like mesothorium, actinouranium, thoron, ..., according to the main family members chemically identified at that time. These denominations are frequently found in older literature. In Table 3.4 the abbreviations of these historical names are assigned to the respective isotopes.

In the four diagrams in Fig. 3.5 all members of the 4 families are given together with their half-lives and decay modes (arranged in a compressed form of the Segrè type of charts of nuclides). For branching decays the main decay branch is given first and indicated by a thick arrow pointing to the daughter nuclide. A thin arrow marks a second weak branch (mostly < 1%) and is also used for subsequent decays in these branches.

Table 3.3. Historical names (abbreviations) of the members of the three natural radioactive families

Thorium family $A = 4n$

^{232}Th	^{228}Ra	^{228}Ac	^{228}Th	^{224}Ra	^{220}Rn	^{216}Po	^{212}Pb	^{212}Bi	^{212}Po	^{208}Tl	^{208}Pb
Th	$MsTh_1$	$MsTh_2$	RdTh	ThX	Tn	ThA	ThB	ThC	ThC'	ThC''	ThD

Actinium family $A = 4n + 3$

^{235}U	^{231}Th	^{231}Pa	^{227}Ac	^{227}Th	^{223}Fr	^{223}Ra	^{219}At	^{219}Rn	^{215}Bi	^{215}Po	^{211}Pb
AcU	UY	—	—	RdAc	AcK	AcX	—	An	—	AcA	AcB
^{215}At	^{211}Bi	^{211}Po	^{211m}Po	^{207}Tl	^{207}Pb						
—	AcC	AcC'	—	AcC''	AcD						

Uranium-radium family $A = 4n + 2$

^{238}U	^{234}Th	^{234m}Pa	^{234}Pa	^{234}U	^{230}Th	^{226}Ra	^{222}Rn	^{218}Po	^{214}Pb	^{218}At	^{218}Rn
UI	UX_1	UX_2	UZ	UII	Io	—	—	RaA	RaB	—	—
^{214}Bi	^{214}Po	^{210}Tl	^{210}Pb	^{206}Hg	^{210}Bi	^{206}Tl	^{210}Po	^{206}Pb			
RaC	RaC'	RaC''	RaD	—	RaE	RaE''	RaF	RaG			

Fig. 3.5a-d (following). Decay chains of the 4 natural radioactive families in a compressed Segrè type representation (*N–Z* over *Z*). In this representation α-decay leads vertical to the second field below the mother nuclide, while β^- decay leads diagonal to the left field above. All chain members are given together with their half-lives and decay modes. For branching decays the main decay branch is given first and indicated by a thick arrow pointing to the daughter nuclide. A thin arrow marks a second mostly very weak branch and is also used for subsequent decays.

***A* = 4n**

N-Z / *Z*	44	46	48	50	52
90			^{228}Th; α 1.913 y		^{232}Th; α $1.405 \cdot 10^{10}$ y
89				^{228}Ac; β^- 6.13 h	
88			^{224}Ra; α 3.66 d		^{228}Ra; β^- 5.75 y
87					
86			^{220}Rn; α 55.6 s		
85					
84	^{212}Po; α 300 ns		^{216}Po; α 0.15 s		
83		^{212}Bi; α, β^- 60.60 m			
82	^{208}Pb stable		^{212}Pb; β^- 10.64 h		
81		^{208}Tl; β^- 3.053 m			

(a) Thorium family

***A* = 4n + 1**

N-Z / *Z*	43	45	47	49	51	53
95					^{241}Am; α 432.2 y	
94						[^{238}U(α,n) ⇒] ^{241}Pu; β$^-$, α 14.35 y
93					^{237}Np; α $2.144 \cdot 10^6$ y	
92				^{233}U; α $1.592 \cdot 10^5$ y		^{237}U; β$^-$ 6.75 d
91					^{233}Pa; β$^-$ 27.0 d	
90				^{229}Th; α 7880 y		[(^{232}Th+n) ⇒] ^{233}Th; β$^-$ 22.3 m
89			^{225}Ac; α 10.0 d			
88				^{225}Ra; β$^-$ 14.8 d		
87			^{221}Fr; α 4.9 m			
86		^{217}Rn; α 0.54 ms				
85			^{217}At; α, β$^-$ 32.3 ms			
84		^{213}Po; α 4.2 μs				
83	^{209}Bi stable		^{213}Bi; β$^-$, α 45.59 m			
82		^{209}Pb; β$^-$ 3.253 h				
81			^{209}Tl; β$^-$ 2.16 m			

(b) Neptunium family

This decay chain is not directly initiated by a primordial nuclide but rather by nuclear reactions of the naturally occurring nuclides ^{238}U and ^{232}Th, respectively, with α-particles and fission neutrons from their decay as indicated in the square brackets.

$A = 4n + 2$

N-Z / *Z*	42	44	46	48	50	52	54
92					^{234}U; α 2.455·10^5 y		^{238}U; α 4.468·10^9 y
91						$^{234g/m}$Pa; β$^-$ 6.7 h/1.17 m	
90					^{230}Th; α 7.54.10^4 y		^{234}Th; β$^-$ 24.10 d
89							
88					^{226}Ra; α 1600 y		
87							
86					^{222}Rn; α 3.825 d		
85				^{218}At; α ≈2 s			
84	^{210}Po; α 138.38 d		^{214}Po; α 164 µs		^{218}Po; α, β$^-$ 3.05 m		
83		^{210}Bi; β$^-$,α 5.013 d		^{214}Bi; β$^-$,α 19.9 m			
82	^{206}Pb stable		^{210}Pb; β$^-$,α 22.3 y		^{214}Pb; β$^-$ 26.8 m		
81		^{206}Tl; β$^-$ 4.2 m		^{210}Tl; β$^-$ 1.3 m			
80			^{206}Hg; β$^-$ 8.15 m				

(c) Uranium-Radium family

In the Uranium-Radium family the decay of ^{234}Th results in the population of the isomeric state (m) as well as the groundstate (g) of ^{234}Pa which in turn decay to ^{234}U.

***A* = 4n + 3**

N-Z / *Z*	43	45	47	49	51
92					^{235}U; α 7.038·10^{8} y
91				^{231}Pa; α 3.276·10^{4} y	
90			^{227}Th; α 18.72 d		^{231}Th; β$^-$ 25.5 h
89				^{227}Ac; β$^-$, α 21.773 y	
88			^{223}Ra; α 11.43 d		
87				^{223}Fr; β$^-$, α 21.8 m	
86			^{219}Rn; α 3.96 s		
85		^{215}At; α 0.1 ms		^{219}At; α, β$^-$ 0.9 m	
84	^{211}Po; α 0.516 s		^{215}Po; α, β$^-$ 1.78 ms		
83		^{211}Bi; α, β$^-$ 2.17 m		^{215}Bi; β$^-$ 7.6 m	
82	^{207}Pb stable		^{211}Pb; β$^-$ 36.1 m		
81		^{207}Tl; β$^-$ 4.77 m			

(d) Actinium family

3.5 Radioactive radiation

3.5.1 Types of radiation

For the identification and investigation of radionuclides as well as for radiation protection the properties and interactions of nuclear radiation have to be known. The most important aspect is the interaction of radiation with matter, which determines the detection methods and the actions on inorganic and organic matter.

Charged particles or photons, such as α-particles, protons, electrons, positrons, γ- or X-rays, induce ionisation processes in gases, liquids or solids and are called, therefore, *ionising radiation*. In addition to the production of excited atoms or molecules also chemical reactions may be induced.

The minimum energy needed for ionisation or excitation of atoms or molecules is of the order of several eV and depends on the type of the atoms or molecules involved. The photons of visible light and neighbouring wavelenghts have energies between about 1 eV (wavelength λ = 1240 nm) and 10 eV (λ = 124 nm). If their energy exceeds the ionisation energy of the absorbing matter, they lose it in a single ionisation process. On the other hand, charged particles have energies in the range of 0.1 to 10 MeV and produce a large number of ions, electrons and excited atoms or molecules in many interaction steps.

3.5.1.1 Particle radiation

This type of radiation comprises the frequently and also naturally occurring α- and β-radiation, the neutron radiation accompanying mainly nuclear fission, and the quite rare types of nucleon- and cluster-emission. The main properties of these particles influencing their physical behaviour like interaction with and absorption in matter are their mass, charge and energy.

Ions and excited atoms or molecules produced in the primary interactions of charged particles with matter give rise to further (secondary) physical and chemical reactions. Many of these secondary reactions are very fast and happen relatively frequently. The concentration of such reaction products along the path of the primary particle is proportional to its energy loss per unit path length. For example, α-particles of 1 MeV loose 190 eV/nm in water, whereas 1 MeV electrons loose only 0.2 eV/nm, resulting in shorter ranges for the α-particles but higher concentrations of reaction products, respectively.

Very high energetic particles may also induce nuclear reactions. Electrons loose in the force field of nuclei part of their energy by generating bremsstrahlung. For electron energies of the order of 1 MeV (e.g. β-radiation), these photons have energies in the X-ray range and for energies >10 MeV the bremsstrahlung photons have the energies of γ-rays.

Since neutrons carry no charge, they can interact with matter via the very short-range nuclear force, and they loose their energy stepwise mainly by collisions with atomic nuclei or they may induce nuclear reactions. This requires a large number of scattering processes and especially light materials to slow down and absorb neutrons.

From range calculations it follows that α-radiation is easily absorbed quantitatively (e.g. by a sheet of paper), for absorption of β-radiation several millimetres or centimetres of material are necessary, and for absorption of γ-radiation thick layers of either lead or concrete are needed taking into account that the absorption of γ-rays follows an exponential law. The ratio of the absorption coefficients for α-, β- and γ-radiation of equal energy is about 10^4:10^2:1.

3.5.1.2 Electromagnetic radiation

The types of electromagnetic radiation considered here are mostly produced during de-excitation processes in the electron shells or the nuclei of atoms. Due to historical reasons, this electromagnetic radiation – despite of similar fundamental properties – is differently denominated according to different origins:

- emission from de-exciting outer electron shells: light
- emission from de-exciting inner electron shells: X-rays
- emission from de-exciting nuclei: γ-radiation
- emission during deceleration of charged particles: bremsstrahlung.

The energy range of X-rays lies within about 100 eV to 100 keV (= wavelengths of about 10 nm to 10 pm), and that of γ-rays within about 10 keV to 10^4 MeV (= wavelengths of about 0.1 nm to 10^{-7} nm). That means there is an overlap in the energy ranges of X-rays and γ-rays. Electrons with energies >10 MeV decelerated in a substance of high atomic number induce the emission of high-energetic ("hard") bremsstrahlung. In contrast to the γ-rays emitted from nuclei, this bremsstrahlung shows a continuous energy distribution.

X-rays are emitted from the electron shell of the nuclides after formation of a hole in one of the inner shells either by an external process (bombardment with photons, electrons or heavy charged particles) or by an internal decay process like electron capture or internal conversion. The X-ray spectrum of an individual nuclide shows a distinct line structure corresponding to the different transition possibilities in the electronic shell and reflecting its structure.

The γ-rays emitted by an excited nucleus have well-defined energies, which correspond practically to the differences in the excitation energies of the nuclei (the recoil energies transferred to the emitting nuclei are very small). Gamma spectroscopy is, therefore, the usual method to investigate the level structure and decay schemes of atomic nuclei.

Generally, the γ-radiation is emitted immediately after a preceding α- or β-decay, since the lifetimes of excited states are of the order of 10^{-13} s. However, if immediate γ-transitions are "forbidden" because of high differences of the nuclear spins of the involved states and the ground state in combination with the conservation laws of nuclear momentum and parity (selection rules), a metastable or isomeric state results which decays with its own half-life different from that of the mother nuclide. The transition from the isomeric to the ground state is called isomeric transition (IT). Some long-lived nuclear isomers in isotopes with a stable ground state are of practical importance as pure γ-emitters.

3.5.1.3 Conversion electrons

If a γ-transition in an excited nucleus is hindered by the selection rules, a certain probability exists increasing with increasing "hindrance" to transfer the excitation energy directly to a shell electron, which is emitted monoenergetically. The energy E_e of these conversion electrons is given by $E_e = E_\gamma - E_B$ where E_γ is the respective γ-energy, and E_B the binding energy of the electron in the respective shell. Since the higher shells L, M, ... have sub-shells with slightly different binding energies, the conversion electron spectra contain 3 lines for L-conversion and 5 lines for M-conversion.

The internal conversion leaves a hole in the electron shell, which by recombination causes the emission of characteristic X-rays or Auger electrons. The latter – in contrast to conversion electrons – gain their energy from transitions between electron shells, which is in general much lower than the nuclear transition energies of conversion electrons.

The relative abundance of internal conversion is given by the total conversion coefficient $\alpha = N_e/N_\gamma$, which is composed from the partial conversion coefficients for the individual electron shells and sub-shells: $\alpha = \alpha_K + \alpha_{LI} + \alpha_{LII} + \alpha_{LIII} + \alpha_M ...$, each defined as the ratio of the number of the respective conversion electrons to the number of γ-rays emitted.

3.5.2 Physical properties of radiation

In order to investigate radionuclides and to develop protection methods against radioactive radiation, one has to deal with their properties and the interaction processes between the different radiation types and matter. In principle, the absorption of electromagnetic radiation like γ- and X-rays is different from that of particles. While the latter lose their energy by successive collisions, photons give off their energy mostly in one process. Because they are chargeless, their interaction with matter is small. The absorption of γ-rays follows an exponential law:

$$I = I_0 \, \mathrm{e}^{-\mu d} \tag{3.5.1}$$

where μ is the absorption coefficient and d the absorber thickness.

The exact validity of this exponential law is, however, restricted to monoenergetic γ-radiation, a narrow beam of γ-rays and a thin absorber.

3.5.2.1 Interaction of charged particles with matter

The primary interactions of fast charged particles with matter can be classified as follows:

a) elastic collisions with atomic electrons
b) inelastic collisions with atomic electrons
c) elastic collisions with nuclei
d) inelastic collisions with nuclei
e) nuclear reactions and interaction with nuclear forces

All these interactions can contribute in principle to deflection and deceleration of incoming particles. Since more than 10^4 collisions are necessary to stop particles with energies in the MeV range, this is a multi-scattering process composed statistically in different ways of the individual processes.

Heavy charged particles (p, d, α, ions) loose most of their kinetic energy by process (b). The electrons of the scattering atom are excited by the energy transfer, most of them to energies high enough to ionise the atoms.

Elastic collisions of heavy particles (protons, deuterons, α-particles, ...) with nuclei (c) are rare in comparison to (b), for light particles (electrons, positrons), however, much more frequent. Inelastic scattering from nuclei (d) also occurs rarely and is negligible for deceleration of heavy particles, for light particles, however, it is important at higher energies giving rise to the emission of bremsstrahlung.

Consequently, there are characteristic differences for the deceleration of heavy and light particles. On the other hand, the detection of all types of charged particles, as well as of electromagnetic radiation, is based in general on their ionisation effects. This holds also for the damaging effects in biological material.

Deceleration of heavy particles

By the inelastic scattering of fast particles from shell electrons the atoms are excited or preferentially ionised. For the resulting loss of kinetic energy per unit path one obtains:

$$-\frac{\mathrm{d}E}{\mathrm{d}s} = \frac{4\pi z^2 e^4}{m_e v^2} N_0 B \tag{3.5.2}$$

In this expression – also called *stopping power* – z means the charge of the fast particle, v its velocity and N_0 the number of nuclei per cm^3 in the absorber material.

The *atomic stopping number B* is a function, depending on the energy of the incoming particle. From a quantum mechanical calculation H. Bethe obtained the following expression for the atomic stopping power *B* under consideration of relativistic effects at high particle energies:

$$B = Z\left[\ln\frac{2m_e v^2}{I} - \ln(1-\beta^2) - \beta^2 - \frac{C_K}{Z}\right] \tag{3.5.3}$$

where Z means the atomic number of the absorber, $\beta = v/c$ and $I = 11.5 \cdot Z$ the average ionisation potential; C_K is a correction factor depending on E and Z, which is of influence only at very low projectile energies and takes values in between 0 and ~1.

For the stopping power it follows:

$$-\frac{\mathrm{d}E}{\mathrm{d}s} = \frac{4\pi z^2 e^4}{m_e v^2} N_0 Z\left[\ln\frac{2m_e v^2}{I} - \ln(1-\beta^2) - \beta^2 - \frac{C_K}{Z}\right] \tag{3.5.4}$$

The schematic course of equ. (3.5.4) shown in Fig. 3.6 demonstrates qualitatively that in the intermediate region for energies much larger than the ionisation potential and much less than the particle rest mass ($I \ll E \ll Mc^2$) the energy loss decreases approximately as $1/E$, since the logarithmic term varies slowly and the relativistic terms are negligible. At high velocities, however, these terms give rise to a slight increase causing a flat minimum at $E \approx 3Mc^2$. At low energies the logarithmic term prevails and the curve decreases steeply below $E \approx 500 \cdot I$. From this behaviour it follows that the ionisation density produced by the decelerated particle in the absorber increases markedly before the end of the path s and decreases steeply behind this point. Furthermore, the $1/v^2$ dependence of the first term in equ. (3.5.4) causes a stretching of the energy scale for different masses of the incoming projectiles (cf. Fig. 3.6).

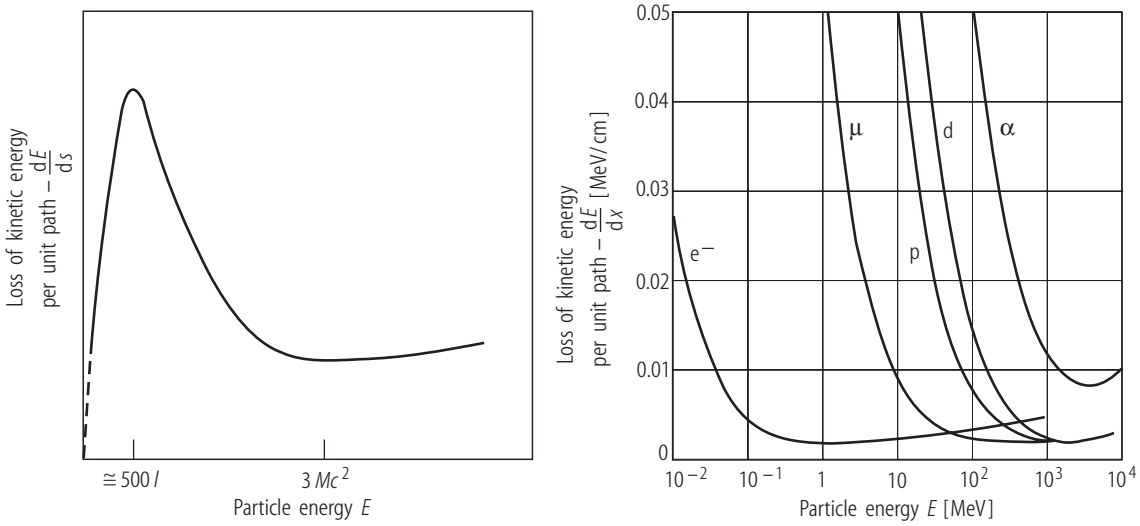

Fig. 3.6. Schematic course of the stopping power with energy (left) and dependence on the projectile type for electrons, muons, protons, deuterons and α-particles (right).

3.5.2.2 Interaction of neutrons with matter

Neutrons are emitted by radioactive decay during spontaneous fission and in rare cases after β-decay of a very neutron-rich nuclide to an excited level of the daughter nuclide. In nuclear reactions and in particular in nuclear fission neutrons are of high importance, especially for the production of radionuclides. Since from radioactive sources mostly only small fluxes can be obtained, neutron sources based on nuclear reactions, like $^{9}Be(\alpha,n)^{12}C$, $^{9}Be(d,n)^{10}B$ or $^{9}Be(\gamma,n)2\alpha$, are used. The highest fluxes of neutrons are available in nuclear reactors.

The interaction of neutrons with matter happens mainly with nucleons and nuclei via elastic and inelastic scattering and nuclear reactions, because they are electrically neutral and interact hardly with electrons. Thus, ionisation by neutrons is negligible. In elastic collisions the total kinetic energy remains constant and only deflection occurs, while in inelastic collisions part of the kinetic energy is used for excitation of the collision partner. Depending on the neutron energy different types of interactions occur and, correspondingly, several energy ranges are distinguished:

- 0-0.1 eV: thermal neutrons (energies similar to those of gas molecules at room temperature)
- 0.1-100 eV: slow neutrons
- 1-10 keV epithermal neutrons
- 0.1-100 keV: neutrons of intermediate energies
- 0.1-10 MeV: fast neutrons

In contrast to charged particles, neutrons do not undergo Coulomb interaction with nuclei. However, low-energy (thermal and slow) neutrons are very effectively absorbed by a great number of nuclei, giving rise to nuclear reactions. Elements such as B, Cd, Sm, Eu, Gd and Dy are used as good neutron absorbers.

Epithermal neutrons are also called resonance neutrons, because there exist absorption maxima or resonances at defined energies for distinct absorber nuclei. Neutrons with energies corresponding exactly to the excitation energies of excited levels of the nucleus are absorbed with very high probability.

Fast neutrons are decelerated mainly by elastic and inelastic collisions. The energy released in one elastic collision depends on the collision angle and the mass number of the target nucleus. The lighter the nucleus, the higher is the energy loss of the neutron. Consequently, hydrogen or hydrogen-containing substances like water or paraffin are very effective to reduce ("moderate") the energy of neutrons. Graphite can also be used as moderating material, but needs larger material thickness due to its higher mass number. After deceleration the "slow neutrons" are captured by nuclei, giving rise to nuclear reactions. High-energy (fast) neutrons may also induce nuclear reactions, but the probability and consequently the contribution of this interaction type is relatively small.

3.5.2.3 Interaction of electromagnetic radiation with matter

Electromagnetic radiation passing a material experiences an intensity attenuation according to equ. (3.5.1) (see above) via interactions with the various components of matter. For instance, γ-rays interact with:

- atomic electrons,
- nuclei,
- electrical fields of the electrons and nuclei,
- meson fields of the nuclei.

These interactions can result in energy losses, alterations of the propagation direction and polarisation. The effects on γ-rays are:

- total absorption,
- inelastic scattering (incoherent),
- elastic scattering (coherent, Thomson scattering).

Though all of the possible combinations of interactions and processes can occur, most of them result in very weak effects, which can normally be neglected for attenuation considerations. Important are:

- the photoeffect,
- the Compton effect,
- the pair formation.

In the *photoeffect* the incoming photon is totally absorbed by an atomic electron of the inner shells (preferably of the K- or L-shells). A free electron cannot take up the total energy of the photon because of the momentum conservation. In contrast, this is possible for the bound atomic electrons, because the atom

takes up the residual momentum as recoil. Consequently, the stronger bound inner electrons exhibit the strongest absorption. The photoelectrons in turn are emitted with a kinetic energy E_{kin} corresponding to the total photon energy $h \cdot \nu$ reduced by the electron's binding energy E_b, which depends on the atomic number of the respective atom:

$$E_{kin} = h \cdot \nu - E_b \tag{3.5.5}$$

Consequently, for high photon energies and light absorbers the photoelectrons carry nearly the total photon energy. In addition, they exhibit a pronounced angular distribution favouring the forward direction with increasing photon energy. Additional (isotropic) radiation is emitted originating from the recombination process of the electron hole left by the emitted photoelectron. During this recombination characteristic X-rays and Auger electrons are released by the energy set free by an outer electron filling up the hole in the inner shell. Furthermore, bremsstrahlung is produced by the photoelectrons especially in heavy materials. All these radiation types connected with the absorption of photons together with the ionised residual absorber atoms can result in a high local energy density.

The *Compton effect* describes the inelastic scattering of photons from outer shell electrons, where the photon looses only part of its energy, which is transferred to the electron. Both, the recoiling electron and the photon are scattered with respect to the original photon direction, and the scattering into forward directions is again favoured with increasing photon energy. The relative energy transfer to the electron increases with increasing photon energy and also with increasing photon scattering angle. On the other hand, the highest backscattering contributions occur at low photon energies.

The *pair formation* occurs in the electric field of an atomic nucleus, where the γ-ray can be spontaneously transformed into an electron-positron pair, if its energy is larger than the sum of the rest masses of the pair: $E_\gamma > 2m_e c^2$ = 1,022 MeV. The nucleus remains unchanged but is necessary for the conservation of momentum. The positron annihilates by recombination with an electron of the absorber atoms, whereby two annihilation γ-quanta are produced with an energy of 511 keV each.

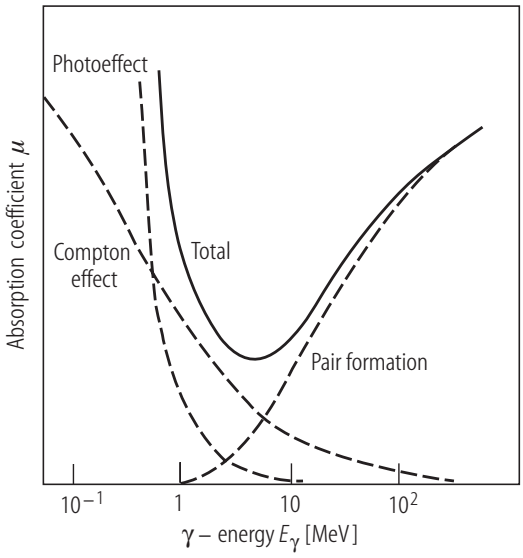

Fig. 3.7. Schematic representation of the partial and total absorption coefficients of a heavy absorber in dependence on the γ-ray energy.

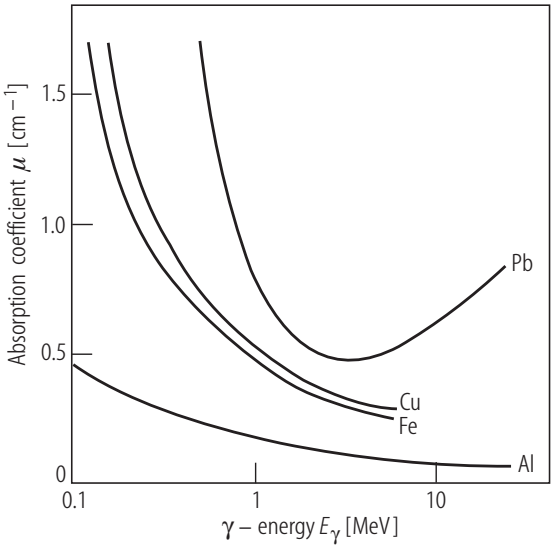

Fig. 3.8. Schematic representation of the total absorption coefficients of different absorbers in dependence on the γ-ray energy.

For the attenuation of photons in matter, mainly the three processes photoeffect, Compton effect and pair formation have to be taken into account. Fig. 3.7 shows schematically their contributions to the total absorption coefficient μ in dependence on the photon energy. A primary photon beam is attenuated by scattering as well as by absorption. The main parameters for the attenuation are the photon energy and the

atomic number and density of the absorber material (cf. Fig. 3.8). At low photon energies, classical coherent (Thomson) scattering predominates, at higher energies incoherent scattering via the Compton effect. Absorption of photon energy occurs mainly via the photoeffect and pair formation and partially via the Compton effect. The lower energy threshold for the photoeffect is given by the binding energy of the inner shell electrons, for the pair formation by the rest mass of the electron-positron pair (1022 keV). At high atomic numbers Z and low photon energies the photoeffect dominates, at high energies the pair formation. For energies between about 1 to 5 MeV the Compton effect dominates at all atomic numbers Z. The resulting Compton electrons give, therefore, the most important contribution to the energy dose in human tissue and to the biological effect of radiation.

3.6 Nuclear fission and fission products

3.6.1 Particle induced nuclear fission

Besides the few heavy nuclides decaying by spontaneous fission many other heavy isotopes undergo fission after bombardment with particles, especially neutrons. Fission of uranium was first observed but misinterpreted in 1934 by Fermi [34Fer] in an attempt to produce transuranium elements by irradiation of uranium with slow neutrons. Similar experiments were performed by several groups, but only in 1937 Hahn, Meitner and Strassmann [37HMS] identified the observed radioactive products of the fission of uranium to have appreciably lower atomic mass, such as ^{140}Ba.

Fission of heavy nuclei always leads to products with a high neutron excess due to the much larger neutron-to-proton ratio of heavy nuclides. The primary fission products are formed in about 10^{-11} s by fission and emission of prompt and β^--delayed neutrons and γ-rays from the highly excited fragments. They always lie on the right hand side of the valley of β-stability and decay by several successive β^--decays following isobaric chains into nuclides of increasing atomic number Z ending up with the first stable isobar in the chain.

The fission process exhibits different features depending mainly on the energy of the inducing particles and on the atomic number Z of the fissioning nuclide. For fission induced by low-energy neutrons with energies up to about 10 MeV two fission products with mass numbers in the range between about 70 and 160 and ≈ 2-3 neutrons are emitted. The energy ΔE released by fission is relatively high (ΔE ≈ 200 MeV), since for the light fission products the binding energy per nucleon is higher than for the heavy fissioning nuclei.

When comparing the naturally occurring uranium isotopes ^{234}U (abundance: 0.0055 %), ^{235}U (0.720 %), and ^{238}U (99.2745 %), it is found that ^{235}U undergoes fission by slow (i.e. thermal) neutron capture with a remarkably large cross section of $\sigma_{n,f} = 586 \times 10^{-24}$ cm^2. This holds also for other odd-mass (even Z, odd N) nuclei like ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu. In all these cases the binding energy of an additional neutron is very high resulting in high cross sections $\sigma_{n,f}$ for fission by slow (thermal) neutrons.

To induce fission in ^{238}U, the neutron must have an energy of 1 MeV. The natural isotope ^{232}Th undergoes fission by 1.1 MeV neutrons. Using higher energetic particles, either neutrons, deuterons, or α-particles, it is possible to induce fission in any element with atomic number larger than 73.

3.6.2 Fission products

One of the most characteristic features of the fission process – very important also for the activities that have to be regarded in nuclear technologies – is the resulting mass distribution of the various fission products. As an example, Fig. 3.9 displays the yield of fission products found after thermal fission of ^{235}U

as a function of the mass number A. The maximum yields are around A = 90 to 100 and A = 133 to 143, respectively. For these mass ranges the fission yields are about 6 %, whereas symmetrical fission around A = 117 occurs with much smaller probability (≈0.01 %). It should be reminded that the sum of the fission yields is 200 %, because each fission produces two fission products.

When investigating the mass distributions from fission with thermal neutrons one observes that those of ^{233}U and ^{239}Pu are similar to that of ^{235}U. In the case of ^{239}Pu, however, the low mass maximum is slightly shifted to higher masses, while the maximum for heavy fission products remains nearly unchanged. This tendency continues with increasing mass of the fissioning nuclei, and in the case of ^{258}Fm the two maxima are superimposed.

An increase of the energy of the neutrons leads to a strong increase of the probability for symmetric fission and gives rise to a flattening of the valley of the mass distribution by up to two orders of magnitude. Increase of symmetric fission is also observed for nuclides with lower atomic numbers Z. For ^{227}Ac (Z = 89) symmetric and asymmetric fission have nearly the same probability, resulting in three maxima in the mass distribution.

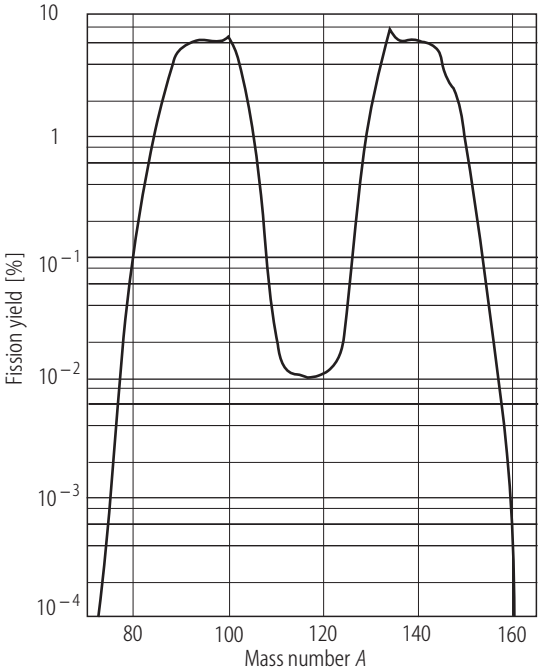

Fig. 3.9. Yields of fission products for the fission of ^{235}U by thermal neutrons.

It should be mentioned that the mass distribution curves as discussed before give the total yields of the decay chains of mass numbers A. The independent yields for individual members of the decay chains, i.e. the yields for direct formation in the fission process, are often difficult to determine, especially if in the precursor chain short half-lives occur.

The total energy ΔE released during fission consists of kinetic energy and excitation energy of the primary fission fragments, where the kinetic energy resulting mainly from the Coulomb repulsion of the two fission fragments gives the main contribution. In the case of low-energy fission the kinetic energy E_{kin} is given by the empirical relation $E_{kin} \sim Z^2 / A^{1/3}$ where Z and A are the atomic number and the mass number of the fissioning nucleus, respectively.

The primary fission fragments release their excitation energy by emission of prompt neutrons with energies between 0 and about 10 MeV (mean value ≈2 MeV) and of prompt γ-rays. The number of prompt neutrons emitted increases with the mass number of the fissioning nuclei and depends mainly on the excitation energy of the primary fission fragments. Furthermore, an average number of 7.5 γ-rays with a mean energy of about 1 MeV are emitted per fission, as well as several low-energy transitions in form of conversion electrons and X-rays.

In very rare cases high-energy charged particles such as p, d, t, α-particles, ^{3}He, ^{7}Li, ^{8}Li, ^{9}Li, ^{9}Be, ^{10}Be and isotopes of B, C, N and O are also emitted at an early stage of the low-energy fission process, when the fission fragments are still very close to each other. The probability of this so-called ternary fission, i.e. formation of three fragments increases strongly with the excitation energy of the fissioning nuclei. For example, high-energy fission of ^{232}Th with 400 MeV argon ions leads to a ratio of ternary to binary fission of about 1:30.

In contrast to low-energy fission, high-energy fission induced by neutrons or other high-energy particles leads to marked changes in the mass distribution of fission products. Among others, the probability of symmetrical fission increases considerably with increasing excitation energy of the target nuclei, resulting in a single flat maximum of the fission yield curve slightly below half the target mass number.

3.6.3 Nuclear reactors

When comparing the energies available from combustion of carbon or carbon compounds (or in general: from a chemical reaction like oxidation) with those from nuclear fission, one has to compare the orders of magnitude of the binding energies in the electron shells and in the atomic nuclei, which are in the order of eV and MeV, respectively. This means a difference of six orders of magnitude: 1 kg of carbon produces an energy of 9.4 kWh, 1 kg of uranium can produce a maximum of 1.85×10^{7} kWh. This high energy release as already observed with the first fission experiments caused immediate considerations to make use of this enormous energy potential. In addition, the neutrons released in each fission process could initiate further fissions leading in principle to a continuous generation of energy. However, it soon turned out that a safe control of a continuously proceeding fission process required the solution of various technological questions and problems. Consequently, the first application of nuclear fission was the construction of a nuclear explosive ("atomic bomb") on the basis of uncontrolled self-amplifying fission in the form of chain reactions.

As mentioned above, the fission process in the first step results in two fragments with high neutron excess and in some prompt high energy neutrons. Furthermore, from the decay chain of the highly excited primary fragments besides β^{-}-particles and γ-rays about 1 % of β-delayed neutrons are emitted which are delayed by at least 0.01 second and about 0.07 % which are delayed by as much as 1 minute. This would be a satisfactory condition for a chain reaction provided the neutrons originating from the fission process have a possibility to react with other atoms of ^{235}U. The neutrons generated during fission are, however, no thermal neutrons but have energies up to the order of 1 MeV. If these neutrons are slowed down to thermal velocities they can excite other ^{235}U atoms to fission, so that the reaction would proceed with increasing amplification as a chain reaction if only ^{235}U would be present.

In natural uranium this process is not possible because of the high concentration of ^{238}U atoms which can capture higher energy neutrons without undergoing fission. In addition, there exist well-defined energies, where a resonance absorption (in a very sharp energy window) of neutrons in ^{238}U occurs with very high cross sections (up to 1200×10^{-24} cm^{2}). Consequently, the fast neutrons have to be effectively slowed down, whereby a sufficient number must escape the resonance absorption in ^{238}U in order to reach thermal velocities. This is achieved by use of a moderator, an element of small atomic weight like deuterium, helium, beryllium, and carbon, whose atoms will not capture the neutrons but rather scatter them elastically.

3.6.4 Nuclear explosives

The high amount of energy released by nuclear fission led very early to the production of nuclear explosives. Since ^{235}U, ^{233}U and ^{239}Pu have sufficiently high fission cross sections for fast neutrons, they can be used as nuclear explosives if the respective critical masses are brought together. Without a neutron

reflector increasing the number of neutrons within the explosive material, a sphere of about 50 kg uranium metal containing 94 % ^{235}U or a sphere of about 16 kg plutonium metal (^{239}Pu) reaches criticality. If a neutron reflector is used, the critical masses are about 20 kg for ^{235}U and about 6 kg for ^{239}Pu. The critical masses for ^{233}U are similar to those for ^{239}Pu.

The use of plutonium in nuclear weapons requires a low concentration of ^{240}Pu in the plutonium, because its presence leads to the production of high numbers of neutrons by spontaneous fission. Consequently; a too high concentration of ^{240}Pu would initiate the neutron multiplication too early with a relatively small multiplication factor and a relatively low energy release. Higher concentrations of ^{241}Pu also interfere, because of its decay into ^{241}Am with a half-life of only 14.4 y. To minimise the formation of ^{240}Pu and ^{241}Pu, Pu for use in weapons is, in general, produced in special reactors with low burn-up (« 20000 MW_{th}d per ton).

Criticality can be reached by shooting two under-critical hemispheres onto each other by means of normal explosives (gun-type) or by compressing an under-critical spherical shell into a supercritical sphere (implosion-type). The bomb ignited over Hiroshima (energy release corresponding to ≈15 kilotons of TNT) was of the gun type using ^{235}U, whereas that ignited over Nagasaki was of the implosion type using ^{239}Pu (energy release corresponding to ≈22 kilotons of TNT). Generally, the fissile core is surrounded by a heavy material, in order to reflect the neutrons and to increase the inert mass and consequently the time in which the super-critical configuration is held together.

The explosion of fissile material leads to temperatures of about 10^8 K which are sufficient to initiate fusion between deuterium and tritium. This is the basis of the development of hydrogen bombs, in which the energy of fission is used for ignition of fusion. LiD serves as a source of D and T, the latter being produced by thermal (^{6}Li(n,αn)d) and by fast neutrons (^{7}Li(n,αn)t). If the temperature is high enough, the D-D reaction can contribute to the energy production. The fast neutrons released by the fusion reactions react very effectively with natural or depleted U initiating fission of ^{238}U. By these kinds of weapons large amounts of fission products are formed (“dirty weapons”). If a surrounding of non-fissile heavy material is used, fission products are released only by the ignition process (“clean weapons”).

3.6.5 Radioactive inventory and nuclear waste

The radioactive inventory in a nuclear power reactor originates from:

- fission products
- uranium and transuranium elements formed by direct neutron induced reactions and their decay chains
- isotopes produced by nuclear reactions in the cladding material of fuel rods, the reactor vessel components and the coolant.

The dominating longer-lived fission products occurring in spent fuel elements are ^{85}Kr, ^{131}I, ^{133}I, ^{133}Xe, ^{135}Xe, ^{134}Cs and ^{137}Cs. Radionuclides produced by nuclear reactions in the coolant are ^{3}H, ^{14}C, ^{13}N, ^{16}N, ^{19}O, ^{18}F and ^{41}Ar. Furthermore, fission products or actinides may leak into the cooling system from faulty fuel elements. Other radionuclides are produced by reactions with metals and their corrosion products in various reactor vessel components like ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{58}Co, ^{60}Co, ^{65}Zn, ^{124}Sb.

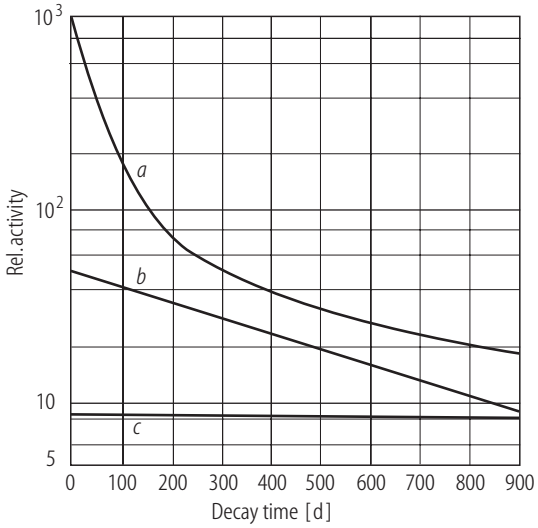

Fig. 3.10. Radioactive decay of the sum of all fission products (a), ^{106}Ru (≈0.5 %, T = 373.6 d) (b), and ^{137}Cs (≈ 6 %, T = 30.17 y) (c).

Shortly after shut-down of a nuclear reactor the activity of the fuel is $\approx 1.7 \times 10^{17}$ Bq per MW of thermal energy produced. ^{237}U (from the reactions ^{235}U(n, γ)^{236}U(n, γ)^{237}U and ^{238}U(n, 2n)^{237}U) causes a relatively high initial uranium activity. Since it decays with a half-life of 6.75 d, it vanishes rapidly after the necessary storage of the discharged fuel elements. A global composition of spent nuclear fuel from light water reactors after storage of 1 year is given in Table 3.5.

The radioactive waste produced during the operation of nuclear reactors is usually classified according to the state of matter (gaseous, liquid or solid) and according to the activity level as low-active waste (LAW), medium-active waste (MAW), and high-active waste (HAW). The largest amount of radioactivity is concentrated in the spent fuel elements representing highly-active waste (HAW).

Table 3.4. Main components of spent nuclear fuel from a light-water reactor with an initial enrichment of 3.3 % ^{235}U, a burn-up of 34000 MWd per ton and a storage time of 1 year (from [97Lie]).

Nuclide	Weight percent	Nuclide	Weight percent
uranium and transuranium elements		*fission products*	
^{235}U	0.756	^{85}K	0.038
^{236}U	0.458	^{90}Sr	0.028
^{237}U	3×10^{-9}	^{129}I	0.09
^{238}U	94.2	^{134}Cs + ^{137}Cs	0.275
^{237}Np	0.05	others	0.19
^{238}Pu	0.018	Stable fission products	3.0
^{239}Pu	0.527		
^{240}Pu	0.220		
^{241}Pu	0.105		
^{242}Pu	0.038		
americium isotopes	0.015		
curium isotopes	0.007		

Reprocessing of nuclear fuel transforms all waste types into liquid solutions and results in the following amounts per ton of U: ≈1 m^3 HAW (fission products and actinides in HNO_3 solution), ≈3 m^3 MAW as organic solution, ≈17 m^3 MAW as aqueous solution, ≈90 m^3 LAW (aqueous solution). By further processing a volume reduction is achieved: ; ≈0.1 m^3 HAW, ≈0.2 m^3 MAW (organic), ≈8 m^3 MAW (aqueous), ≈3m^3 LAW (aqueous). After respective storage times the HAW solutions are transformed by calcination or vitrification into stable forms like ceramics or glasses suitable for long-term disposal and also in order to reduce the volume.

After one year of intermediate storage and reprocessing, the initial activity of the HAW solutions is of the order of 10^{14} Bq/l from which the activity due to ^{90}Sr and ^{137}Cs is about 10^{13} Bq/l; after 10 y the activity of the HAW solution decreases approximately with the half-life of these nuclides (28.64 y and 30.17 y, respectively, cf. Fig. 3.10). After 1000 y the residual activity (of the order of 10^4 Bq/l) is determined by long-lived fission products like ^{99}Tc, ^{129}I and actinides.

Solid MAW and LAW originate from structure material of the fuel elements, undissolved, dispersed and filtered particles of metals or metal oxides (e.g. Ru, Rh, Mo, Tc), and gaseous components like tritium (as T_2 or HTO), ^{14}C (as CO_2), ^{85}Kr, ^{129}I or ^{106}Ru (as RuO_4) after adsorption in special adsorbents.

Computer codes have been developed to calculate the inventory of all radionuclides in spent fuel rods after discharge and after several periods of storage. As an example in the Tables 3.6 and 3.7, taken from [95SSK], the activities of actinides, fission products and light elements from surrounding material are listed, which were obtained from a calculation with the code KORIGEN for a given reactor type, initial ^{235}U enrichment, and fuel burn-up at charge time, discharge time and several storage times. Further information on these calculations and their results as well as on general reprocessing, recycling and disposal concepts can be found e.g. in [83FiW], [93Wie], [95SSK], [80Clo] and [97Lie].

Table 3.5. Inventory of selected relevant radionuclides in Bq for a pressurized water reactor with a thermal power of 3.733 MW after an operation time of 1 day [95SSK].

	Time after discharge				
Nuclide	0 h	1 h	6 h	24 h	120 h
^{85}Kr	4.08×10^{13}	4.29×10^{13}	4.96×10^{13}	5.50×10^{13}	5.53×10^{13}
^{85m}Kr	1.43×10^{18}	1.24×10^{18}	5.72×10^{17}	3.53×10^{16}	1.25×10^{10}
^{87}Kr	2.86×10^{18}	1.68×10^{18}	1.10×10^{17}	6.02×10^{12}	0.0
^{88}Kr	4.03×10^{18}	3.16×10^{18}	9.32×10^{17}	1.15×10^{16}	7.60×10^{05}
^{133}Xe	2.89×10^{17}	3.10×10^{17}	4.04×10^{17}	6.15×10^{17}	5.82×10^{17}
^{135}Xe	1.74×10^{18}	2.09×10^{18}	2.91×10^{18}	1.72×10^{18}	2.04×10^{15}
Σ Kr-Xe	1.03×10^{19}	8.47×10^{18}	4.93×10^{18}	2.38×10^{18}	5.84×10^{17}
^{131}I	2.36×10^{17}	2.43×10^{17}	2.46×10^{17}	2.38×10^{17}	1.80×10^{17}
^{132}I	9.66×10^{17}	9.61×10^{17}	9.26×10^{17}	7.92×10^{17}	3.38×10^{17}
^{133}I	4.16×10^{18}	4.15×10^{18}	3.57×10^{18}	1.96×10^{18}	8.01×10^{16}
^{134}I	8.76×10^{18}	6.53×10^{18}	2.63×10^{17}	2.23×10^{11}	0.0
^{135}I	6.76×10^{18}	6.09×10^{18}	3.60×10^{18}	5.46×10^{17}	2.32×10^{13}
Σ Iodine	2.09×10^{19}	1.80×10^{19}	8.61×10^{18}	3.54×10^{18}	5.98×10^{17}
^{89}Sr	7.29×10^{16}	7.42×10^{16}	7.41×10^{16}	7.33×10^{16}	6.94×10^{16}
^{90}Sr	4.30×10^{14}	4.32×10^{14}	4.31×10^{14}	4.31×10^{14}	4.31×10^{14}
^{91}Sr	5.51×10^{18}	5.14×10^{18}	3.57×10^{18}	9.59×10^{17}	8.71×10^{14}
^{90}Y	1.08×10^{14}	1.12×10^{14}	1.29×10^{14}	1.83×10^{14}	3.44×10^{14}
^{91}Y	4.11×10^{16}	4.37×10^{16}	5.47×10^{16}	7.28×10^{16}	7.60×10^{16}
^{95}Zr	7.87×10^{16}	7.95×10^{16}	7.93×10^{16}	7.87×10^{16}	7.53×10^{16}
^{97}Zr	4.28×10^{18}	4.11×10^{18}	3.35×10^{18}	1.60×10^{18}	3.12×10^{16}
^{95}Nb	7.68×10^{14}	8.32×10^{14}	1.15×10^{15}	2.29×10^{15}	7.93×10^{15}
^{99}Mo	1.58×10^{18}	1.56×10^{18}	1.48×10^{18}	1.23×10^{18}	4.48×10^{17}
^{99m}Tc	9.38×10^{17}	9.86×10^{17}	1.14×10^{18}	1.15×10^{18}	4.32×10^{17}
^{103}Ru	6.80×10^{16}	6.81×10^{16}	6.78×10^{16}	6.69×10^{16}	6.24×10^{16}
^{105}Ru	1.40×10^{18}	1.24×10^{18}	5.67×10^{17}	3.41×10^{16}	1.05×10^{10}
^{106}Ru	1.22×10^{15}	1.22×10^{15}	1.22×10^{15}	1.22×10^{15}	1.21×10^{15}
^{105}Rh	4.10×10^{17}	4.28×10^{17}	4.68×10^{17}	3.81×10^{17}	5.88×10^{16}
^{127}Sb	2.27×10^{16}	2.32×10^{16}	2.36×10^{16}	2.09×10^{16}	1.02×10^{16}
^{129}Sb	7.54×10^{17}	6.53×10^{17}	2.93×10^{17}	1.63×10^{16}	3.32×10^{09}

	Time after discharge				
Nuclide	0 h	1 h	6 h	24 h	120 h
^{127}Te	1.07×10^{16}	1.13×10^{16}	1.41×10^{16}	1.77×10^{16}	9.85×10^{15}
^{127m}Te	1.07×10^{13}	1.16×10^{13}	1.59×10^{13}	3.06×10^{13}	8.18×10^{13}
^{129}Te	6.70×10^{17}	6.43×10^{17}	3.42×10^{17}	2.08×10^{16}	1.37×10^{15}
^{129m}Te	1.79×10^{15}	1.87×10^{15}	2.11×10^{15}	2.27×10^{15}	2.10×10^{15}
^{131m}Te	1.72×10^{17}	1.70×10^{17}	1.52×10^{17}	1.00×10^{17}	1.09×10^{16}
^{132}Te	9.48×10^{17}	9.42×10^{17}	9.01×10^{17}	7.68×10^{17}	3.28×10^{17}
^{134}Cs	3.67×10^{10}	3.77×10^{10}	4.02×10^{10}	4.13×10^{10}	4.12×10^{10}
^{136}Cs	2.99×10^{14}	2.98×10^{14}	2.95×10^{14}	2.83×10^{14}	2.29×10^{14}
^{137}Cs	4.56×10^{14}	4.58×10^{14}	4.58×10^{14}	4.58×10^{14}	4.58×10^{14}
^{140}Ba	3.83×10^{17}	3.82×10^{17}	3.78×10^{17}	3.63×10^{17}	2.92×10^{17}
^{140}La	7.20×10^{16}	7.73×10^{16}	1.02×10^{17}	1.74×10^{17}	2.90×10^{17}
^{141}Ce	1.07×10^{17}	1.13×10^{17}	1.30×10^{17}	1.40×10^{17}	1.29×10^{17}
^{143}Ce	2.66×10^{18}	2.65×10^{18}	2.39×10^{18}	1.63×10^{18}	2.18×10^{17}
^{144}Ce	1.52×10^{16}	1.52×10^{16}	1.52×10^{16}	1.52×10^{16}	1.50×10^{16}
^{143}Pr	7.23×10^{16}	7.78×10^{16}	1.04×10^{17}	1.74×10^{17}	2.68×10^{17}
^{239}Np	1.50×10^{19}	1.51×10^{19}	1.43×10^{19}	1.15×10^{19}	3.53×10^{18}
^{238}Pu	8.38×10^{06}	9.42×10^{06}	1.44×10^{07}	2.99×10^{07}	7.04×10^{07}
^{239}Pu	6.18×10^{11}	6.67×10^{11}	9.09×10^{11}	1.67×10^{12}	3.79×10^{12}
^{240}Pu	5.25×10^{09}	5.51×10^{09}	5.79×10^{09}	5.81×10^{09}	5.81×10^{09}
^{241}Pu	3.60×10^{09}	3.60×10^{09}	3.60×10^{09}	3.60×10^{09}	3.60×10^{09}
^{241}Am	3.87×10^{03}	4.53×10^{03}	7.82×10^{03}	1.97×10^{04}	8.28×10^{04}
^{242}Cm	2.15×10^{02}	2.67×10^{02}	4.95×10^{02}	1.01×10^{03}	1.41×10^{03}
^{244}Cm	9.62×10^{-04}	1.26×10^{-03}	2.46×10^{-03}	4.55×10^{-03}	5.40×10^{-03}
Σ Aerosols	3.52×10^{19}	3.46×10^{19}	3.00×10^{19}	2.05×10^{19}	6.37×10^{18}
Total	6.27×10^{20}	1.46×10^{20}	7.09×10^{19}	3.29×10^{19}	7.96×10^{18}

Table 3.6. Inventory of selected relevant radionuclides in Bq for a pressurized water reactor with a thermal power of 3.733 MW after an operation time of 333 days [95SSK]

	Time after discharge				
Nuclide	0 h	1 h	6 h	24 h	120 h
^{85}Kr	1.61×10^{16}	1.61×10^{16}	1.61×10^{16}	1.61×10^{16}	1.61×10^{16}
^{85m}Kr	1.19×10^{18}	1.04×10^{18}	4.78×10^{17}	2.95×10^{16}	1.05×10^{10}
^{87}Kr	2.27×10^{18}	1.33×10^{18}	8.72×10^{16}	4.78×10^{12}	0.0
^{88}Kr	3.22×10^{18}	2.53×10^{18}	7.45×10^{17}	9.19×10^{15}	6.08×10^{05}
^{133}Xe	7.62×10^{18}	7.62×10^{18}	7.60×10^{18}	7.35×10^{18}	4.75×10^{18}
^{135}Xe	1.83×10^{18}	2.20×10^{18}	3.07×10^{18}	1.82×10^{18}	2.16×10^{15}
Σ Kr-Xe	1.61×10^{19}	1.47×10^{19}	1.20×10^{19}	9.22×10^{18}	4.77×10^{18}
^{131}I	3.50×10^{18}	3.50×10^{18}	3.45×10^{18}	3.25×10^{18}	2.33×10^{18}
^{132}I	5.18×10^{18}	5.16×10^{18}	4.99×10^{18}	4.27×10^{18}	1.82×10^{18}
^{133}I	7.63×10^{18}	7.50×10^{18}	6.40×10^{18}	3.52×10^{18}	1.43×10^{17}
^{134}I	8.38×10^{18}	6.00×10^{18}	2.33×10^{17}	1.96×10^{11}	0.0
^{135}I	7.14×10^{18}	6.43×10^{18}	3.81×10^{18}	5.77×10^{17}	2.45×10^{13}
Σ Iodine	3.18×10^{19}	2.86×10^{19}	1.89×10^{19}	1.16×10^{19}	4.30×10^{18}

	Time after discharge				
Nuclide	0 h	1 h	6 h	24 h	120 h
^{89}Sr	4.49×10^{18}	4.48×10^{18}	4.47×10^{18}	4.43×10^{18}	4.19×10^{18}
^{90}Sr	1.26×10^{17}	1.26×10^{17}	1.26×10^{17}	1.26×10^{17}	1.26×10^{17}
^{91}Sr	5.42×10^{18}	5.05×10^{18}	3.50×10^{18}	9.42×10^{17}	8.55×10^{14}
^{90}Y	1.30×10^{17}	1.30×10^{17}	1.30×10^{17}	1.29×10^{17}	1.27×10^{17}
^{91}Y	5.58×10^{18}	5.57×10^{18}	5.57×10^{18}	5.54×10^{18}	5.29×10^{18}
^{95}Zr	6.70×10^{18}	6.70×10^{18}	6.68×10^{18}	6.63×10^{18}	6.35×10^{18}
^{97}Zr	6.52×10^{18}	6.26×10^{18}	5.10×10^{18}	2.44×10^{18}	4.75×10^{16}
^{95}Nb	6.55×10^{18}	6.55×10^{18}	6.55×10^{18}	6.55×10^{18}	6.55×10^{18}
^{99}Mo	6.91×10^{18}	6.84×10^{18}	6.49×10^{18}	5.37×10^{18}	1.96×10^{18}
^{99m}Tc	6.05×10^{18}	6.05×10^{18}	5.95×10^{18}	5.14×10^{18}	1.89×10^{18}
^{103}Ru	4.88×10^{18}	4.88×10^{18}	4.86×10^{18}	4.80×10^{18}	4.47×10^{18}
^{105}Ru	2.80×10^{18}	2.47×10^{18}	1.13×10^{18}	6.82×10^{16}	2.10×10^{10}
^{106}Ru	6.57×10^{17}	6.57×10^{17}	6.57×10^{17}	6.56×10^{17}	6.51×10^{17}
^{105}Rh	2.66×10^{18}	2.66×10^{18}	2.57×10^{18}	1.91×10^{18}	2.93×10^{17}
^{127}Sb	2.58×10^{17}	2.57×10^{17}	2.50×10^{17}	2.19×10^{17}	1.07×10^{17}
^{129}Sb	1.01×10^{18}	8.69×10^{17}	3.89×10^{17}	2.17×10^{16}	4.42×10^{09}
^{127}Te	2.50×10^{17}	2.50×10^{17}	2.49×10^{17}	2.32×10^{17}	1.30×10^{17}
^{127m}Te	2.80×10^{16}	2.80×10^{16}	2.80×10^{16}	2.80×10^{16}	2.79×10^{16}
^{129}Te	9.89×10^{17}	9.52×10^{17}	5.49×10^{17}	1.20×10^{17}	8.65×10^{16}
^{129m}Te	1.47×10^{17}	1.47×10^{17}	1.46×10^{17}	1.44×10^{17}	1.33×10^{17}
^{131m}Te	4.86×10^{17}	4.77×10^{17}	4.25×10^{17}	2.81×10^{17}	3.05×10^{16}
^{132}Te	5.12×10^{18}	5.08×10^{18}	4.86×10^{18}	4.14×10^{18}	1.77×10^{18}
^{134}Cs	8.26×10^{16}	8.26×10^{16}	8.26×10^{16}	8.25×10^{16}	8.22×10^{16}
^{136}Cs	6.90×10^{16}	6.89×10^{16}	6.81×10^{16}	6.55×10^{16}	5.30×10^{16}
^{137}Cs	1.51×10^{17}	1.51×10^{17}	1.51×10^{17}	1.51×10^{17}	1.51×10^{17}
^{140}Ba	6.87×10^{18}	6.86×10^{18}	6.78×10^{18}	6.51×10^{18}	5.24×10^{18}
^{140}La	6.95×10^{18}	6.94×10^{18}	6.93×10^{18}	6.85×10^{18}	5.91×10^{18}
^{141}Ce	6.45×10^{18}	6.45×10^{18}	6.44×10^{18}	6.35×10^{18}	5.83×10^{18}
^{143}Ce	6.18×10^{18}	6.09×10^{18}	5.49×10^{18}	3.76×10^{18}	5.01×10^{17}
^{144}Ce	3.25×10^{18}	3.25×10^{18}	3.25×10^{18}	3.25×10^{18}	3.21×10^{18}
^{143}Pr	6.10×10^{18}	6.10×10^{18}	6.10×10^{18}	6.04×10^{18}	5.21×10^{18}
^{239}Np	6.25×10^{19}	6.21×10^{19}	5.85×10^{19}	4.69×10^{19}	1.45×10^{19}
^{238}Pu	5.76×10^{14}	5.76×10^{14}	5.77×10^{14}	5.80×10^{14}	5.88×10^{14}
^{239}Pu	8.83×10^{14}	8.84×10^{14}	8.85×10^{14}	8.88×10^{14}	8.96×10^{14}
^{240}Pu	5.91×10^{14}	5.91×10^{14}	5.91×10^{14}	5.91×10^{14}	5.91×10^{14}
^{241}Pu	1.10×10^{17}	1.10×10^{17}	1.10×10^{17}	1.10×10^{17}	1.10×10^{17}
^{241}Am	4.16×10^{13}	4.16×10^{13}	4.17×10^{13}	4.21×10^{13}	4.40×10^{13}
^{242}Cm	4.73×10^{15}	4.74×10^{15}	4.75×10^{15}	4.76×10^{15}	4.71×10^{15}
^{244}Cm	2.92×10^{13}	2.92×10^{13}	2.93×10^{13}	2.94×10^{13}	2.95×10^{13}
Σ Aerosols	1.67×10^{20}	1.65×10^{20}	1.55×10^{20}	1.30×10^{20}	7.50×10^{19}
Total	7.75×10^{20}	3.12×10^{20}	2.33×10^{20}	1.75×10^{20}	9.69×10^{19}

3.6.6 Release of radionuclides from the radioactive inventory of a nuclear reactor

In case of an accident of a less dangerous category mainly gaseous or volatile fission or decay products are released. Heavier elements or compounds are more or less retained, since they are hardly transported over longer distances. Consequently, the respective risk assessment studies show the highest risk potentials for the gaseous fission or decay products like: noble gases, iodine, radon, volatile elements and compounds (cf. e.g. [95SSK]).

Furthermore, it has to be taken into account that the chemical and physical behaviour of many fission products may change considerably after nuclear transformations within their radioactive decay chains (e.g.: Ra $\rightarrow$ Rn). This often leads to consequences for the retention behaviour, release of and filter effectiveness for various nuclides in the radioactive inventory.

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