RADIOCHEMISTRY

EXTENDED LECTURE NOTES

Contents

R1. The atomic nucleus
- R1.1. Classification of the nuclides

R2. Radioactivity
- R2.1. Types of the radioactive decay
  - Isomeric transition
  - Beta (β) decays
    - β^- decay
    - β^+ decay
    - Electron capture
  - Alpha (α) decay
  - Spontaneous fission

R3. Kinetics of the radioactive decay
- R3.1. Simple decay
- R3.2 Decay chains

R4. Interaction of radiation with matter
- R4.1. Classification of the particles
- R4.2. Potential partners during the interactions
- R4.3. Mechanisms
- R4.4. Interaction of ionizing radiation with matter
  - Quantitative description of the interaction
  - Ionisation interactions of alpha radiation
  - Ionisation interactions of beta radiation (electron or positron)
  - Ionisation interactions of gamma radiation
    - Compton scattering
    - Photoelectric effect
    - Pair production
  - Neutrons
- R4.5. Nuclear reactions
  - Kinetics
  - Neutron interactions
    - (n,f) fission

R5. Nuclear instruments and measurements
- R5.1. Nuclear radiation detectors
  - Gas ionization detectors
    - Geiger-Müller (GM)-counters
  - Semiconductor detectors
  - Scintillation detectors

References

The goal of this chapter is to understand the nuclear forces acting in the nucleus of the atoms, the kinds and source of nuclear radiations. Moreover, their interactions with their surroundings and their various applications including radioactive labelling, source of power, etc.

R1. The atomic nucleus

The **nucleus** is the very dense region consisting of protons and neutrons at the centre of an atom. These particles are further composed of subatomic fundamental particles known as quarks bound by the strong interaction. From the point of radiochemistry: protons and neutrons. The size of an atom, the nucleus as well as that of the protons and neutrons are illustrated in Figure R1.

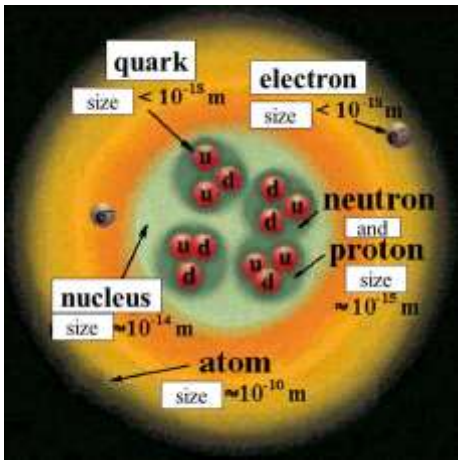


Figure R1. The main building elements of the nucleus
(after http://astronomyonline.org/Science/Images/Mathematics/AtomicStructureSmall.jpg)

Protons and neutrons are nucleons. Their number is given by Z and N, respectively. The mass of an atom is determined by the number of the protons and neutrons, therefore, their total (A) is also called mass number, atomic mass number or nucleon number.

The mass number (A), also called atomic mass number or nucleon number, is the total number of protons (Z) and neutrons (N), together known as nucleons in an atomic nucleus:

$$A=Z+N$$

Z is also called as the atomic number because the number of protons in a nucleus uniquely identifies an element.

Information concerning the properties of the nucleus are written as indices on the left side of the element symbol (X) in the following way: ${}^{A}_{Z}\mathrm{X}$. As each element is defined by its atomic number, *Z* is often omitted.

The basic properties of these particles are compared to the electrons orbiting around the nucleus are listed in Table R1. According to Einstein's mass–energy equivalence equation:

$$E=mc^2$$

where E is energy, m is mass, and c is the speed of light (299,792,458 m/s). Thus, the mass of these particles also can be given in energy units.

Table R1. Fundamental properties of protons and neutrons

particle	symbol	charge*	mass, g	energy, MeV**
proton	p	+1	1.6726×10^{-24}	938.27
neutron	n	0	1.6749×10^{-24}	939.55
electron	e^-	-1	9.109×10^{-28}	0.51

*The elementary charge is $1.602176565\times10^{-19}$ C
**mega electron volt = 10^6 electron volt (symbol eV). eV is a unit of energy equal to approximately 1.6×10^{-19} J. It is the amount of energy gained (or lost) by the charge of a single electron moved across an electric potential difference of one volt.

The total electrical charge of the nucleus which is spread fairly uniformly within the nucleus, is defined by the number of protons. Protons unlike neutrons are stable particles:

$$n \rightarrow p + e^- + 0.8 \text{ MeV}$$

The decay of neutrons is an exothermic process. You may easily follow the energy (or mass) balance of the process.

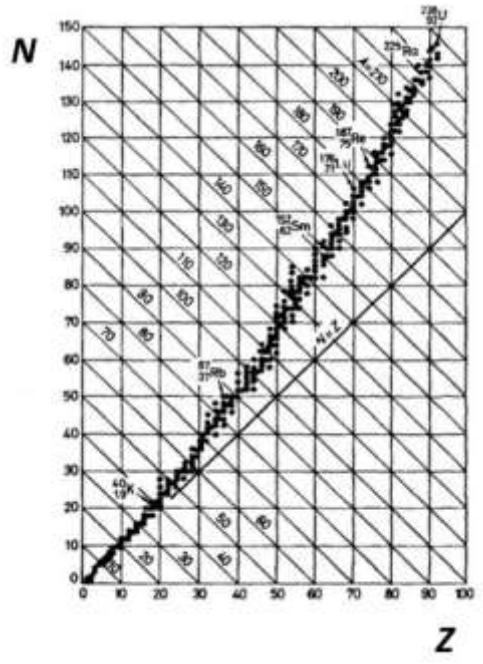


Figure R2. Comparison of the Z and N values of the stable nuclides

An atomic species is characterized by the specific constitution of its nucleus, i.e., by its number of protons *Z*, its number of neutrons *N*, and its nuclear energy state are called nuclide. The total of the mass of the protons and neutrons is less than the mass of a nuclide. Thus, the nuclear force can be calculated from this mass defect Δm:

$$\Delta m = (Z \cdot m_p + N \cdot m_n) - (\text{measured mass of nucleus})$$

which can be easily converted into the binding energy using the mass–energy equivalence equation. Figure R3 shows the change of the specific binding energy, i.e., the ratio of the binding energy and the mass number as a function of the mass number.

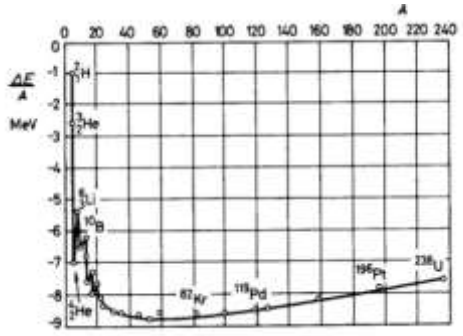


Figure R3. The change of the specific binding energy with the mass number

Figure R3 shows the energy being released when a nuclide is formed. The same energy has to be invested to break up a nuclide into its building blocks. Features:

- asymmetric curve
- a flat minimum around A≈60: most stable nuclides
- at low A: steep drop →fusion of small nuclei results in a large energy release; technically difficult to handle (H-bomb)
- at high A: shallow increase: fission of a nuclide with a large A into smaller ones release only a limited energy, but technically controllable
- note the extremely large binding energy of ^4_2He.

R1.1. Classification of the nuclides

Isotopes are the elements having same atomic number but different mass number. They have the same atomic number because the number of protons inside their nuclei remains the same. The difference in their mass number is due to the difference in their number of neutrons. Thus, isotopes are chemically same and physically different.

Isobars are atoms of different elements having the same atomic mass but different atomic number. Since their number of electrons is different, their chemical properties are different. but physically same.

Isotones are elements having the same number of neutrons.

Although isotopes are chemically identical (Z is the same) but due to their different mass they may show different behaviour from kinetic, spectroscopic, etc., aspects: **isotope effect**. An isotopic substitution can modify e.g., the boiling point or the reaction rate when the isotopic replacement is in a chemical bond that is broken or formed.

Applications of the isotope effect:

IR spectroscopy

MS

NMR

contrast matching e.g., in neutron scattering

enrichement/separation

compound specific isotope analysis (CSIA)

Radioactive vs. stable isotopes

R2. Radioactivity

or radioactive decay: a spontaneous transformation of the unstable nucleus. The properties of the nucleus change in time and energy is lost. All the conservation laws are met. A material that spontaneously emits this kind of radiation - which includes the emission of energetic alpha particles, beta particles, and/or gamma photons - is considered radioactive. Radioactive decay is a spontaneous process that cannot be influenced from the exterior and that leads to the formation of new nuclei from the originally existing unstable ones.

R2.1. Types of the radioactive decay

The most important types of decay are discussed here.

As it is said above during the radioactive decay the properties of the nucleus spontaneously change. This may involve the change of Z, A and/or N.

Isomeric transition

or gamma (γ) decay. Decay of excited metastable nuclear isomers, where the energy is emitted in form of high energy photons released from the nucleus, i.e., gamma (γ-photons).

Scheme:

$$^{A*}_{Z}X \rightarrow {}^{A}_{Z}X + \gamma$$

* or occasionally m stands for showing the metastability. Note that no nuclear transmutation (creation of an atom of a new element) occurs.

The emitted energy is equal to the energy of the gamma photon:

$$\Delta E = h\nu,$$

therefore, the energy distribution function of the photons (spectrum) consist of discrete lines with well defined energy (Figure R4) . The emitted energy is characteristic to the emitting nucleus. Examples are given in Table R2.

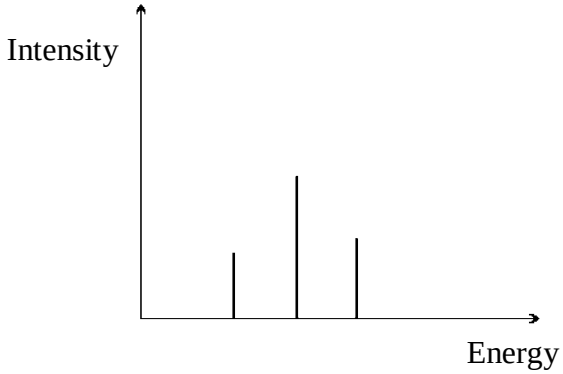


Figure R4. Typical (differential) nuclear line spectrum

Table R2. Isomeric transition, examples

nuclide	Half life*, $T_{1/2}$	E_{γ}, MeV
^{60m}Co	10.5 min	0.059
^{99m}Tc	6.0 h	0.143

* see later

Beta (β) decay

The most frequent type of decay. Almost all the elements throughout the periodic table have instable isotope(s) showing beta decay.

A family of 3 different decays, but common properties: isobaric transition ($\Delta A=0$, $\Delta Z=\pm 1$, neutrino, ν or antineutrino, $\tilde{\nu}$ is emitted concurrently)

β^--decay

Scheme:

$$^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y + \beta^- (\equiv e^-) + \tilde{\nu} + [\gamma]$$

[γ] means optional gamma radiation. Note that the transmutation is also noted with a new symbol for the element. $\tilde{\nu}$ is the symbol for antineutrino (see later).

The elementary process: $n \rightarrow p + \beta^- + \tilde{\nu}$. Exothermic. Note the conservation of mass and charge. Examples are given in Tables R3 and R4.

Table R3. Pure β^- emitters

nuclide	β energy, MeV	$T_{1/2}$
^{3}H	0.018	12.26 y
^{14}C	0.159	5730 y
^{32}P	1.71	14.3 d
^{35}S	0.167	88 d
^{90}Sr	0.54	28.1 y
^{90}Y	2.25	64 h

^{3}H, ^{14}C, ^{35}S have low beta energy. Such nuclides are called soft beta emitter and need a special detection technique.

Table R4. Mixed β^- - γ emitters

nuclide	$T_{1/2}$	β-energy, MeV	γ-energy, MeV
^{60}Co	5.27 y	0.31	1.17; 1.33
^{131}I	8.07 d	0.61	0.36
^{137}Cs	30.23 y	0.51	0.662

β^+-decay

Scheme:

$$^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + \beta^+ (\equiv e^+) + \nu + [\gamma]$$

β^+ is the symbol of positron, ν is the symbol for neutrino (see later).

The elementary process: $p \rightarrow n + \beta^+ + \nu$. Endothermic. Examples are given in Table R5.

Table R5 Positron emitters

nuclide	$T_{1/2}$	$E_{positron}$, MeV
^{11}C	20.3 min	0.97
^{13}N	9.97 min	1.2
^{15}O	124 s	1,7
^{18}F	109.7 min	0.064

Neutrino and antineutrino are very important particles for physicists, but they are neglected in radiochemistry due to their negligible interaction with matter.

Electron and positron share the same mass and elementary charge (see earlier), unless electron is negative (-1) and positron is positive (+1). Due to the latter, when it is emitted, it can penetrate in the matter as long as its excess energy compared to its environment is lost (due to collisions and continuous electromagnetic radiation called bremsstrahlung). Such positrons are easily captured by the any electron found in the matter:

$$e^- + e^+ = 2\gamma$$

The process is called positron annihilation. The energy of each gamma photon is 0.51 MeV and they move in opposite (180 °) direction. This process is the basis of the positron emission tomography and can be used in porosity measurements.

Electron capture

Scheme:

$$e^- + {}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y^{*+} + \nu + [\gamma]$$

The elementary process: $p + e^- \rightarrow n + \nu$. The electron is captured most often from the K shell. The process is endothermic but less than the positron decay. The product Y is an instable ion, as the electron vacancy is in an internal (most often K) shell. A cascade reorganisation of the electrons (from the outer shells to the internal ones) will stabilise the system. This process results in the emission of photons. The energy of these photons is determined by the difference of the energy levels of the atomic electron levels and falls in the X-ray range of the spectrum of the electromagnetic radiations. As the energy levels are defined by the emitting atom (and thus it can be used for its identification), the name of this radiation is characteristic X-ray. That is, electron capture is always followed by the emission of X_{char}. Examples are given in Table R6.

Table R6. Electron capture (EX) isotopes

Nuclide	$T_{1/2}$	E_γ MeV
^{54}Mn	303 d	0.84
^{125}I	60 d	0.035

The electron/positron spectrum is continuous (Figure R5), as the two particles concurrently emitted (electron + antineutrino or positron + neutrino) share the quantized energy of the nuclear energy transition.

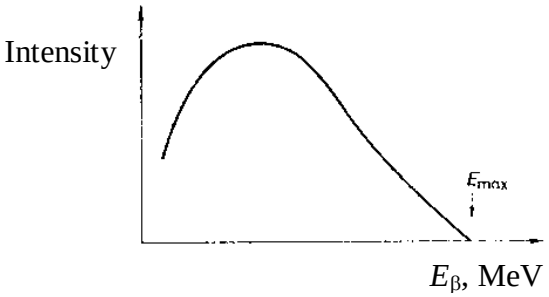


Figure R5. Typical beta spectrum

Alpha (α) decay

Recall the outstandingly high binding energy of $^4_2\mathrm{He}$.

$$^A_Z X \rightarrow {}^{A-4}_{Z-2}Y + \alpha(\equiv {}^4_2\mathrm{He}^{2+}) + [\gamma]$$

[γ] means optional gamma radiation. Such decay is typical for nuclides of high N/Z ratio Examples are given in Table R6. The energy of the alpha particle is 4-9 MeV: heavy particle (four nucleons) with high energy. The spectrum is line spectrum (see Figure R4.)

Table R6. Examples for alpha decay

nuclide	$T_{1/2}$
^{235}U	7.1E8 y
^{226}Ra	1600 y
^{222}Rn	3.8 d

Figure R7 shows that there are radionuclides which may decay in several ways

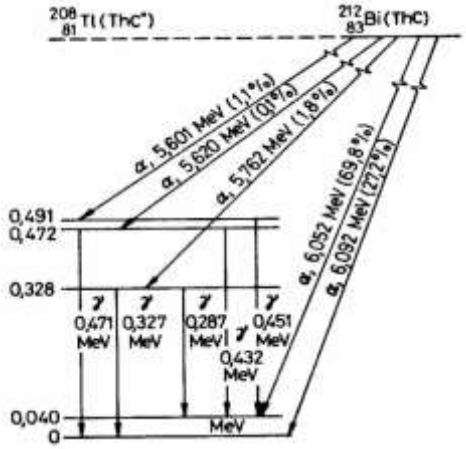


Figure R7. Alternative decay paths of $^{212}_{83}$Bi

Spontaneous fission

As discussed earlier the heavy nuclides at the end of the periodic table are less stable. Although they have a large N/Z ration, as an alternative of the alpha decay they may split into smaller nuclides in order to get into an energetically favourable state. This process is called spontaneous fission. As an example: In each g of a natural uranium 25 nucleus suffer fission within an hour, while the number of uranium nuclides involved in alpha decay is ca. $45 \cdot 10^6$. Remember: the disintegration of a single radioactive nuclide may produce more than one elementary particle or gamma photon.

R3. Kinetics of the radioactive decay

Radioactive decay is a stochastic (i.e., random) process at the level of single atoms, in that, it is impossible to predict when a particular atom will decay. However, the chance that a given atom will decay is constant over time. For a large number of atoms, the decay rate for a ***macroscopic*** amount is computable.

R3.1. Simple decay

The description is similar to a first order chemical reaction: N is the number of radioactive nucleus present, t is time. The rate of the decay is characterized by the activity, A and defined as:

$$A \equiv -\frac{dN}{dt}$$

$$A = \lambda N$$

λ here is the decay constant and in spite of the analogy with the rate factor in chemistry, it cannot be influenced by either temperature or pressure. Integration in the 0 - *t* interval (Figure R8) provides

$$N = N_0 e^{-\lambda t}$$
$$A = A_0 e^{-\lambda t}$$

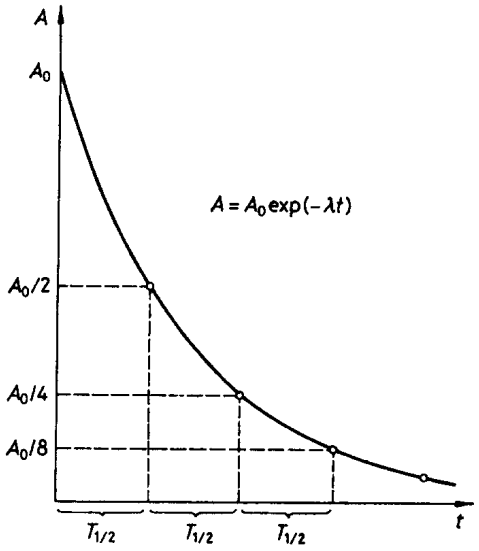


Figure R8. The simple radioactive decay can be described with an exponential function

The semi-logarithmic plot of both the $N(t)$ and $A(t)$ functions therefore will provide a straight line with a negative slope (-λ) and the intercept is $\ln N_0$ and $\ln A_0$, respectively:

$$\ln N = \ln N_0 - \lambda t$$
$$\ln A = \ln A_0 - \lambda t$$

Instead of the decay constant half-life (time needed for the decay of the initial number of nuclides) is more often used:

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

$T_{1/2}$ can range from nearly instantaneous to as much as 10^{19} years or more.

The unit of activity is becquerel. A source has an activity of one becquerel if one disintegration takes place in it in one second:

$$\frac{1\ \text{disintegration}}{\text{sec}} = 1\ \text{becquerel} = 1\ \text{Bq}$$

The former, traditional unit was curie. This unit has a historical background. It corresponds to the activity of 1 g uranium. $1\ \text{Ci} = 3.7\times10^{10}\ \text{Bq}$.

It should be noted that radioactivity can be followed only through the interaction of the emitted particles (alpha, beta, gamma, etc.) with the matter (the detector). Therefore, we strictly have to distinguish the measured quantity from the radioactivity or shortly activity. If the number of signals (pulses) collected within time t is n, we can define the counting intensity $I = \frac{n}{t}$. The unit of I is counts/min (cpm) or counts/s (cps) and should be strictly distinguished from activity A measured in Bq.

$$I = k\eta A$$

η is the overall efficiency of the detection and k is the number of particles sensed by the detector from the nuclear decay.

R3.2. Decay chains

Most radioactive elements do not decay directly to a stable state, but rather undergo a series of decays until eventually a stable isotope is reached. The radioactive decay of different discrete radioactive decay products as a chained series of transformations is called decay chain. Its members are referred to by their relationship. A parent isotope is one that undergoes decay to form a daughter isotope. The daughter isotope may be stable or it may decay to form a daughter isotope of its own. The daughter of a daughter isotope is sometimes called a granddaughter isotope. Let's suppose the following decay chain:

$$X \xrightarrow[T_{1/2,X}]{\lambda_X} \rightarrow Y \xrightarrow[T_{1/2,Y}]{\lambda_Y} \rightarrow Z_{stable}$$

The activity of X can be calculated as shown above. The activity of Y is

$$A_Y = \lambda_Y N_Y = A_{X,0}\frac{\lambda_Y}{\lambda_Y - \lambda_X}\left(\mathrm{e}^{-\lambda_X t} - \mathrm{e}^{-\lambda_Y t}\right),$$

i.e., its activity depends on the initial activity of the parent isotope X and on the ratio of the decay constants of the parent and the daughter isotopes.

Four main decay chains exist in nature. The so called the thorium series, the radium series, and the actinium series after the long-lived starting isotopes of these three isotopes, respectively thorium-232 ($^{232}_{90}\text{Th}$), uranium-238 ($^{238}_{92}\text{U}$), and uranium-235 ($^{235}_{92}\text{U}$). All of them end in three different, stable isotopes of lead. These so called decay chains have existed since

the formation of the earth. The fourth chain, the neptunium series ($A = 4n + 1$), due to the quite short half-life of its starting isotope neptunium-237 ($^{237}_{93}\mathrm{Np}$, 2.14×10^{-06} years), is already extinct in nature, except for the final rate-limiting step, decay of bismuth-209. The ending isotope of this chain is thallium-205. Some older sources give the final isotope as bismuth-209, but it was recently discovered that it is radioactive, with a half-life of 1.9×10^{19} years.

As an example, we will consider two decay chains of environmental relevance. Note, that the half-life of the parent is several order of magnitude longer than that of the daughters, i.e., $T_{1/2,X}>>T_{1/2,Y}$.

1) $^{90}\mathrm{Sr} \xrightarrow[28\,a]{\beta^-} {}^{90}\mathrm{Y} \xrightarrow[64\,h]{\beta^-} {}^{90}\mathrm{Zr}$

Consider the chemical nature of Sr and the role of ca int he body.

2) $^{226}_{88}\mathrm{Ra} \xrightarrow[1620\,a]{\alpha} {}^{222}_{86}\mathrm{Rn} \xrightarrow[3,83\,d]{\alpha} \ldots {}_{82}\mathrm{Pb}$

Consider that Ra is solid, Rn is a noble gas (mobility!!!) and Pb is a heavy metal.

R4. Interaction of radiation with matter

R4.1. Classification of the particles

Table R7. Classification of the particles according their mass and charge

I. mass and charge		II. mass, no charge	III. no mass, no charge
a	b		
may induce nuclear reaction	no nuclear reaction		
p	e^+	n	γ
α	e^-		X-ray

Recall the mass and the nature of these particles (see above the mass and energy represented by them as well as their charge).

R4.2. Potential partners during the interactions

1. Electromagnetic field
2. Electron
3. Field of the nucleus
4. Nucleus

R4.3. Mechanisms

Mechanism	Effect in the radiation	Effect in the matter
absorption	change in the intensity and the energy	kinetic energy + excitation
coherent scattering (only the direction of the radiation)	change in the intensity	-
incoherent scattering i) elastic ii) inelastic	change in the intensity and the energy	kinetic energy kinetic energy +excitation

R4.4. Interaction of ionizing radiation with matter

The first step of the ionizing radiation in the matter:

1. Neutral excitation

$A + \text{radiation} \rightarrow A^* + \text{radiation'}$

2. External ionization

$A + \text{radiation} \rightarrow A^+ + e^- + \text{radiation'}$

$A_2 + \text{radiation} \rightarrow A^+ + A^- + \text{radiation'}$

$A_2 + \text{radiation} \rightarrow A_2^+ + e^- + \text{radiation'}$

$A_2 + \text{radiation} \rightarrow 2\,A\cdot + \text{radiation'}$

3. Internal ionization

$A + \text{radiation} \rightarrow A^{*+} + e^- + \text{radiation'}$

$A^{*+} \rightarrow A^+ + X_{char}$

$A^{*+} \rightarrow A^{2+} + e^-$ (Auger)

X_{char} is the characteristic X-ray radiation, characterizing the atom A. Its energy is quantized.

4. Bremsstrahlung = "braking radiation" or "deceleration radiation" (only with charged particles)

$$A + \text{radiation}^{\pm} \rightarrow A + X_b + \text{radiation}^{\pm'}$$

X_b is the braking X ray radiation with a continuous spectrum.

These interactions are the basis of the detection of the nuclear radiations.

Quantitative description of the interaction

A typical set-up for the detection of nuclear radiation is shown in Figure R9.

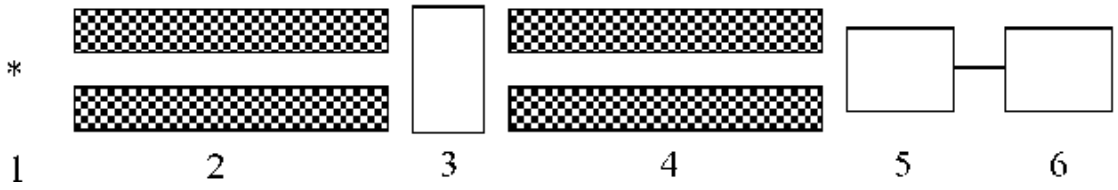


Figure R9. Set-up for detection of radioactive radiation. 1: point-source, 2 and 4: collimators (to obtain parallel radiation), 3: the absorbing material (thickness x, atomic density ρ_A and macroscopic density ρ), 5: detector, 6: data processor

The number of the particles dn absorbed within a distance dx a can be defined as

$$-dn = \sigma(E) n \rho_A dx$$

where n is the number of particles passing through the matter, ρ_A is the atomic density of the matter and σ is the nuclear cross section. The nuclear cross section of a nucleus is used to characterize the probability that a nuclear process will occur. The concept of a nuclear cross section can be quantified physically in terms of "characteristic area" where a larger area means a larger probability of interaction. The SI unit of nuclear cross section is m². Very often it is expressed in barn, which is equal to 10^{-28} m² or 10^{-24} cm² (compare to the area of a single nucleus). The latter strongly depends on the energy of the entering particle. If n_0 is the number of the entering particles (at x=0) then the

$$n = n_0 e^{-\sigma(E)\rho_A x}$$

The intensity of these particles can be defined as

$$\frac{n}{t} = I$$

and thus

$$I = I_0 e^{-\sigma(E)\rho_A x} = I_0 e^{-\mu x}$$

which states that the intensity of the radiation decreases from the original value of I_0 to a value of I after proceeding through a layer of the absorbent of thickness x. $\mu = \sigma(E)\rho_A$ is the so called linear absorption coefficient (unit: m, cm, mm, etc.), which strongly depends on the energy E of the nuclear radiation. In practice, the use of mass-adsorption coefficient μ_m is more frequent. The two constants can be converted easily to each other knowing the density of the absorbent, ρ: $\mu_m = \frac{\mu}{\rho}$. The absorption of radioactive radiation in matter can be thus calculated by the expression

$$I = I_0 e^{-\mu_m d}$$

i.e., the intensity of the radiation decreases from the original value of I_0 to a value of I after proceeding through a layer of the absorbent of surface mass $d=\rho x$ (unit: kg/m^2 or g/cm^2, etc.). The thickness of the absorbing layer which reduces any I to its half can be calculated as

$$x_{1/2} = \frac{ln2}{\mu} \text{ or } d_{1/2} = \frac{ln2}{\mu_m}$$

Ionizing particles or gamma photons bring about ion-pairs along their track when penetrating into the medium. The number of ion-pairs related to unit path-length is called specific ionization. The energy delivered to (or left in) the medium in unit path-length is called linear energy transfer (LET) and is given in eV/nm (keV/μm) (Figure R10.). In a given medium LET depends on the energy ($\frac{dE}{dx} \approx \frac{1}{E}$) and the mass of the particles.

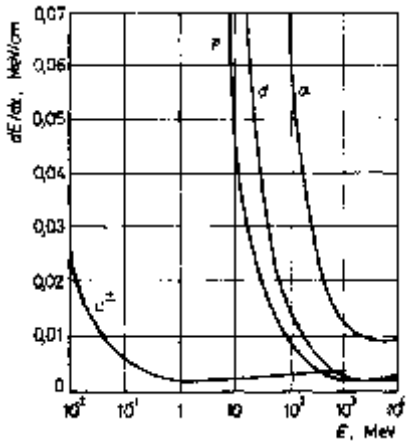


Figure R10. Linear energy transfer (LET) of various particles in air

The efficiency of the absorption also depends on the material. A qualitative comparison is shown in Figure R11.

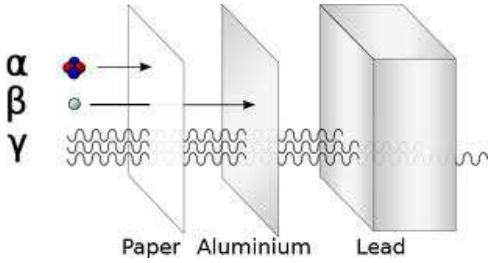


Figure R11. Comparison of the *penetration potential* of the three main nuclear radiations (http://hyperphysics.phy-astr.gsu.edu/hbase/nuclear/imgnuc/radpen.gif)

Ionisation interactions of alpha radiation

Charged (2+) and heavy particles (4 nucleons)

Interactions i) with the electrons incoherent scattering

ionization and excitation of the matter ca. 50-50 %

the energy and the direction of the alpha particles will change

ii) with the nucleus

see the classical Rutherford experiment

nuclear reaction (see later)

iii) interaction with the electromagnetic field: Charged particle: bremsstrahlung

The path length of the alpha radiation is short and strongly depends on the density of the exposed material (Figure R12.)

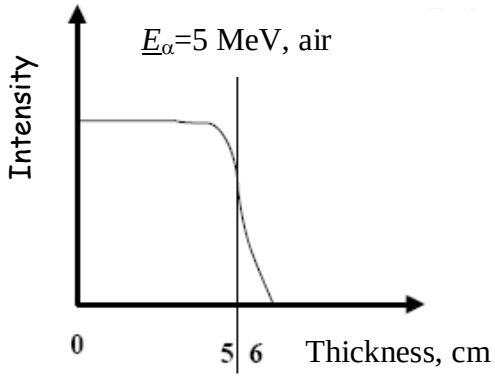


Figure R12. The path length of the alpha particles

Ionisation interactions of beta radiation (electron or positron)

charged, small (light)

With the electrons: incoherent scattering; ionisation (external and internal) + excitation

Change in energy and direction

With the nuclear field: incoherent scattering

charged particles: bremsstrahlung.

The ratio of the energy released in form of X-ray (r) and ionisation (ion) can be estimated as

$$\frac{\left(\frac{d\underline{\underline{E}}}{dx}\right)_r}{\left(\frac{d\underline{\underline{E}}}{dx}\right)_{ion}} = \frac{\underline{\underline{EZ}}}{800}$$

where E is the beta energy. Due to the continuous beta-spectrum, its intensity reduces exponentially throughout the material during the interaction. A comparison of beta radiation and mono-energetic electron radiation is given in Figure R13.

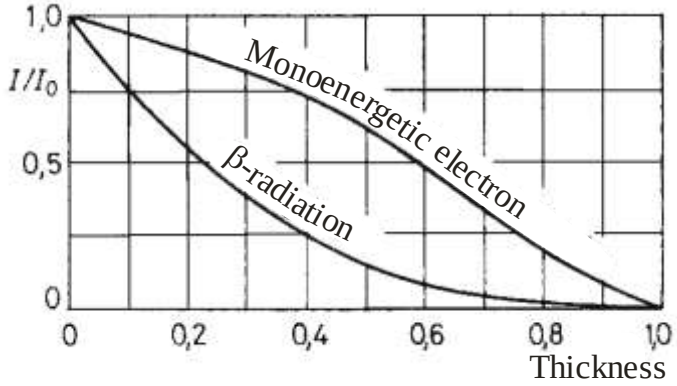


Figure R13.Comparison of the absorption properties of monoenergetic electron- and beta radiations

The attenuation of beta-radiation can be given as $\frac{I}{I_0} = e^{-\mu x}$ (see later).

Remember: β^- and β^+ are similar except the positron's annihilation after being thermalized. Electron–positron annihilation occurs when an electron (e−) and a positron (e+, the antiparticle of the electrone) collide. The result of the collision is the annihilation of both the electron and positron, and the creation of two gamma photons:

$$e- + e+ \rightarrow \gamma + \gamma$$

The process must satisfy a number of conservation laws, including the conservation of energy ($\approx$ mass), electric charge (the net charge before and after the annihilation is zero), etc.

Therefore, the energy of each photon is 0.51 MeV

Ionisation interactions of gamma radiation

no mass, no electric charge; high energy electromagnetic radiation

The most important mechanisms:

Compton scattering

Compton scattering is an inelastic scattering of a gamma photon by an electron. It results in a decrease in energy (decrease in frequency) of the Part of the energy of the photon is transferred to the scattering electron. Practically independent on Z.

Photoelectric effect

Electrons can absorb energy from gamma photons when irradiated. All of the energy from one photon must be absorbed and used to liberate one electron from atomic binding, or else the energy is re-emitted. If the photon energy is absorbed, some of the energy liberates the electron from the atom, and the rest contributes to the electron's kinetic energy as a free particle. This leads to the internal ionisation of the atom. Therefore, photoelectric effect is always succeeded by characteristic X-ray radiation (occasionally by the emission of an Auger electron, depending on energy and Z):

$$A + \gamma \longrightarrow A^{*+} + e^-_{\mathrm{f}}$$

$$A^{*+} \longrightarrow A^{+} + X_{\mathrm{k}}$$

$$A^{*+} \searrow A^{2+} + e^-_{\mathrm{A}}$$

Depends on $Z^{4\text{-}5}$.

Pair production

This process occurs when a high-energy photon interacts with a nucleus. The energy of this photon can be converted into the mass of an electron plus a positron. The photon must have enough energy to create the mass. The energy corresponding to the rest mass of an electron is 0.511 MeV, the same as a positron, i.e., a threshold energy of 1.02 MeV is needed!!! Depends on $Z^{\sim 2}$.

As an overall result

$$I = I_0 e^{-\mu d} = I_0 e^{-(\mu_C + \mu_f + \mu_p)d}$$

where C, f and p refers to Compton, photoelectric and pair production, respectively.

The energy dependence of the mass absorption coefficient is illustrated in gamma photon – germanium interactions in Figure R14. At low energy ($E_\gamma < \sim 0.2$ MeV) photoelectric effect,

in 0.2 – 2 MeV Compton scattering and above 2 MeV pair production has the highest probability.

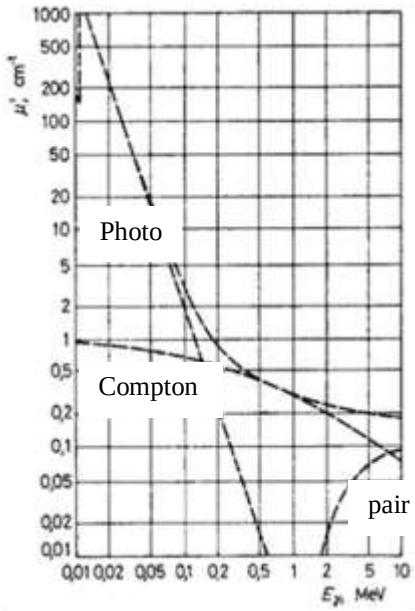


Figure R14. The interaction of gamma photons with germanium.

The *E* and *Z* dependence of the gamma interaction are summarized in Figure R15.

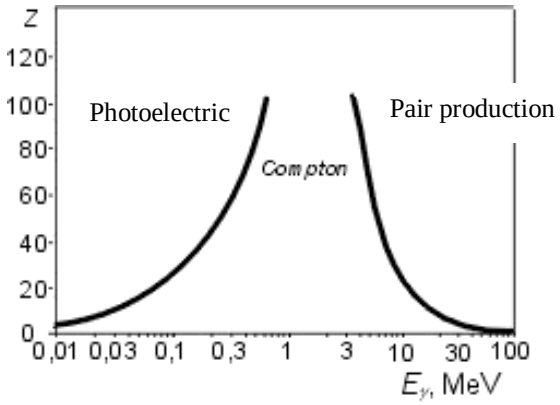


Figure R15. The strong *Z* and energy dependence of the gamma interactions

Neutrons

No direct ionization occurs.

R4.5. Nuclear reactions

The transient nucleus

$^{10}B + \alpha \rightarrow$ $\rightarrow {}^{10}B + \alpha$

$^{14}N^{*}$ $\rightarrow {}^{13}C + p$

$^{12}C + d \rightarrow$ $\rightarrow {}^{13}N + n$

The reaction equation, in a way analogous to a chemical equation, one may in addition give the reaction energy *Q* on the right side:

$$X + a \rightarrow Y + b\ (+Q)$$

The short form: X (a,b) Y

b is for the prompt particle formed in the process. The particles emitted due to the radioactive nature of the product nucleus is not shown in these equations.

All the usual conservation laws are met.

There are two main types of rnergy dependence of the cross section of nuclear reactions (Figure R16.)

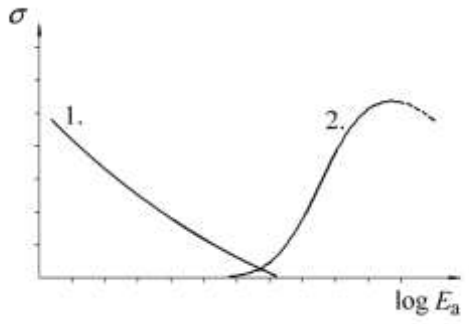


Figure R16. The two types of energy dependence of the probability of a nuclear reaction

Nuclear reactions of energy dependence 1 and 2:

1. (n,γ)
   (n,f) ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu
   $^{10}B(n,\alpha)$
   $^{6}Li(n,\alpha)$ tunnel effect

2. (γ,n)
   (n,2n)
   (n,α)
   (p,)
   (d,)

Kinetics

$$\frac{dN^*}{dt} = \sigma_a N\phi - \lambda N^*$$

$$N^* = N^*_\infty \left[1 - \exp(-\lambda t)\right]$$

$$A = A_\infty \left[1 - \exp(-\lambda t)\right]$$

$$A_\infty = \lambda N^*_\infty = \phi \sigma_a N$$

here N is the number of target nuclei, N* is the number of activated nuclei, ϕ is the particle flux (i.e., the rate of particle flow per unit area, which has the dimensions $\frac{\text{particle}}{\text{time} \cdot \text{area}}$, e.g., $\frac{1}{\text{s} \cdot \text{cm}^2}$), σ_a is the cross section of the activation, λ is the decay constant of the radioactive nuclide produced and *t* is the duration of the irradiation (Figure R17.)

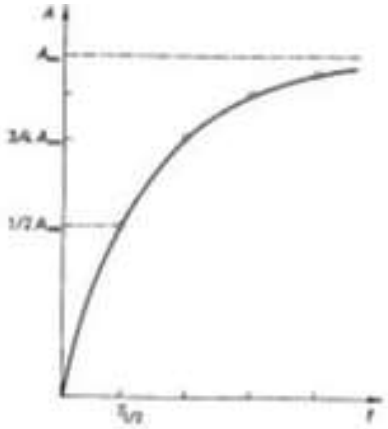


Figure R17.Time dependence of the activation

When the irradiation is stopped after time *t*, the radioactive nuclid obtaind continues to decay, a process sometimes called “cooling”:

$$A' = \lambda N^* =$$

$$= A_\infty \left[1 - \exp(-\lambda t)\right] \exp\left(-\lambda t_h\right)$$

Here t_h is the time of “cooling” (Figure R18.).

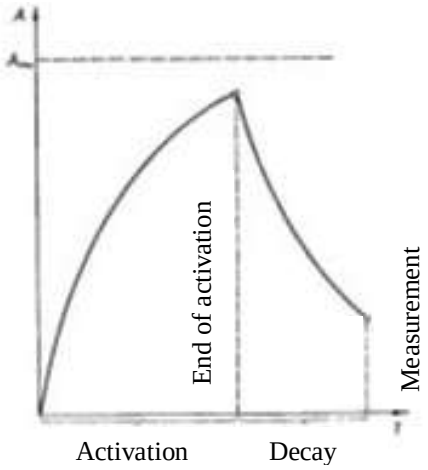


Figure R18. The change of the activity during the irradiation for time t and the cooling time t_h

Neutron interactions

elastic scattering

inelastic scattering (excited nucleus, hν)

The energy loss of neutrons when scattering with various light elements is characterized in Table R8.

Table R8. The energy absorption efficiency of light elements

$(E_0 = 2\ \text{MeV}, E = kT)$

Element	$\Delta\bar{E}$, keV	n
^{1}H	1000	18
^{2}D	888	24
^{4}He	640	41
Be	360	50
C	284	111
Al	137	240

E is the mean energy transferred in one step and n is the number of steps needed for thermalization.

Examples of practical importance:

(n,γ) $^{113}\text{Cd}(n,\gamma)^{114}\text{Cd}$ σ=6.31×10^{-24}m^2

$^{135}\text{Xe}(n,\gamma)^{136}\text{Xe}$ σ=2.7×10^{-22}m^2

$^{149}\text{Sm}(n,\gamma)^{150}\text{Sm}$ σ=6.6×10^{-24}m^2

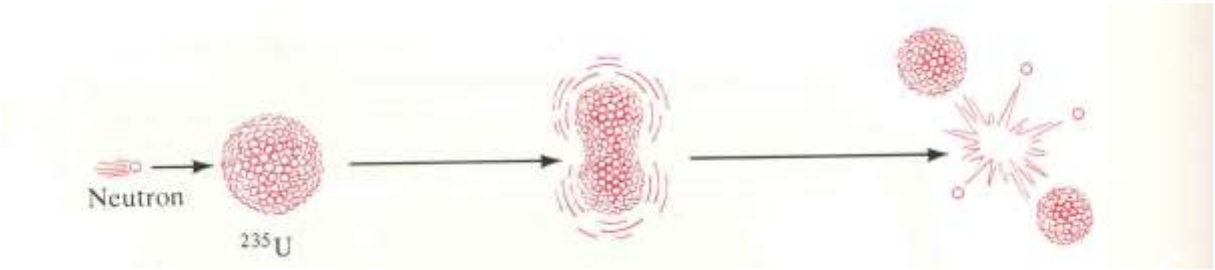
$^{157}\text{Gd}(n,\gamma)^{158}\text{Gd}$ σ=4.6×10^{-23}m^2

(n,α) $^{10}\text{B}(n,\alpha)^{7}\text{Li}$ σ=3×10^{-25}m^2

(n,f)

(f=fission).

The fission reaction of ^{235}U:



$$^{235}U + n \xrightarrow{[^{236}U]} 3n + {}^{90}Kr + {}^{143}Ba \ +200\ MeV$$

Only a limited number of nuclides is available and able to participate in such reactions (Table R9).

Table R9. Nuclides showing fission reaction

Fuel	Source of the fuel	Neutron energy needed
^{235}U	natural uranium	0.025 eV<$E_{neutron}$< 0.44 eV (thermal)
^{233}U	from thorium with n absorption	0.025 eV<$E_{neutron}$< 0.44 eV (thermal)
^{239}Pu	from ^{238}U with n absorption	0.025 eV<$E_{neutron}$< 0.44 eV (thermal)
^{241}Pu	from ^{238}U with n absorption	0.025 eV<$E_{neutron}$< 0.44 eV (thermal)
^{238}U	natural uranium	0.5 MeV<$E_{neutron}$< 10 MeV (fast)
^{232}Pu	natural thorium	0.5 MeV<$E_{neutron}$< 10 MeV (fast)

Figure R19. shows the distribution of the products (the fission takes place in about 50 routes producing a mixture of 300 isotopes of about 35 elements).

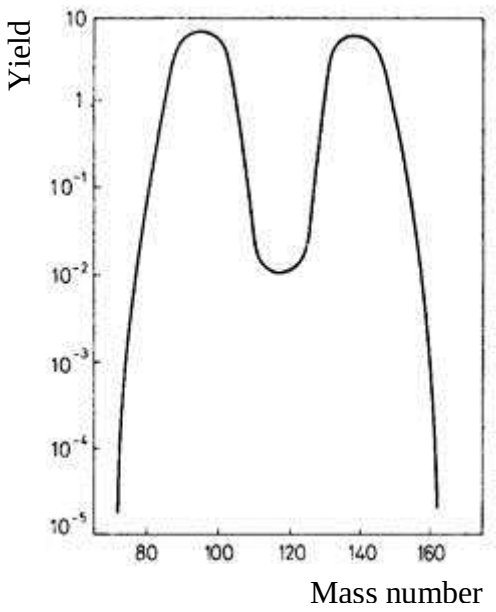


Figure R19.The distribution of the fission products of ^{235}U

Most of the products are radioactive resulting in further radioactive isotopes:

$$^{90}Kr \xrightarrow[33\ s]{\beta^-} {}^{90}Rb \xrightarrow[2,7\ min]{\beta^-} {}^{90}Sr \xrightarrow[28\,a]{\beta^-} {}^{90}Y \xrightarrow[64\,h]{\beta^-} {}^{90}Zr$$

The influence of the neutron energy on the probability of this fission reaction is shown in Figure R20:

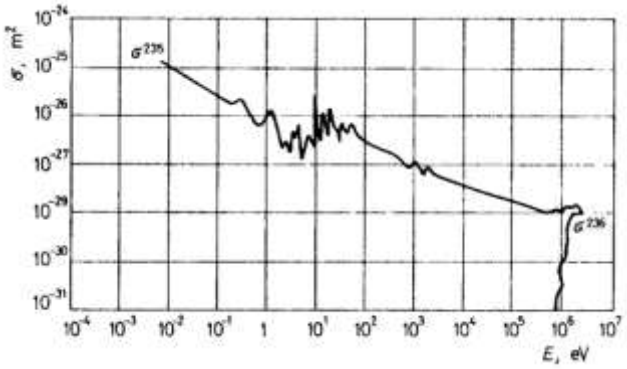


Figure R20. The energy dependence of the ^{235}U (n,f) nuclear reaction

The distribution of the 200 MeV:

kinetic energy of the fission products:	≈ 160 MeV
kinetic energy of the neutrons:	≈ 5 MeV
energy of the γ radiation:	≈ 5 MeV
energy of the secondary radioactive decays:	≈ 20 MeV
energy from neutron absorption:	≈ 10 MeV

A controlled reduction of the number and energy of these high energy neutrons produced opens the avenue for a sustained chain reaction in nuclear reactors for energy production.

R5. Nuclear instruments and measurements

Industrial application of isotopes utilizes the radiation of radioactive materials (radiation sources). Depending on the mode of industrial application, information is in most cases obtained through the effects of materials on radiation, about, e.g., the qualitative and quantitative material properties. The fundamental requirement to obtain and process the desired information about technological process is the detection and numerical evaluation of radiation.

Radioactive radiation cannot be sensed in a direct way; however, the interaction between radiation and suitably chosen media placed in the radiation path and within its effective range induces effects which can be used to determine the intensity and, in some cases, the energy of the radiation by electrical or electronic techniques, using appropriate instrumentation. The component of the total measuring system sensing and converting radiation into electrical signals is the radiation detector, the unit processing and recording signals from the detector is the measuring equipment or instrument.

The principal purpose of nuclear measurement technology is to determine either the integral radiation parameters or the partial parameters of particles or photons constituting certain portions of radiation.

R5.1. Nuclear radiation detectors

The stringent requirements set against radiation detectors for nuclear instruments are;

- sensing radiation with high efficiency;
- performance independent of or proportional to the energy of radiation;
- providing high-level electrical signals to make the electronic equipment as simple as possible;
- requiring the lowest possible supply voltage;
- operation not sensitive to supply voltage fluctuation;
- operation not sensitive to changes of ambient temperature and of other climatic parameters;
- high resistance against shock and vibration;
- long service life with stable operation;
- relatively low costs.

-

At present, not all the requirements listed above are satisfied simultaneously by the detectors in general use. The type of detector should always be chosen in accordance with the nature of a given problem to meet the most suitable conditions for that particular problem.

The most extensively utilized effects for radioactive radiation detection are:

- ionization: The atoms of gases or solids (e.g., semi-conductors) penetrated by radiation are ionized at a degree proportional to the intensity of radiation;
- luminescence: The absorption of radiation induces light flashes or scintillations in certain substances.
- In additional to the above effects, other physical phenomena can also be used for detecting radiation (radiation-induced blackening of X-ray films, track detection methods, etc.).
- The nuclear radiation detectors used for industrial isotope measurements can be classified according to their operation principles, i.e., to the nature of the interaction. The main detector types and their important characteristics discussed below are summarized in Table R10.

Table R10. Features of the main detector types

Properties	GM counter	Scintillation detector	Semiconductor detector
Field of application	Primarily for particle radiation measurements	Measurements of any radioactive radiation types	Measurements of any radioactive radiation
Measurement efficiency	For particle radiation near 100% for electromagnetic radiation 1 or 2%	Generally good	Generally good strongly temperature dependent at some types
Dead time	< 1 ms	<1 μs	<0.1 μs
Energy selectivity	Non-selective	Selective	Very selective
Costs	Low	High, due to accessories	High
Other aspects	Limited but usually long time	High counting rates	For drifted semiconductors, cooling required both for measurement an storage

Gas ionization detectors

Two major detector types can be distinguished according to the media where the ionization takes place, e.g., gas ionization and solid state (semiconductor) detectors.

The common structural design of the various gas ionization detectors is that they consist of two metallic electrodes connected to a power supply with a gas between them which can be ionized by radioactive radiation. The neutral gas molecules are ionized by the incoming radiation causing the production of pairs of positively charged ions and negatively charged electrons. In the electric field between the electrodes the positive ions and the electrons migrate to the negatively and positively biased electrodes, respectively.

The performance of gas ionization detectors depends strongly on the electric field between the electrodes.

Geiger-Müller (GM)-counters

Geiger-Müller (GM)-counter is one type of ionization detectors applying high internal electric field – this having been chosen to induce high multiplication factors (10^8 to 10^9) within the detector generally filled with argon. The quantitative change in the multiplication factor invokes also a qualitative one: the primary ions are accelerated by the voltage difference up to a value where an avalanche ionization process develops in the gas space which will propagate along the anode wire to the total length of the tube within several hundred nanoseconds.

In addition to ionization, a secondary effect of the partial recombination of ions leads to the appearance of excited gas atoms; they release their excess energy by photon emission. As a result of the combined effect, a gas discharge appears between the electrodes and the pulse height will be independent of the number of primary ions. However, the resistivity between the electrodes will be reduced and if no provisions for maintaining the potential difference were to be made the ionization would continue even without the arrival of new particles or photons. It means that the tube would be inadequate for further radiation detection because no further ionization process could be initiated in the "dead time". The quenching of the discharge process and thus the reduction of dead time can be realized by detectors filled by special gas mixtures (addition of alcohol or other volatile organic compounds or halogens). The dead time of the GM-tube is 50-200 μs. The dead time effect is disadvantageous if the number of particles to be detected in unit time is high.

The operation of the counter tube is represented by the characteristics shown in Figure R21.

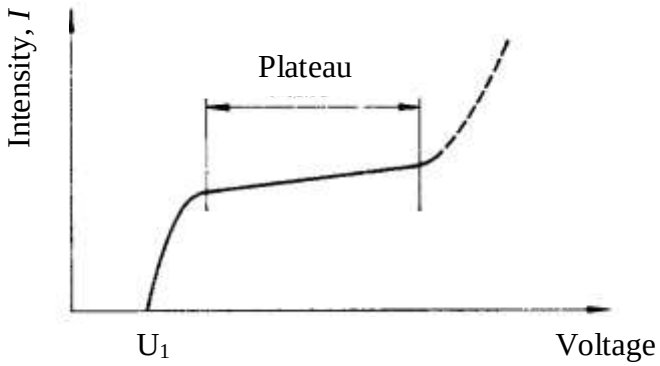


Figure R21. The characteristics of the GM tube

The voltage U_1 represents the counting threshold where the gas amplification starts. By increasing the voltage, the number of detected pulses changes slightly in the region U_1-U_2. This region is called the plateau of the counter. The longer the plateau and the less its slope, the better the counter. The operational characteristics should be defined in accordance with the parameters of a given application.

The detection efficiency of GM-counter tube for α- and β-particles is close to 100 % but for electromagnetic radiation (γ- or X-rays) it rarely exceeds 1 or 2 %.

GM-counters are the most suitable for constructing the simplest detector systems. These counters work on a few 100 V power supply whose instability has an almost negligible effect on the performance of the detector, provided that the instruments are appropriately set to the conditions. The performance of the detector is only slightly depends on temperature; it provides reasonably large electric signals, thus the requirements for signal processing are rather moderate. Its disadvantages lie in the low sensitivity for γ-radiation, the inapplicability for the discrimination of energy and its finite life-time. In spite of these disadvantages, the GM-counter is the most widely used detector, mainly because of the low costs.

Semiconductor detectors

The basic principle of radiation sensing by semiconductor detectors is the ionization interaction process identical to that is gas ionization detectors. The semiconductor detector can also be considered as a planar-electrode ionization chamber with a semiconducting crystal substituting the filling gas. The stopping power of solid-state detectors prepared from semiconductor materials is several times higher than that of the gas filling in ionization detectors, due to the difference in specific weights of these materials. An elementary particle incident on a germanium or silicon single crystal semiconductor detector and causing ionization there is stopped rapidly, within 10^{-11} s. During slowing down, its energy will be

transferred to the electrons of crystal atoms (ionization) which in turn can give rise to secondary ionization if the absorbed energy is high. In the semiconducting material charge carrier pairs (electrons and holes) are produced directly by electrically charged particles (α- and β-particles) or indirectly by X-ray, γ- or neutron radiation, in the latter cases through the charged particles from some radiation interaction process. The energy needed to generate an electron-hole pair is 3.61 eV and 2.98 eV in silicon and germanium, respectively.

The carrier collection time taking into account the detector geometry and carrier mobility is inversely proportional to the field strength. To increase the field strength, a relatively high detector voltage of several hundred volt is applied. To reduce noise effects accompanying the operation of detectors they are, in general, cooled below -190 °C by liquid air or nitrogen.

Semiconductor detectors have been developed rapidly in the past years utilizing recent advances in semiconductor device technology. It means that their growing use can be anticipated both in scientific research and engineering application. Their principal advantages are:

- excellent energy discrimination;
- linear energy-to-pulse height conversion (for total absorption);
- short pulse generation;
- non-sensitivity to magnetic fields;
- low supply voltage requirement;
- vibration and impact resistance;
- long service life.

On the other hand, there are some drawbacks of semiconductor detectors, such as

- relatively high, supply voltage dependent self-capacity;
- disturbances in operation by lattice defects in the crystal;
- the signal-to-noise ratio at room temperature is low compared with other detector types;
- surface effects of atmospheric contaminations can increase the inherent noises;
- long term irradiation can cause damage in the crystal structure leading to the degradation of detector characteristics;
- the geometrical dimensions are limited.

In spite of these disadvantages the field of semiconductor detector applications is growing, mainly due to its feature of good energy discrimination for particles or photons constituting

the radiation. In other words, the "energy resolution" of semiconductor detectors is the best of all counter types (Figure R22.).

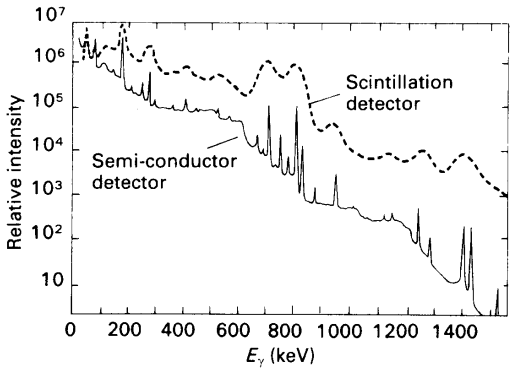


Figure R22. Comparison of a scintillation and a semi-conductor detector spectrum

Scintillation detectors

The operation principles of scintillation detectors are based upon the specific properties of some organic and inorganic substances, i.e., light-flash generation or luminescence induced by radioactive radiation. The weak flashes or scintillations are detected and amplified by a photomultiplier used as a light-to-electric signal transducer. The scintillation is proportional to the incident particle energy; thus, from the voltage output of the photomultiplier, due to its linear characteristics, information on particles energies can be obtained. Substances possessing this luminescence property are called scintillators in nuclear measurement techniques. The scintillation mechanism is a complex process. The scintillator absorbs radiation energy resulting excited or ionized states of some of the atoms. They will fall back into their ground state in a very short time accompanied by light, or by photon emission. The higher the energy absorption from the traversing particle in the material, the more scintillating atoms will be excited. The energy transfer from individual particles of photons will depend on the interaction between the detector material and the radiation. There are three groups of scintillator materials, inorganic single crystals, liquid scintillators and plastic scintillators.

During the well-known interaction processes of γ-photons such as photoelectric effect, Compton scattering and pair production (see earlier), electrons are created which ionize and excite the atoms similarly to the direct effect of β-rays. Also a secondary radiation may occur which can also be absorbed through additional interaction processes by the scintillator. Thus, provided the crystal sizes are adequate, the total energy of γ-radiation will be expended to excitation through several intermediate process. The most frequently used inorganic scintillator single crystal is thallium-activated sodium iodide. Due to its relatively high

density, it is suitable mainly for the detection of strongly penetrating electromagnetic γ-radiation (Table R 10.).

The penetration depths of α-particles is small through their energy can be several MeV, therefore the energy transfer takes place in a very thin layer. They ionize atoms along their paths strongly, producing secondary electrons which excite the atoms. A thin ZnS layer is the most frequently applied scintillator material for alpha-radiation.

The penetration depths of β-particles can be several mm depending on their energies. During penetration they ionize more or less strongly, causing secondary electron production which in turn excites the atoms. The detection efficiency for electrons is essentially 100% for most scintillators. But because electrons can make large angle scatterings (sometimes back-scatterings), they can exit the detector without depositing their full energy in it. The back-scattering is a rapidly increasing function of the atomic number Z of the scintillator material. Organic scintillators, having a lower Z than inorganic crystals, are therefore best suited for the detection of low-energy (< 10 MeV) beta particles. Since the neutron is not charged it does not interact via the Coulomb force and therefore does not ionize the scintillation material. It must first transfer some or all of its energy via the strong force to a charged atomic nucleus. The positively charged nucleus then produces ionization. Neutron radiation can be detected by scintillators containing boron or lithium where the neutrons produce α-radiation which gives rise to scintillation in the way described earlier.

The arrangement known as a scintillation counter includes a scintillator and an optically coupled photomultiplier. The detection of a particle takes place in several steps. The scintillator crystal should be selected according to the actual radiation type (α, β, γ or neutron radiation). The crystal size, encapsulation and its incorporation in the sensor are determined by the radiation energy. The electron multiplication process and, consequently, the multiplier performance are sensitive to temperature and magnetic field variations. The efficiency, i.e., the counting probability of scintillation detectors is high. For α- and β-rays it can be as high as 100% but it can also reach 20 to 30% for γ-radiation.

In several cases the energy of the emitted radiation is too low to reach the detector (soft β-radiation, in particular). In such cases liquid scintillators are applied. The specimen tested is dissolved in the liquid allowing high efficiency. The most important liquid scintillator compounds are 2,5-diphenyloxazol (PPO), 2-phenyl-5-(4-diphenyl)-1,3,4-ocadiazol (PBD), 2-[4'-tert-butylphenyl-5-(4''-diphenyl)]-1,3,4-oxadiazol (butyl-PBD), 1,-4-di(5-phenyloxazol-2-yl) benzene (POPOP), and 1,4-bis-2(4-methyl-5-phenyloxazolite) benzene (dimethyl-

POPOP). They are used in solutions of cyclic hydrocarbons (mixed with toluene or xylene or cyclohexane, for example). The energy of the soft beta radiation is primarily absorbed by the solvent molecules and their emitted energy will excite the scintillator materials dissolved. The specific position of the photomultipliers around the sample holder may enhance the efficiency of the detection.

The fact that the performance of the scintillation detector tends to be dependent on the working conditions is highly disadvantageous, mainly in industry, because of its features listed as follows:

- a quite good time-resolution;
- the proportionality of the signal to the energy;
- the great amplitude of the electric signal;
- the high efficiency detection (also for γ-radiation);
- their (practically) infinite lifetime.

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