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BASICS OF RADIOCHEMISTRY. LECTIONS

ПЕТРОВ
ВЛАДИМИР ГЕННАДЬЕВИЧ

ФИЗФАК МГУ

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1. Lecture 1. Introduction to radioactivity

Terminology

Radioactivity is a physical phenomenon, defined as the property of certain nuclei to spontaneously fragment or rearrange, resulting in the emission of radiation.

Matter consists of **atoms** which in turn consist of a very small **nucleus** with positively charged **protons** and neutral **neutrons** (together they are called **nucleons**) inside it and negatively charged electrons orbiting around. Almost all mass of an atom is concentrated inside the nucleus.

The number of protons inside a nucleus define the position of an element in the periodic table of the elements (fig. 1.1).

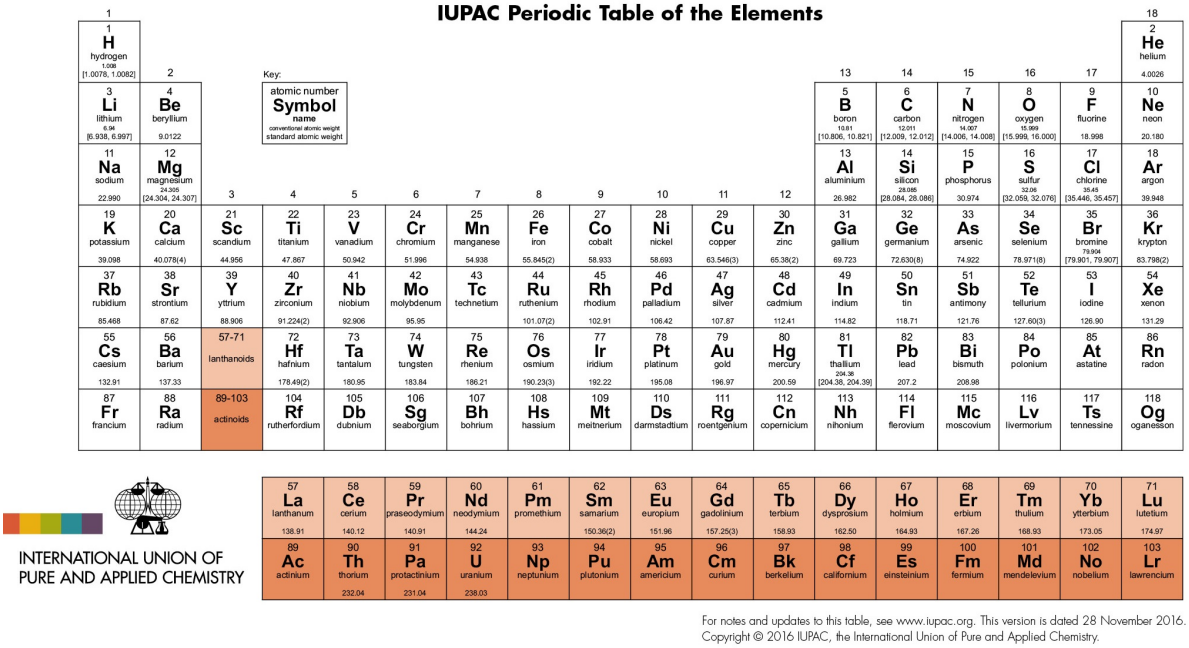


Figure 1.1. The periodic table

Comparison of proton, neutron and electron masses respectively:

$$\begin{cases} m_p = 1\ a.m.u. \approx 1836\ m_e \\ m_n \approx 1\ a.m.u. \approx 1839\ m_e \\ m_e \approx \frac{1}{1800}\ a.m.u. \end{cases} \tag{1.1}$$

where $1\ a.m.u. = 1/12$ of mass of ^{12}C isotope $= 1.66 \cdot 10^{-27}$ kg.

Isotopes are nuclei of the same element (same number of protons) but having different numbers of neutrons (hence different atomic mass). The chemical properties of isotopes are the same, but some physical properties may differ.

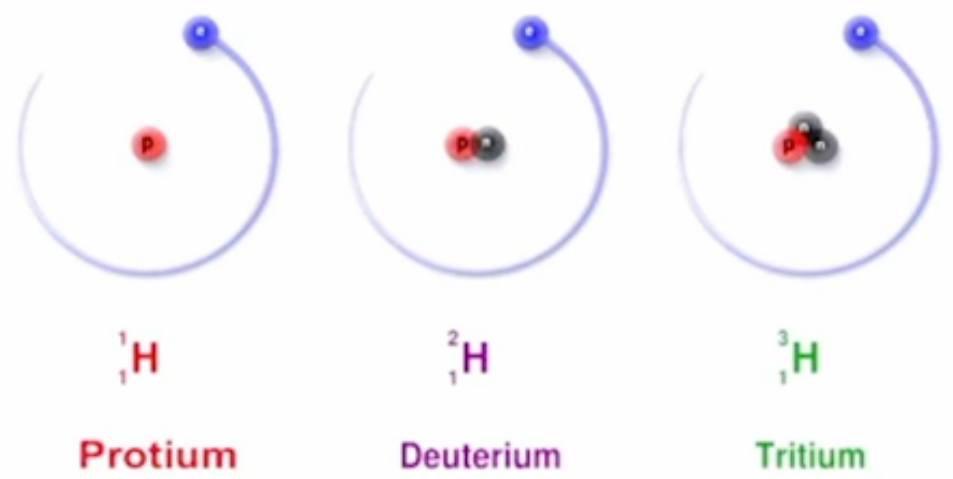


Figure 1.2. Isotopes of hydrogen

Protium and deuterium in fig. 1.2 are stable, whereas tritium is radioactive. We call radioactive isotopes **radionuclides**.

Just like electrons in atoms nucleons in nuclei have energy levels. If an energy level in a nucleus is completely filled with nucleons ⇒ this nucleus will be stable. The numbers of nucleons that fully fill energy levels in nucleus are called **magic numbers**.

How do protons and neutrons exist in a nucleus when protons should have pushed each other away due to electrostatic repulsion? When protons and neutrons come sufficiently close to each other there activates another type of interaction - **nuclear force**. This force is much stronger than electrostatic one and responsible for binding nucleons together. When we talk about nuclear interaction there is no mass conservation law (but energy conservation still works).

The famous Einstein's equation (1.2) gives the relationship between energy and mass:

$$E = mc^2 \tag{1.2}$$

where c is the speed of light.

The mass of a nucleus is always less than the sum of the masses of its constituents (protons and neutrons). The difference between them is called the **mass defect**. Using a mass defect and equation (1.2) we can calculate energy of nuclear interaction inside a nucleus (**binding energy**).

Taking **binding energy per nucleon** and plotting it for nuclei with different number of nucleons we can obtain the curve shown in fig. 1.3. Left part of the plot (before iron) shows that we can release energy by combining light nuclei with each other in the process called **fusion**. Nuclei heavier than iron can split into lighter nuclei producing energy in the process called **fission**.

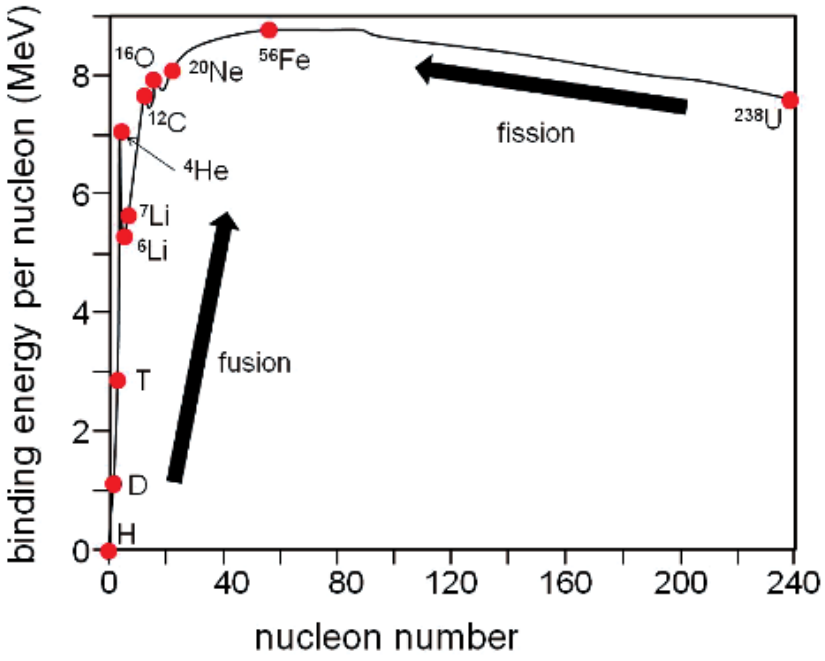


Figure 1.3. Binding energy per nucleon VS nucleon number

Discovery of radioactivity

The phenomenon of radioactivity was discovered in 1896 by Henri Becquerel during his studies of the luminescence of uranium salt. Becquerel found out that uranium irradiated with sunlight emitted some hitherto unknown rays that could penetrate through paper and had an effect on photographic plate. Once he put the uranium salt together with the photographic plate in the drawer of his table and in a couple of days he discovered, that the salt emitted radiation in the shade of the drawer even without being exposed to the sunlight. Therefore Becquerel suggested that it was the internal property of uranium to emit this unknown rays.

Followers of Becquerel (Pier and Marie Curie) continued to study radiation and later discovered other elements that emitted those unknown rays such as radium and polonium. The term radioactivity was coined by Marie Curie.

Discovery of nucleus, proton and neutron

The discovery of radioactivity and ionizing radiation led to many other findings, among which was Rutherford's scattering experiment. In that experiment Rutherford bombarded thin golden foil with $\alpha-$particles most of which scattered on very small angles, but much to Rutherford's surprise a small fraction of $\alpha-$particles bounced backwards at a large angles close to 180°. The only explanation to this could be that inside an atom of foil there was tiny but very heavy positively charged nucleus and electrons revolved around that nucleus. It is, in a nutshell, Rutherford's planetary model.

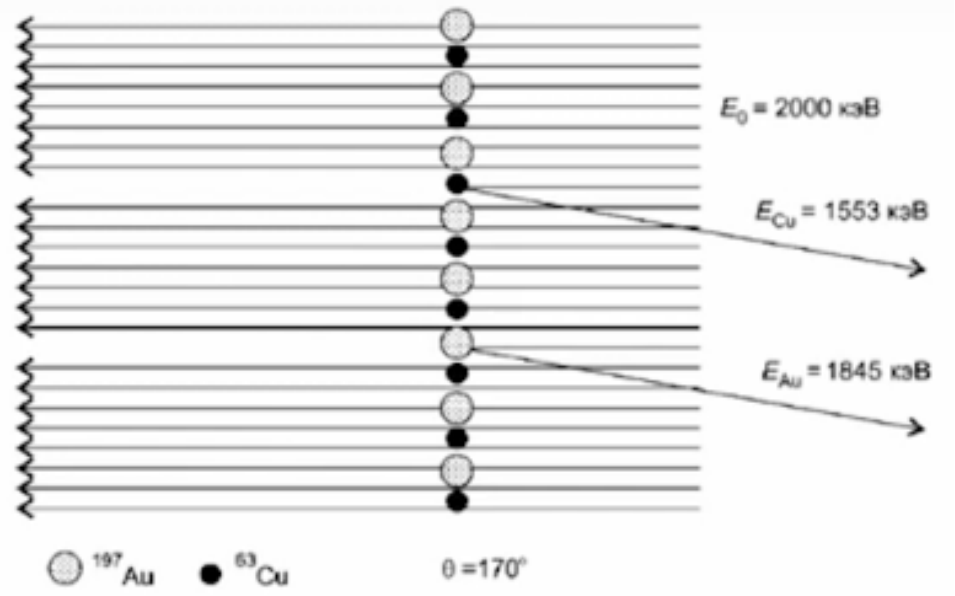


Figure 1.4. Rutherford's experiment

Discoveries of Becquerel, Curie and Rutherford paved the way to more pioneering works in this field. Short history of discoveries goes like this:

- By 1910 forty natural radionuclides had been discovered.
- In 1932 the neutron was discovered by G. Chadwick.
- In 1934 the artificial radioactivity was discovered by Irène Joliot-Curie and Frédéric Joliot-Curies. They made aluminum radioactive by bombarding it with $\alpha-$particles.
- The discovery of spontaneous fission of ^{235}U in 1940
- In 1945 the first atomic bomb was tested

Application of nuclear energy

Nuclear energy can also be used in a peaceful way, for example **power plants**, that can produce electricity and thermal energy (fig. 1.5)

Figure 1.5. Power plants

Another application of ionizing radiation in modern world is **nuclear medicine**. Doctors can diagnose diseases more accurately using MRI technology (fig. 1.6), radiopharmacy uses radionuclides as a component of medicine, accelerators of charged particles are used to treat cancer.

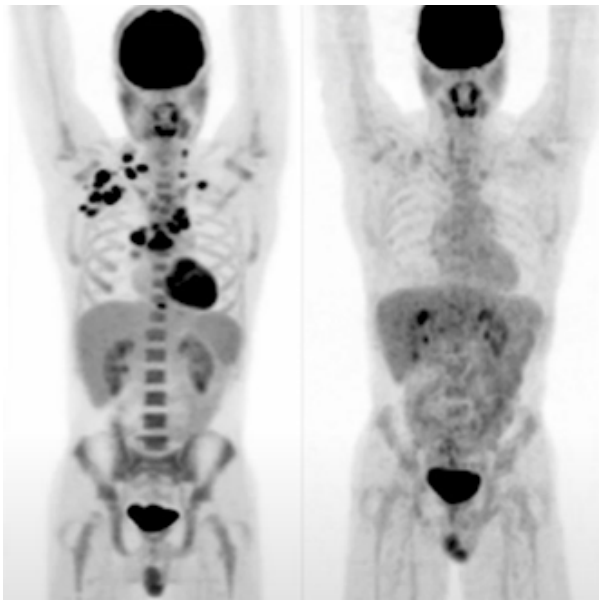


Figure 1.6. Magnetic resonance imaging (MRI)

Yet another contribution of radiation technologies is **waste treatment**:

- **Gas waste**
  - removal of NO_x and SO_x from flue gas of coal power plants (fig.1.7)
  - waste incinerator off-gases treatment
- **Water**
  - treatment of drinking water to remove pesticides and pharmaceuticals
  - sterilization of municipal waste into nitrogen and phosphorus rich fertilizers vs. pollution
- **Oil and gas**
  - conversion of natural gas to liquid hydrocarbons
  - removal of NO_x and SO_x from flue gas
- **Nuclear**
  - destruction of long-lived nuclear waste via ADS (accelerator-driven system) (fig.1.8)
- **Solid**
  - infected medical wastes, polluted soil, etc

- **Pest control**
  - radiative sterilization of male insects (fig.1.9)

For removal of nitrogen and sulfur oxides special radiation technique is used: radicals that are produced inside a gas stream attack nitrogen and sulfur to oxidize them to higher oxides which in turn are removed by absorbing in water or other sorbents.

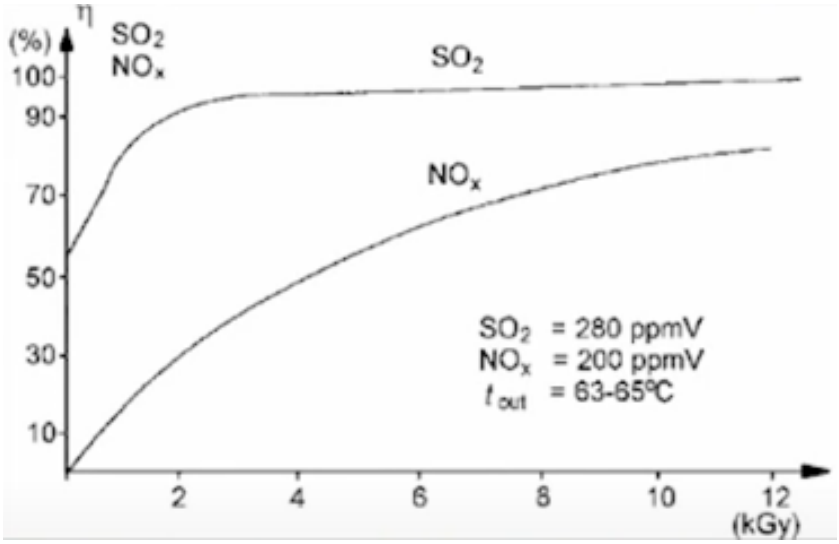


Figure 1.7. NO_x and SO_2 removal efficiency VS dose. The results obtained in the pilot plant experiments.

Accelerator-driven systems allows to decrease the number of radioactive waste by transforming long-lived radionuclides into short-lived radionuclides or even stable nuclei.

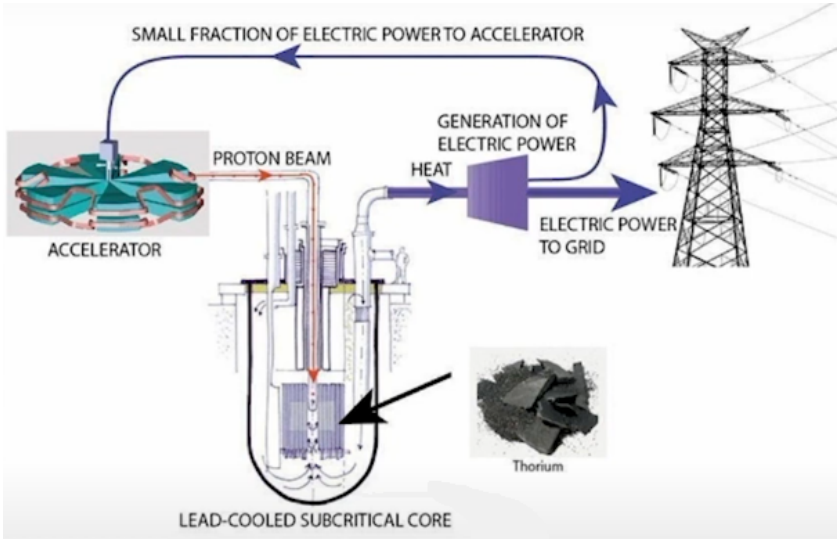


Figure 1.8. The scheme of an accelerator-driven system

The pest control is used to control some infections that are carried by insects. Part of the male population of insects are taken from the nature and irradiated by ionizing radiation ⇒ they become sterile. Then the sterile insects are released back in the nature when they try to reproduce but obviously fail. As a result the entire population of insects decreases and consequently the spread of the diseases they carry also drops down.

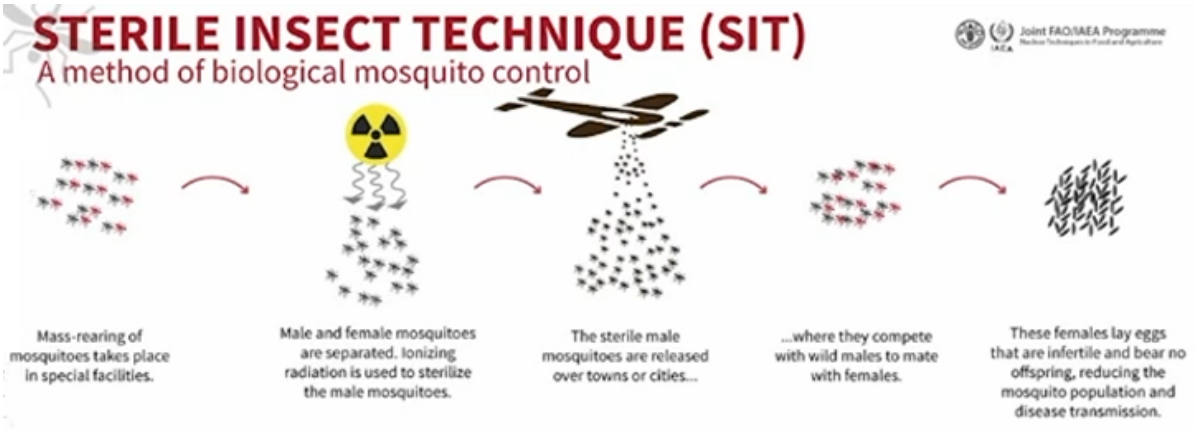


Figure 1.9. Insect sterilization scheme

Indirect contribution of radiation techniques involves decreasing of use of organic solvents and other chemicals in production of polymers, semiconductors, etc.

Branch of radiation technology	Process or irradiated products	Positive indirect effect
Sterilization of medical products	Disposable syringes, blood transfusion systems, etc.	Elimination of ethylene oxide or compressed steam
Treatment of food products	Grains, potato, spices, poultry meat, vegetables, fruits, etc.	Elimination of fumigants (such as methyl bromide), killing of pathogenic microorganisms and parasites, rotting control, etc.
Curing of coatings	Paint films, printing inks, etc.	Elimination of organic solvents and energy savings
Degradation of spent polymer materials and products	Butyl rubber, Teflon, and polyethylene	Recycling
Irradiation of cellulose pulp in viscose production	Partial degradation of cellulose	Decreased consumption of reagents (including CS_2)
Vulcanization	Natural rubber latex	Manufacture of vulcanized products without harmful impurities
Graft polymerization	Preparation of an amidoxime adsorbent	Adsorption of uranium from seawater
Sterilization and insect disinfestation of plant materials	Wood, nutrient media for mushrooms, soil, etc.	Killing of microorganisms, insects, weeds, etc.
Genetics	Rice, wheat, soybean, manioc, pear, etc.	Production of new varieties with increased crop capacity

Figure 1.10. Indirect contribution of radiation techniques

One more contribution of radiation techniques is their **impact on society**. Radiation technologies require highly qualified specialists ⇒ the quality of higher and school education increases to meet the demand. This in turn leads to development of scientific and technology industries which in the long run will improve the quality of life. The more educated the people the more they are concerned about environment and therefore more willing to preserve it.

Nuclear weapon can be considered as direct impact of radiation technology. Despite many people being scared of it, the end of 20th century and the beginning of 21st century is the most peaceful period in the history of the mankind. There is a special website called "Bulletin of the Atomic Scientists" where nuclear scientists publish their papers and reports and also make predictions about the time remaining before the so-called nuclear midnight occurs when one country attacks another with nuclear weapon. This time represents how far away we are from that devastating event.

In conclusion, it goes without saying that radiation technology (as well as any other) must be used wisely. It is not the instrument that is to blame for the disasters in the past, but ourselves who used in the wrong way.

2. Lecture 2. Radioactive decay

Types of decay

To understand about the structure and relationship between different radionuclides and how they transform into each other via different types of radioactive decay let's have a look at the nuclide map (fig.2.1).

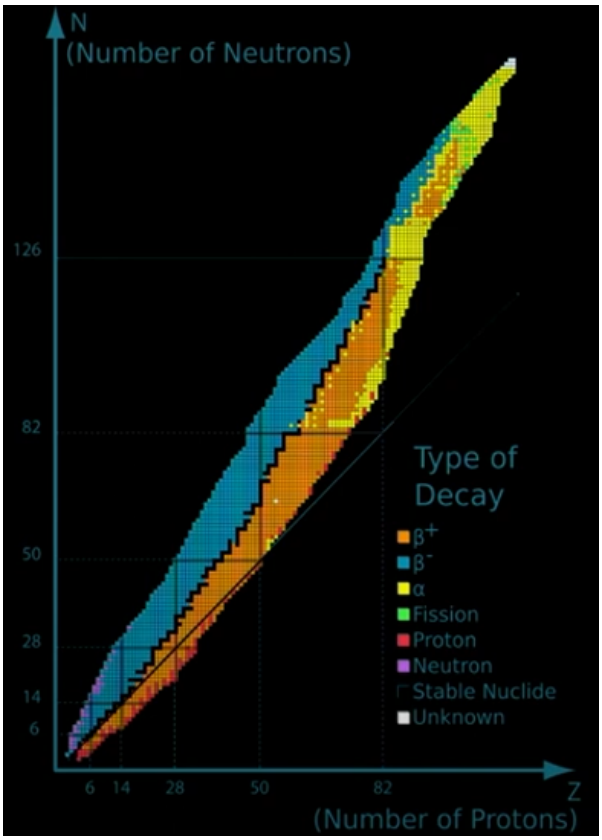


Figure 2.1. Nuclide map

The X- and Y- axis of the nuclide map are the numbers of protons and neutrons correspondingly. It means that the isotopes of a given element are located in vertical lines. Colors show which type of decay occurs for a given nuclide. There are around 3000 nuclides known so far but only a small fraction of them is stable (shown in black line).

There are four main types of decay:

- Alpha-decay (α-decay)
- Beta-decay (β-decay)
- Gamma-decay (γ-decay)
- Spontaneous fission(we will talk about it later when we discuss nuclear energy and power plants)

Alpha-decay is an emission of an alpha-particle by radionuclide. Alpha-particle consists of 2 protons and 2 neutrons and basically is a nucleus of ^4He. As a result of alpha-decay the mass of the nuclide decreases by 4 and the charge decreases by 2, which corresponds to moving 2 cells to the left in the periodic table (fig.2.2).

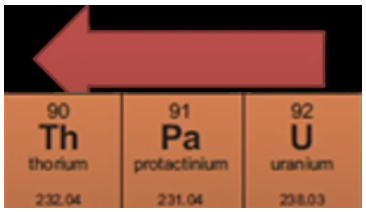


Figure 2.2. Moving left in the periodic table

The equation of alpha-decay looks like this:

$$^A_ZX \rightarrow ^{A-4}_{Z-2}Y + ^4_2\alpha \tag{2.1}$$

where X, Y are the initial and recoil nuclides, α is the alpha-particle.

Alpha-decay is typical of heavy nuclei starting from ^{83}Bi. In the nuclide map (fig.2.1) alpha-decay is shown with the yellow color and concentrated in the area of heavy nuclei.

The total energy of the alpha-decay is given by the following equation:

$$Q = 931.5 \cdot (m^A_Z - m^{A-4}_{Z-2} - m_\alpha) \tag{2.2}$$

This energy is distributed between the recoil nucleus and the α-particle:

$$Q = E_R + E_\alpha \tag{2.3}$$

where $\frac{E_R}{E_\alpha} = \frac{m_\alpha}{m_R}$ because there only 2 final particles produced.

As long as there is strict relationship between the energies of the alpha-particle and recoil nucleus $\Rightarrow$ the energy spectrum of alpha-particles looks like discrete peaks (fig.2.3).

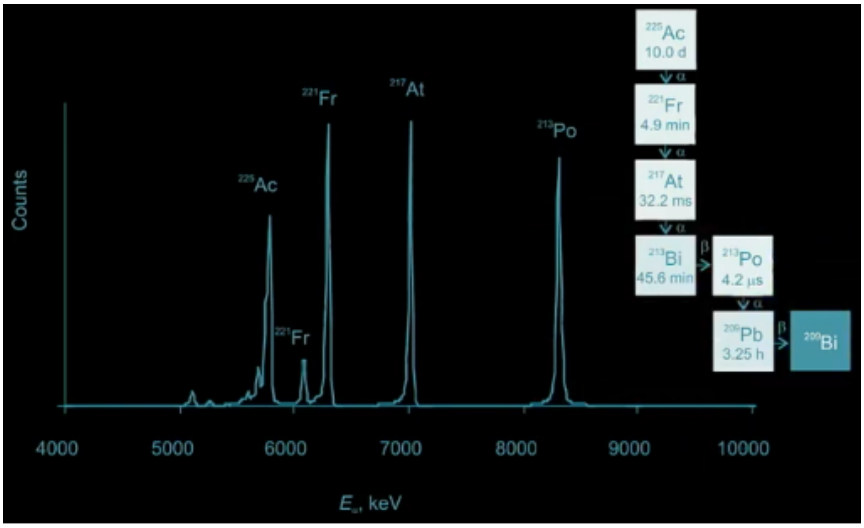


Figure 2.3. Energy spectrum of alpha-particles

There are three types of the **beta-decay**:

- β^--decay is given by this equation:

$$ {}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y + {}^{0}_{-1}e + {}^{0}_{0}\bar{\nu}_e \tag{2.4} $$

  where ${}^{0}_{-1}e \equiv \beta^-$ is an electron, ${}^{0}_{0}\bar{\nu}_e$ is an electronic anti-neutrino.

  The decay itself happens to a neutron inside the nuclide:

$$ {}^{1}_{0}n \rightarrow {}^{1}_{1}p + e^- + \bar{\nu}_e \tag{2.5} $$

  One act of the β^--decay corresponds to moving 1 cell to the right in the periodic table.

- β^+-decay is very similar to the β^--decay and given by the following equation:

$$ {}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + {}^{0}_{+1}e + {}^{0}_{0}\nu_e \tag{2.6} $$

  where ${}^{0}_{+1}e \equiv \beta^+$ is a positron, ${}^{0}_{0}\nu_e$ is an electronic neutrino.

  The decay itself happens to a proton inside the nuclide:

$$ {}^{1}_{1}p \rightarrow {}^{1}_{0}n + e^+ + \nu_e \tag{2.7} $$

  One act of the β^+-decay corresponds to moving 1 cell to the left in the periodic table.

- During electron capture the nucleus captures an electron from the nearest electronic shell (K-shell) of the atom. Basically what happens is a proton inside the nucleus consumes an electron and turns into a neutron with emission of an electronic neutrino:

$$ p + e^- \rightarrow n + \nu_e \tag{2.8} $$

  One act of the electron capture corresponds to moving 1 cell to the left in the periodic table as the charge of the nucleus decreasing by 1.

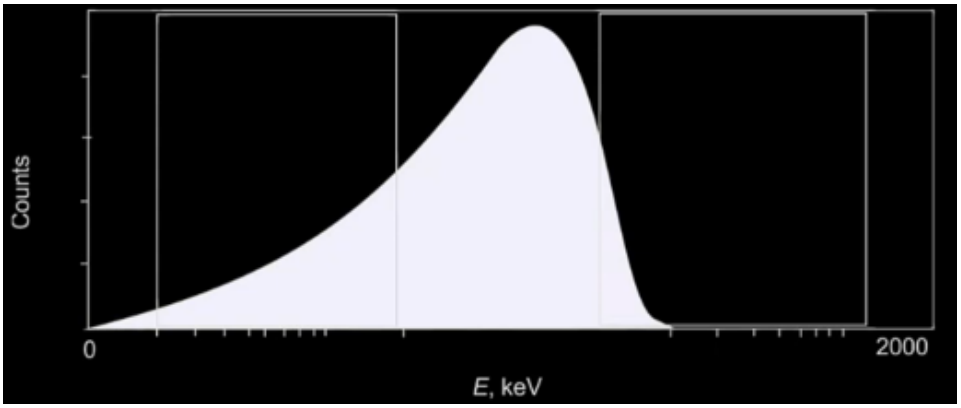


Figure 2.4. Energy spectrum of beta-particles

Electronic neutrino (or anti-neutrino) is the third final particle in the reactions of β^+ and β^- decay, that takes some energy from the reaction. Thus the spectrum of beta-particles will not be discrete as in the case of alpha-decay but will look as it is shown in fig.2.4.

The above mentioned neutrinos are extremely light neutral particles. Neutrinos can easily penetrate through all kinds of matter so it is very difficult to detect them. Electronic anti-neutrino was detected for the first time in 1956 in the Reines-Cowan experiment. The detection system consisted of 3 detectors and the space between them was filled with water solution of cadmium salt. The chain of nuclear reactions started with an anti-neutrino hitting a proton of water molecule:

$$\bar{\nu} + p \rightarrow n + e^+ \tag{2.9}$$

The positron from the reaction (2.9) annihilated with electrons (almost instantly) which resulted in emission of 2 γ-quanta, each with the energy of 511 KeV:

$$e^+ + e^- \rightarrow 2\gamma \tag{2.10}$$

The neutron from the reaction (2.9) wandered in the volume of the solution until it found a cadmium nucleus (it takes time $\Delta t \approx 5\ \mu s$) and interacted with it producing single γ-quantum :

$$^{108}Cd + n \rightarrow ^{109}Cd + \gamma \tag{2.11}$$

As a result Reunes and Cowan observed two γ-peaks from reactions (2.10) and (2.11) separated by 5 μs in time. That was solid evidence to existence of neutrinos.

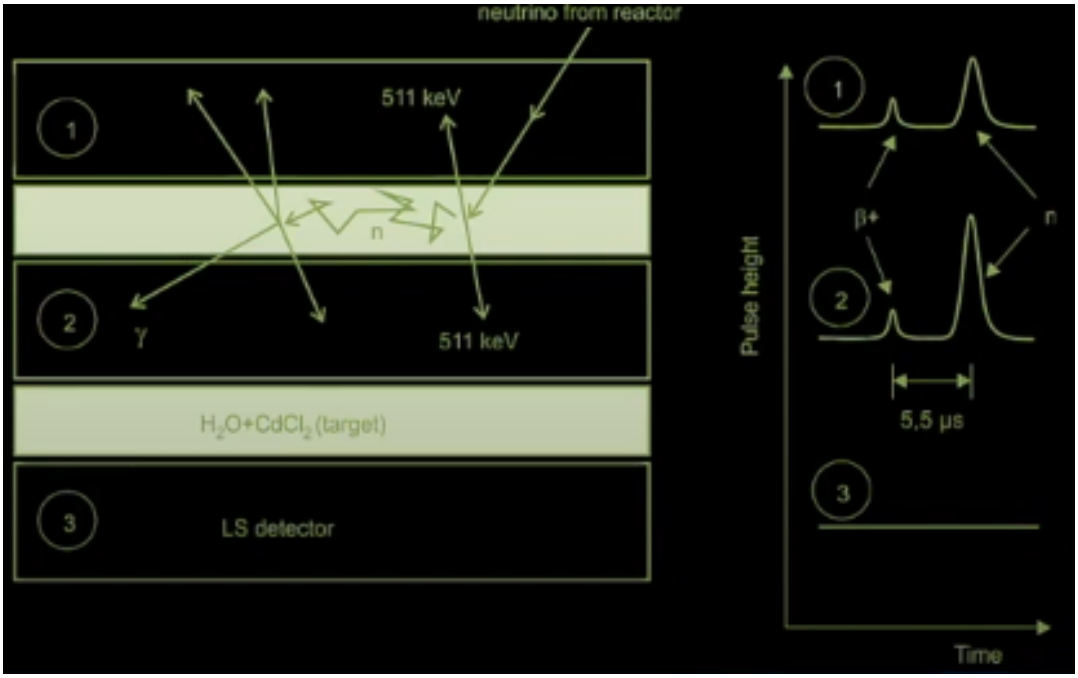


Figure 2.5. The scheme of the Reines-Cowan experiment

When an electron goes into the nucleus in the process of electron capture there appears a "hole" in the inner electron shell which will be filled with the electrons from outer shells. So there occurs transition of electrons from higher shells to the lowest one which leads to 2 processes (fig.2.6):

- **Emission of X-rays** (a form of electromagnetic radiation) with the energy equal to the difference of electron energy levels. X-rays can be used to identify chemical composition of matter.
- **Auger process** involves the emission of Auger electrons from the electron shell.

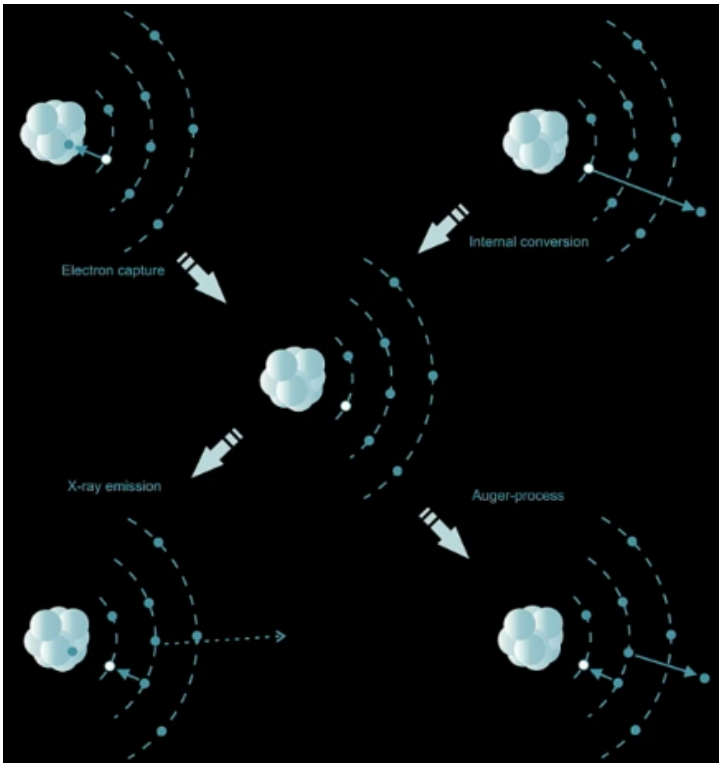
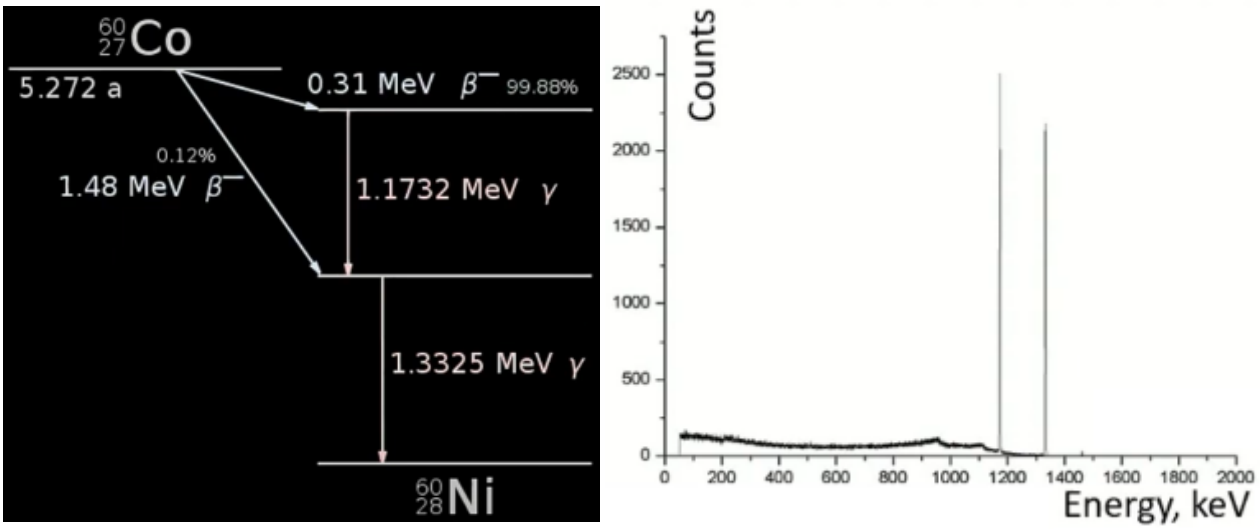


Figure 2.6. What can happen during electron capture

Gamma-decay or (**gamma-transition**) occurs only after another type of decay (alpha or beta).

After some type of decay the final nucleus may not be in the ground state but in the excited state. The relaxation from the excited state to the ground state occurs via the emission of γ-quanta that are also called **photons**. During gamma-transition there is no change in the composition of the nucleus: the number of protons and neutrons stays the same but the energy of the nucleus drops (see example in fig.2.7 **a)**). As long as nuclear energy levels are discrete the gamma spectrum of the emitted photons will also be discrete (fig.2.7 **b)**).



a) **b**)

Figure 2.7. **a**):The process of ^{60}Co decay; **b**): Gamma spectrum

The energy of the excited nucleus can be transferred to nearest electrons that will be emitted. These are so-called **conversion electrons**, their spectrum will also be discrete (fig. 2.8)

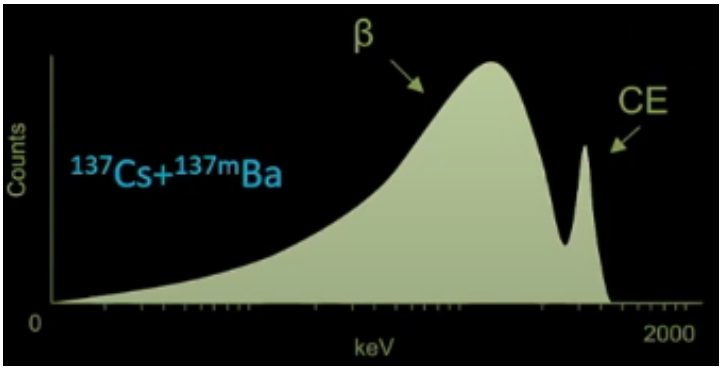


Figure 2.8. Beta spectrum and conversion electron spectrum

The law of radioactive decay

- The probability of a decay does not depend on time, temperature, pressure, electromagnetic fields, etc and is an intrinsic characteristic of a nucleus
- All nuclei are identical and independent
- Decay is a statistical event governed by Poisson statistics

The **activity**(A) of a nuclide is measured in becquerel (Bq) or curie (Ci) and given by the following equation:

$$A \equiv -\frac{dN}{dt} = \lambda N \tag{2.12}$$

where N is current number of nuclides, λ is the **decay constant**.

Activity is the quantitative measure of radioactivity: the number of nuclear decays occurring in a given quantity of matter over a certain time interval, divided by that time interval.

Solving differential equation (2.12) we obtain the law of radioactive decay:

$$N(t) = N_0 \cdot e^{-\lambda t} = N_0 \cdot 2^{-t/T_{1/2}} \tag{2.13}$$

where N_0 is the initial number of nuclides, $T_{1/2} = \frac{\ln(2)}{\lambda}$ is the **half-life** time - the amount of time needed for approximately a half of the current nuclides to decay.

The mean lifetime is $\tau = \frac{1}{\lambda}$.

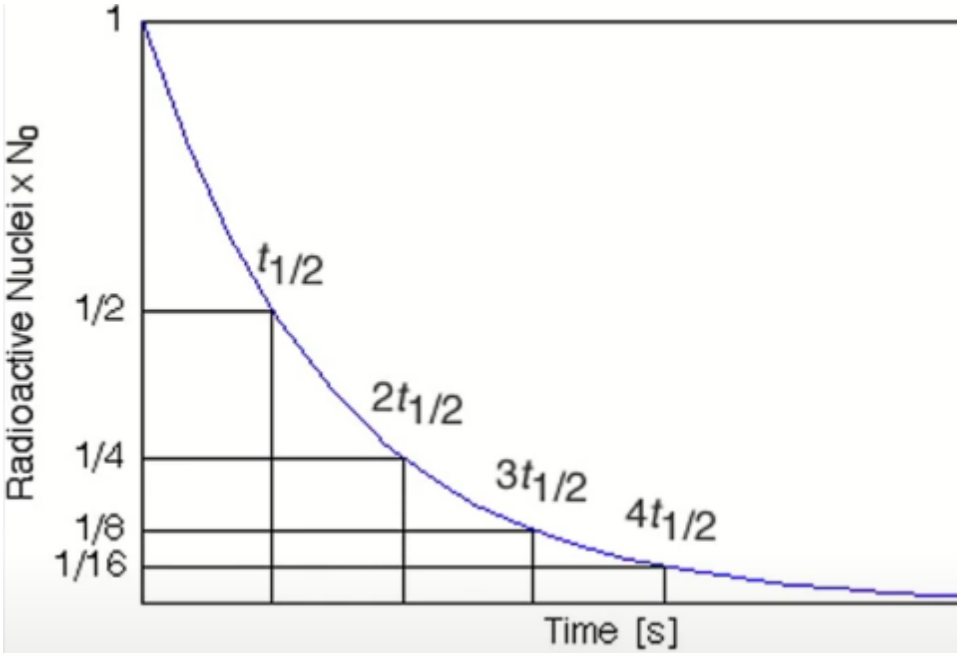


Figure 2.9. The law of radioactive decay

There is a simple relationship between the activity (A) and the mass of the nuclide (m):

$$A = \lambda N = \frac{mN_A \ln(2)}{T_{1/2} A_r} \tag{2.14}$$

where $N_A = 6.02 \cdot 10^{-23}$ mol^{-1} is the Avogadro constant, A_r - the atomic mass of the element.

Quite often radionuclides disintegrate not into stable nuclides, but into other radionuclides $\Rightarrow$ a chain of radioactive decays appears, for example:

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C_{\text{stable}} \tag{2.15}$$

There are three cases of relationship between the half-lives of the parent and daughter radionuclides:

- $T_{1/2_A} < T_{1/2_B}$:

$$^{56}_{28}Ni \xrightarrow{T_{1/2}=6.4 \text{ days}} {}^{56}_{27}Co \xrightarrow{T_{1/2}=77.3 \text{ days}} {}^{56}_{26}Fe_{\text{stable}} \tag{2.16}$$

First decay of the parent radionuclide, then decay of the daughter radionuclide only. There is no equilibrium between A and B radionuclides. The time of accumulation of maximum activity of the daughter radionuclide: $t_{max} = \frac{\ln(\lambda_B/\lambda_A)}{\lambda_B - \lambda_A}$

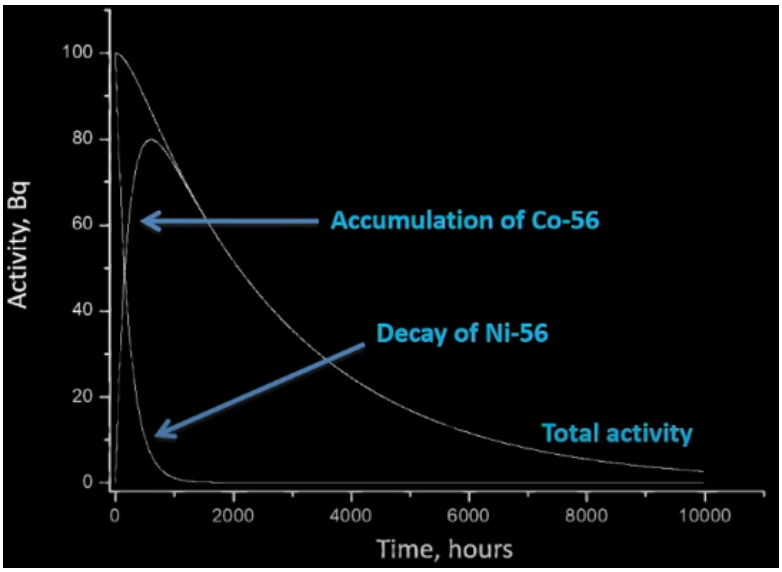


Figure 2.10. 1st case

- $T_{1/2_A} > T_{1/2_B}$:

$$^{100}_{46}Pd \xrightarrow{T_{1/2}=96 \text{ hours}} {}^{100}_{45}Rh \xrightarrow{T_{1/2}=21 \text{ hours}} {}^{100}_{44}Ru_{\text{stable}} \tag{2.17}$$

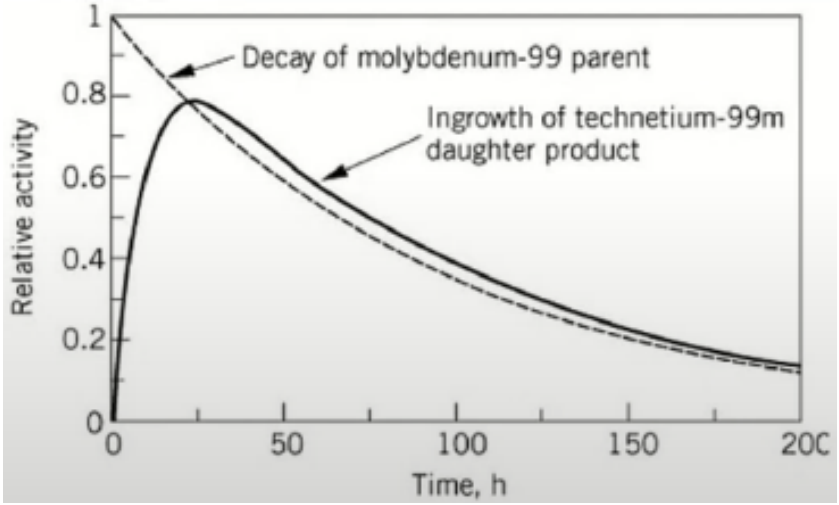


Figure 2.11. 2nd case

There is accumulation and then disintegration of the daughter radionuclide A:

$$\begin{cases} \frac{dN_A}{dt} = -\lambda_A N_A \\ \frac{dN_B}{dt} = \lambda_A N_A - \lambda_B N_B \end{cases} \xrightarrow{\text{if } A_{B,0}=0} \begin{cases} A_B = \frac{\lambda_B A_{A,0}}{\lambda_B - \lambda_A}(e^{-\lambda_A t} - e^{-\lambda_B t}) \\ \frac{A_B}{A_A} = \frac{\lambda_B}{\lambda_B - \lambda_A} \end{cases} \tag{2.18}$$

This is a case of **transient equilibrium**.

- $T_{1/2_A} \gg T_{1/2_B}$:

$$^{90}_{38}Sr \xrightarrow{T_{1/2}=30 \text{ years}} {}^{90}_{39}Y \xrightarrow{T_{1/2}=64 \text{ hours}} {}^{90}_{40}Zr_{\text{stable}} \tag{2.19}$$

This case is very similar to the previous one but here $\lambda_A \ll \lambda_B \Rightarrow A_A = A_B$.

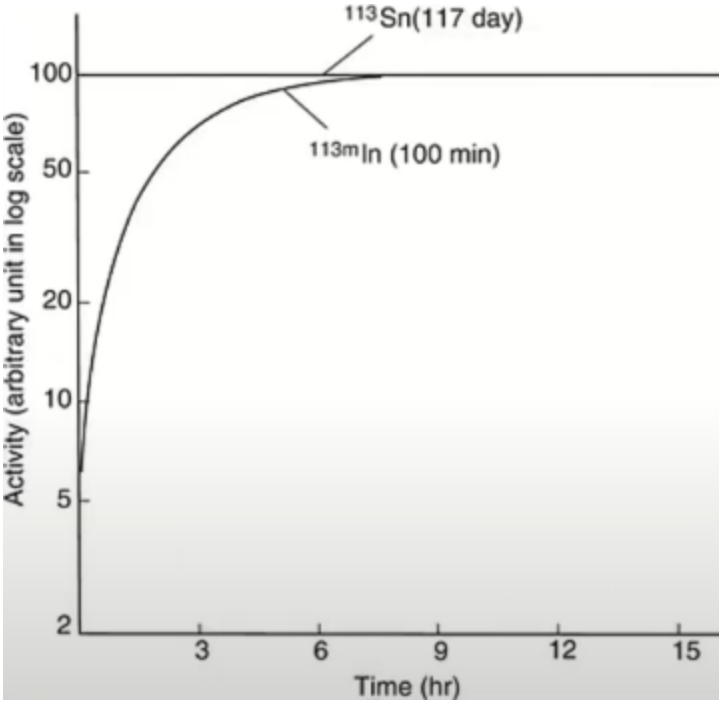


Figure 2.12. 3rd case

This is called secular equilibrium.

Radioactive equilibrium is a constant ratio of the parent and daughter radionuclides activities. It can be used in so-called **isotope generators**. Isotope generators are the systems where parent radionuclide has longer half-life than the daughter one (cases of transition or secular equilibrium). Over time the activity of the daughter radionuclide is accumulated and at a certain point we can remove it (fig.2.13) and use it for medical or scientific purposes for instant.

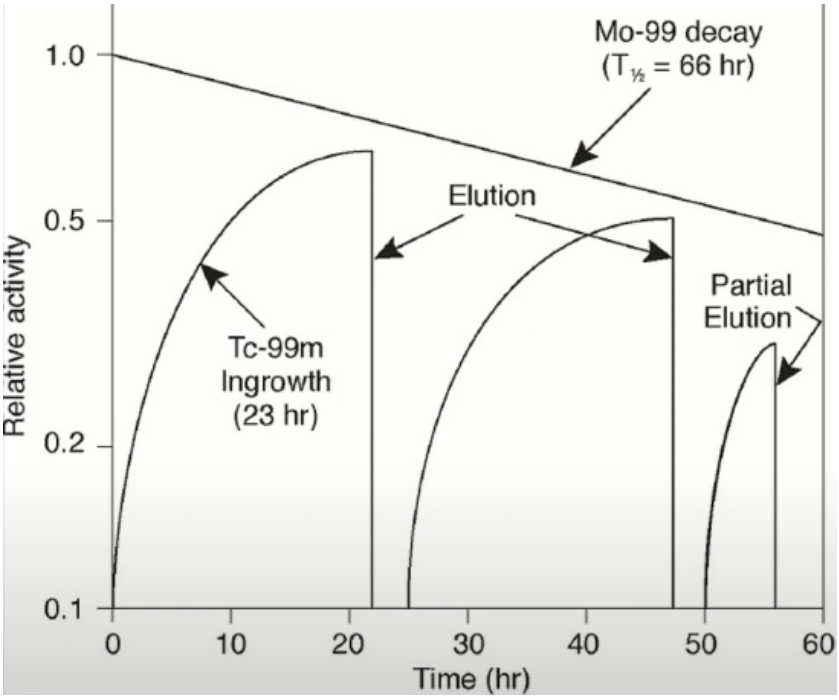


Figure 2.13. Isotope generators

The chains of radioactive decay also occur in nature and start with uranium-238, uranium-235 and thorium-232 (fig.2.14).

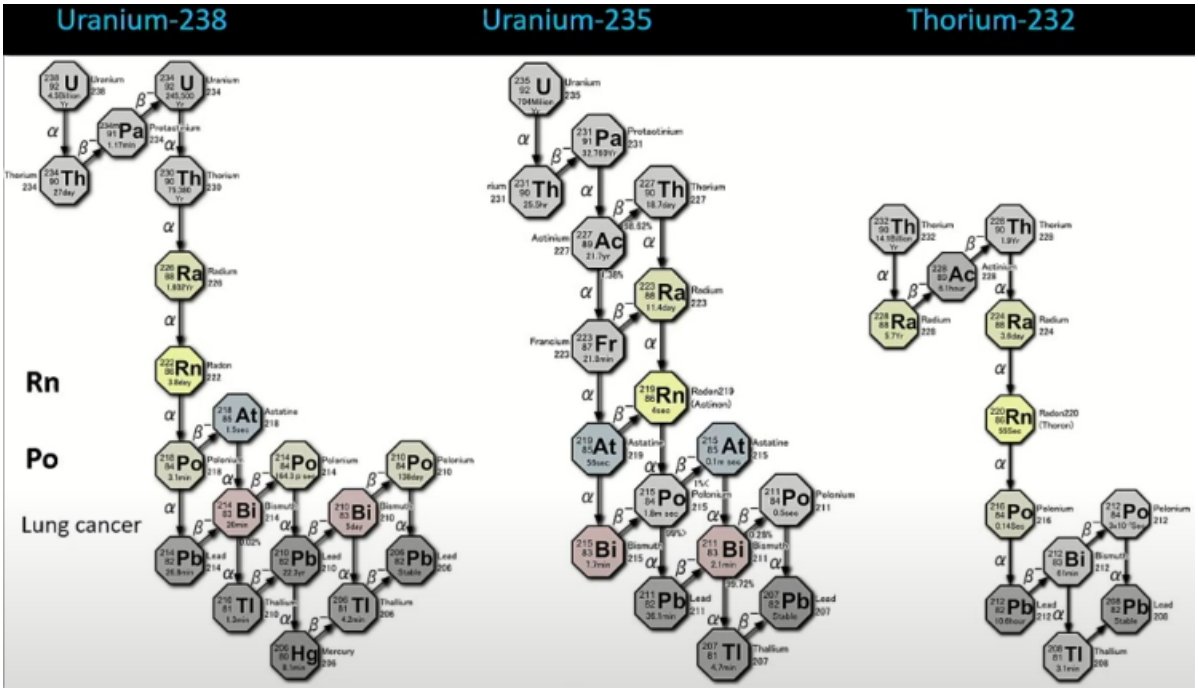


Figure 2.14. Radioactive natural chains

3. Lecture 3. Interactions of ionizing radiation with matter

Types of interactions

Studying the interactions of ionizing radiation with matter is of utmost importance as it determines the effects of ionizing radiation on both materials and living creatures so we can learn how to protect people from it and use it for our advantage.

Basically there are two types of interactions:

- **Elastic**: there is only change in momentum.
- **Inelastic**: Change in momentum, internal state, charges, etc
  - **Excitation**: transformation of atoms and molecules into a state with higher energy. It occurs when the energy deposited to the matter (ΔE) is less than the energy of ionization (E_I):

$$\Delta E < E_I \tag{3.1}$$

  - **Ionization**: transformation of neutral atoms and molecules into positively charged ions. Happens when

$$\Delta E > E_I \tag{3.2}$$

Heavy charged particles

Heavy charged particles are basically α-particles, protons, deutrons and nuclei. Heavy charged particles tend to leave straight tracks in the matter as they traverse. The loss of energy of a heavy charged particle when it travels through the matter is given by the Bethe-Bloch formula:

$$-\frac{dE}{dx} = \frac{4\pi}{m_e c^2} \cdot \frac{nZ^2}{\beta^2} \cdot \left(\frac{e^2}{4\pi\varepsilon_0}\right)^2 \cdot \left[\ln\left(\frac{2m_e c^2\beta^2}{I \cdot (1-\beta^2)}\right) - \beta^2\right] \tag{3.3}$$

where c - the speed of light, $m_e c^2 = 0.511$ MeV - the rest energy of an electron, Z - the charge of the particle, $\beta = \frac{v}{c}$, v - the speed of the particle, ε_0 - vacuum permittivity, n - electron density of the material, I - mean excitation potential.

There are two types of ionization:

- **Direct**: the electrons of matter that are released due to direct interaction with heavy charged particles are called δ-electrons and have energies $\sim 100 - 200$ MeV. The fraction of direct contribution is $\sim 20\%$ of the total ionization.
- **Indirect** ionization is caused by the δ-electrons themselves. The fraction of indirect contribution is $\sim 80\%$ of the total ionization.

The combined effect of direct and indirect contributions results in widening of the track of an α-particle (fig.3.1).

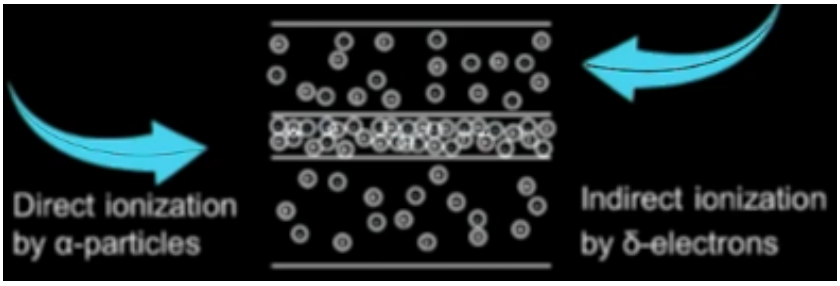
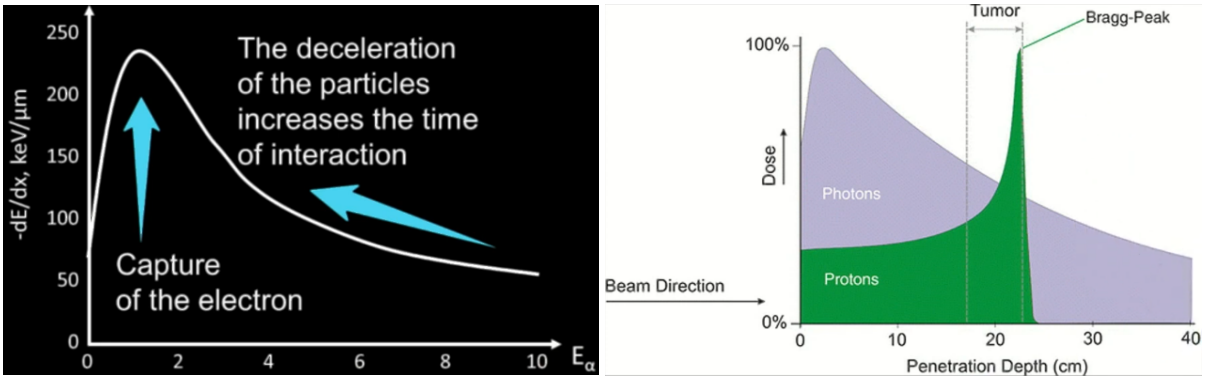


Figure 3.1. Track of an α-particle

The amount of ionized atoms depending on the energy of an alpha-particle is described by the curve in fig.3.2 **a)**.



a) **b**)

Figure 3.2. **a**):The loss of energy per distance VS the energy of an α-particle; **b**): The Bragg curve

It is easy to understand, that when an α-particle moves at a very high energy it moves really fast and there is not much time for it to interact with the electrons $\Rightarrow$ the loss of energy is small at a high energy region. As the α-particle decelerates it starts to interact with the electrons more intensively and might, at a certain energy capture one electron and than another. As soon as the α-particle has captured 2 electrons it becomes neutral and the ionization plummets to zero. Therefore, almost all energy of the α-particle is lost at the very end of its track.

The fact that after the maximum distance there are no heavy charged particles (like protons) and all their energy (the dose) is absorbed in a small area at the end of that distance (fig.3.2 **b)**) is widely used in medicine for the treatment of cancer and tumors. Such approach does not work with photons as there is no maximum range of absorption for them, their flux continuously decreases but does not go to zero.

Electrons

Equation (3.3) can also be applied to electrons in 2 regimes: slow electrons and ultra relativistic electrons. Apart from the above mentioned direct and indirect interactions in the case of electrons there also exist another process. The **bremsstrahlung radiation** occurs when an electron decelerates in the matter as any charged particle moving

with acceleration (or deceleration) emits electromagnetic waves. The intensity of such radiation is proportional to the square of acceleration:

$$W \sim a^2 = \left(\frac{1}{m} \cdot \frac{Z_A z e^2}{r^2}\right) \tag{3.4}$$

Notice, that $W \sim \frac{1}{m^2}$ so for light $\beta-$particles radiation is the second most significant channel of energy loss.

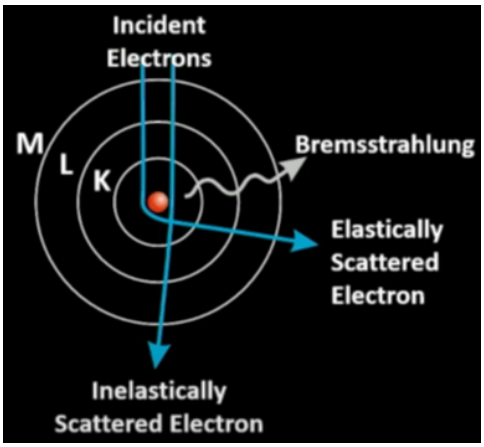


Figure 3.3. The bremsstrahlung radiation

It is wise to use light materials such as carbon or aluminum to shield from electrons because the intensity of the bremsstrahlung radiation, which is proportionate to square charge number of the element (Z_A^2) would not be high, as opposed to heavy materials like lead in which case the intensity of the radiation will be much higher.

The spectrum of the bremsstrahlung radiation is continuous as shown in fig.3.4

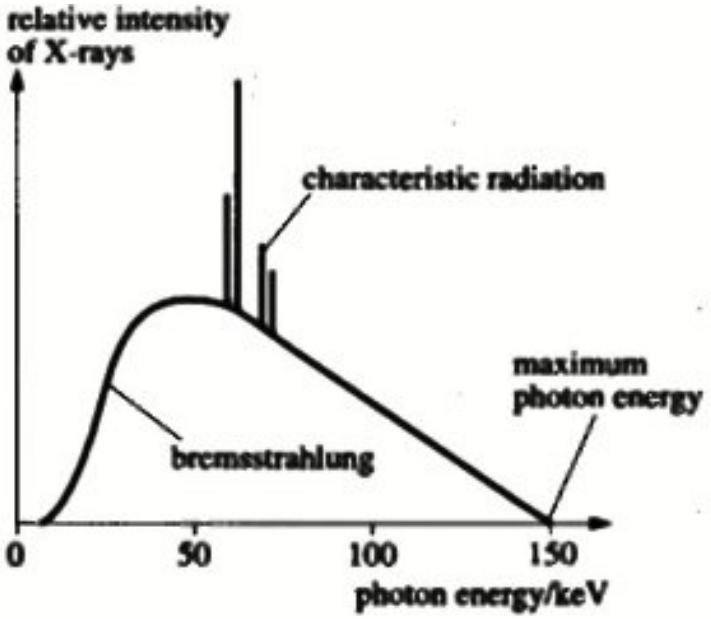


Figure 3.4. Spectrum of the bremsstrahlung radiation

Another type of interaction of electrons with matter is **Cherenkov radiation**. This radiation occurs when the velocity of an electron or other charged particle (v) is higher than the speed of light in the matter (c'):

$$v > c' = \frac{c}{n} \tag{3.5}$$

where c is the speed of light in vacuum, n is the refractive index for given matter.

For water $n = 1.33 \Rightarrow \frac{v}{c} = 0.75 \Rightarrow$ the energy of an electron should be $E_e > 0.267$ MeV for Cherenkov radiation to occur. Many natural $\beta-$radioactive radionuclides can emit electrons with even higher energies.

In layman's terms Cherenkov radiation can be explained as shock waves produced by a particle moving faster than light in given matter. The process is very similar to formation of a Mach cone around a supersonic aircraft (fig.3.5).

Figure 3.5. The Mach cone

The equation (3.6) gives the average square of the multi-scattering angle:

$$\langle\theta^2\rangle =\sim \frac{z^2 Z^2}{(pv)^2} \cdot l \tag{3.6}$$

where $z,\ Z$ are charges of the particle and the target nucleus, $p,\ v$ are the momenta and the velocity of the particle, l is thickness of the target.

The bigger the charge of the target and the thicker it is the larger the multi-scattering angle. The momenta $p = mv$, so the lighter the particle, the larger the multi-scattering angle.

Almost all alpha-particles stop at the same distance, but that is not the case for beta-particles. Due to multi-scattering different electrons stop at different distances and there is the **maximum range** of electron penetration.

Positrons

When we talk about ionizing radiation and beta-particles, we should remember that beta-particles are not only electrons but also positrons. Basically, positrons interact with

matter in the same way as electrons, with a slight caveat. At sufficiently low energies (having traveled long enough range) positrons can annihilate with electrons generating electromagnetic radiation (fig.3.6)

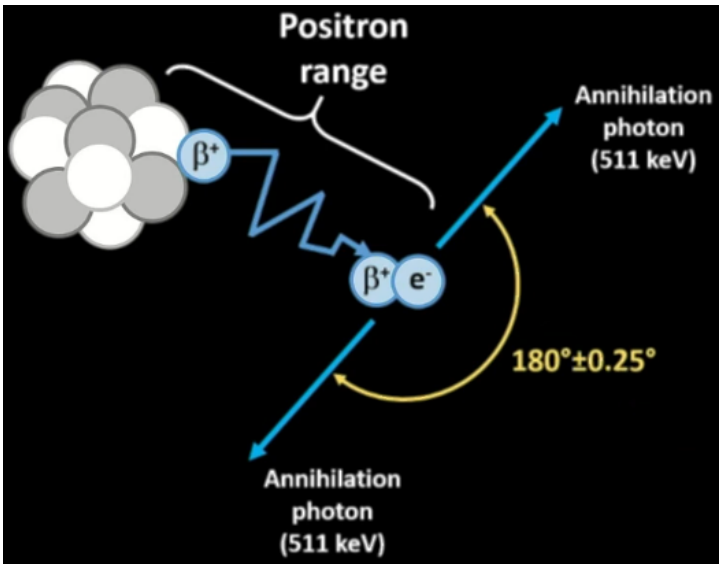


Figure 3.6. Electron-positron annihilation

Gamma-radiation

Photons have no mass and no charge and therefore can go through matter with very low level of interaction. There are three types of interaction of high-energy electromagnetic radiation with matter (fig.3.7)

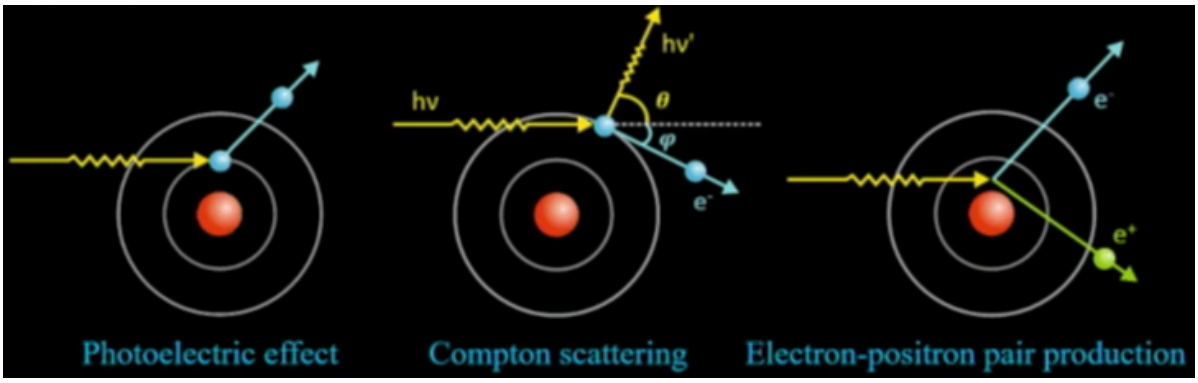


Figure 3.7. Interaction of $\gamma-$quanta with matter

In the process of the **photoelectric effect** the entire energy of a γ-quantum is transferred to an electron in the inner atomic shell $\Rightarrow$ as a result the electron is kicked out of the shell. The cross-section (probability) of such reaction depends on the charge of the matter and on the photon's energy (fig. 3.8).

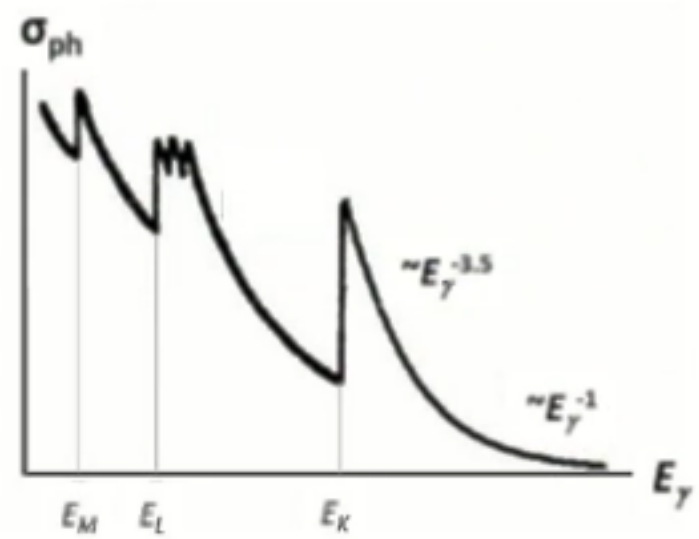


Figure 3.8. The cross-section of the photoelectric effect

$$\begin{cases} \sigma_{ph} \sim \frac{Z^5}{E^{3.5}_{\gamma}}, \text{ if } E_\gamma < 511\ KeV \\ \sigma_{ph} \sim \frac{Z^5}{E_\gamma}, \text{ if } E_\gamma > 511\ KeV \end{cases} \tag{3.7}$$

The vacancy created in the K shell is filled by an electron from higher levels (for example, the L-shell). Excesses of energy can be dissipated by X-ray photons (3.9 left) or transferred to another electron (3.9 right).

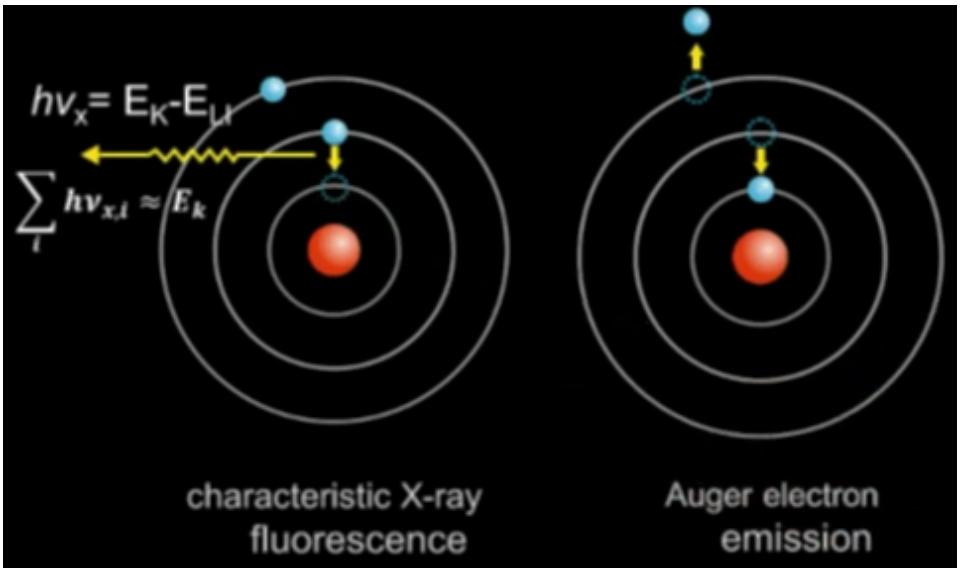


Figure 3.9. The ways of disposing of the energy excess

When an electron goes from a higher level to a lower it emits electromagnetic radiation with the energy equal to the energy difference between these two levels. This phenomenon can be used to analyze chemical composition of matter. Auger emission

can also be used for chemical composition analysis as well as for the determination of the valence state and local surroundings of the atom.

Another type of interaction of γ-quanta with matter is **Compton scattering**. In this case not all the energy of a photon is transferred to an electron but part of it is emitted as a photon with different energy. And there is also emission of the electron from the shell.

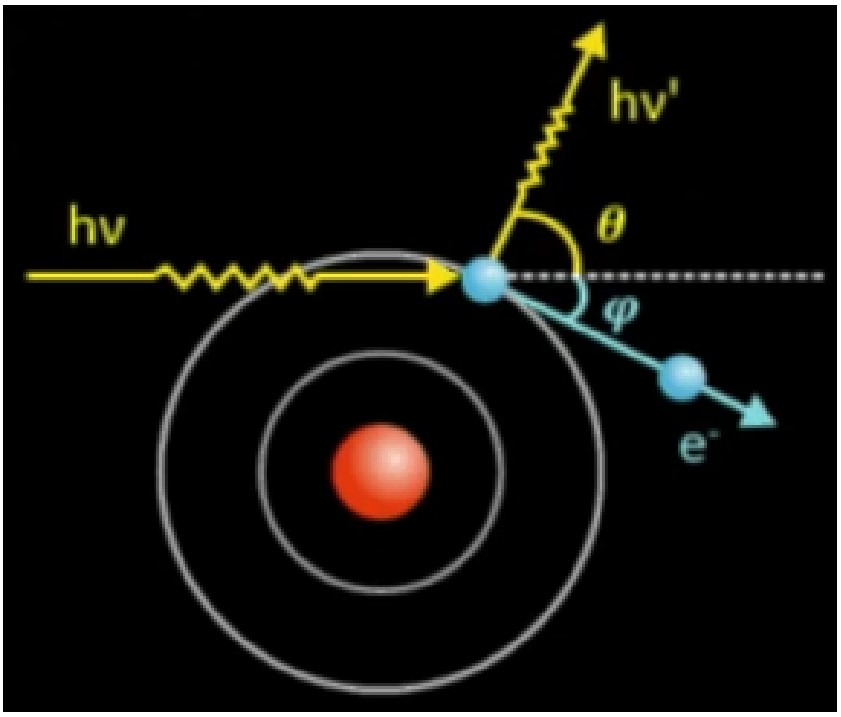


Figure 3.10. Compton scattering

The kinetic energy of the emitted electron may vary from 0 to E_{Compt}, where

$$E_{Compt} = E_\gamma[1 + (511)/2E_\gamma] \tag{3.8}$$

The cross-section of Compton effect is given by:

$$\sigma_c \approx \frac{Z}{E_\gamma} \tag{3.9}$$

The third type of interaction is the **electron-positron pair production** from a $\gamma-$quantum (3.11). The emitted positron will further annihilate with electrons of matter. This process is possible if the energy of the $\gamma-$quantum $E_\gamma > 1022$ KeV. The cross-section of this process is given by:

$$\sigma_{ep} \sim Z^2 \cdot \ln(E_\gamma) \tag{3.10}$$

Notice, that unlike the previous types of interactions, the cross-section of the electron-positron pair production grows with the photon energy E_γ.

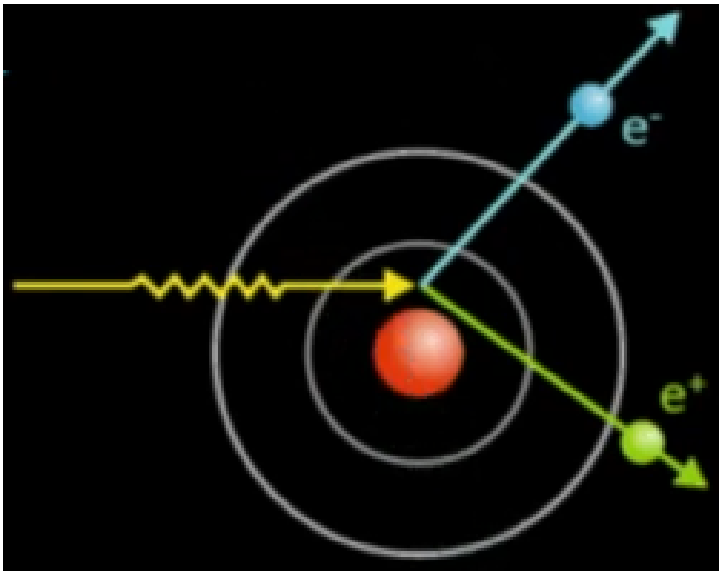


Figure 3.11. The electron-positron pair production

The comparison of the three types of interaction is shown in fig.3.12.

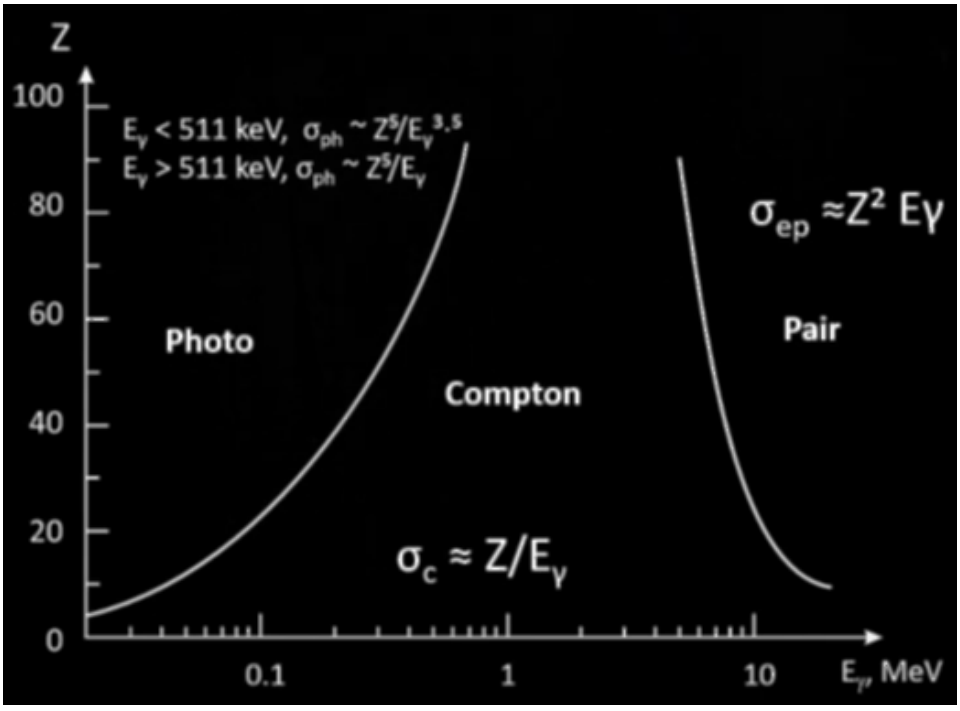


Figure 3.12. The comparison of the three types of interaction

We can clearly see, that the higher the energy of a photon E_γ the more probable Compton effect becomes. Photoelectric effect is more common for low-energy $\gamma-$quanta, that is why we usually use X-rays for chemical analysis. For the matter with very small charge the probability of the photoelectric effect is low and the main type of interaction in this case is Compton effect. In the photoelectric effect all energy of a

photon is transferred to an electron so there are no other photons produced. The similar situation occurs in the case of pair production. But in Compton effect there is emission of secondary photons. If we want to stop the radiation of photons we should use materials for which the photoelectric effect is more probable, that is to say heavy, dense materials like lead.

We can summarize the energy dependence of the reactions:

- Low energy photons: mainly photoelectric effect
- Medium energy photons: mainly Compton effect
- High energy photons: pair production

There are also other types of interaction of $\gamma-$radiation with matter such as the Mossbauer effect and photoinduced nuclear reactions.

For the $\gamma-$radiation there is no maximum range of radiation. The penetration ability of $\gamma-$quanta is described by the exponential law:

$$F_x = F_0 \cdot e^{-\mu/x} \tag{3.11}$$

where F_0 and F_x are the fluxes before and after the absorber, x is the thickness of the absorber in cm, μ is the attenuation coefficient in cm^{-1}.

The attenuation coefficient depends on the energy of photons and chemical composition of the absorber. The total attenuation coefficient can be represented as a sum of attenuation coefficients responsible for photoelectric effect, Compton scattering and pair production:

$$\mu = \mu_{ph} + \mu_c + \mu_{ep} \tag{3.12}$$

The fact that different matters have different of attenuation coefficient is used in X-ray scanning (fig.3.13).

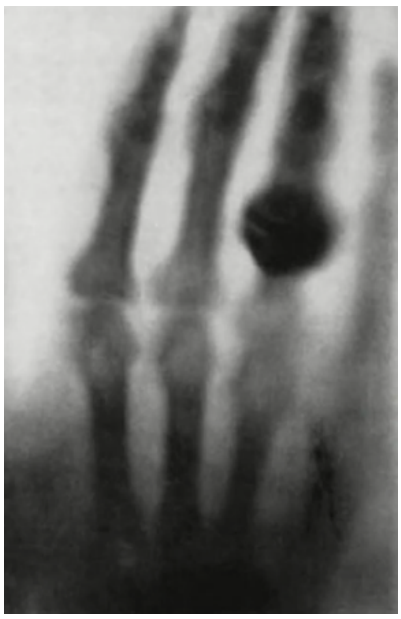


Figure 3.13. The x-ray picture of a hand with a ring on

Neutrons

Neutrons can be sorted by their energies in the increasing order:

1 Slow: $E_n \leq 100$ KeV

- 1a Ultra-cold: $E_n < 10^{-7}$ eV
- 1b Cold: $10^{-7} < E_n < 0.01$ eV
- 1c Thermal: $0.01 < E_n < 0.1$ eV
- 1d Epithermal: 0.1 eV $< E_n \leq 10$ KeV
- 1e Intermediate: $10 < E_n \leq 100$ KeV

2 Fast: 100 KeV $< E_n <$ 100 MeV

3 Relativistic: $E_n >$ 100 MeV

Cross-sections for neutron interactions with nuclei are, on average, inversely proportional to the speed of neutrons ($\sim 1/v$). All the neutrons produced are initially fast and we need to slow them down for practical applications.

Having no charge, neutrons cannot effectively interact with electrons, they mostly interact with atomic nuclei which is a rare event ⇒ neutrons go through matter with very high efficiency. When a neutron meets a nucleus inelastic scattering occurs (fig.3.14)

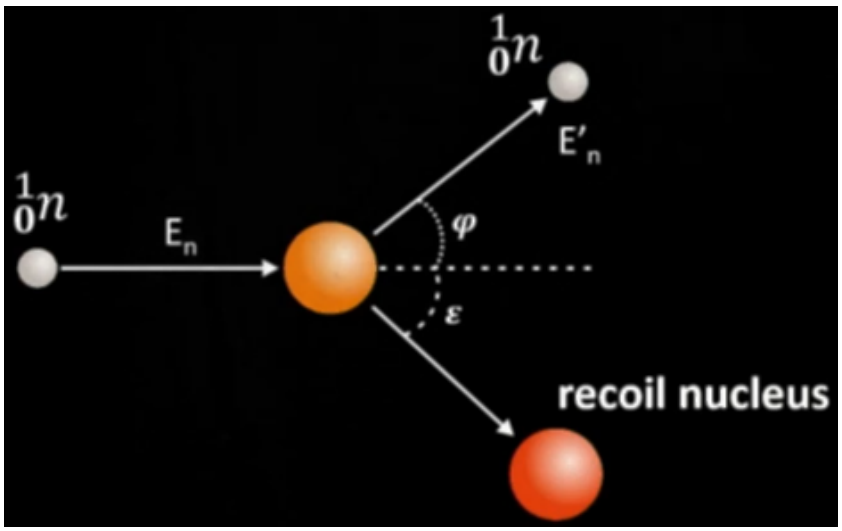


Figure 3.14. Neutron energy loss in a collision

The difference between the remaining and the initial energy of a neutron after the collision is:

$$\Delta E = E_{n,0} \cdot \frac{2Am_n}{(A+m_n)^2} = E_{n,0} \cdot \frac{2A}{(A+1)^2} \tag{3.13}$$

where $E_{n,0}$ is the initial energy of a neutron, A is the mass number of the matter, m_n is the mass of the neutron.

Therefore, light nuclei are the best at decelerating neutrons.

4. Lecture 4. Detection of ionizing radiation

Principles of ionizing radiation measurement

All detection methods are based on interactions of ionizing radiation with matter.

Mechanisms	Response	Detectors
Ionization	Detection of electric current	Ionization
Excitation	Detection of visible light	Scintillation
Production of defects	Analysis of defects	Track detectors

The modes of operation of such detectors can be either **pulsed** (measuring discrete responses of each event) or **integral** (collecting the signal from many events and then analyze it).

It is crucial to remember, that there exists **background radiation** everywhere. Therefore, when we measure the counting rate we never directly measure the activity of the sample.

The counting rate I_c of the specimen with background as indicated by the detector is given by:

$$I_c = p\varphi A + B \tag{4.1}$$

where p is probability of a certain decay mode for given radionuclide, φ is the coefficient of registration, A is absolute activity of the specimen, B is background.

The coefficient of registration (or detection efficiency) φ shows that not all of the particles that came to the detector can be detected. The coefficient of registration depends on other parameters:

$$\varphi = \varepsilon\eta kSq \tag{4.2}$$

where ε is detector efficiency - the probability that a particle that deposited some of its energy into the atoms of the detecting medium will be detected, η is geometry factor - the relative position of the specimen and the counter, k is the attenuation - absorption and scattering of particles by matter between the source and the detector, S is self-attenuation - absorption and scattering of particles by the material of the source itself, q is the back-scattering coefficient - accounts for the scattering of particles from the substrate material.

Another very important parameter is called the **"dead time"**, which is the time between two events that still can be detected as separated events. When a detector registers a particle it needs some time (the "dead time") to return to the initial state and get ready for another particle coming. So if a particle comes before the detector resets this particle will not be detected. For this reason a correction for the counting rate is introduced:

$$I_r = \frac{I_c}{1 - \tau \cdot I_c} \tag{4.3}$$

where τ is the "dead time".

Having taken into account all the parameters we are ready to write the general formula for calculation of the activity:

$$A = \frac{1}{p\varphi} \cdot \left(\frac{I_c}{1 - \tau \cdot I_c} - B \right) = \frac{1}{p\varepsilon\eta kSq} \cdot \left(\frac{I_c}{1 - \tau \cdot I_c} - B \right) \tag{4.4}$$

Track detectors

The first track detectors detectors historically were photographic plates. **Radiography** in radiochemistry is a 2D research method. There two types of radiography:

- Passive radiography (without external source) - detection of the own $\alpha-$, $\beta-$, $\gamma-$ radiation
- Induced radiography - under the influence of external excitation ($\gamma-$, $n-$ activation) we detect $\alpha-$, $\beta-$, $\gamma-$ radiation

Combining both of these methods helps to distinguish between uranium and plutonium. Plutonium has higher cross section for nuclear fission and thus we can first use passive radiography analyzing the distribution of $\alpha-$emitting radionuclides (both plutonium and uranium in fig.4.1 left), then we use induced radiography to see where exactly the plutonium is located (fig.4.1 right) and the rest of the signal will be attributed to uranium.

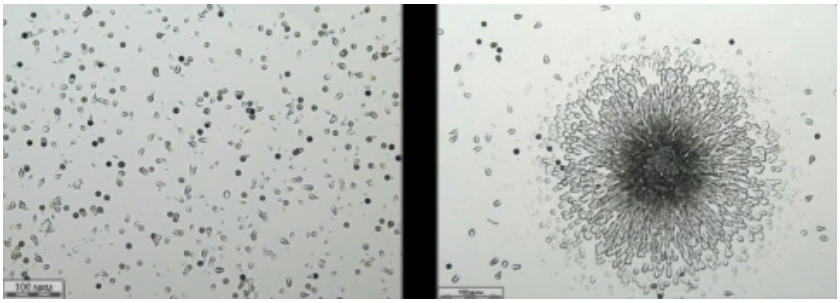


Figure 4.1. Incorporation of passive and induced radiography methods

For radiography we can use **solid-state nuclear track detectors**. Heavy charged particles produce damage to the microstructure of the detector material as they pass through it. Those defects are made visible by chemical etching. Then we analyze the tracks using microscope, count the number of tracks and, in case of alpha-particles, judging by the shape of the track we can even determine the energy of an alpha particle.

Ionization detectors

Ionization detectors can be divided into two groups:

- **Gas-filled** detectors consist of a cylindrical chamber filled with inert gas. There are two electrodes connected to the chamber: the anode and the cathode with high voltage applied to these electrodes. Without ionizing radiation in the chamber there is no electric current. But once ionizing radiation comes inside the chamber, it ionizes the gas in it creating free charges that in turn go to corresponding electrodes and produce electric current. By analyzing the electric current we can estimate the amount of particles and even their energies. Thus we have information about radionuclide composition of our sample.

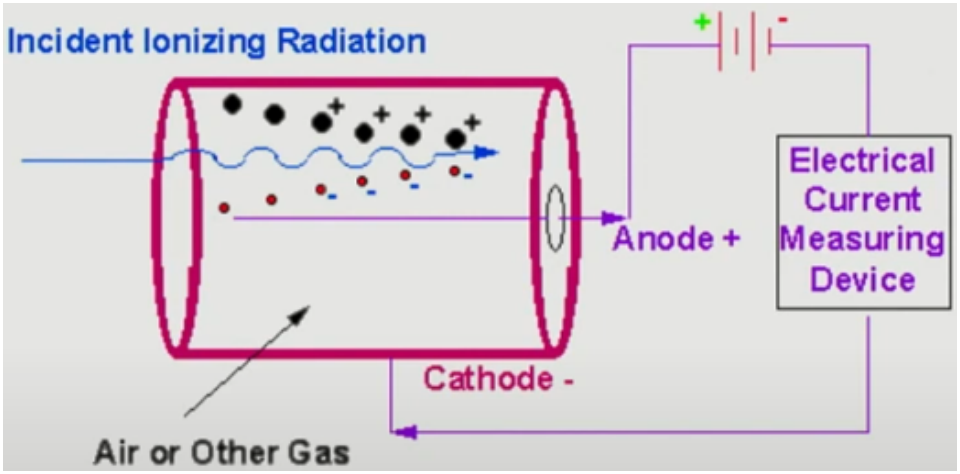


Figure 4.2. The scheme of a gas-filled detector

There is special dependence of generated charges inside the chamber on the applied voltage. The plot can be divided into regions (fig.4.3).

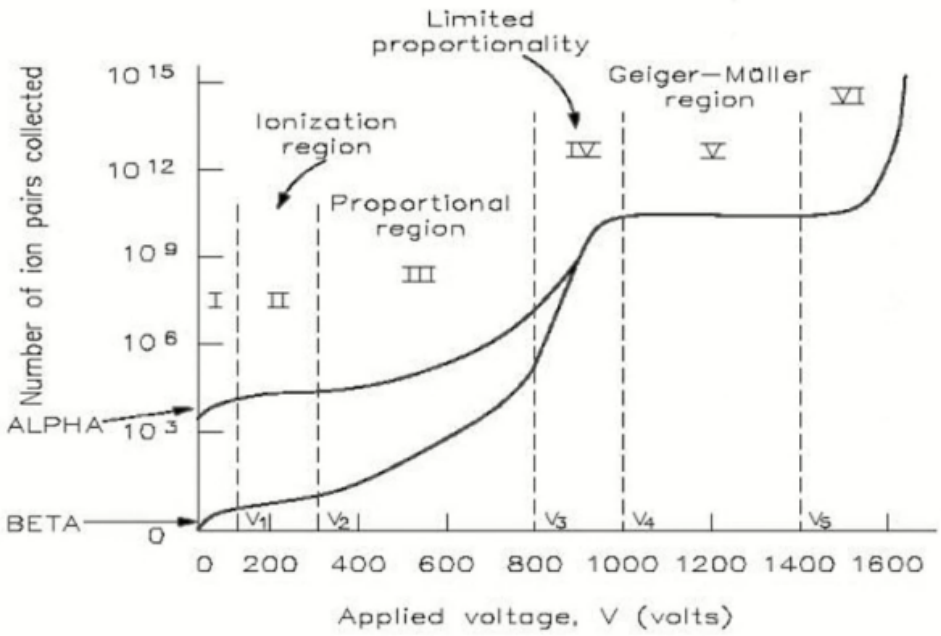


Figure 4.3. Number of generated charges VS voltage

In the 1st region ionizing radiation produces charges that move to the electrodes. But charges move slowly due to low voltage and may recombine and decrease the electric current current. The dependence of current on the voltage is in accordance with the Ohm law. As the voltage increases the speed of charges also increases and there is not enough time for them to recombine, so all charges reach the electrodes and produce maximum possible current. This is the ionization region (fig.4.4) and there are detectors called **ion chambers**, that work in this region. The created electric current directly depends on the energy of a particle. The disadvantages of

ion chambers are low signal and the fact that the amplitude depends in the entry point of a particle. Such detectors are used for alpha spectroscopy and individual dosimetry.

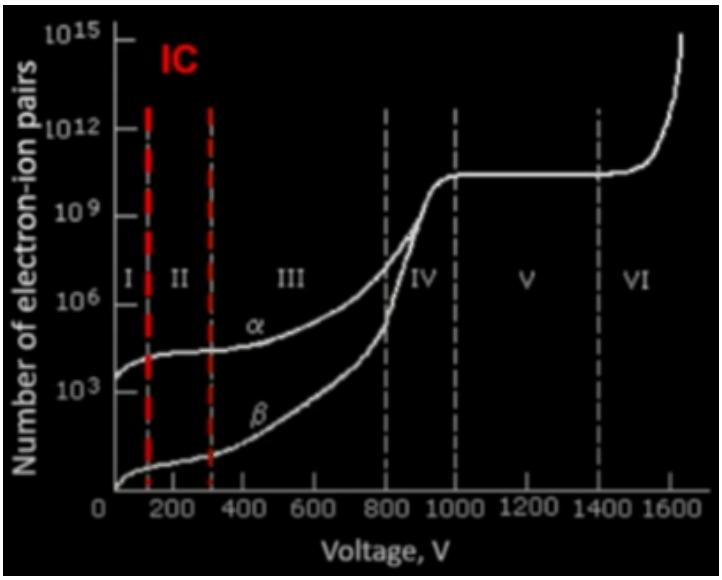


Figure 4.4. Ionization region

The next region is the proportional region (fig.4.5). In this region the charges are accelerated to such velocities that they themselves can ionize the gas in the chamber. The charge amplification may reach $10^3 - 10^6$ times. The amplification coefficient is constant at a certain voltage and does not depend on the energy of an initial particle: the higher its energy the higher the generated current is. The **proportional counters** are used in spectroscopy but their main drawback is low energy resolution.

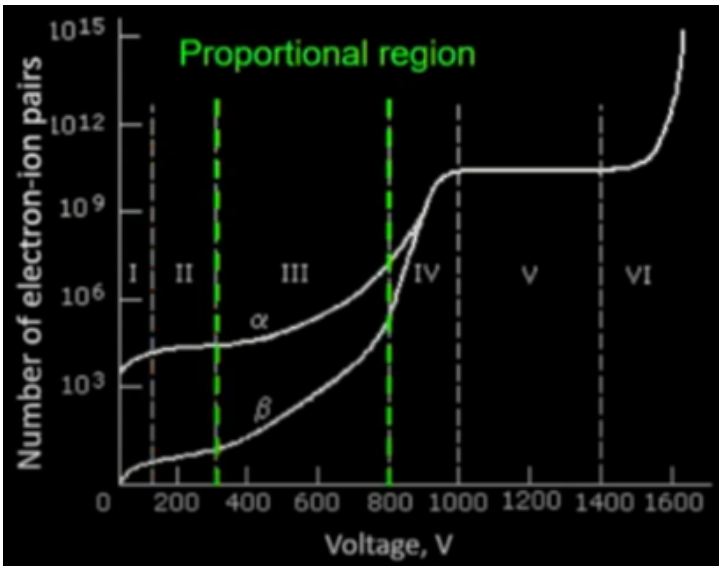


Figure 4.5. Proportional region

In the limited-proportionality region (number IV in fig.4.3) the amplification coefficient is no longer a constant and as a consequence this region is not useful for spectroscopy.

The Geiger-Mueller region (frig.4.6) is used in **Geiger-Mueller counters**. In this region the accelerated particles have such high energies, that they ionize the whole gas in the chamber and the corona discharge occurs. The ionization of the whole chamber takes place even for a single detected particle. The charge amplification is $10^8 - 10^{10}$. One drawback is that we cannot say anything about the energy of the initial particle nor what particle it is: beta, photon or something else. Another disadvantage is the long "dead-time" of the counter - the amount of time after registering a particle needed for relaxation to the initial state. During this time the counter is unable to detect new particles. Geiger-Mueller counters are used in dosimetry and beta-counting. We cannot register alpha-particles with them since the walls of the detector are thicker than an alpha-particle can penetrate.

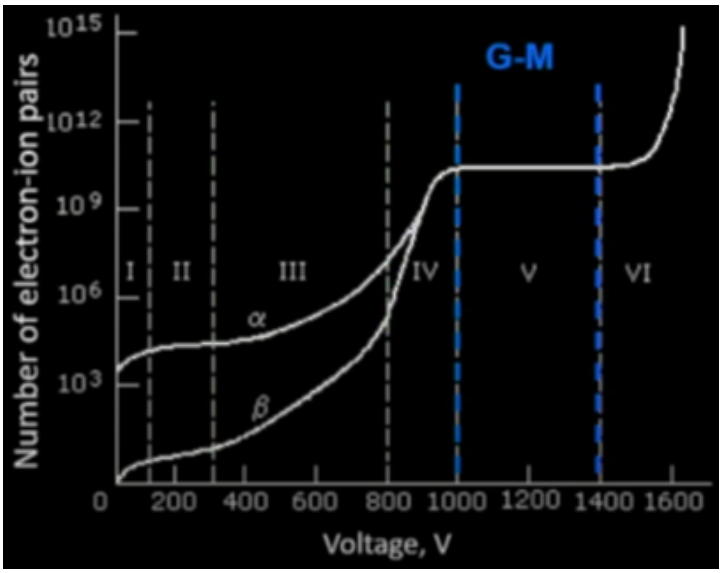
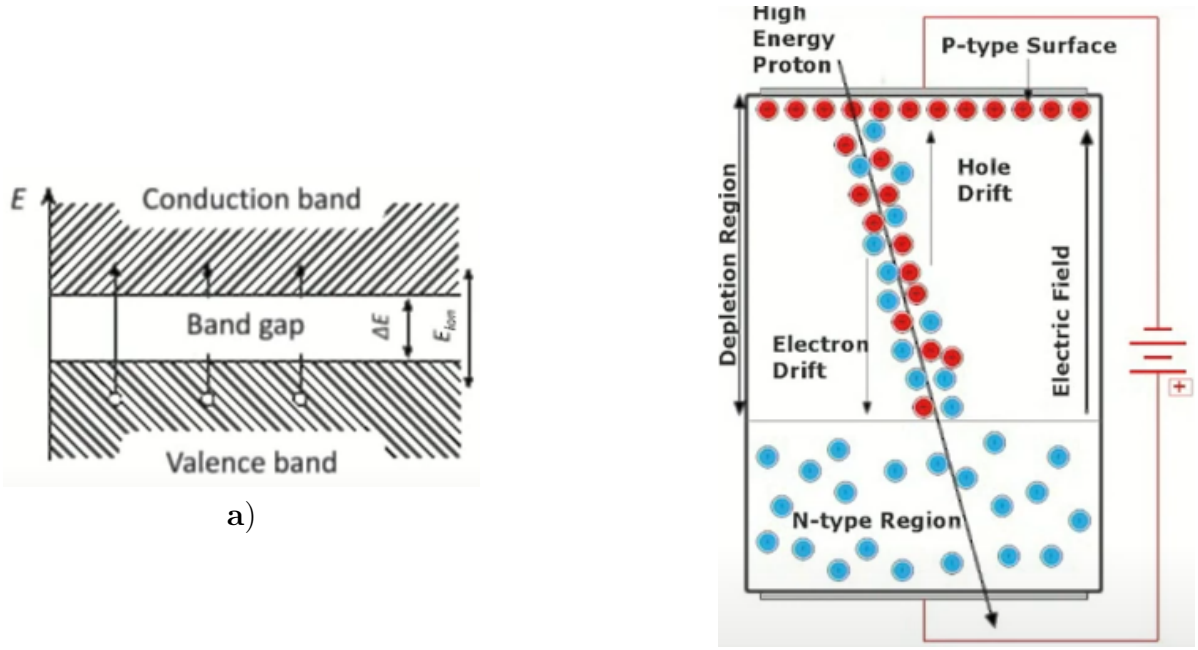


Figure 4.6. Geiger-Mueller region

- The second type of ionization detectors is **semiconductor detectors** which are basically ionization chambers with a semiconductor crystal instead of gas. Semiconducting materials are defined according to the band gap size (2-5 eV) (fig.4.7 **a)**). Semiconductor detectors have high resolution but lower than the gas-filled chamber efficiency. The principal of working is pretty much the same: initially there are no free charges inside the detector, but as the ionizing radiation comes it creates electrons and positively charged vacancies called holes. These charges then go to corresponding electrodes and produce electrical current (fig.4.7 **b)**).



a)

b)

Figure 4.7. **a**):Band gap for semiconductors; **b**): The scheme of a semiconductor detector

There are two main types of detectors, depending on the geometry - coaxial and well detectors. In the case of coaxial detectors the sample is placed in front of the detector, as opposed to the well type where the sample is located inside the detector (fig.4.8).

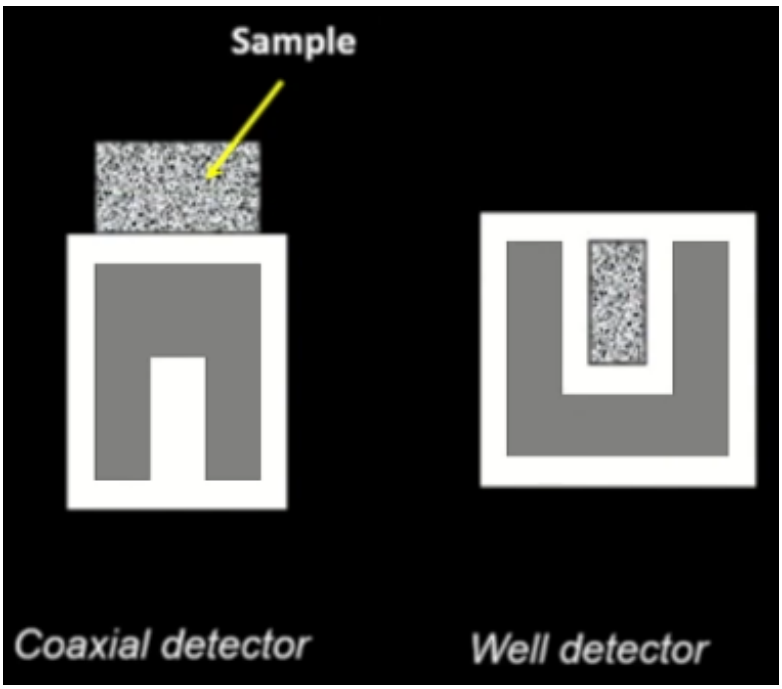


Figure 4.8. Coaxial and well detectors

Semiconductor detectors are mainly used for alpha- (silicon detectors) and gamma-spectrometry (germanium spectrometry). Due to low penetration ability of alpha particles, to detect them we have to create vacuum inside our system. We also have to shield the detection system of the background radiation as shown in fig.4.9

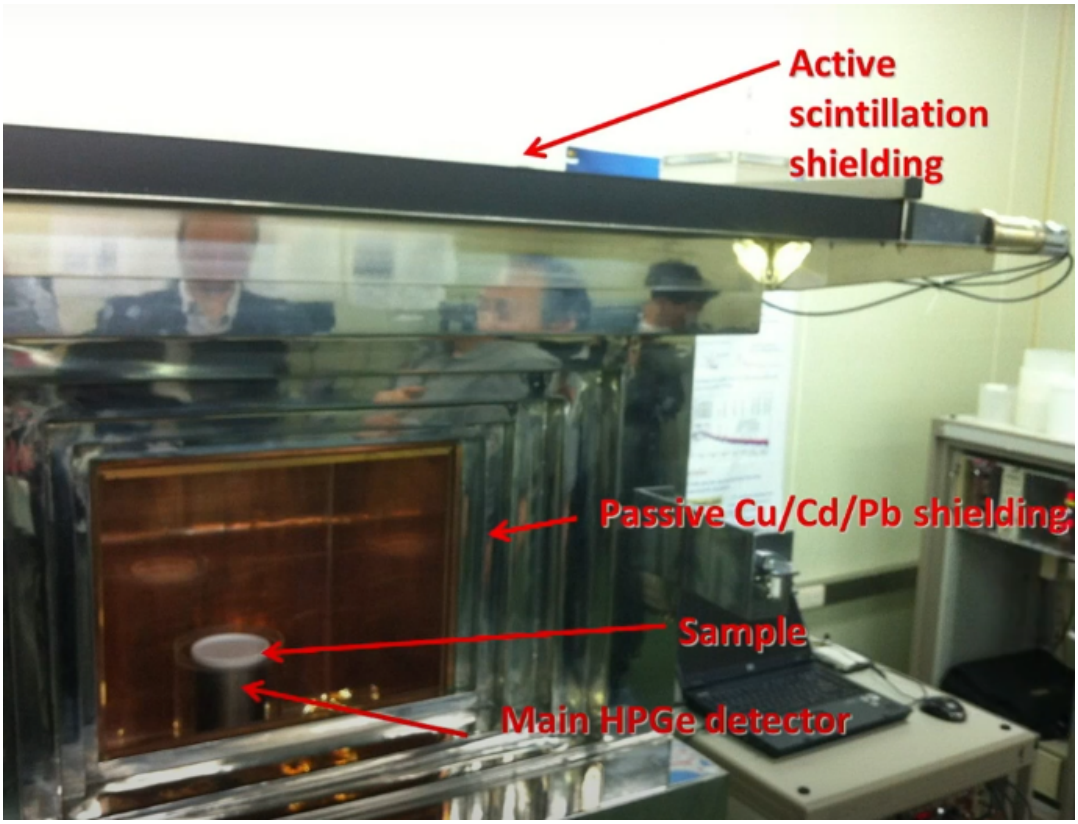


Figure 4.9. The example of shielding of the detection system

Scintillation detectors

- Solid scintillation detectors
  - Organic: Anthracenes, stilbene, naphtalene
  - Inorganic: $NaI(Ti)$, $LaBr_3(Ce)$, $ZnS(Ag)$, $Bi_4Ge_3O_{12}$, etc
- Liquid mixture: 2,5-diphenyloxazole, pseudocumene, etc

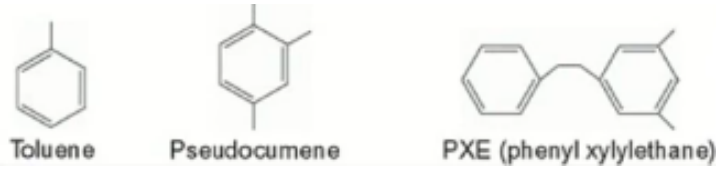


Figure 4.10. The organic molecule

The scheme of the scintillation detector is shown in fig.4.11. The main idea is that the scintillator generates visible light by the action of ionizing radiation. As he visible light reaches the photo multiplier tube it induces the photoelectric effect and creates a photo electron. Then the photo electron interacts with special electrodes called dynodes which accelerate the primary photo electron and produce more electrons in the ionization process. Thus, we amplify the signal. As a result, a signal in form of electric current is created. The more the energy of a registered particle, the higher current we get at the end - scintillation detectors are proportional detectors and can be used for spectrometry.

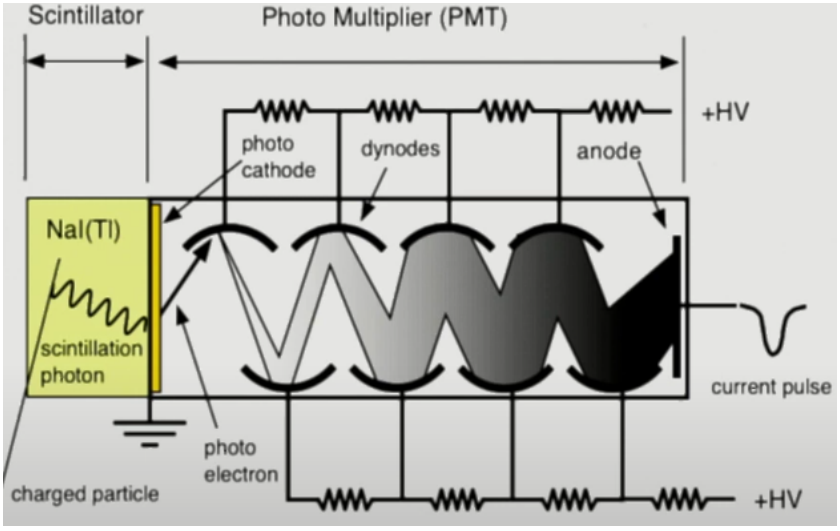


Figure 4.11. The scheme of the scintillation detector

Photo multiplier tube creates so called "dark current" - the current generated without the action of light and is a background noise. This "dark current" can be subtracted by virtue of using a special system called coincidence scheme (fig. 4.12).

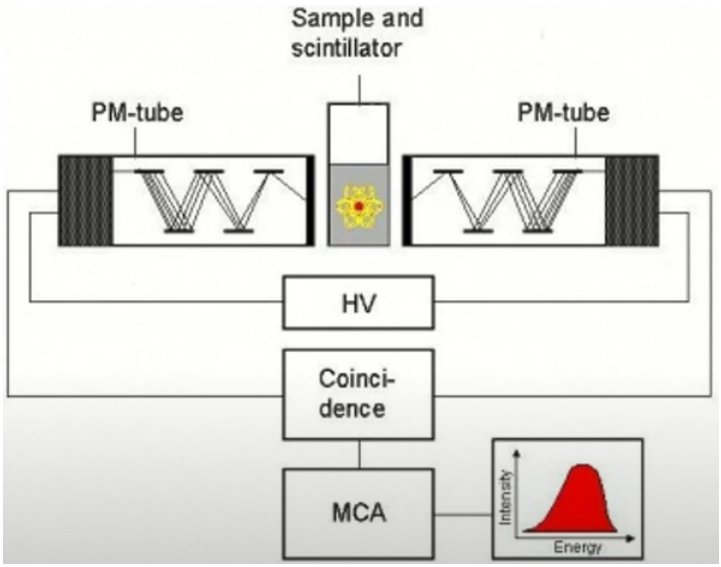


Figure 4.12. The coincidence scheme

If there is a signal generated by one PM-tube and no such signal coming from the other one the event will be rejected. True event, such as radioactive decay creates light propagating in all directions and thereby affects both PM-tubes.

Liquid Scintillation Counting (LSC) uses implies mixing the radioactive sample with the so called "scintillator cocktail" that consists of solvent and scintillator (the minor fraction of the cocktail). Molecules of the solvent meet the ionizing radiation first. The solvent molecules then meet and transfer their energy to the scintillator molecules that in turn emit visible light going to the PM-tubes. In the way of solvent transferring its energy to the scintillator there might be chemical quenching of the signal. If there are colored molecules that can capture the visible light the color quenching will occur (fig.4.13).

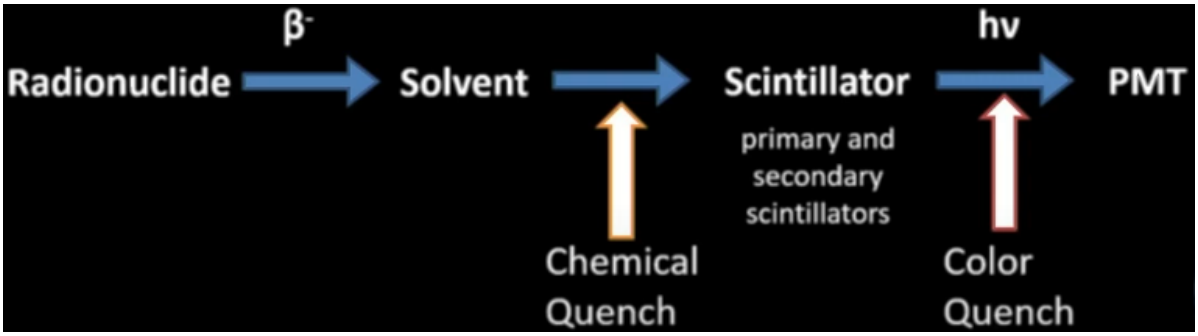


Figure 4.13. The LSC scheme

As a result of both chemical and color quenching fewer photons reach the PM-tubes and to the detector the fewer photons mean lower energy initially transferred to the system. As a result, we observe the shift of the energy peak to the lower values of energy (fig.4.14).

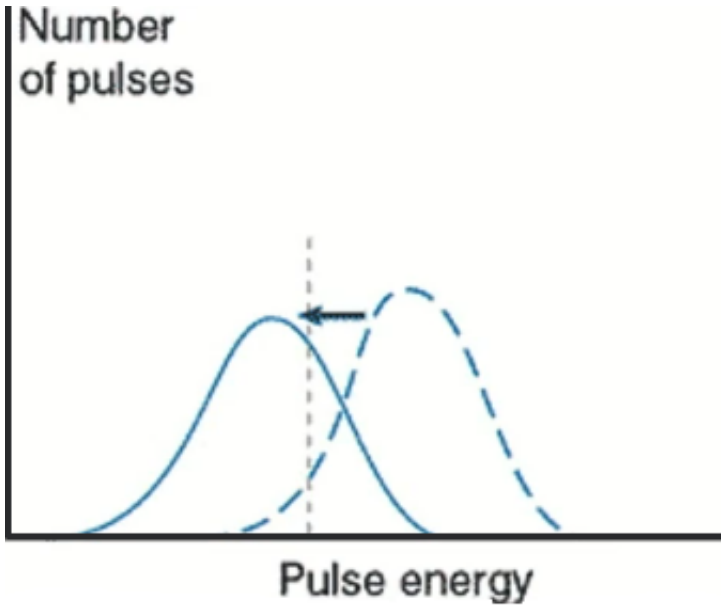


Figure 4.14. The energy shift

Not only does the peak shift to the left but also the area under the peak that represents the number of events will decrease due to the color quenching. The whole process is shown in fig. 4.15.

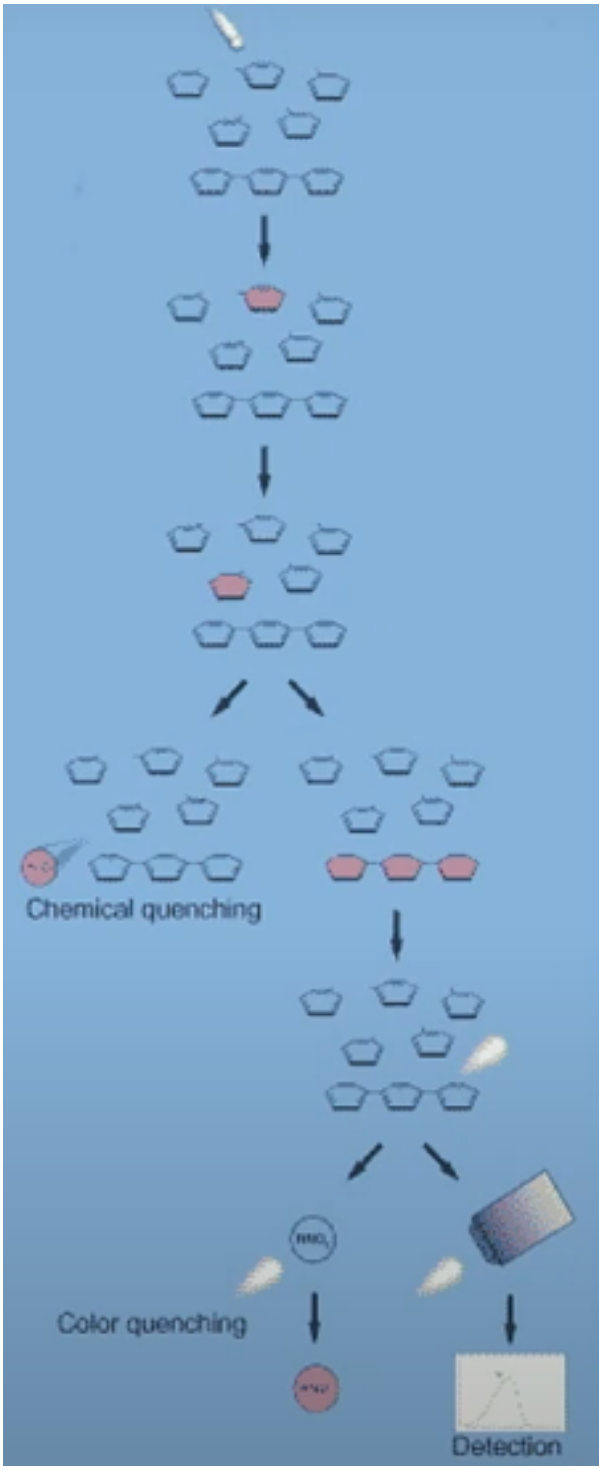


Figure 4.15. The LSC process

5. Lecture 5. Methods for measuring alpha-, beta-, gamma- radiation and neutrons

Gamma spectrometry

Gamma spectrometry is a very convenient method for measuring for measuring the activity of gamma-emitting radionuclides in matter because it does not require complex chemical preparation of the sample. We can measure a wide variety of different gamma-radioactive nuclides over a broad kinematic area.

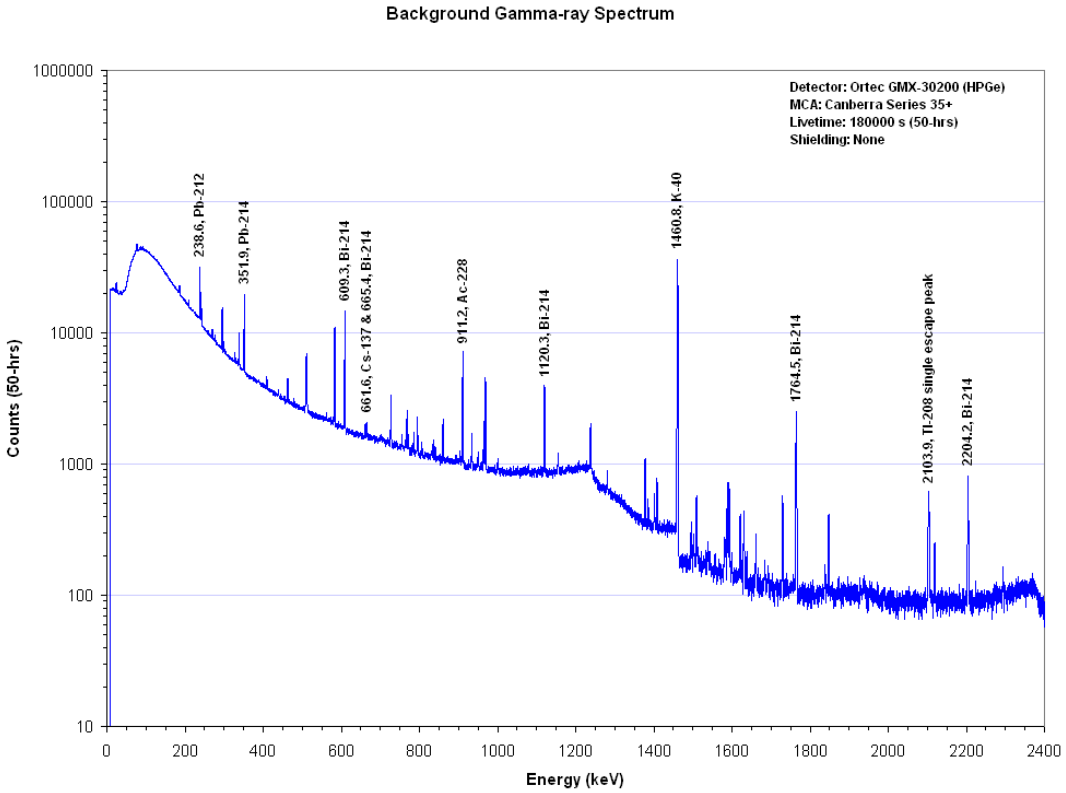


Figure 5.1. Gamma spectrum of background radiation

What peaks can we find in a gamma spectrum?

- **Total absorption peak** E_γ

  All processes of interaction of a gamma-quantum with matter (photoelectric effect, Compton scattering, electron-positron pair production) are quite fast ($\sim 10^{-12}\ s$) and are detected as a single event due to the limited time resolution of the detector. So all the energy of the initial gamma-quantum is transferred to the detector and we observe a single peak (fig.5.2).

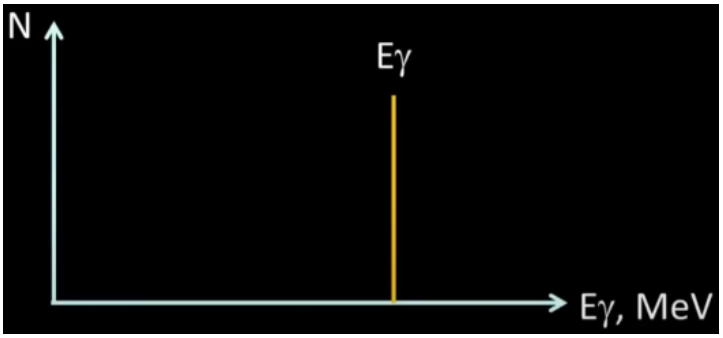


Figure 5.2. The total absorption peak

- **Compton continuum** from 0 to $E_\gamma \cdot \left[1 + \left(\frac{511}{2E_\gamma}\right)\right]^{-1}$ KeV.

  During Compton scattering only part of the initial photon's energy is transferred to an electron. Compton scattering is a stochastic process ⇒ even distribution of the scattered energy (fig.5.3).

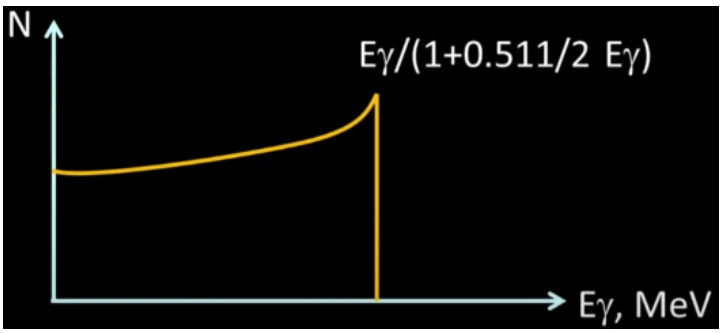


Figure 5.3. The Compton continuum

- **Escape peaks**

  If $E_\gamma > 1022$ KeV ⇒ production of e^+e^- pairs is possible. A positron can annihilate in matter creating two secondary $\gamma-$quanta each with the energy 511 KeV. Then three cases are possible:

  1) Both $\gamma-$quanta are absorbed by the matter, hence all the energy of the initial photon is registered (fig.5.4 right), resulting in the peak of total absorption (fig.5.5 the right peak)
  2) One of the released $\gamma-$quanta is absorbed, but the other escapes the matter volume undetected (fig.5.4 middle) ⇒ we lose 511 KeV of energy ⇒ we observe a single escape peak at the energy value equal to $E_\gamma - 0.511$ MeV (fig.5.5 the middle peak)
  3) Both $\gamma-$quanta escape the matter volume undetected (fig.5.4 left) ⇒ we lose 1022 KeV of energy ⇒ we observe a double escape peak at the energy value equal to $E_\gamma - 1.022$ MeV (fig.5.5 the left peak)

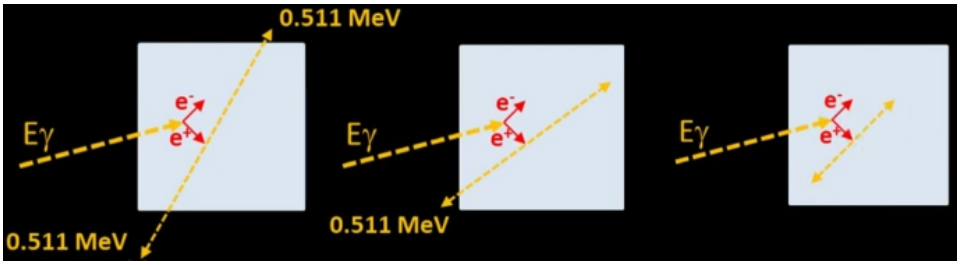


Figure 5.4. e^+e^- pair creation and positron annihilation

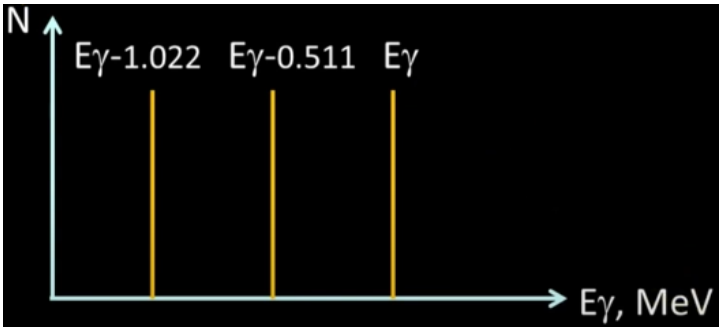


Figure 5.5. Escape peaks

- **Annihilation photon peak** and **511 KeV peak**

  Th annihilation process takes some tine as the positron has to decelerate to the velocities of the thermal motion. Hence the processes of the absorption of the initial $\gamma-$quantum and the release of the annihilation $\gamma-$quanta are separated in time (fig.5.6).

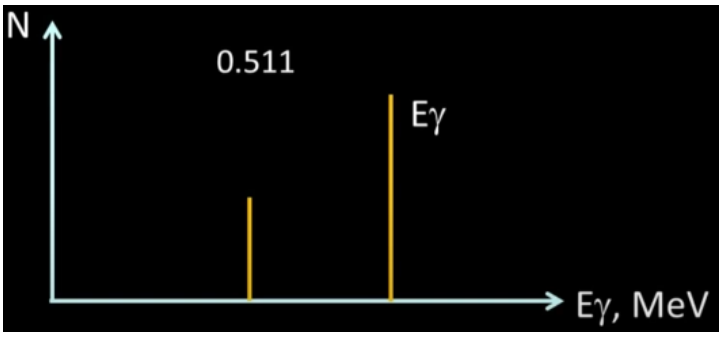


Figure 5.6. Annihilation photon peak, 511 KeV peak

- **Backscattering peak** and **Shielding characteristic radiation peak**

  1) Backscattering peak is directly related to the initial energy of the $\gamma-$quantum. Usually we surround our sample by lead shielding to eradicate the background radiation . The $\gamma-$quanta emitted from the sample go in all directions thus many will reach the lead shielding and sometimes scatter back to the detector (fig.5.7 right). Such photons will off course lose some energy and have energy

equal to $E_\gamma \cdot \left[1 + \left(\frac{511}{2E_\gamma}\right)\right]^{-1}$ KeV and we will observe the middle peak in fig.5.8.

2) Shielding characteristic radiation peak (fig.5.8 the left peak) appears when the initial photon is absorbed in the lead shielding in the process of the photoelectric effect. Then a lead atom emits a photo electron from the K-shell with the energy of 88 KeV (fig.5.7 left).

Notice, however, that the shielding characteristic radiation peak does not depend on the initial photon's energy and only depends on the material of the shielding. Thus we can reduce this peak by putting additional shielding made of lighter elements such as copper that will absorb the initial photon and not emit anything back.

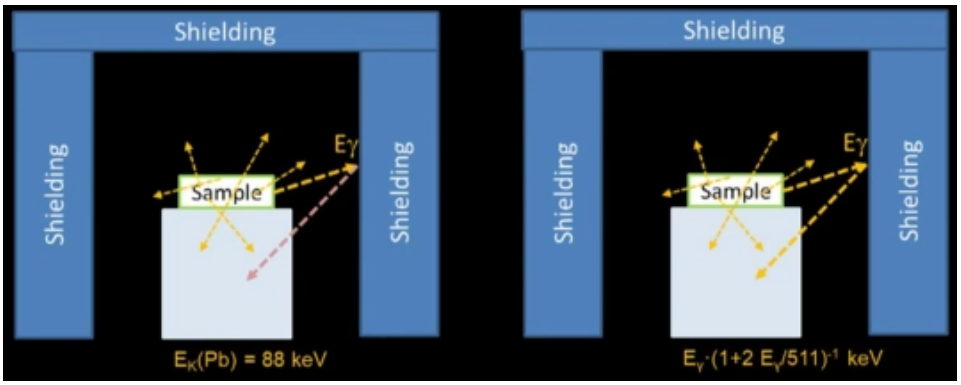


Figure 5.7. Consequences of lead shielding

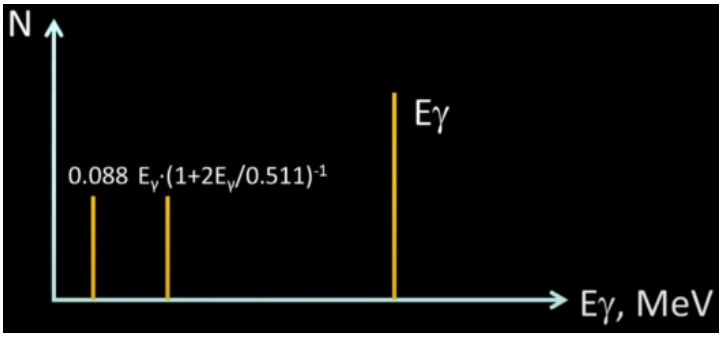


Figure 5.8. Backscattering peak and Shielding characteristic radiation peak

- **Characteristic escape peak of the detector material**

In this case a photo electron or a Compton electron is emitted in the matter of the detector and we cannot register it, but we observe the scattered $\gamma-$quantum with the energy of $E_\gamma - E_x$, where E_x is the energy if the electron binding in the atom (fig.5.9), thus we observe the corresponding peak (fig.5.10 the left peak).

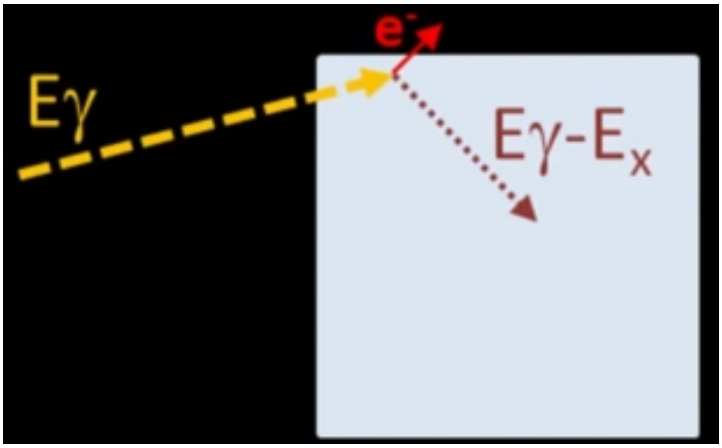


Figure 5.9. The scattered photon with the energy of $E_\gamma - E_x$

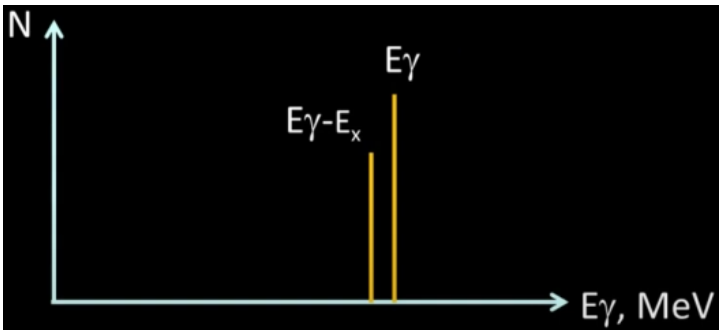


Figure 5.10. Characteristic escape peak of the detector material

- **Coincidence peaks of cascade photons**

  If our parent radionuclide transforms into excited states of the daughter radionuclide and there can be two $\gamma-$transitions one by one (fig.5.12). Detector may not separate the photons from the two consequent transitions $\Rightarrow$ the two $\gamma-$quanta are registered as a single event with the energy $E_\gamma^1 + E_\gamma^2$ (fig.5.11 the right peak). If the detector manages to separate these two events, there will be two more peaks with the energies E_γ^1 and E_γ^2 correspondingly (fig.5.11 the left and middle peaks)

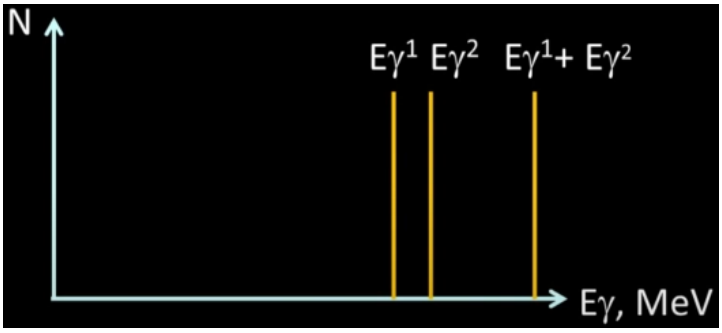


Figure 5.11. Coincidence peaks of cascade photons

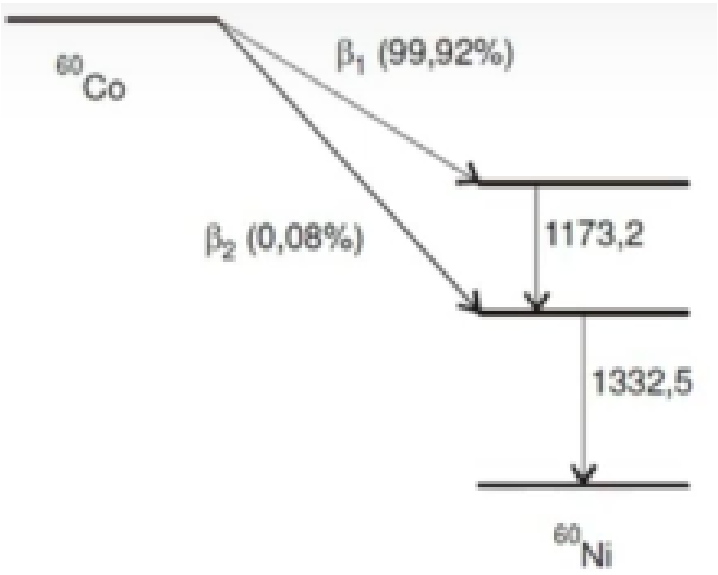


Figure 5.12. The $\gamma-$cascade

If we compare different materials for $\gamma-$radiation measurements we see the striking difference in energy resolution (fig.5.13). The blue color shows a semiconductor (high purity germanium) detector with high energy resolution, the green color represents a scintillator detector with much worse resolution and the red color is for another scintillator with slightly better resolution but still not as good as in the case of the semiconductor.

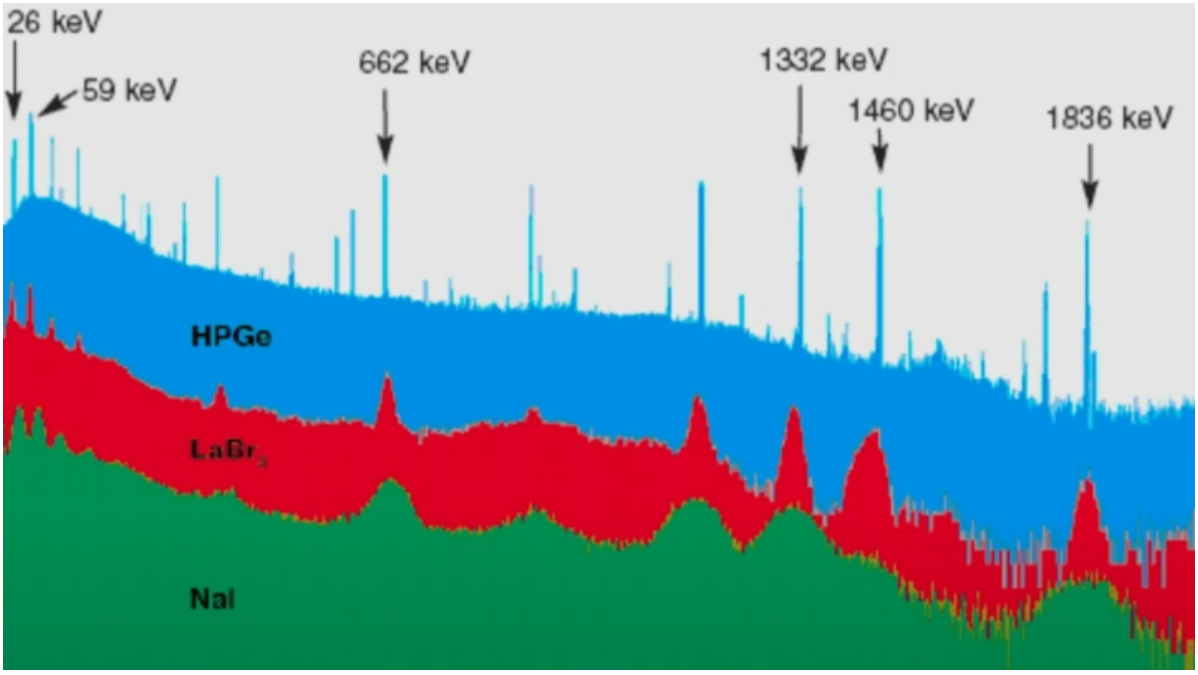


Figure 5.13. The comparison of three detectors

Alpha spectrometry

The use of high-resolution semiconductor detectors allow to distinguish between different radionuclides. We have to prepare vacuum in the experiment as alpha-particles have very low penetrating ability as well as do some chemical preparation - to extract pure alpha-emitting radionuclides from our sample.

As alpha-particles are efficiently absorbed by any matter even by air we can study the effect the air pressure has on the alpha spectrum. The denser the matter is the more energy an alpha-particle loses as it travels through. Thus, increasing the air pressure in the chamber from 20 mbar to 100 mbar leads to the alpha-spectrum shifting to lower energies (fig.5.14).

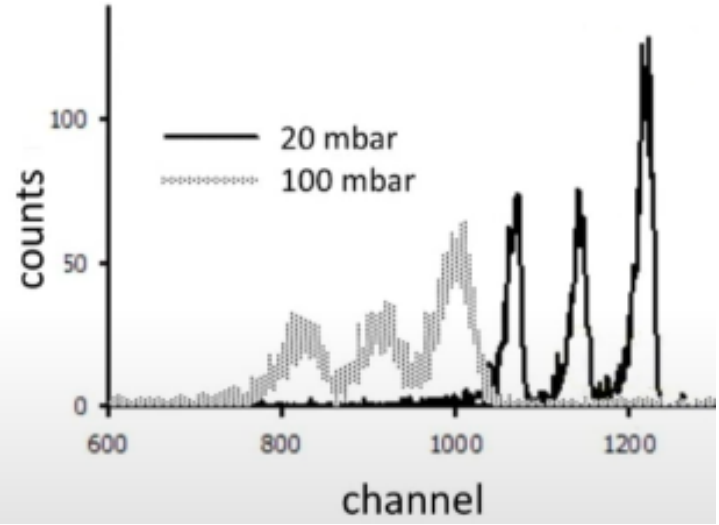


Figure 5.14. The influence of air pressure on the alpha-spectrum

It is also crucial to prepare a very thin alpha-emitting sample, because in the case of the thick one, alpha-particles emitted from the deeper layers are absorbed by the matter of the sample (fig.5.15).

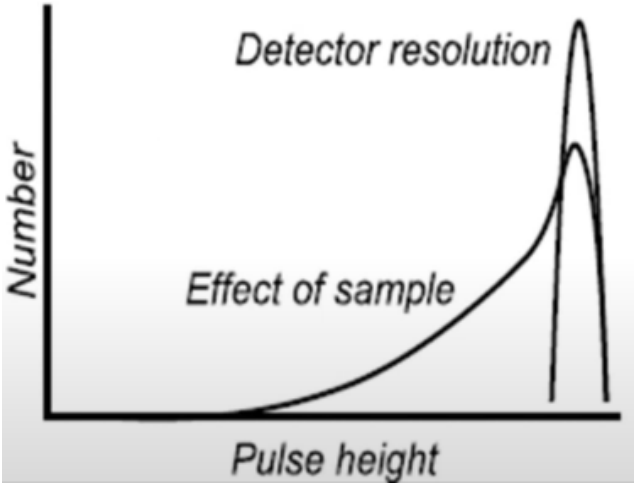


Figure 5.15. The influence of the thickness of the sample on the alpha-spectrum

Liquid scintillation counting (LSC)

LSC method also allows to distinguish between alpha- and beta-radioactivity by studying the pulse shape: processes associated with $\beta-$particles are faster than those with $\alpha-$particles, hence the difference in the pulse shape (fig.5.16).

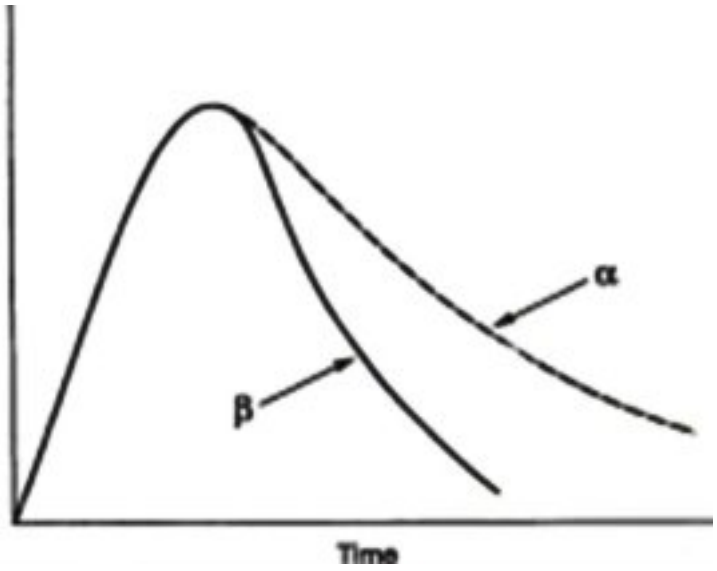


Figure 5.16. The shapes of α and β pulses

LSC is suitable for measuring alpha- and beta- radioactivity but not gamma radioactivity. By putting the sample in the "scintillating cocktail" we get high registration efficiency. Naturally, the chemical and color quenching should be taken into account. LSC method can be used for separation of alpha- and beta-spectrum.

The simplest way to correct for the quenching effects is to use the **internal standard** (E), which means measuring our sample when we add the known amount of the same radionuclide in the system and measure the sample again.

$$E = \frac{C_{s+i} - C_s}{D_i} \tag{5.1}$$

where $C_{s+i},\ C_s$ is the count rate of the sample after and before adding the fixed amount of the radionuclide correspondingly, D_i - the fixed amount of the radionuclide.

Using the internal standard we can determine the efficiency of our measurements and recalculate the activity of our radionuclide. As you can see, we will have to measure each sample twice which is not very convenient.

Another way is to analyze the spectrum of the studied radionuclide considering the composition of the system. We can divide the spectrum in regions and calculate the ratio of counts in these regions for solution with and without quenching agents and therefore calculate the efficiency of our measurement.

A more sophisticated method is the analysis of **spectral indexes of the sample (SIS)**:

$$SIS = K \cdot \frac{\sum_{X=L}^{U} X \cdot n(x)}{\sum_{X=L}^{U} n(x)} \tag{5.2}$$

where X is a channel number, n is number of counts in X, L, U are the lowest and the highest count height limit (channel), K is the fixed factor for non-extinction standard.

Or we can use **spectral indexes of external standard (SIE)** with the idea being to measure the quenching effect not for the certain radionuclide but for the whole system.

Neutron detection

Neutron detection might be very tricky because neutrons do not have a direct ionization effect on matter. The key here is to measure photons or charged particles produced by neutron-induced nuclear reaction such as:

$$^{10}B(n,\alpha)^{7}Li \tag{5.3}$$
$$^{3}He(n,p)^{3}H \tag{5.4}$$
$$^{6}Li(n,\alpha)^{3}H \tag{5.5}$$

So all previously discussed methods for detecting charged particles are valid in this case.

Background reduction methods

For gamma-spectrometry:

- To use lead (10-15 cm) protection around the detector and the sample (fig.4.9).
- Internal surfaces of passive shielding are often lined with material with a lower effective atomic number. Old lead is preferable as it does not contain radioactive nuclei of ^{210}Pb as opposed to the modern lead.
- To get rid of radon from the sample as it is a major source of radiation.

For LSC:

- To use double match scheme (coincidence circuit if fig.4.12)
- To use anti-coincidence circuit (protection from cosmic rays)

Operating procedure for measuring the activity of radioactive samples

The measurement process itself consists of the following stages:

1. Energy calibration of the spectrometer (in the case of α and γ spectrometry);
2. Efficiency calibration of the spectrometer (in the case of α and γ spectrometry) or determining the quench parameter dependence of efficiency (in the case of LSC)
3. Background measurement or blank experiment:
4. Test sample spectrum measurement, peak identification and peak area determination.
5. Calculation of radionuclide activity based on the given background and efficiency.

The order of steps 2-4 can be altered, as their results are mutually independent.

6. Lecture 6. Dosimetry and radiation safety

Influence of radiation on a human body

The consequences of ionizing radiation on human health:

- Cataract
- Skin damage
- Cancer

The sources of radiation:

- Nuclear facilities
- Medical equipment
- Natural background radiation

Even natural radionuclides can be harmful if handled in the wrong way due to lack of knowledge and non-compliance with the safety standards.

Different types of ionizing radiation have different penetrating ability (fig.6.1).

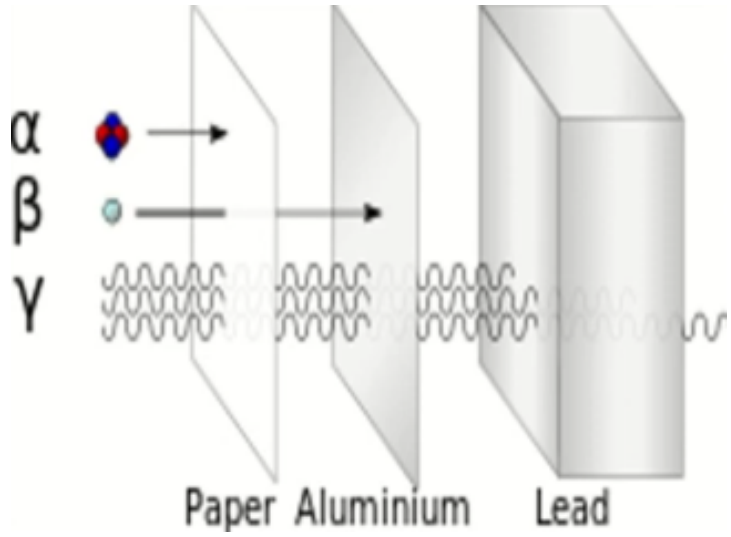


Figure 6.1. Penetrating ability of different types of ionizing radiation

Alpha-particles have the smallest penetration range and can be stopped with a mere sheet of paper, beta-particles have a longer range and gamma-radiation has very high penetrating power.

If the source of radiation is **outside**, alpha and beta emitters are virtually harmless, however if alpha or beta radiation sources get **inside** the body, they produce the main dose.

Radiation influences a human body in two main ways: **radical production** and **changes in biomolecules** resulting in mutations or death.

It is crucial to know some basic definitions:

Radiochemical yield (G) - the quantity of created or destroyed particles (molecules, radicals, ions) or change in parameters of substance (number of polymer chain breaks or cross-links, the angle of rotation of the plane of polarization, etc) tr unit energy absorbed.

We can write:

$$G = \frac{dX}{dE} \tag{6.1}$$

where X is the amount of the substance (mol) or a quantitative value of a changing parameter, E is the energy absorbed (J).

Radiochemical yield is measured in mol/J (in SI) or in molecules/100 eV: 1 molecule/100 eV = 0.1038 μmol/J.

Under the influence of ionizing radiation, molecules are ionized and excited and radicals are formed. The created particles are highly reactive and enter into various chemical reactions (fig.6.2).

$$H_2O \xrightarrow{h\nu} H_2O^+ + e^-$$

$$H_2O \xrightarrow{h\nu} H_2O^* \rightarrow H\cdot + HO\cdot$$

$$H_2O^* \rightarrow H_2 + O\cdot$$

$$H_2O^+ + H_2O \rightarrow H_3O^+ + HO\cdot$$

$$e^- + H_2O^* \rightarrow H_2O^- \rightarrow H\cdot + OH^-$$

$$HO\cdot + HO\cdot \rightarrow H_2O_2$$

$$H_2O_2 + HO\cdot \rightarrow HO_2\cdot + H_2O$$

etc.

Figure 6.2. Creation of radicals and chemical reactions with them

In living organisms, intermediate radicals and ions react with biomolecules changing them (oxidation, substitution of functional groups, destruction, cross-linking of polymers). As a result there appears disruption of the normal activity of cells, organs and the body as a whole.

The most affected by radiation structure of our body is undoubtedly the DNA and RNA. Deep disturbances in vital functions are caused by negligible amounts of absorbed energy. A lethal dose of radiation corresponds to the absorbed energy, which would lead to heating our body by a mere 0.001 $^\circ C$.

The effect on the DNA molecule can be either **direct** or **indirect**. In the first case, radiation hits the DNA itself and may break one or two of its chains. In the second case, there occurs the creation of radical molecules from water and they in turn attack the DNA (fig.6.3). The damaging of the DNA and RNA often lead to mutations.

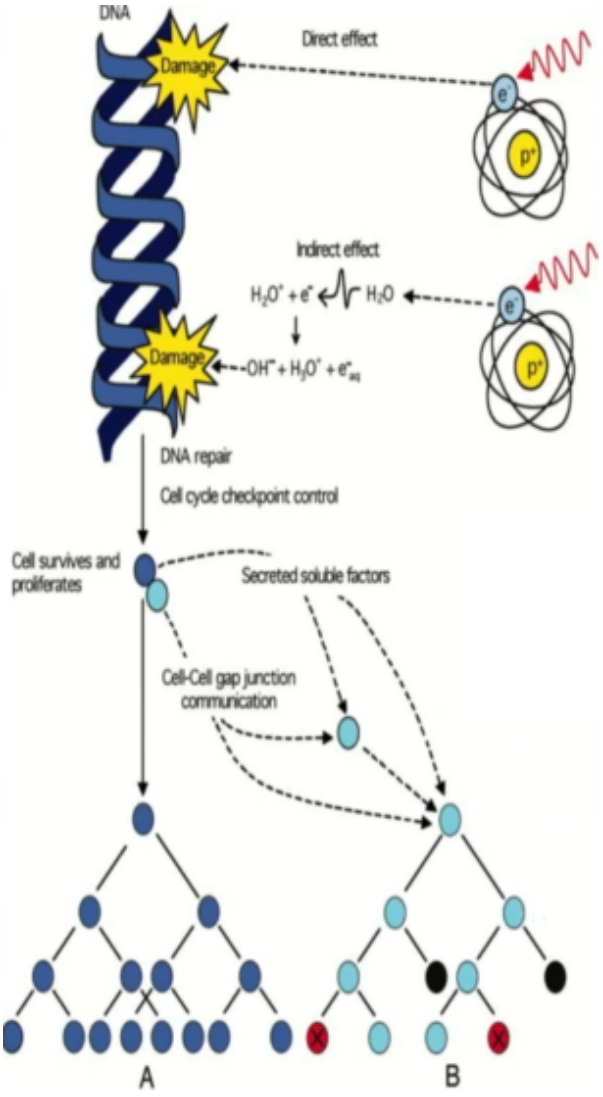


Figure 6.3. The effects of radiation on the DNA

Radiation protection

All radiation protection methods are based on these three postulates:

- **Justification**. If we can perform some procedures without ionizing radiation we should go for it. When the use of harmful radiation is inevitable, the positive effects should outweigh any negative ones.
- **ALARA** (As Low As Reasonably Achievable). We should use as small doses of radiation as we can, having taken into account all economical, social, etc effects.

- **Dose limits.** We should strictly moderate the amount of radiation we get.

There are several relations between different **dosimetry quantities** shown in the scheme in fig.6.4.

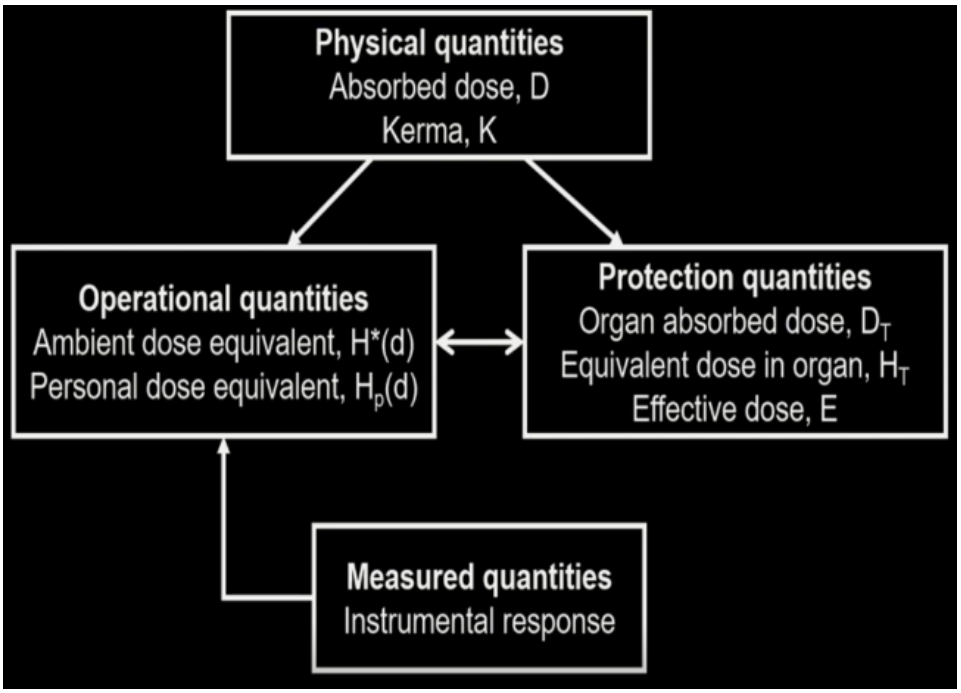


Figure 6.4. Dosimetry quantities

Measured quantities are just signals (like electric current) that are coming from a detector. **Protection quantities** and **operational quantities** are used to describe the safety level of an operation. They all arise from fundamental **physical quantities**.

The main quantity is the **absorbed dose** (D), which is basically the amount of energy absorbed in the matter divided by the mass of that matter:

$$D = \frac{dE}{dm} \tag{6.2}$$

The absorbed dose is used for any kind of radiation (α, β, γ, etc) and absorbing matter (solid, liquid, gas). The unit of absorbed dose in SI is J/kg$\equiv$ gray (Gy).

Another quantity **KERMA** (Kinetic Energy Released per unit Mass):

$$K = \frac{dE_{tr}}{dm} \tag{6.3}$$

where dE_{tr} is the kinetic energy of all charged ionizing particles produced by indirectly ionizing radiation (like $\gamma-$quanta).

Radiation exposure:

$$X = \frac{dQ}{dm} \tag{6.4}$$

where dQ is the total charge generated in air by indirectly ionizing radiation. The unit of radiation exposure is C/kg.

Absorbed dose in an organ or tissue T (D_T):

$$D_T = \frac{1}{m_T} \int Ddm = \frac{E_T}{m_T} \tag{6.5}$$

Equivalent dose in an organ or tissue T (H_T) is used to take into account the fact that different types of radioactivity harm our body to differently:

$$\sum_R w_R \cdot D_{T,R} \tag{6.6}$$

where w_R is a weighing factor for radiation of type R (RBE quality factor).

Values of weighing factors for different types of radiation are shown in fig.6.5.

Radiation type	W_R, Sv/Gy
Photons of any energy	1
Electrons and muons of any energy	1
α particles, fission fragments	20
Neutrons (a non-monotonous relationship with a maximum at 0.5-2 MeV).	2.5 to 20

Figure 6.5. Weighing factors for radiation

Effective dose (E) is useful when a body is irradiated unevenly:

$$E = \sum_T w_T \cdot H_T = \sum_T w_T \sum_R w_R \cdot D_{T,R} \tag{6.7}$$

where w_T is a weighing factor for a tissue T: $\sum_T w_T = 1$

The table in fig.6.6 shows different sensitivity towards radiation of different tissues.

No.	Tissue	w_T	Σw_T
1	Bone marrow (red), large intestine, lungs, stomach, breast, other 13tissues*	0.12	0.72
2	Gonads	0.08	0.08
3	Bladder, oesophagus, liver, thyroid gland	0.04	0.16
4	Bone surface, brain, salivary glands, skin	0.01	0.04
		Total	1

Figure 6.6. Weighing factors for tissues

* The other 13 tissues are: adrenal glands, gallbladder, heart, kidneys, muscle extra-thoracic respiratory organs, oral mucosa, pancreas, small intestines, spleen, thymus, uterus/prostate

Ambient dose equivalent ($H^*(d)$) and **personal dose equivalent** ($H_p(d)$):

$$H = Q \cdot D \tag{6.8}$$

where H is external irradiation use dose equivalent, Q is the mean quality factor for radiation, D is the absorbed dose.

We obtain these quantities looking at the effect of radiation on a tissue-equivalent phantom at depth d (fig.6.7).

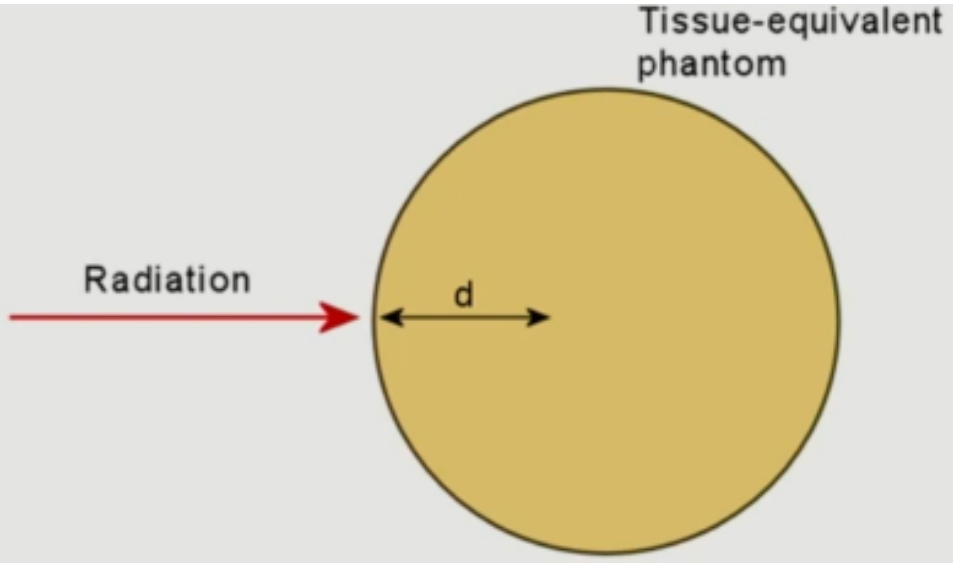


Figure 6.7. The tissue-equivalent phantom

We calculate the dose rate from a point-like source of $\gamma-$radiation at distance r like this:

$$P = K_\gamma \frac{A}{r^2} \tag{6.9}$$

where K_γ is the dose rate of $\gamma-$radiation created by a point-like source of radioactivity 1 Bq in a medium at a distance of 1 m. This constant can be calculated knowing the energy of photons, intensity and absorption coefficient in the air. A is the activity.

Dependence on the penetration depth

The accumulated dose of different kinds of radiation is dependent on the penetration depth as shown in fig.6.8. When we irradiate matter with an external source of $\gamma-$radiation we are interested in KERMA value. The dependence of KERMA and absorbed dose on the distance from the body surface is shown in fig.6.9. Notice, that at some point (d_{max}) KERMA and the accumulated dose become equal and after that the dose decreases.

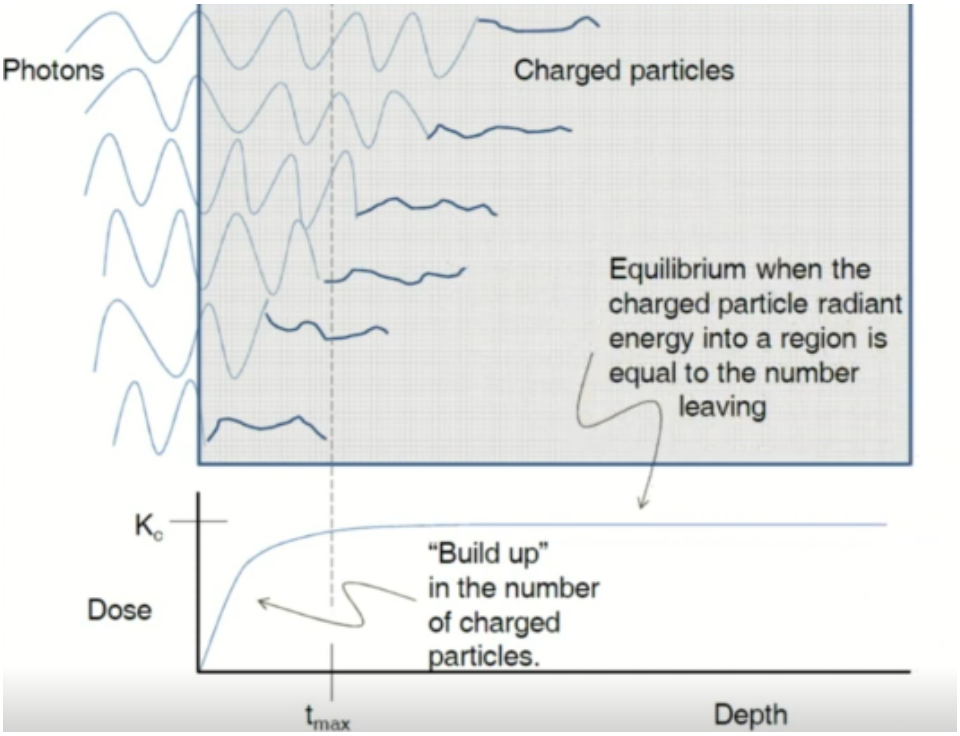


Figure 6.8. The dependence of dose on the penetration depth

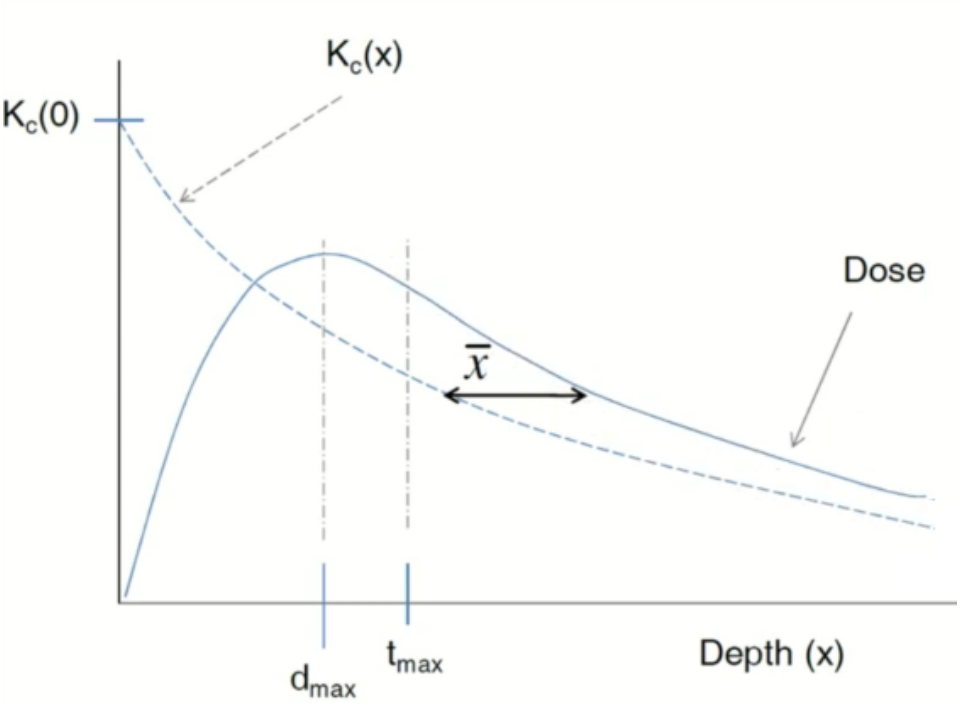


Figure 6.9. The dependence of KERMA and absorbed dose on the distance from the body surface

Dose vs. effect

There are two classes of effects the radiation has on out body: **deterministic** and **stochastic** (fig.6.10). The first means, that after a certain relatively high dose ($> 0.1\ Gy$) we can definitely say what exact harm was done - skin damage, eye damage, etc. The second class is probabilistic - we can only predict the risks caused by radiation, not a certain outcome.

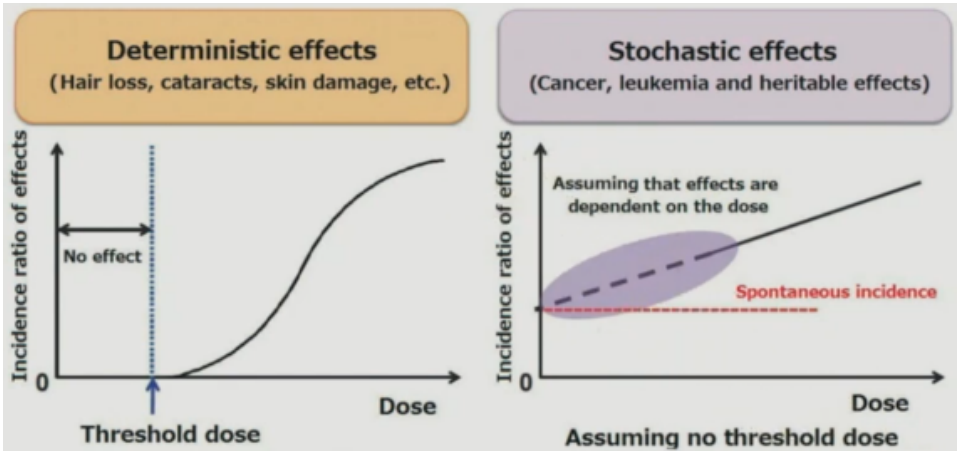


Figure 6.10. Deterministic and stochastic effects of radiation

The stochastic effects are typical for low doses of radiation. There are several models for determination of negative effects caused by radiation. The best among others is considered to be the linear non-threshold model (fig. 6.11).

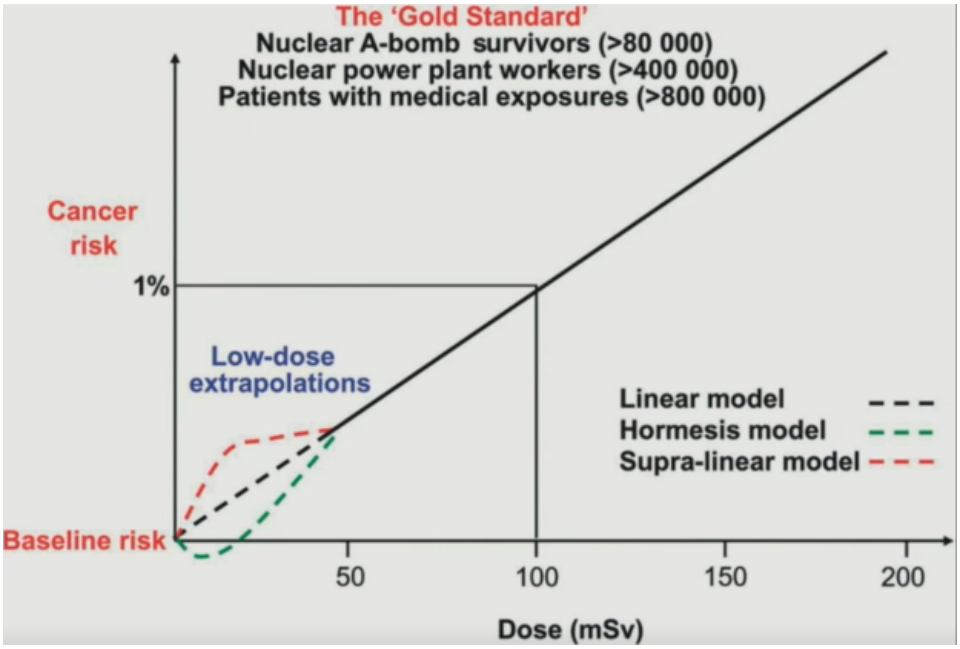


Figure 6.11. The comparison of different models

We have little knowledge of risks at very low doses ($< 50\ mSv$) as it is difficult to separate the studied effects from the natural background radiation. The data for the plot was taken from atomic bombing survivors, nuclear plants workers and patients from hospitals.

Deterministic effects are summarized in the table in fig.6.12

Dose, Gy	Effect
0.1	Doubling rate of the probability of gene mutations
1,0	Acute radiation sickness dose
3-5	Without treatment, 50% of those exposed to radiation die within 1-2 months due to disruption of the activity of bone marrow cells
10-50	Death occurs in 1-2 weeks due to harms of the gastrointestinal tract
100	Death occurs after a few hours or days due to damage to the central nervous system

Figure 6.12. The deterministic effects

Dosimeters

Dosimeters are devices used to control the dose rates. There are several types of dosimeters:

- **Ionization chambers**. They measure charge and current and require calibration.
- **Thermo-luminescent Dosimeter (TLD)**. They absorb the energy of ionizing radiation, heat up and then emit light. Such dosimeters have small size and are reusable.
- **Optically Stimulated Luminescence Dosimeter (OSLD)**. These dosimeters exploit luminescence induced by optical light. They have small size and are reusable but the dose needs recalculating, adjusting for biological tissues.
- **Chemical dosimeters** are suitable for measurement of high doses.
- **Diode dosimeters** are very compact.
- etc...

The three pillars of protection from ionizing radiation:

- **Distance**. As the dose of ionizing radiation decreases as $\frac{1}{r^2}$ the further we are from the source the safer we are.

- **Time**. If we cannot increase the distance between ourselves and the source of radiation we at least should spend as little time as possible near the source.
- **Shielding** is used as the last resort when the first two approaches are unavailable. We use different shielding material for different types of radiation. Against alpha-radiation we can use glass or cloth, against beta radiation we use light materials like plastic and aluminum, against gamma-radiation the most suitable options are lead and high-density concrete, against neutrons we should use materials containing a lot of hydrogen like water, plastic, wood.

7. Lecture 7. Nuclear reactions

Mechanism of nuclear reactions

As was mentioned previously (see Lecture 3), there are two ways ionizing radiation can interact with matter: elastic and inelastic interaction. Inelastic interaction can result not only in excitation and ionization, but also in a **nuclear reaction**.

The first nuclear reaction was conducted by Rutherford in 1911. He bombarded nitrogen with alpha particles producing oxygen and hydrogen

$$^4He +^{14} N \rightarrow^{17} O +^1 H \iff {}^{14}N(\alpha, p)^{17}O \tag{7.1}$$

Diagram in fig.7.1 shows how a nucleus can transform as a result of radioactive decay and nuclear reaction.

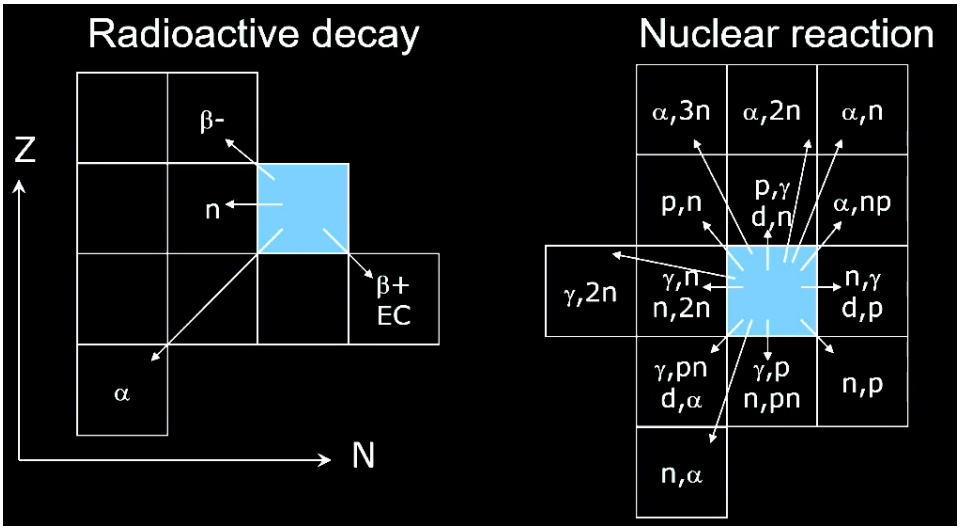


Figure 7.1. Nuclear transformation in radioactive decay and nuclear reaction

The general mechanism of a nuclear reaction can be explained as following. Suppose we have a nucleus A irradiated with a particle a. Given the energy is sufficient, they form a compound nucleus Aa. The further process depends on the composition of the compound nucleus and its energy state (fig.7.2):

- If in the final state after decomposition of the compound nucleus we have the same nucleus and particle it is an elastic scattering.
- If the final particle and nucleus are the same, but the nucleus is in the excited state it is an inelastic scattering.
- The compound nucleus might disintegrate into other particles b, B that are different from the initial ones. It might e result of the fission process.
- There also might be three particles in the final state.

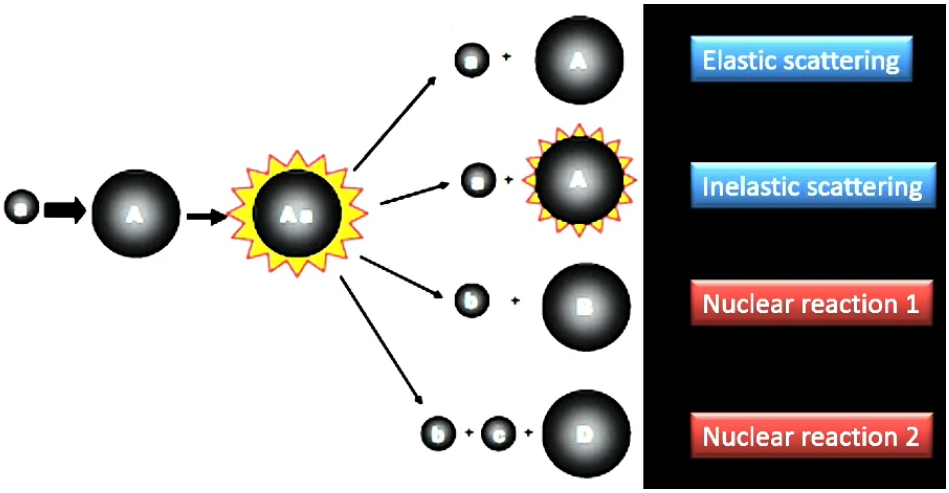


Figure 7.2. Mechanism and examples of nuclear reactions

The yield of the reaction products is determined by the excitation energy of the compound nucleus and does not depend on the components from which it is obtained.

Cross section

To quantitatively estimate the result of our nuclear reaction we use the cross section σ. The cross section is the probability that a system of two interacting particles will transfer from the initial state to the final state as a result of interaction. The higher this probability the more interactions will happen. The cross section is given by the following expression:

$$N = \sigma N_0 n \tag{7.2}$$

where N is the number of interactions, N_0 is the number of nuclei in the unit area, n is the number of particles passing through this area.

Cross sections are measured in barn: 1 barn $= 10^{-24}$ cm^2, typical values of cross sections are 0.1 - 10^4 barn. A way to visualize a cross section is to assume that it is the effective area of a nucleus. Indeed, 1 barn is approximately the cross-section area of e medium-sized nucleus.

When we represent the cross section as the area of interaction between particles A and B it can be estimated as the overlap area:

$$\sigma = \frac{\pi(d_A + d_B)^2}{4} \tag{7.3}$$

where d_A, d_B are the diameters of the interacting particles (see fig.7.3).

Microscopic particles can be represented as waves whose wavelength λ depends on the energy of the particle E:

$$\lambda = \frac{h}{\sqrt{2mE}} \tag{7.4}$$

h is the Planck's constant, m is the mass of the particle

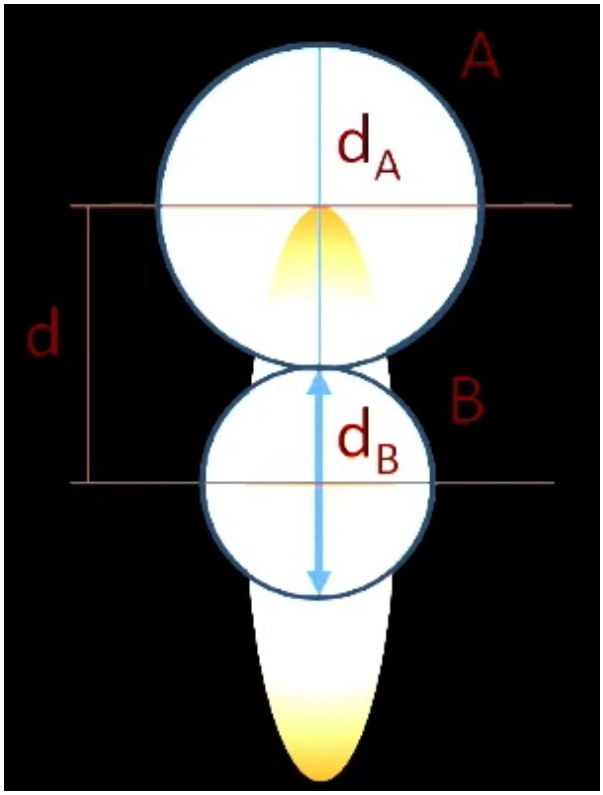


Figure 7.3. The estimation of the cross section

The overlap area depends on the wavelength. As we can see from the equation (7.4) the wavelength of the particle is inversely dependent on its energy, hence the faster the particle moves the smaller the cross section and vice versa. Such tendency is very typical for neutrons (fig.7.4).

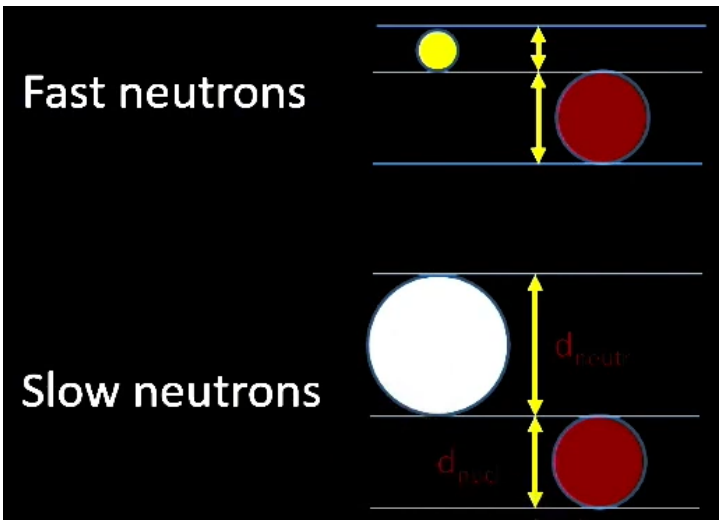
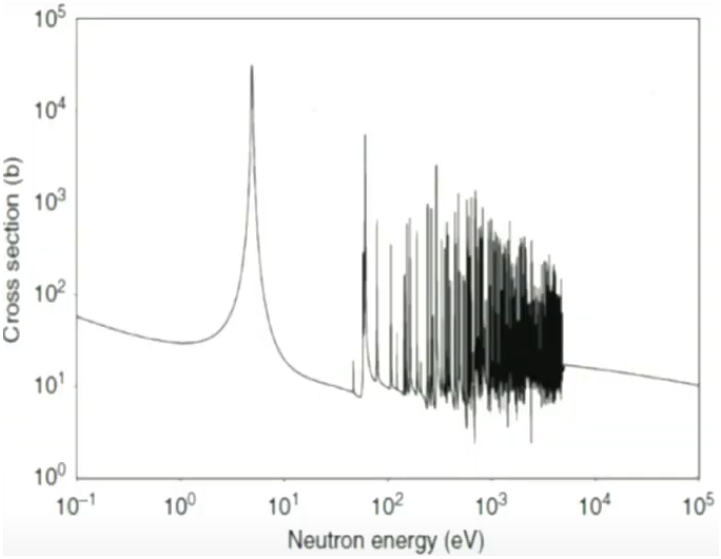


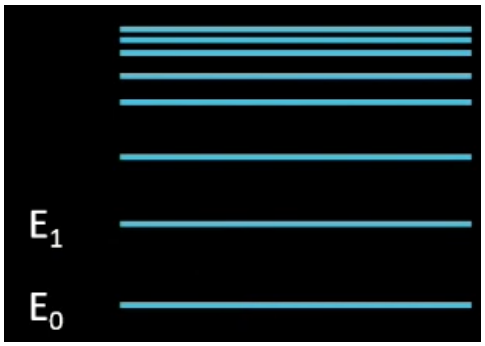
Figure 7.4. The overlap area of fast and slow neutrons

On the other hand, the slower the particles move the more time they spend near each other hence the probability of interaction increases.

As a main trend, the cross section of reactions with neutrons decreases with energy, but on top of that there might be sharp peaks called **resonances** at certain values of energy that correspond to energy levels of the excited nucleus. It is convenient for a nucleus when it consumes a neutron to transfer to an excited energy state, hence the increase in the cross section (fig.7.5 **a**).



a)



b)

Figure 7.5. **a**):The resonance structure of cross section for $^{197}Au(n,\gamma)^{198}Au$ reaction; **b**): The energy levels of a nucleus

For relatively heavy nuclei (A = 100 - 150) the energy levels near the ground state are separated by intervals of ~ 0.1 MeV. In excited nuclei the distance between levels is few eV and at energies ~ 15 MeV the energy levels become practically continuous (fig. 7.5 **b**). For this reason in fig.7.5 **a)** we observe a clear resonance peak corresponding to the first energetic level in fig.7.5 **b)** and a few more but then the peaks start to blend in as we approach continuum area.

Energy effect of a reaction

For a reaction of type $X(a,b)Y$ we can calculate the amount of energy released (Q value) as:

$$Q = [(M_x + M_a) - (M_y + M_b)] \cdot 931501 = \Delta m \cdot 931501 \text{ (KeV)} \tag{7.5}$$

where M_i are the exact masses (in a.m.u.) of particles and nuclei participating in the reaction.

If $Q > 0$ we have an **exoenergetic** reaction for which there is no threshold. If $Q < 0$ it is an **endoenergetic** reaction. In case of an endoenergetic reaction the kinetic energy

of the particle a ($T_{a,min}$) must be at least:

$$T_{a,min} = |Q| \cdot \frac{M_x + M_a}{M_x} \approx |Q| \cdot \frac{A_x + A_a}{A_x} \approx |Q| \cdot \frac{A_Y + A_b}{A_x} \tag{7.6}$$

However, the equation (7.6) is only suitable for neutral particles. If the particles have electric charge there interferes the Coulomb barrier. So positive particles must overcome the forces of electrostatic repulsion, therefore for a particle with charge Z_2e, radius R_2 and a nucleus with charge Z_1e, radius R_1 the height of the Coulomb potential barrier is given by:

$$E_{Coul} \equiv B = \frac{Z_1 Z_2 e^2}{(R_1 + R_2)} \approx \frac{Z_1 Z_2 e^2}{R_0(A_1^{1/2} + A_2^{1/2})} \tag{7.7}$$

where R_0 is a constant equal to 1.0-1.4 fm (1 fm = 10^{-15} m).

Up to a point the higher the energy of a bombarding proton the higher the cross section of this particular reaction is but then it starts to decrease as new channels for the reaction become available (fig.7.6). This phenomenon occurs due to the fact that the probabilities of different channels add up to a limited value.

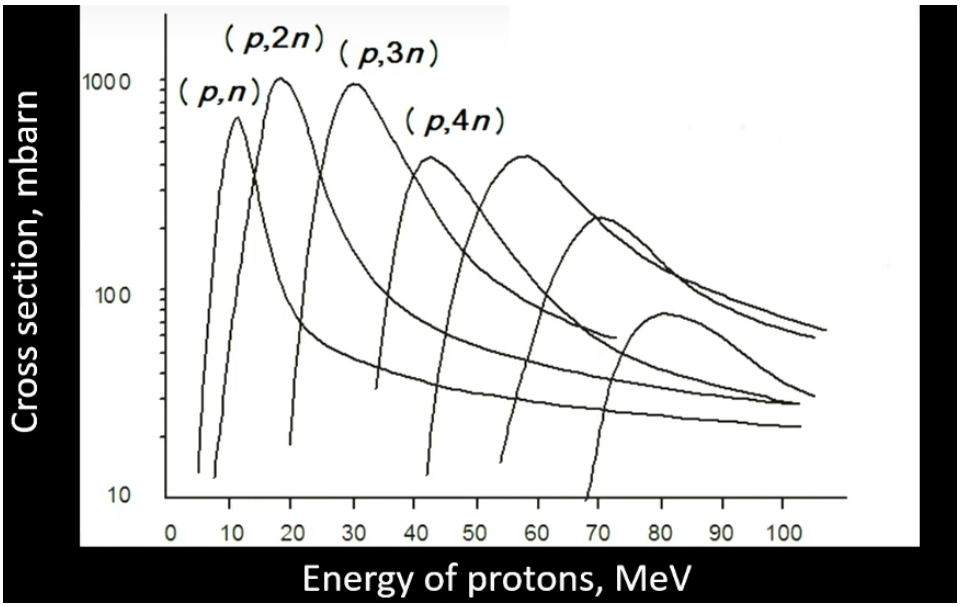


Figure 7.6. Cross sections for $^{125}Te(p, xn)^{125-x}I$ reactions

A nuclear reaction will be effective if the kinetic energy of a bombarding particle is higher than the height of the Coulomb barrier $E_{kin} > B$. But even if $B > E_{kin} > E_{threshold}$ the particle might go into the nucleus due to the **tunnel effect** (fig.7.7). The tunnel effect is probabilistic, therefore the cross section which is determined by the transparency of the barrier will be small.

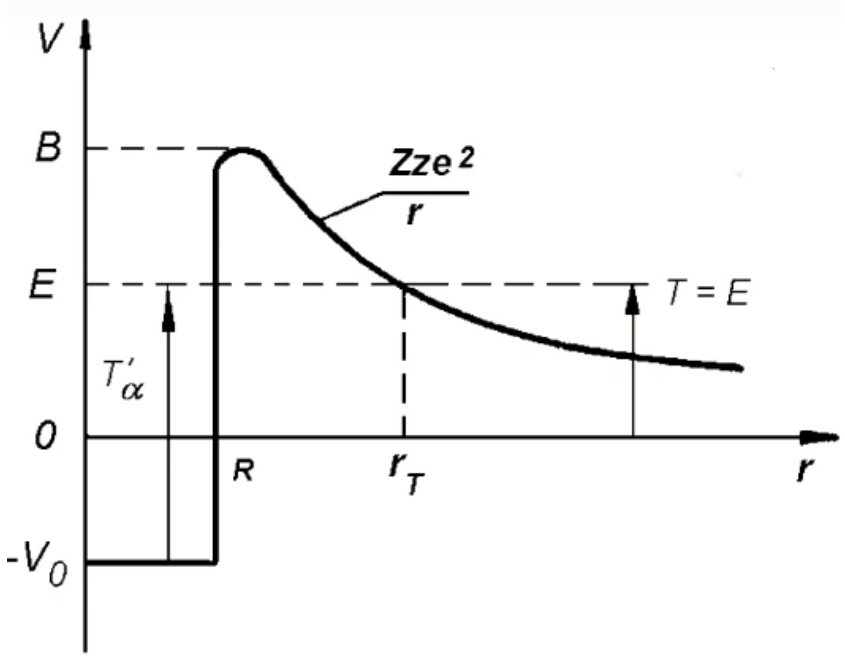


Figure 7.7. Coulomb barrier for charged particles and the tunnel effect

Emitted particle energy

Energy $(E_{kin,a}+Q)$ in reactions like $X(a,b)Y$ is converted mainly into $(E_{kin,b}+E_{kin,Y})$. Examples:

- (n,γ)-reactions on fast neutrons.

  The amount of kinetic energies of particles and nuclei: $E_T = E_n + Q = E_\gamma + E_R \approx E_\gamma + E_\gamma^2/(1862 \cdot M)$ where M is the mass of the compound nucleus, E_R is the energy of the recoil nucleus. Because $E_R \ll E_\gamma \Rightarrow E_\gamma = Q + E_n$

- (n,γ)-reactions on thermal neutrons.

  $Q = E_\gamma + E_R \approx E_\gamma + E_\gamma^2/(1862 \cdot M) \Rightarrow E_\gamma = Q - E_\gamma^2/(1862 \cdot M) \approx Q$

- (γ,n) Photo-nuclear reactions.

  Using the energy conservation law: $E_\gamma + Q = E_n + E_\gamma$ and the momentum conservation law for the compound nucleus and emitted neutron we get:
  $1 \cdot V_n = M_\gamma \cdot V_\gamma \Rightarrow 1 \cdot E_n = M_\gamma \cdot E_gamma \Rightarrow E_n = \frac{(E_\gamma+Q)\cdot M_\gamma}{M_\gamma+1}$

- (n,b)-reactions on thermal neutrons, where b is some particle: p, α, etc.

  The energy of the particle b: $E_b = \frac{Q\cdot M_\gamma}{M_\gamma+M_b}$

Neutron production methods

There are three main ways to produce neutrons:

- **Nuclear reactors**

  We get huge fluxes of neutrons due to the nuclear fission reaction. The average energy of fast neutrons is 2 MeV, the major part of neutrons is thermal neutrons.

- **Generators of neutrons**

  We use accelerated particles to induce nuclear reactions where neutrons are released as final particles. We can vary the energy of the irradiating particle to obtain neutrons with different energies.

- **Neutron sources** like $Ra - Be$ and others.

  In this method we also use induced nuclear reactions, for example a short chain of reactions that produces neutrons (see fig.7.8):

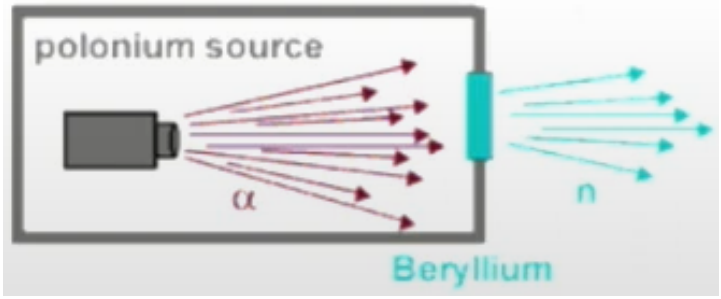


Figure 7.8. A source of neutrons: $^{210}Po \rightarrow ^{206} Pb + \alpha \Rightarrow ^{9} Be + \alpha ^{12}C + n$ ($E_n = 4$ MeV)

The main types of nuclear reactions, initiated by neutrons are summarized in the table in fig.7.9

Reaction	Decay	Comments
(n,γ)	Predominantly β^-, rarely β^+, EC	High yield for thermal neutrons
(n,2n)	Predominantlyo β^+, sometimes β^-	Always endothermic, neutron energy > 10 MeV
(n,p)	Predominantly β^-	Usually endothermic, exceptions ^{14}N (n, p) ^{14}C, ^{35}Cl(n, p) ^{35}S
(n,α)	Predominantly β^-	Predominantly endothermic, exceptions ^{6}Li (n,α) ^{3}H, ^{10}B(n, α)7Li
(n, f)	Predominantly β^-, rarely β^+, EC	Nuclides with atomic number > 90 for ^{233}U, ^{235}U, ^{239}Pu for thermal neutrons

Figure 7.9. Nuclear reactions initiated by neutrons

Radionuclide accumulation rate in reactors

Activity of the nuclide accumulated in the target by the end of the irradiation is given by:

$$a = \lambda N = \Phi S \cdot [1 - e^{-nl\sigma}] \cdot [1 - e^{-\lambda t}] \tag{7.8}$$

where a is activity (Bq), Φ is the flux density of particles falling on the target along the normal to its surface (particles/cm^2·s), S is the target area (cm^2), n is the number of activates nuclei in 1 cm^3 of the target, l is the thickness of the target (cm), σ is the cross section (barn), t is the exposure time (s), λ is the decay constant (s^{-1}).

The equation (7.8) is only valid if:

1) The thickness of the irradiated target allows us to neglect the change in Φ in the target itself, which occurs due to absorption and scattering.

2) It is possible to neglect the loss of stable target nuclei and the "burnout" of the radionuclude.

If $nl\sigma < 0.1$ the target is considered thin, therefore we can re-write (7.8) as:

$$a = \Phi\sigma n_0[1 - e^{\lambda t}] \tag{7.9}$$

where n_0 is the total number of activated nuclei in the target.

With prolonged irradiation of the target nuclei with a large cross section σ, it is necessary to take into account not only the accumulation and radioactive decay of the product, but also the consumption of the target nuclide and the reaction product ("burnup"). In this case the accumulation rate of our radionuclide will also depend on the cross section of the reaction with our radionuclide (σ_2):

$$\frac{dN_2}{dt} = \Phi\sigma_1 N_1 - \lambda_2 N_2 - \Phi\sigma_2 N_2 \tag{7.10}$$

the first term in (7.10) reflects the accumulation of the reaction product, the second - its radioactive decay, the third - the product burnup. Index 1 refers to the target nucleus, 2 - to the reaction product nucleus.

As a result, the activity of the radionuclide:

$$a_2 = \lambda_2 N_2 = \frac{\lambda_2 \Phi\sigma_1 N_{01}}{\Lambda_2 - \Phi\sigma_1} \cdot (e^{-\Phi\sigma_1 t} - e^{-\Lambda_2 t}) \tag{7.11}$$

where $\Lambda_2 = \lambda_2 + \Phi\sigma_2$ is the effective decay constant.

The example of accumulation of ^{90}Y during irradiation of ^{89}Y by neutron flux 10^{14} cm^{-2}s^{-1} is shown in fig.7.10. At a certain point we reach the maximum or saturated activity A_{sat}.

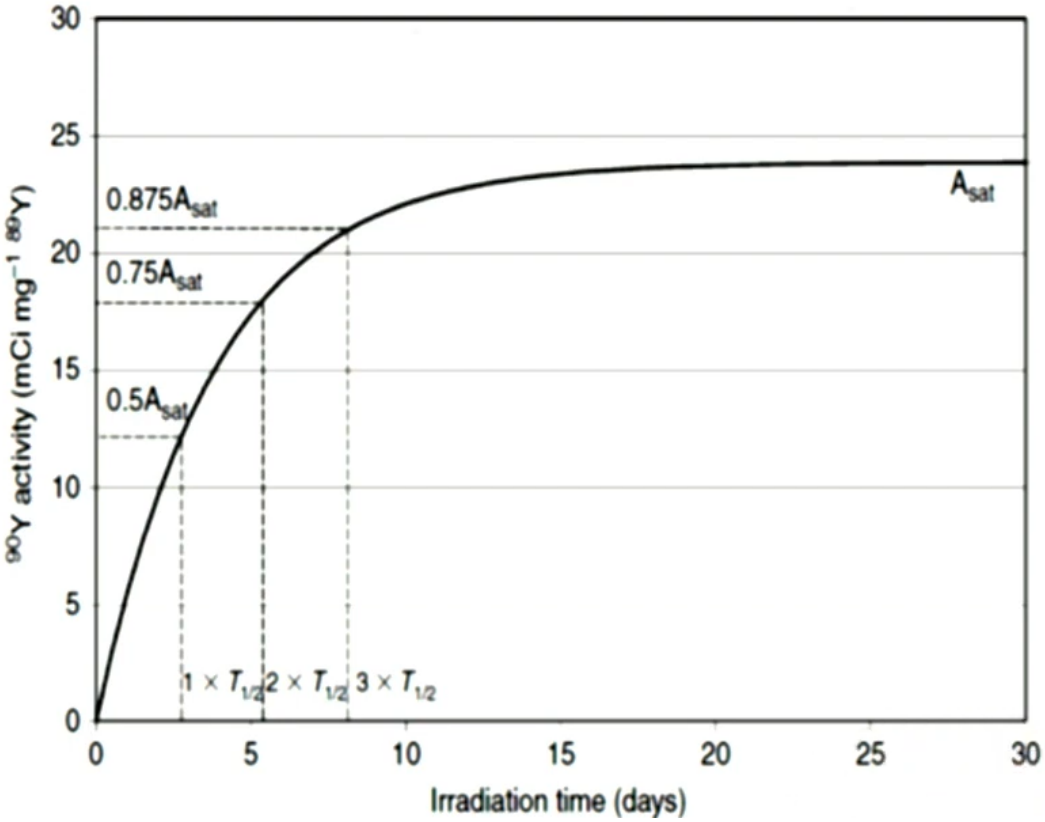


Figure 7.10. The of accumulation of ^{90}Y

Activity of a radionuclide obtained at a particle accelerator

In this case, to calculate the activity, we use the value of the beam current strength I in amperes instead of the flux Φ. Absolute activity of a nuclide in a thin target by the end of irradiation:

$$a = \lambda N = \lambda \cdot 6.24 \cdot 10^{18} \cdot It\sigma nI/Z \tag{7.12}$$

In practice, the "thick target yield" is commonly used. The yield Y is the activity of the radionuclide per unit of charge transferred to the target. The Y value can be found in reference books. The activity of the radionuclide at the moment t is equal to:

$$A = Yi(1 - e^{-\lambda t})/\lambda \tag{7.13}$$

where Y is yield (Bq / (μAh)), i is the beam current (μA), λ is the decay constant of the produced radionuclide (h^{-1}).

The expression (7.13) takes into account the accumulation and decay of the radionuclide during irradiation.

It is crucial to remember, that if we use neutron sources or charged particles to induce our nuclear reactions, when we stop irradiation and take away the target the accumulation of the radionuclide also stops and it begins to decay (fig.7.11).

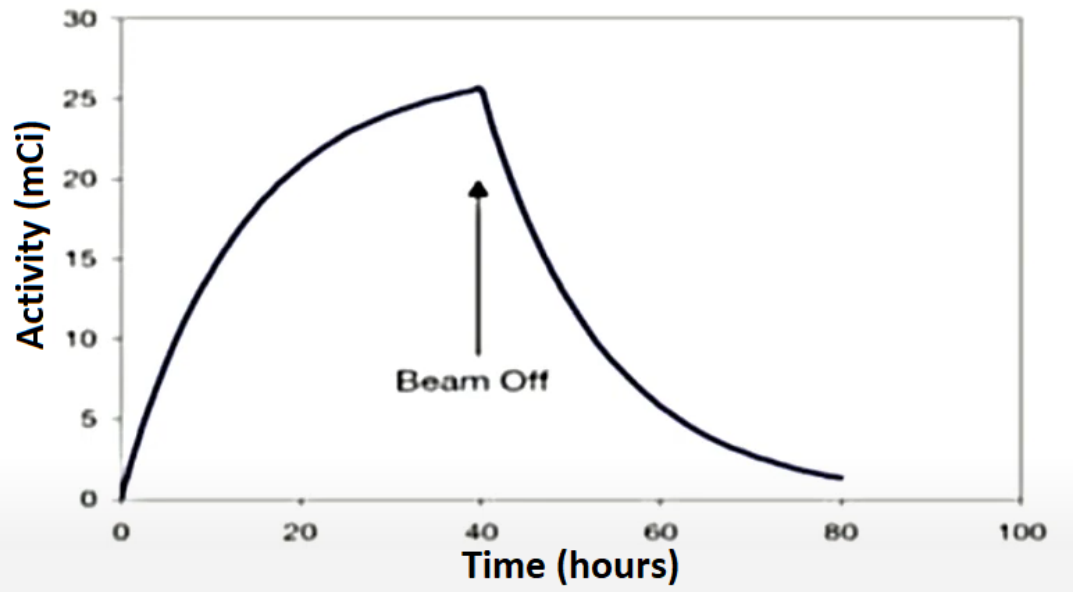


Figure 7.11. Decay of the produced radionuclide

8. Lecture 8. Basics of nuclear fuel cycle

Energy production

What is the situation with the energy in the world? According to the data at hand 1 billion people live in poverty and do not have any access to energy and about the same amount get no health care due to the lack of energy. Approximately 40% of the world's population rely on biomass. There exist 2 million excess deaths per year due to exposure to indoor air pollution that occurs as a consequence of using coal or oil for production of energy. The paramount goal to achieve in the future is to cut down on the carbon-based fuel in production of electricity.

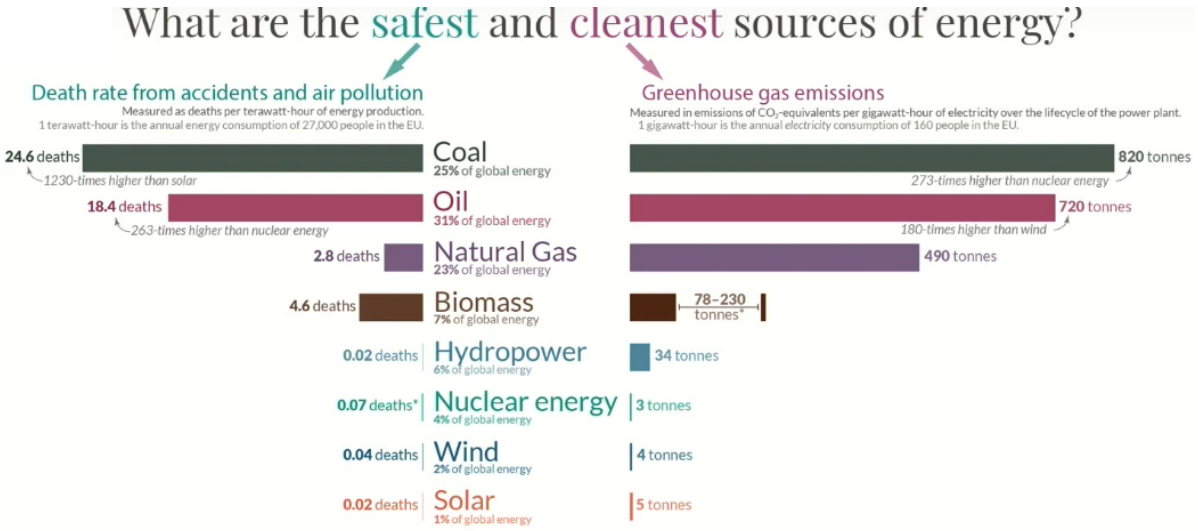


Figure 8.1. Decay of the produced radionuclide

The safest and cleanest sources of energy that have the smallest **death rate from accidents and air pollution** and **greenhouse gas emission** are **hydropower**, **nuclear energy**, **wind** and **solar energy** (see fig.8.1). As the nuclear force is 5-6 orders of magnitude stronger than any chemical bond the energy production by the nuclear fuel is much more efficient than that of coal or gas. For instance, burning 26 m^3 of gas produces more ore less the same amount of energy as using 26 milliliters of nuclear fuel. To produce 1000 MW of energy using nuclear power $\leq$ 1 square mile of land is required to build a nuclear power plant, whereas for wind turbines and solar panels we will need 360 and 75 times lager area correspondingly to produce the same amount of electricity.

The concept of so-called "green square" implies using the four above mentioned ways of safe and clean energy production to achieve the desired goal. Nuclear power has great potential for efficient, long-term production of energy - nuclear power plants can operate within 30-50 years and even longer. Nowadays there are quite a lot of radiophobic people who do not understand radioactivity and therefore are afraid of any manifestations of it, thus it is important to spread awareness of nuclear energy benefits among public. We can do so by explaining how the harmful radioactive waste is dealt with, for example.

Nuclear fuel cycle

In reality a nuclear power plant is just one part in the nuclear fuel cycle (NFC). NFC starts with uranium ore mining, then goes refinement and enrichment of uranium with further production of nuclear fuel that is put in nuclear power plants where electricity is produced. The spent of fuel is stored in special pools and then we either dispose of the spent nuclear fuel (**open NFC**) or we re-process it, extract valuable components to produce fresh fuel that can we be used again in power plants (**closed NFC**).

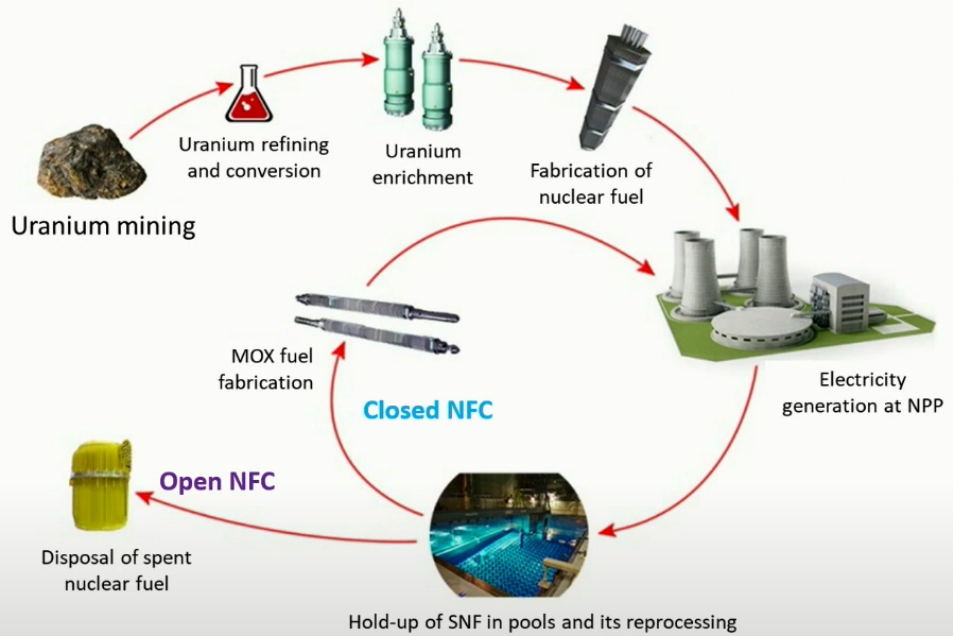


Figure 8.2. The nuclear fuel cycle

Radioactive waste is material that contains or contaminated with radionuclides at concentrations (or activities) greater than **clearance levels** established by individual countries, and for which no use is currently foreseen. It is important to remember, that the hazard from radioactive waste is finite as radionuclides decay until the stable nuclides are reached.It is not the case for other types of waste like heavy metals (mercury, cadmium, etc.) whose toxicity does not decrease with time.

Radioactive waste is generated at each step of the NFC. In general, working with radioactive material in scientific, industrial or medical facilities also produces radioactive waste.

Let's have a deeper look into each step of the NFC.

Mining

There are several types of uranium ore mining:

- In **open careers** and **mines** - the classical methods. The extracted uranium ore is piled up in pyramids, then sulfuric acid that dissolves the ore is poured on top. At the end the liquid containing uranium is collected and transported to a power plant where pure uranium is extracted (fig.8.3).

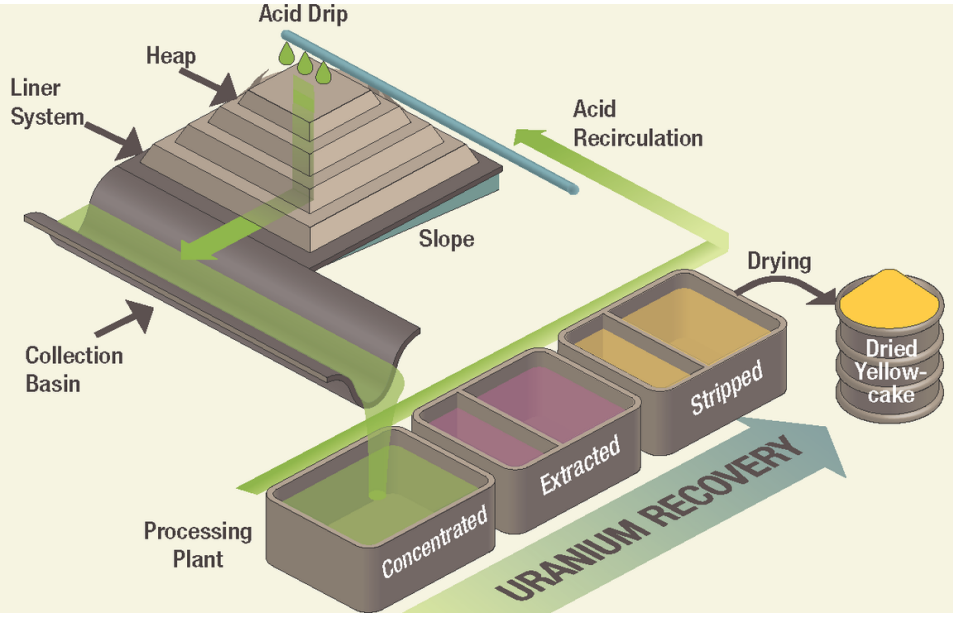


Figure 8.3. Extraction of uranium ore from careers and mines

As by-product, during uranium mining in careers and mines we produce quite a lot of tailings that contain natural radionuclides (first of all, radium). These hazardous radionuclides can reach people's homes though water which is a big problem (fig.8.4).

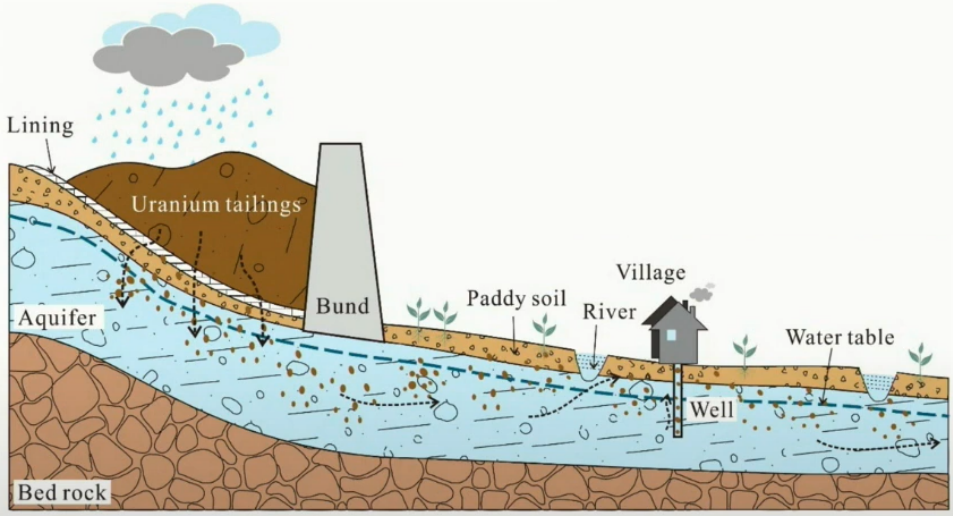


Figure 8.4. Uranium tailings

- Through **underground leaching**, which is considered to be one of the safest methods. Basically, we bury several injection wells in the ground, through which sulfuric acid solution that dissolves uranium ore is pumped in. Recovery wells pump out the solution containing uranium which is further extracted (fig.8.5).

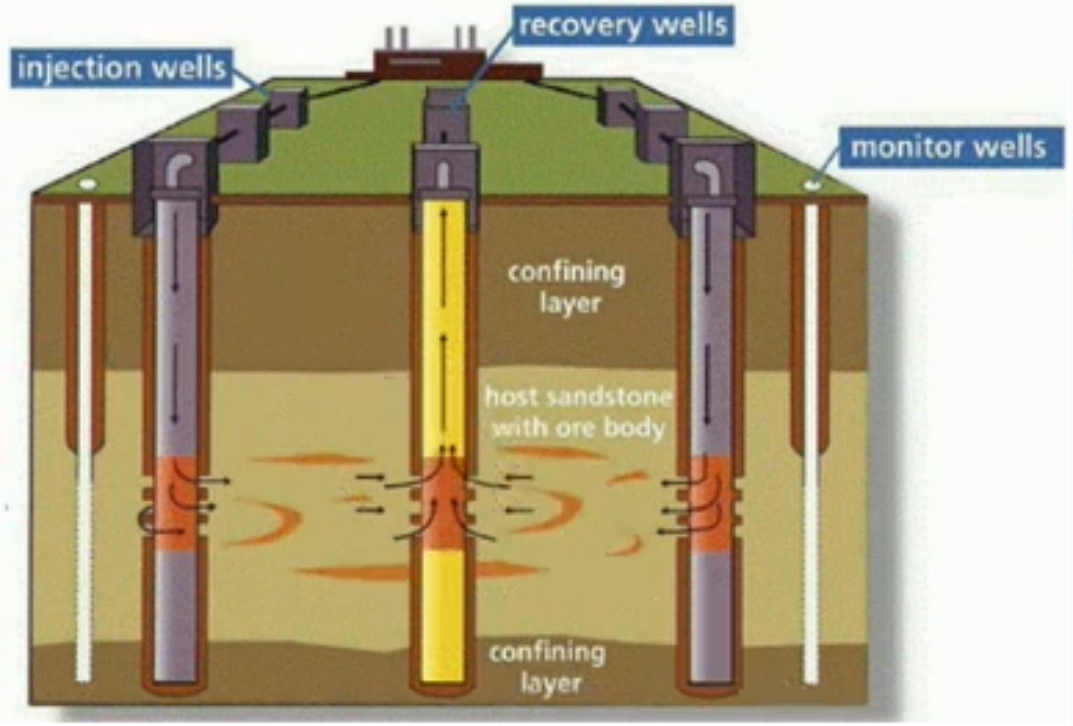


Figure 8.5. Underground leaching

Refinement and conversion

After uranium is extracted it needs further purifying by ammonium diuranate precipitation, for example. Then we need to enrich uranium so that it contains 2-5% of ^{235}U (necessary requirement for reactors). To do this we convert uranium into UF_6 because it has quite low sublimation temperature ($T = 56.4\ ^\circ C$). UF_6 goes from solid state straight to gas, and in gaseous state in is convenient to separate isotopes of uranium (fig.8.6).

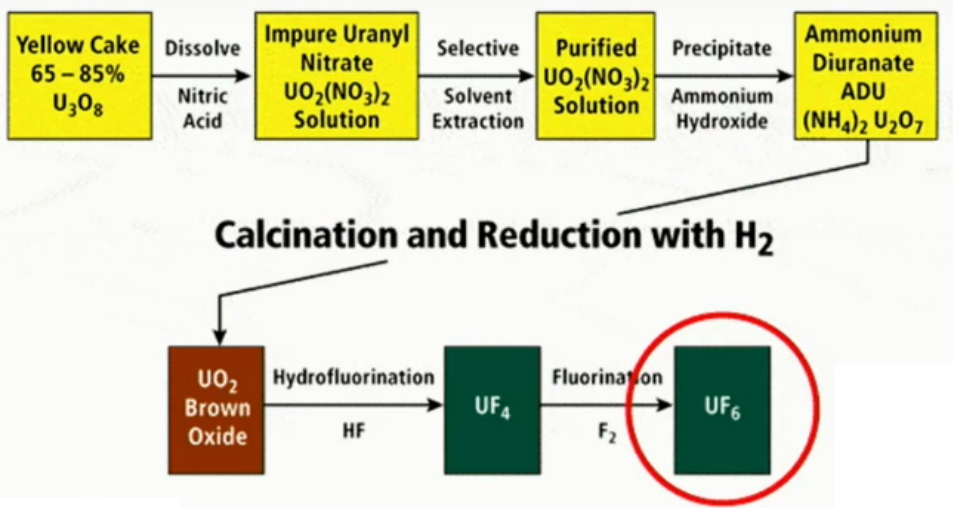


Figure 8.6. Conversion of uranium

Enrichment

Uranium can be classified by the degree of enrichment (proportion ^{235}U):

- Natural: 0.72%
- Depleted: <0.72%
- Low-enriched: up to 20% (2-5% for fuel)
- Highly enriched: over 20%

There are several methods for uranium enrichment:

- **Magnetic separation of different isotopes**. This approach requires a lot of energy and nowadays is no longer in use.
- **Gas diffusion**. This method exploits the difference in diffusion of $^{235}UF_6$ and $^{238}UF_6$ through a membrane with a pore diameter of 10-100 nm.

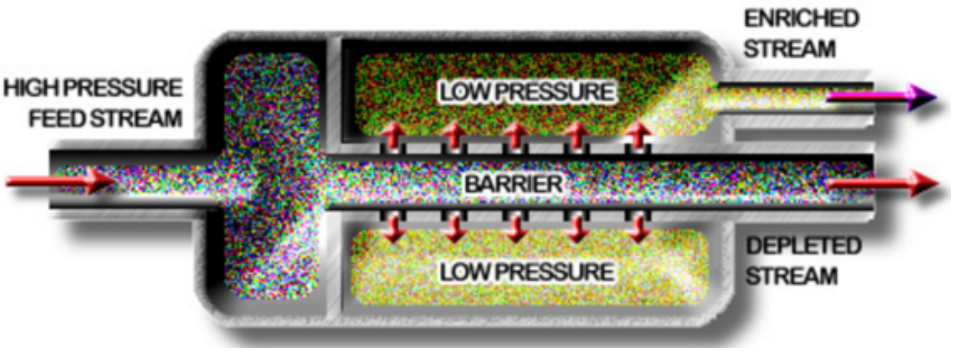


Figure 8.7. Gas diffusion method

- **Gas centrifuge**. This method uses the difference in the speeds of $^{235}UF_6$ and $^{238}UF_6$ in the field produced by centrifugal forces.

The main waste produced as a by-product of uranium enrichment is depleted UF_6 which is stored in barrels outside and more chemically toxic than radioactively. It so happens, that technically depleted UF_6 is not classified as waste, because there are nuclear reactors that can operate using depleted uranium.

Fuel fabrication

Main components of nuclear fuel are:

- Uranium dioxide (UO_2) and mixed oxide (PuO_2 + UO_2). It is stored in the form of small cylinders 8 mm in diameter and 2-2.5 cm in height. Then the cylinders and undergo heat treatment, after that they are placed in zirconium claddings 3-6 m in height. The claddings are organized in square-shaped assemblies which in turn go to the reactor.

- Metals and alloys
- Carbides of U and Pu
- Nitrides of U and Pu
- Liquids (melts, solutions)
- Composite

Fuel with defects, old fuel, etc. are dissolved, purified, re-fabricated and the liquid of radioactive waste of complex composition is formed.

Electricity generation at nuclear power plants

A nuclear reactor includes an active zone containing nuclear fuel and a neutron moderator, a neutron reflector, a heat carrier for heat removal, a chain reaction control system, reactor protection and its control.

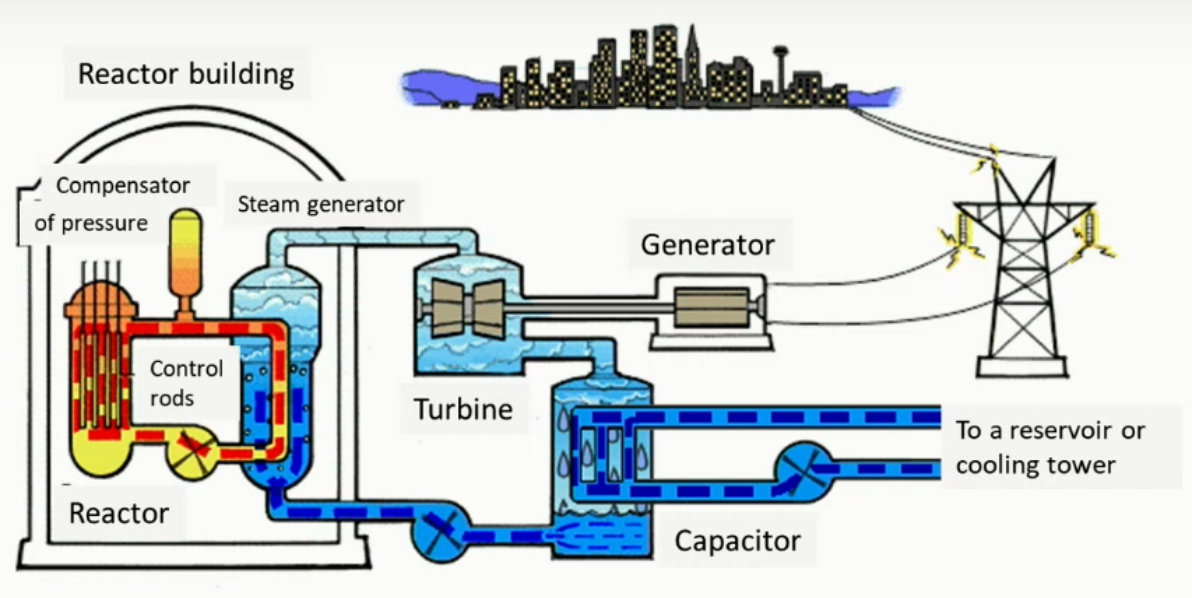
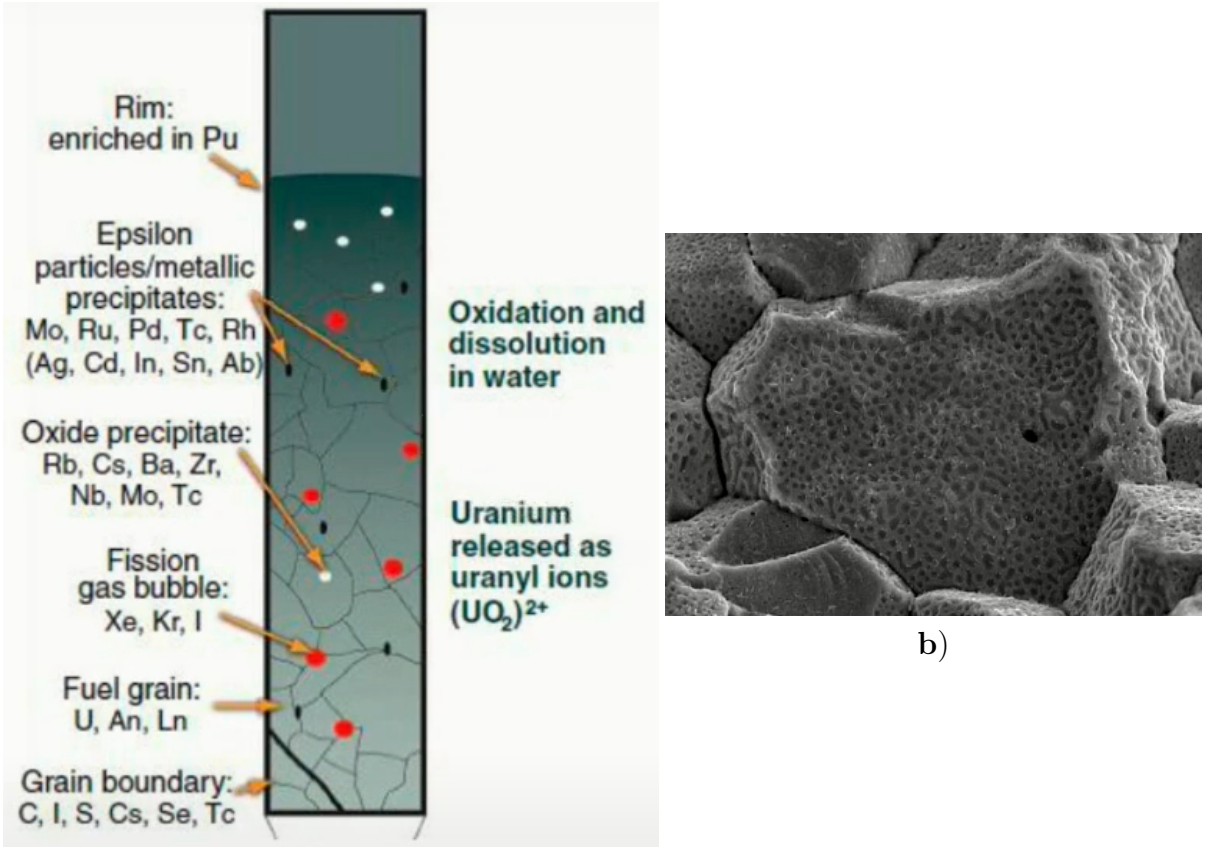


Figure 8.8. The scheme of a nuclear reactor

Inside the active zone nuclear fuel is put in assemblies and ongoing nuclear reactions produce thermal energy. The water, that circulates through the assemblies is heated to $\sim 300°C$ (the water is also under big pressure). This hot water then goes to the heat exchanger where it heats up water in another circuit turning it into steam, which rotates the turbines and hence the electricity is produced. The steam then condensates back into water and the cycle repeats (fig.8.8). The main waste at nuclear power plants are radioactive resins that purify water in circuits.

As far as the energy is produced due to the fission of uranium, a lot of elements are produced that are different from the contents of the fuel. The noble metals form so-called epsilon phase, the fission products may form complex compounds with uranium, salts

and therefore fuel is no longer homogeneous material (fig.8.9 **a)**). Due to the heating of this material there might appear some cracks and when gaseous phase escapes from the fuel through channels it creates bubbles on its surface (fig.8.9 **b)**).



a)

b)

Figure 8.9. **a**):Phases of the spent nuclear fuel; **b**): Bubbles on the fuel surface

Re-processing of the spent fuel

Around 95% of the spent fuel can be re-used to produce new fuel in an open NFC. The graph in fig.8.10 demonstrates the benefits of uranium fuel re-processing.

The upper red line labeled "Total" shows how the radiotoxicity of fresh nuclear fuel decreases with time if we do not re-process it. The green line depicts the constant radiotoxicity of natural uranium ore. It is easy to see, that around 200000 years are required for nuclear fuel to reach the toxicity level of the natural uranium ore - to attain **radio-equivalency**. However, if we take away long-living radionuclides like actionoids from the spent fuel so that we only have fission products such as cesium, strontium, etc. radio-equivalency will be reached within 300 years. Thereby, we only need to ensure the safety of the disposal site for a much shorter period of time.

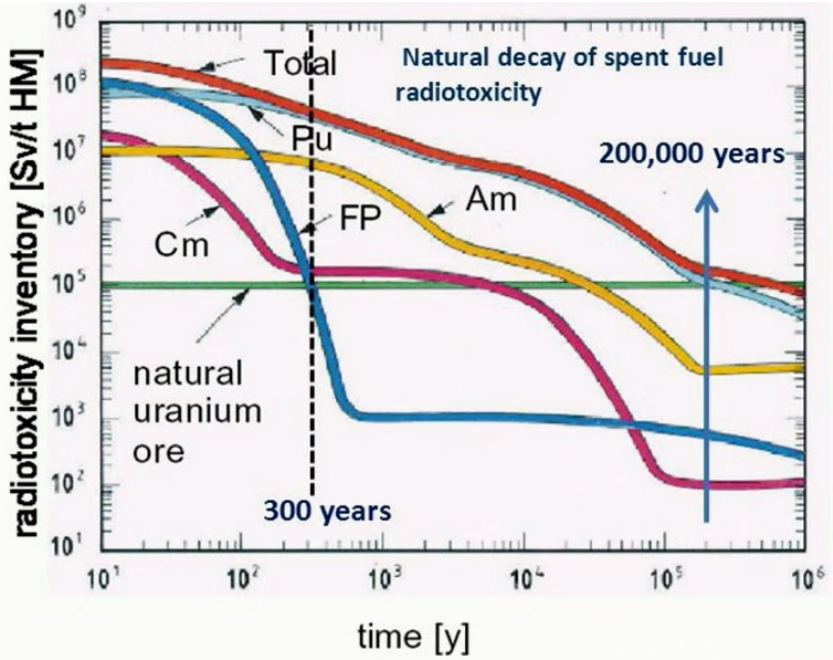


Figure 8.10. Relative radiotoxicity vs tine in years (logarithmic scale)

The extracted long-lived radionuclides undergo the **transmutation** procedure - we induce fission of the nuclei by neutrons and transform them into short-lived radionuclides or even stable elements.

The separation of uranium and plutoniun from other parts of the fuel is done using the **liquid-liquid extraction method** which is based on the difference in **RedOx reactions** for different components (fig.8.11).

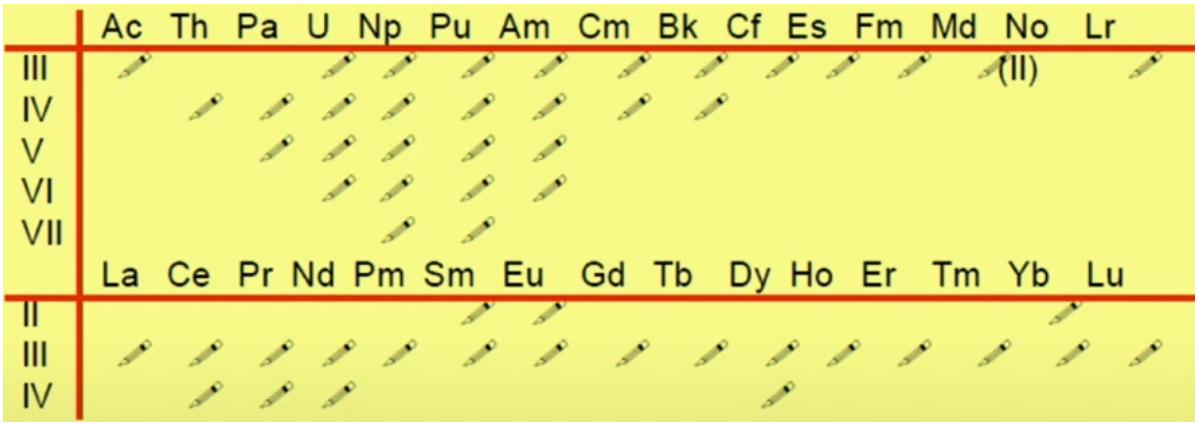


Figure 8.11. RedOx reactions for different components

The PUREX (Plutonium-Uranium extraction process) consists of several steps:

1) The spent nuclear fuel is dissolved in nitric acid.

2) Extraction of plutonium and uranium using organic solution of TBP in kerosene, where uranium and plutonium go to the organic phase.

3) Washing of the organic phase with the aqueous phase containing a reducing agent (Fe(II)) $\Rightarrow$ plutonium transfers to the aqueous phase, uranium remains in the organic phase.

4) Thus, we separated uranium and plutonium from other component and from one another.

Disposal of the spent fuel and high-level waste

During the re-processing of spent fuel a huge amount of liquid waste is generated and stored in natural reservoirs and cement tanks, which may leak consequently causing pollution. The key to the right disposal is the use of the **multi-barrier concept** which implies changing radioactive material into a compact, resistant to any external influence form and putting it behind many layers of protection shells (fig.8.12).

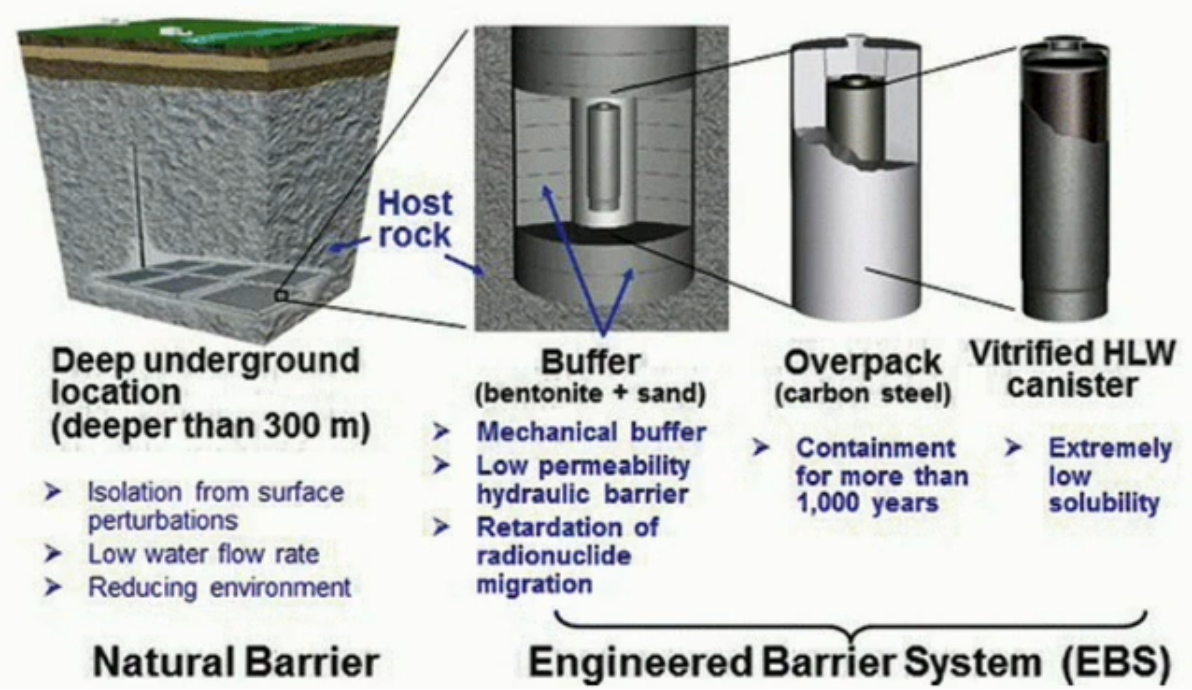


Figure 8.12. The multi-barrier concept for the disposal of radioactive waste

9. Lecture 9. Basics of radio-pharmaceutical chemistry

A quick reminder on shielding against main types of ionizing radiation (fig.9.1).

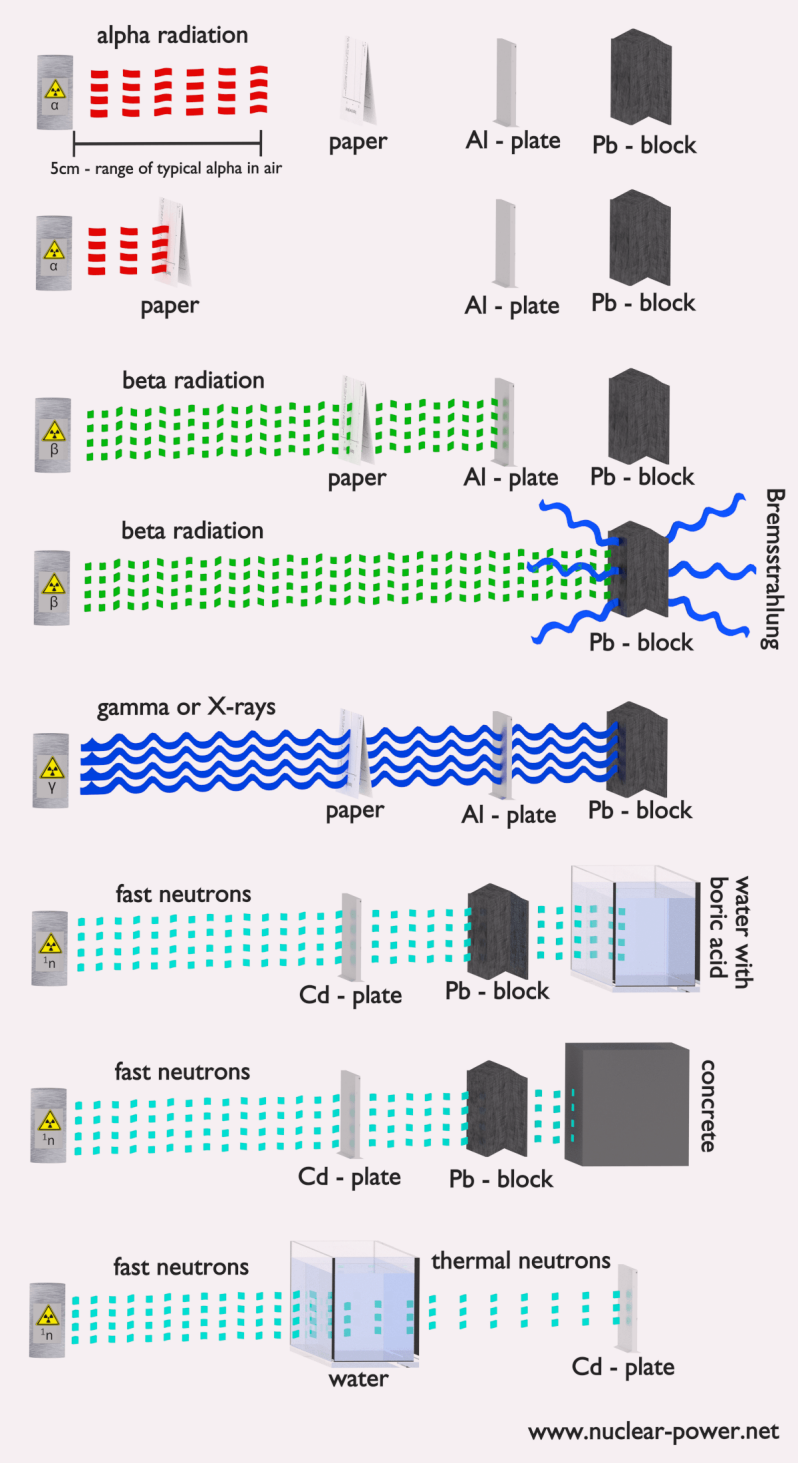


Figure 9.1. Shielding against different types of ionizing radiation

Nuclear medicine

When α or β radiation sources get inside a human body, alpha- and beta-particles are completely absorbed transferring energy to internal organs, tissues and cells. As for $\gamma-$radiation, having high penetration ability it goes out from the human body that has many applications in diagnostics.

Nuclear medicine is a branch of modern medicine that exploits radioactive substances and properties of atomic nuclei for diagnostics and therapy of different diseases. Diagnostics implies identification of structural and functional changes in organs and tissues at the cellular level, thereby enabling the diagnosis of a diseases at the earliest stages. This significantly saves money on treatment and increases the chances of recovery. Therapy uses accumulation of absorbed doses of ionizing radiation in pathological tissues, allowing the death of unhealthy cells with minor side effects and minimal damage to normal tissues.

Radioisotope diagnostics has plenty different applications:

- assessment of the degree of dilution of a radioactive tracer in body fluids (determination of the volume of circulating blood, water exchange, exchange of potassium, sodium, etc.)
- determination of changes (in time) in activity in the organs (study of central and peripheral hemodynamics, hepatography, renography, radio pneumography, determination of the intrathyroid stage of iodine metabolism, study of the dynamics of the relative level of phosphorus metabolism, etc.)
- visualization of the distribution of a radioactive tracer injected into the body (methods of scanning and gamma-scintigraphy of organs and systems: brain, thyroid gland, lungs, liver, kidneys, bone marrow, bones, lymphatic system, etc.)
- determination of the removal of radioactive drugs from the body or their redistribution in biological media (determination of gastrointestinal bleeding, protein-bound iodine in the blood, absorption of neutral fats, etc.)
- interaction "in vitro" of labeled compounds with components of biological media of the body (without introducing radioactive drugs into the body), in particular, interaction of the "antigen-antibody" type (determination of the thyroxine-binding capacity of serum, the concentration of various hormones in the blood, etc.).

Nowadays, basic diagnostic methods (medical imaging) are:

- Sonography (ultrasound). A very safe method to investigate the morphology of the internal organs.
- X-ray methods are widely used for imaging of bones and other organs,
- Computed X-ray tomography (CT)
- Magnetic resonance imaging (MRI). This method does not use of any kinds of ionizing radiation but relies on the response from atomic nuclei in the magnetic field.

- Planar (gamma) scintigraphy (PS). This method along with the next two uses radionuclides injected in a human body (radionuclide diagnostics methods).
- Single photon emission computed tomography (SPECT)
- Positron Emission Tomography (PET)
- and others

Tomography

Tomography is a method that involves of non-destructive layer-by-layer investigation of the internal structure of an object by the means of multiple trans-illumination in various intersecting directions (fig.9.2).

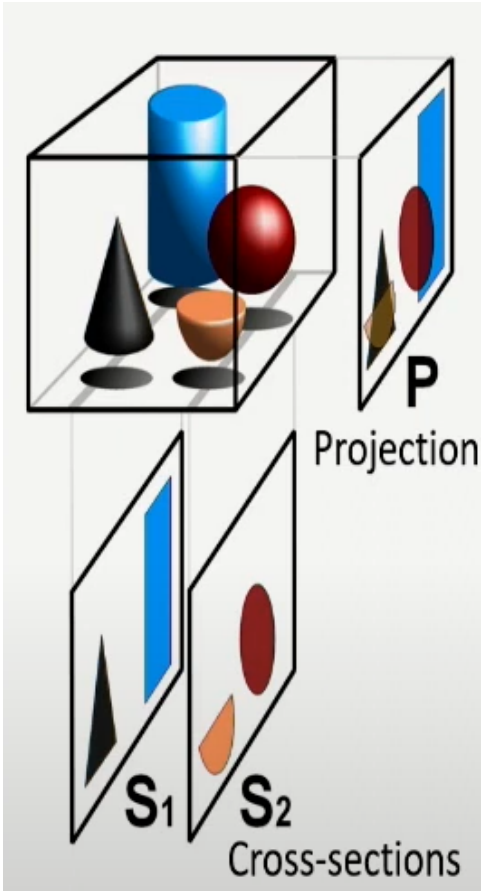


Figure 9.2. Two ways to obtain an image: projection and cross-section

The obtained cross-sections of an object can be combined to get a 3D model.

CT scanning

CT scanning uses a source of X-rays rotating together with a detector around a human body. After a full turn one cross-section is obtained, then the body is shifted along the

rotation axes and the procedure repeats until the sufficient number of cross-sections is acquired and a 3D model of the internal structure of very high resolution is build (fig.9.3). The drawback of such procedure is that a patient is exposed to the dose of radiation of 2-20 mSv whereas the average annual dose is 2 mSv. However, according to the postulate from Lecture 6, since the benefits of CT scanning overshadow the negative effects, its use is completely justified.

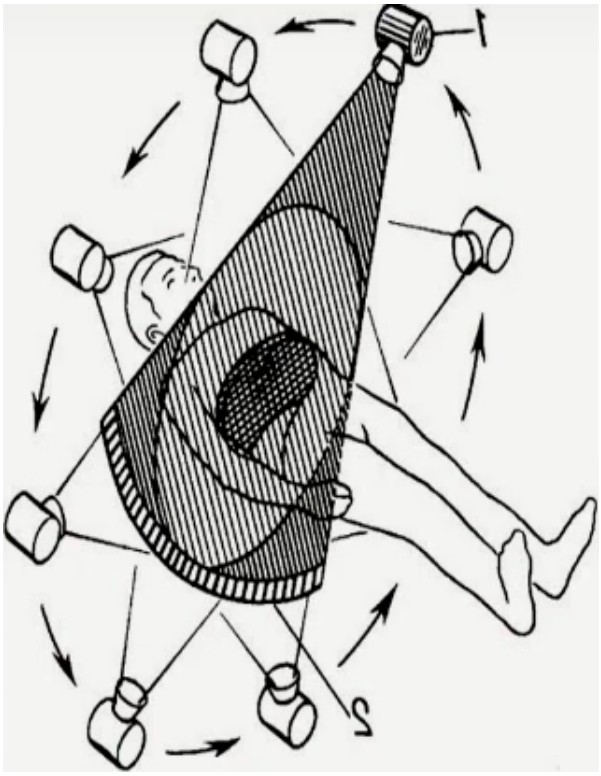


Figure 9.3. CT scanning

Planar scintigraphy

In this method, the radio-pharmaceutical (a drug containing radionuclides succ as ^{123}I, ^{111}In, etc.) is injected in the human body and then a set of detectors register gamma-rays emitted from the body so we can see the distribution of the radionuclides inside. Although we do not get the 3D model of the internal structure, it is often sufficient to obtain projections of some organs.

Single Photon Emission Computed Tomography (SPECT)

In this method we inject gamma-emitting radionuclide (more often than not ^{99m}Tc) inside the human body, that emits photons with a certain energy. A rotating system of collimated detectors registers photons moving at a certain angle with respect to the detectors. The information about intensity of the signal at different angles is collected

and using that distribution of the radionuclides inside the body is obtained. This allows to get cross sections of the internal body structure.

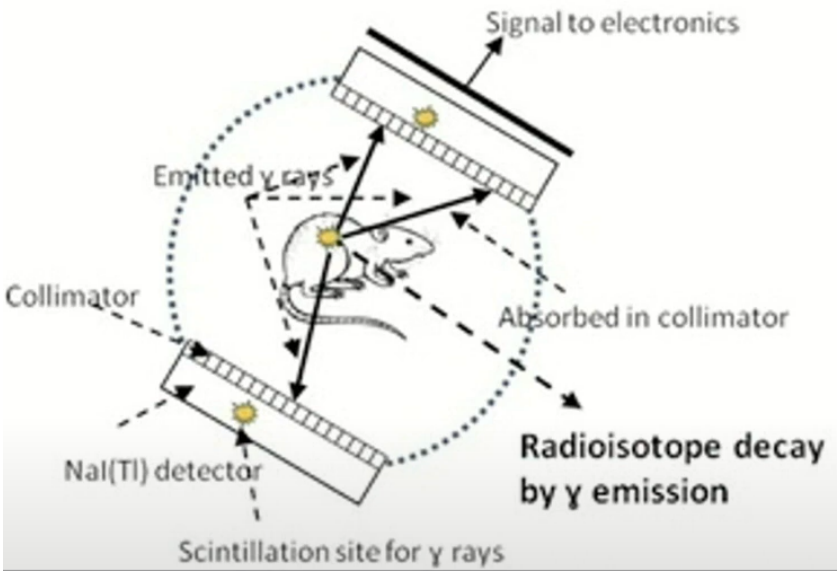


Figure 9.4. SPECT method

Positron emission tomography (PET)

This method exploits β^+ decay and electron capture that are characteristic of neutron-deficient nuclei. In such processes positrons are emitted, that then annihilate with electrons producing two gamma quanta moving in opposite directions. The detector system registers them using a coincidence scheme and reconstructs the point, from which they were emitted.

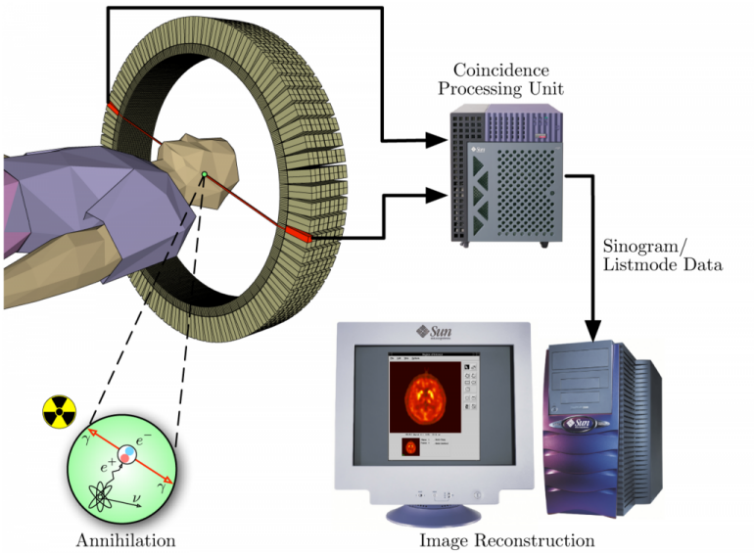


Figure 9.5. PET method

As a result, cross sections of the internal organs are obtained, pretty much like in normal tomography. Different radionuclides emit positrons with different energies, therefore the positrons travel different distances until they annihilate. Those distances are in the range of few millimeters which means the resolution of the image is limited to $\sim 2-3$ mm.

In practice, the combination of the above-mentioned methods is used to achieve accurate diagnostics.

Requirements for radionuclides to be used in diagnostics:

- Suitable energy of gamma-quanta and positrons (100-250 KeV).
- Minimum dose from other types of radiation, no hard gamma-lines
- Short half-life of the used radionuclides
- For PET: high yield and low decay energy of positrons.

Classifications of radiotherapy

We can classify different methods of radiotherapy by the **type of radiation** we use:

- **Corpuscular**
  - $\alpha-$particles
  - $\beta-$particles
  - neutrons
  - protons
- **Wave**
  - X-ray radiation
  - $\gamma-$radiation

and by the **position of the source of ionizing radiation**:

- External beam radiation therapy (Teletherapy)
- Contact radiation therapy (Brachytherapy)
- Radionuclide therapy

Radionuclide radiotherapy

In this method particles with short penetration range are used: Auger electrons (~ 10 nm), $\alpha-$particles ($\sim 50-80$ μm), low-energy $\beta-$particles ($\sim 1-10$ μm). The main idea is to irradiate unhealthy, damaged cells in the human body and destroy their DNA.

Radiopharmaceuticals are radioactive isotopes or their compounds with various inorganic or organic substances, intended for biomedical research, radioisotope diagnostics

and treatment of diseases, mainly for radiation therapy of malignant tumors. In other words, radiopharmaceuticals are diagnostics therapeutic agents in a ready-to-use form containing one ore more radionuclides.

Radionuclides can be injected inside a human body in different forms: gas, salt ($^{223}RaCl_2$), radionuclides coupled with chelators, compound with chelators and biomolecules, nanoparticles, modular nanotransporters.

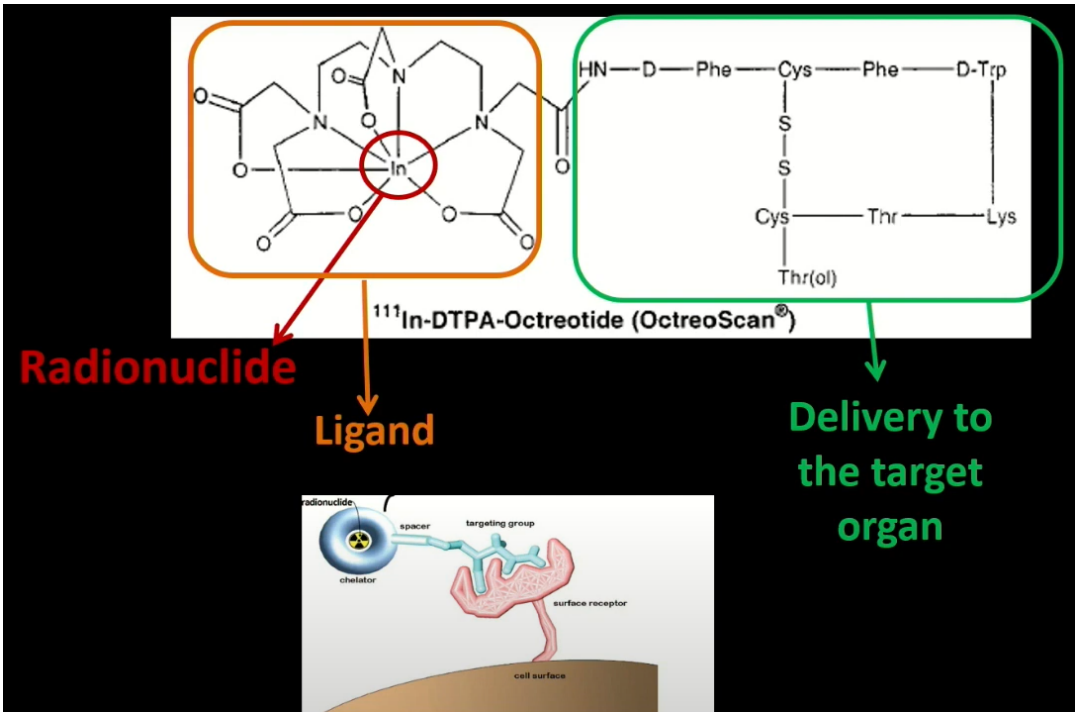


Figure 9.6. Radionuclides compound with chelators and biomolecules

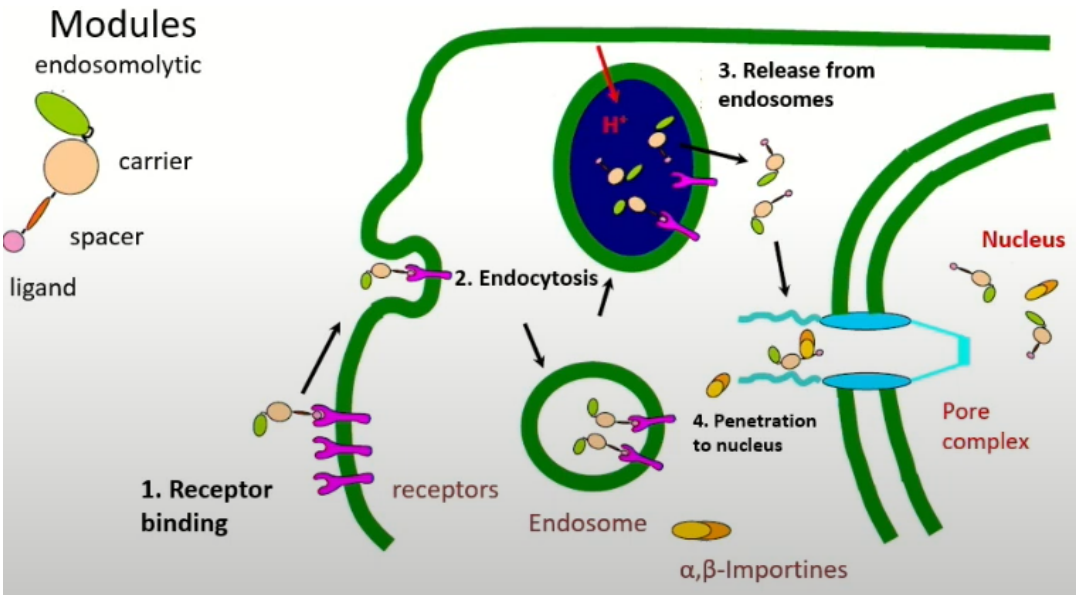


Figure 9.7. Modular nanotransporters

Brachytherapy

Brachytherapy is a type of radiation therapy when the radiation source is introduced into the affected organ (fig,9.8).

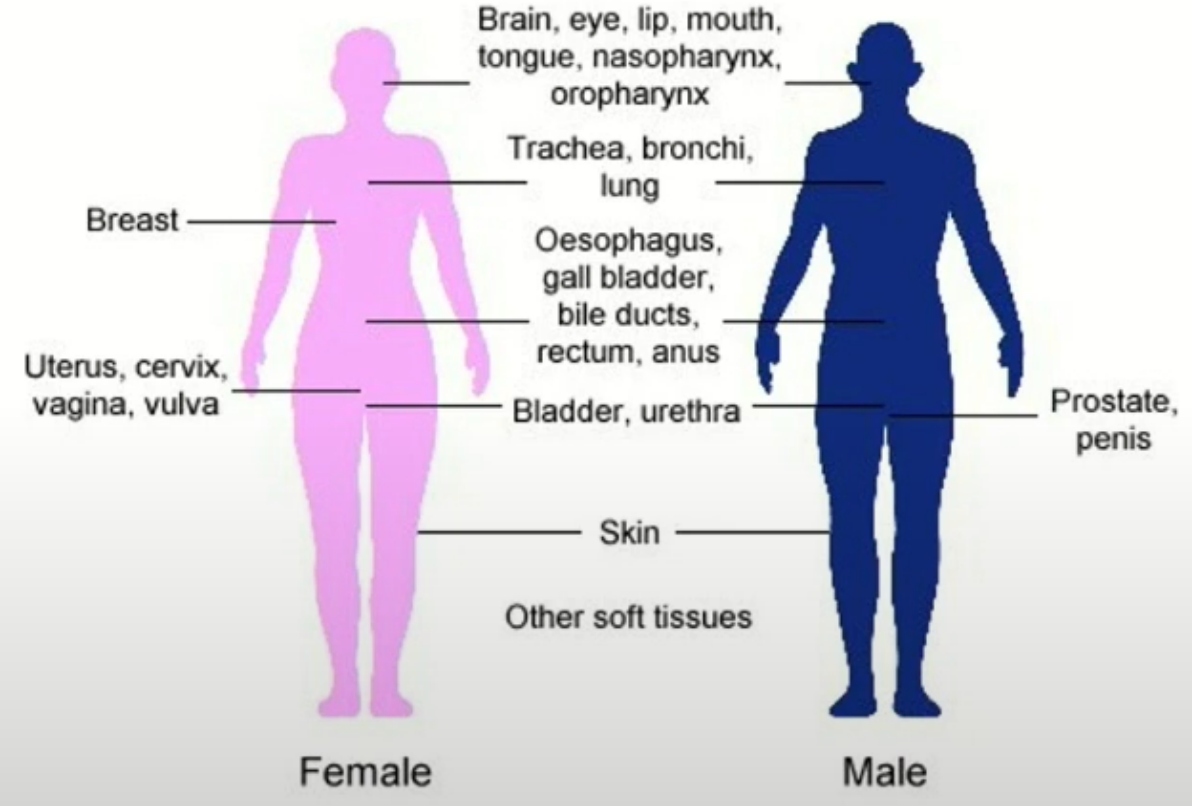


Figure 9.8. Organs, that might be treated with brachytherapy

We can combine both therapy and diagnostics. This approach is called **theranostics** and the following radionuclides are widely used for this purpose (see fig.9.9):

Radionuclide	$T_{1/2}$	Energy
Lu-177	6,6d	$E_{\beta^-}{}^{max}$=0,49MeV; $E_{\gamma}{}^{>10\%}$=208keV
Cu-64	12,7h	$E_{\beta+}{}^{max}$=0,65MeV; $E_{\beta^-}{}^{max}$=0,58MeV; $E_{\gamma}{}^{>10\%}$=511keV
Cu-67	2,6d	$E_{\beta^-}{}^{max}$=0,58MeV; $E_{\gamma}{}^{>10\%}$=185кэВ, 93keV
Pb-212	10,6h	$E_{\beta^-}{}^{max}$=0,57MeV; $E_{\gamma}{}^{>10\%}$=239keV

Figure 9.9. Radionuclides used in theranostics

Synthesis of radiopharmaceuticals

Radionuclide production

- Reactor
- Cyclotron
- Radionuclide generators

Radionuclide purification

- Ion exchange
- Extraction

Synthesis of chelator and conjugate

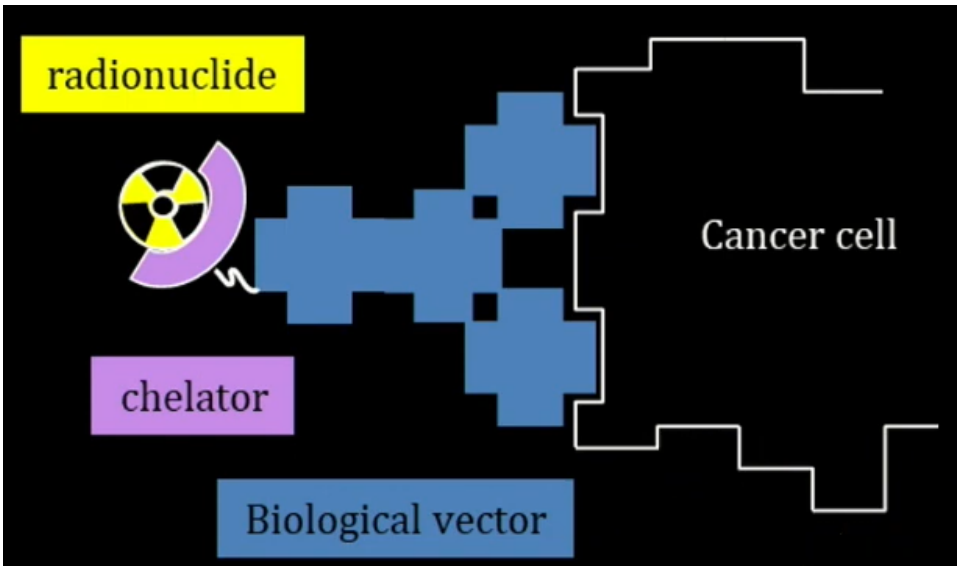


Figure 9.10. Synthesis of chelator and conjugate

The task of radiochemists is to develop a simple and effective synthesis method.

Basic requirements for radiochemical synthesis

- If multi-stage synthesis is required, then the tracer should be introduced at the last stages if possible (radiation safety and economic considerations).
- It is necessary to use small amounts of reagents (microsynthesis).
- Requires special safety measures.
- Sometimes it is necessary to take into account the effect of ionizing radiation on the course of the reaction (radiolysis of the starting compounds and reaction products).

Basic requirements for ligands

- Ligands must quantitatively bound radionuclide
- Be synthesized at room temperature
- Be synthesized as quickly as possible

In conclusion, nuclear medicine is only possible owing to medics, physicists, chemists and biologists working side by side.

10. Lecture 10. Engineering safety barriers

Nuclear waste repositories and storage facilities

The map in fig.10.1 shows how many nuclear facilities (nuclear power plants, reprocessing facilities, nuclear waste repositories, etc.) are located around the world.

Figure 10.1. Distribution of nuclear facilities around the globe.

The International Atomic Energy Agency (IAEA) uses the classification on radioactive waste shown in fig.10.2. The majority of facilities are dedicated to storage and disposal of intermediate, low and very low level waste. There are no licensed facilities for disposal of high-level waste.

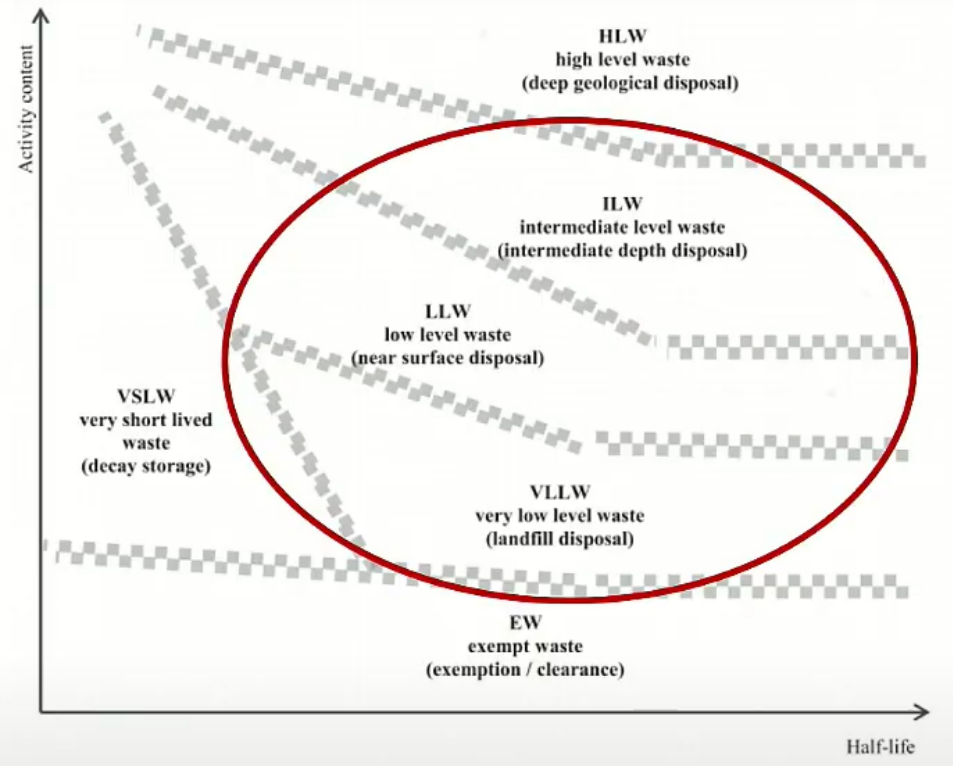


Figure 10.2. Different types of radioactive waste

According to IAEA classification for different classes of waste different types of repository facilities are used. They must meet the requirements regarding the depth of storage underground (fig.10.3).

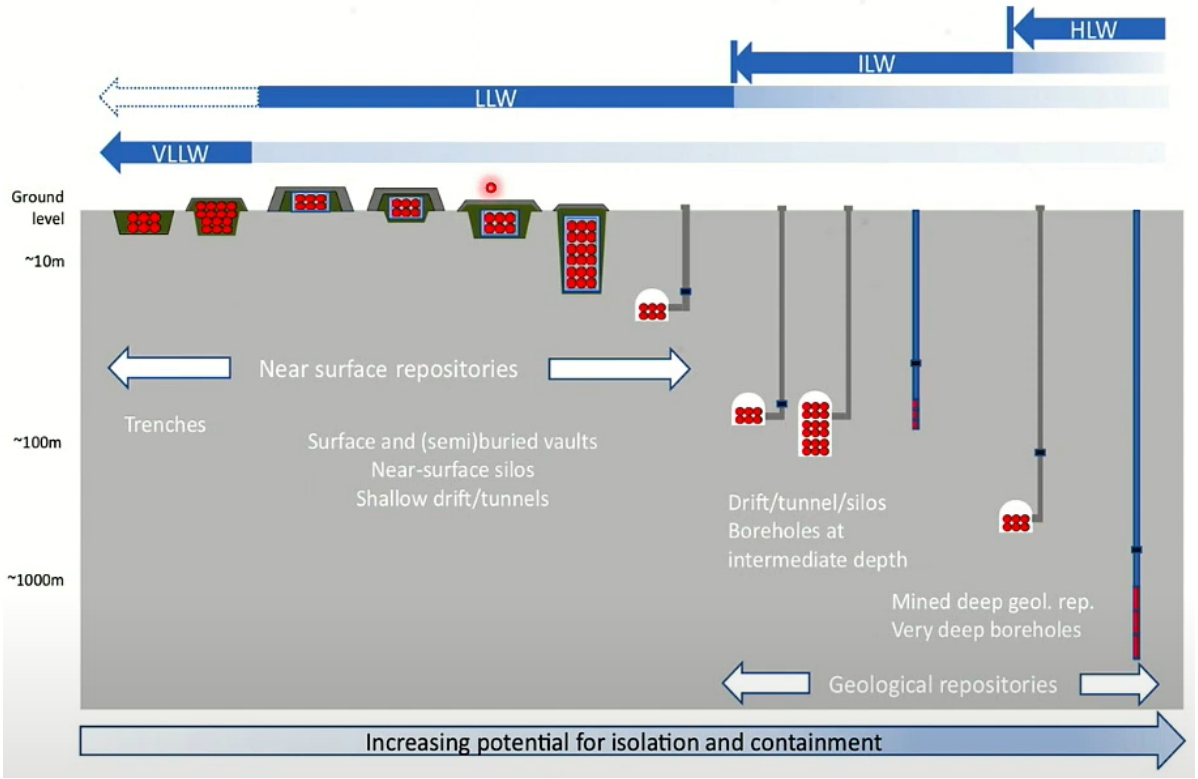


Figure 10.3. Depth requirements for radioactive waste storage

Driving forces that cause the migration of RN:

- Filtering of atmospheric precipitation
- Capillary influx of moisture to the surface as a result of evaporation, thermal transfer under the influence of temperature gradient
- Water movement over surface
- Diffusion of anions and cations, if water enters repository
- Transport on colloidal particles

Structure of nuclear waste repositories

For the storage of low and very low-level waste IAEA recommends using trenches with several layers of protection: supporting geotextile, granulated bentonite, covering geotextile. For intermediate and high-level waste bentonite blocks are placed between containers and bentonite pellets fill the tunnels. The main material for containers in many countries is metal and reinforces concrete.

Metal safety container is designed for:

- the collection and intermediate storage of solid or solidified RAW of low and medium specific activity;
- transporting radioactive waste to the processing and conditioning facilities;
- the preparation of conditioned radioactive waste packages by impregnating the solid waste with cement mortar;
- storage, long-term storage (up to 50 years), transportation and disposal of conditioned radioactive waste.

General requirements for engineering barriers:

- Limit contact of radioactive waste packages with environmental water
- Prevent the destruction of radioactive waste packages under natural and technogenic impacts
- Prevent the destruction of radioactive waste packages under the influence of surrounding rocks
- Limit distribution of radionuclides into the surrounding rocks within the limits established in the project

The relationship between the roles, objectives of the barrier and its properties is summarized in the table in fig.10.4.

Objectives	Properties
To stop/ minimize leakage of contaminant beyond the barrier	low filtration and diffusion properties of the barrier High sorption characteristics, low radionuclide desorption
Impossibility of formation of rapidly migrating forms of radionuclides during the exploitation of the barrier	Low ability to form colloids, lack of substances capable of forming complex compounds with RNs
No need to maintain the barrier during the entire period of operation	Material stability over time (hundreds to tens of thousands of years)
Resistance to tectonic movements and seismic events	Barrier plasticity
Fire and explosion safety and low toxicity of the barrier components	Determined by the choice of barrier material
Acceptable cost, availability of materials and technology	Determined by the choice of barrier material

Figure 10.4. Objectives and properties of the barrier

Bentonite's features:

- Low water permeability
- Ability to "self-heal"
- Long term stability of properties
- High sorиtion properties, low desorиtion of radionuclides
- Low diffusion

Bentonite's features:

- Effective sorption of radionuclides in case of probable depressurization of waste tanks
- Creating conditions under which mass exchange between waste and groundwater is possible only through diffusion
- Restriction of water access to radioactive waste
- Sealing of open cracks and large pores in the rocks due to high swelling capacity
- Stability during operation
- Plasticity

The scheme of a low level radioactive waste repository is shown in fig.10.5

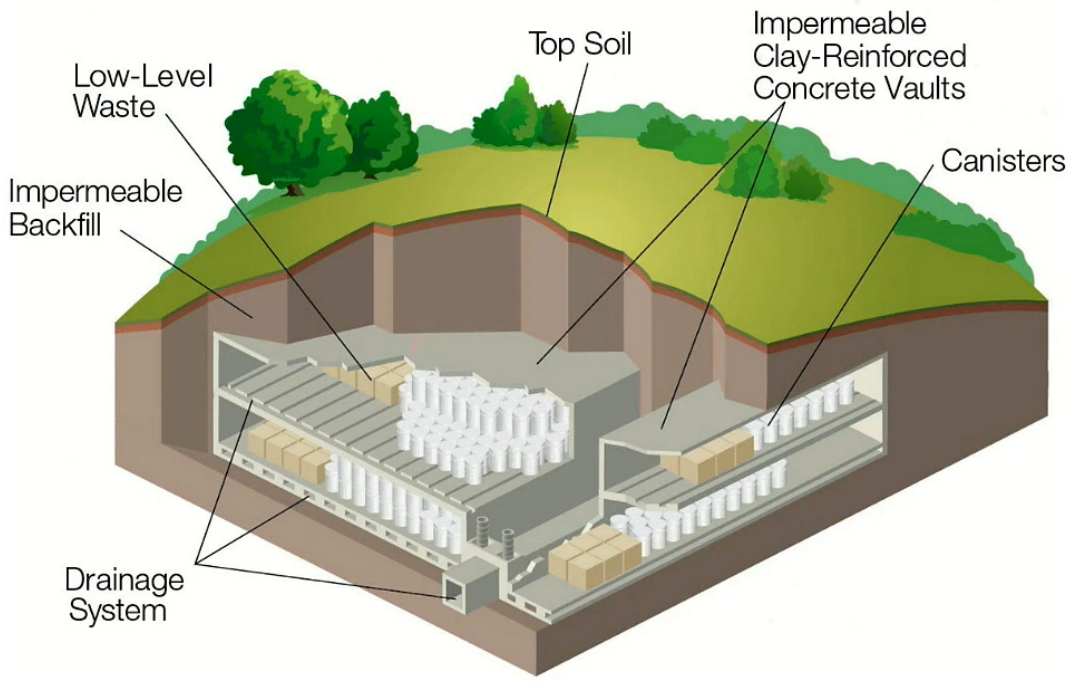


Figure 10.5. Low level radioactive waste repository

The project of disposal of long-lived low-level and intermediate level radioactive waste in concrete structures, placed in crystalline rocks at a depth of about 300 m. Four variants of the concept are considered, let us consider two of them.

a) The waste is placed inside a rectangular concrete structure with walls 0.5 m thick using bentonite blocks as backfill material

b) The waste is placed inside a cylindrical bunker with two-layer walls of concrete. The voids between the two walls on the top and sides are filled with bentonite, and on the bottom — with a mixture of sand and bentonite. The bunker is installed on a gravel base, the space between the bunker walls and the host rocks is filled with gravel.

Another approach to improve safety of storage facilities is to use the **wall-in-soil technology**. A rectangular-shaped well is created and inside put a mixture of clay and other materials. Then several more walls are created (fig.10.6).

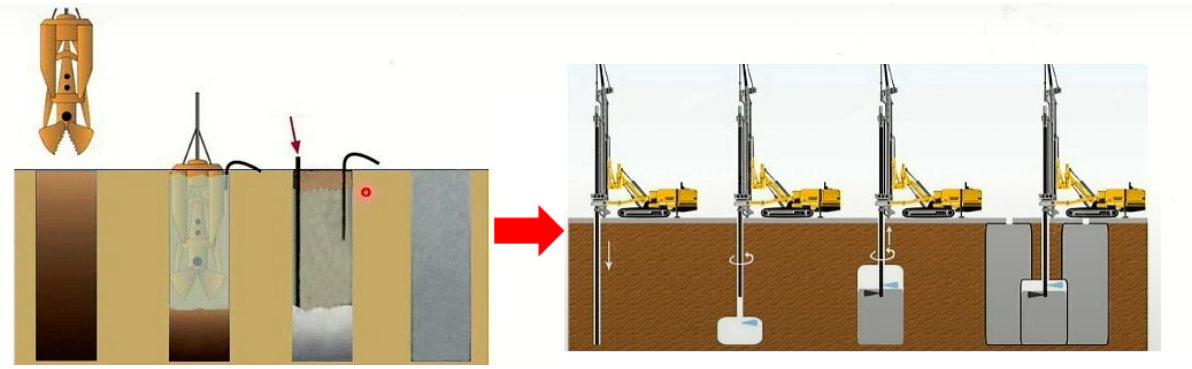


Figure 10.6. The wall-in-soil approach

Modeling shows, that safety barriers prevent uranium leakage for 1000 of years.
Characteristics of barrier material:

- Technological
  - Granulometric, mineral and chemical composition
  - Bulk density, moisture, colloidality, moisture capacity, specific surface
  - Flexibility and deformation of disperse materials in dry (or wet) state
  - Swelling pressure, permeability and water-saturated deformation
- Functional
  - Sorbtion/desorbtion
  - Diffusion
  - Swelling pressure
  - Organic matter content
  - Evaluation of bacteriological activity on material degradation

– Formation of colloids
– Evolutionary properties under storage conditions (highly dependent on composition, temperature, cyclic freezing)

The scheme in fig.10.7 shows possible chemical interactions of radionuclides when they leak into underground water.

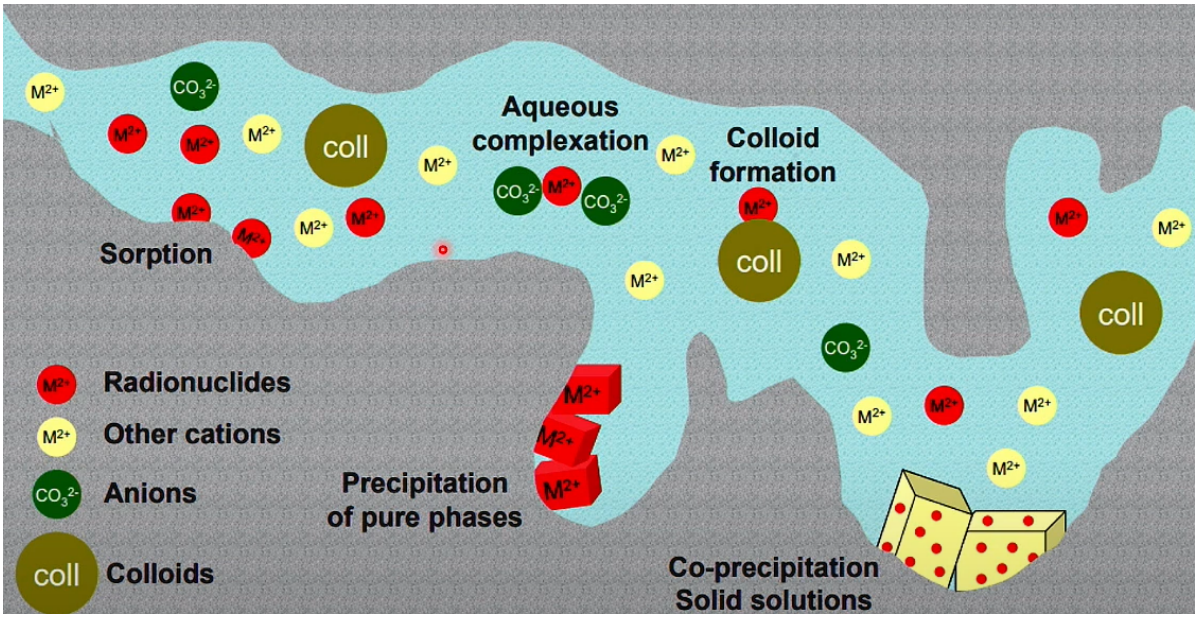


Figure 10.7. Interactions of radionuclides

11. Lecture 11. Labeled compounds. Basic concepts.

Method of radioactive tracers

Method of radioactive tracers uses labeled compounds: a foreign substance mixed with or attached to a given substance to enable the distribution or location of the latter to be determined subsequently.

There are several types of tracers which are used:

I) A physical tracer is one that is attached by physical means to the object being traced;

II) A chemical tracer is a chemical with properties similar to those of the substance being traced with which it is mixed homogeneously;

III) An isotopic tracer (only differs in isotopic composition from the substance to be traced) is a unique isotope, either radioactive or an enriched, uncommon stable isotope, of the element to be traced;

IV) A radioactive tracer is a physical or chemical tracer having radioactivity as its distinctive property which allows detection at small concentrations and hence after large transport distances.

Nomenclature of radioanalytical chemistry. Labeled molecules with altered isotopic composition are divided into two groups:

- **Isotopically substituted compounds**

  With the increasing use of isotopically substituted (i.e. "carrier-free") compounds, especially in studies of the steric course of reactions, the conventions for specifying such compounds may now be of more importance to biochemists than they have been.

  The symbol for the isotope may be placed in the structural formula or in round brackets immediately before the name of the compounds, for example: $^{14}CH_4$ or (^{14}C)methane, CH_3-$^{12}COOH$ or (1-^{14}C)acetic acid, $^{14}CH_3$-$^{12}COOH$ or (2-^{14}C)acetic acid.

- **Isotope labeled compound**

  Chemical substance in which part of the molecules are labeled with an isotope so that observations the isotopic composition make it possible to follow the compound or its fragments through physical, chemical, or biological processes.

  We write the isotope in square brackets before the name of the compound within a formula: [^{18}O]ethanol or CH_3-CH_2-[^{18}O]H, [2-[^{18}O]]glycolic acid or H[^{18}O]-CH_2-COOH

Types of isotopically labeled compounds

There are several types of isotopically labeled compounds listed in fig.11.1.

Isotopically labelled compound	Formular and name
Specifically labeled	$C[^2H_3]CH_2CH_3$ $[1\text{-}^2H_3]$propane
Selectively labeled or Nominally labelled	$[1\text{-}^2H]CH_3CH_2CH_3$ $[1\text{-}^2H]$propane
Non-selectively labeled	$[^2H]CH_3CH_2CH_3$ $[^2H]$propane
Uniformly labeled	$[U\text{-}^2H]CH_3CH_2CH_3$ $[U\text{-}^2H]$propane
Generally labelled	$[G\text{-}^2H]CH_3CH_2CH_3$ $[G\text{-}^2H]$propane
Isotopically deficient	$[def\,^2H]CH_3CH_2CH_3$ $[def\,^2H]$propane

Figure 11.1. Types of isotopically labeled compounds

- **Specifically labeled tracer**

  A tracer in which the label is present in a specified position. An isotopically labeled compound is designated as specifically labeled when a unique isotopically substituted compound is formally added to the analogous isotopically unmodified compound. In such a case, both position(s) and number of each labeling nuclide are defined.

Isotopically substituted compound	when added to	Isotopically unmodified compound	gives rise to	Specifically labelled compound
$^{13}CH_4$		CH_4		$[^{13}C]H_4$
$CH_2{}^3H_2$		CH_4		$CH_2[^3H_2]$

Figure 11.2. Specifically labeled compound

Notice, that although the formula for a specifically labeled compound does not represent the composition of the bulk material, which usually consists overwhelmingly of the isotopically unmodified compound, it does indicate the presence of the compound of chief interest, the isotopically substituted compound.

- **Selectively (nominally) labeled compound**

  A tracer in which the label is present mainly in a specified position. An isotopically labeled compound is designated as selectively labeled when a mixture of isotopically substituted compounds is formally added to the analogous isotopically unmodified compound in such a way that the position(s) but not necessarily the number of each labeling nuclide is defined. A selectively labeled compound may be considered as a mixture of specifically labeled compounds.

  A selectively labeled compound cannot be described by a unique structural formula; therefore it is represented by inserting the nuclide symbols preceded by any necessary locant(s) (letters and/or numbers) but without multiplying subscripts, enclosed in square brackets directly before the usual formula or, if necessary, before parts of the formula that have an independent numbering. Identical locants are not repeated. When different nuclides are present, the nuclide symbols are written in alphabetical order according to their symbols, or when the atomic symbols are identical, in order of increasing mass number.

Mixture of isotopically substituted compound	when added to	Isotopically unmodified compound	gives rise to	Selectively labelled compound
$CH_3{}^3H$, $CH_2{}^3H_2$, CH^3H_3, C^3H_4 or any two or more of the above		CH_4		$[^3H]CH_4$ [^{3}H]Methane not $[^3H_4]$Methane

Figure 11.3. Selectively labeled compound

  The name of a selectively labeled compound is formed in the same way as the name of a specifically labeled compound, except that the multiplying subscript following the atomic symbols are generally omitted. Identical locants corresponding to the same element are not repeated. The name of a selectively labeled compound differs from the name of the corresponding isotopically substituted compound in the use of square brackets surrounding the nuclide descriptor rather than parentheses and in the omission of repeated identical locants and multiplying subscripts.

- **Non-selectively labeled tracer**

  An isotopically labeled compound is designated as non-selectively labeled when the position(s) and the number of the labeling nuclide(s) are both undefined.

  – Non-selectively labeling is indicated in the formula by inserting the nuclide symbol, enclosed in square brackets, directly before the usual line formula with no locants or subscripts: $[^{13}C]CH_3$-CH_2-CH_2-COOH.

  – The name of a non-selectively labeled compound is formed in the same way as the name of a selectively labeled compound but contains neither locants

nor subscripts in the nuclide descriptor: [^{13}C]butyric acid.

- **Uniformly labeled tracer**

  A tracer in which the label is uniformly distributed over its possible positions.

  Symbol "U" describing a labeled compound in which the label is distributed in a statistically uniform, or nearly uniform, manner between all the possible positions in the molecule.

  [U-^{14}C]propionic acid = mixture of equimolar amounts of (1-^{14}C)propionic acid, (2- ^{14}C)propionic acid and (3- ^{14}C)propionic acid as well as non-labeled propionic acid.

- **Generally labeled tracer**

  A tracer in which the position of the label is not defined.

  Symbol "G" describing the labeling of a molecule in such a way that a radionuclide may be present at any or all [but not necessarily all) possible positions: [G-^{3}H]lysozyme

- **Isotopically deficient tracer**

  An isotopically labeled compound may be designated as isotopically deficient when the isotopic content of one or more elements has been depleted, i.e., when one or more nuclide(s) is (are) present in less than the natural ratio.

  – Isotopic deficiency is denoted in the formula by adding the italicized syllable "def" immediately preceding, without a hyphen, the appropriate nuclide symbol: [def^{14}C]$CHCl_3$
  – The name of an isotopically deficient compound may be formed by adding the italicized syllable "def" immediately preceding, without a hyphen, the appropriate nuclide symbol, both enclosed in square brackets and cited before the name or that part of the name that is isotopically modified: [def^{14}C]Cloroform

Methods for preparing labeled compounds

- Chemical synthesis
- Isotope exchange
- Biosynthesis
- Nuclear chemical methods:
  – "Hot" fusion (method of recoil atoms)
  – β-decay of labeled compounds
- Physical chemical methods:
  – photolysis
  – chemical accelerators and discharge

– radiation chemical synthesis

Basic requirements for radiochemical synthesis

- If multi-stage synthesis is required to obtain a compound, then the label should be introduced, if possible, at the last stages, both from the standpoint of radiation safety and for economic reasons. Labeled compounds are usually more expensive than any complex unlabeled precursor, and in the case of multistage synthesis with the participation of a labeled compound, losses are inevitable due to the fact that the chemical yield is never 100%.
- When synthesizing compounds with high specific radioactivity, it is necessary to use small amounts of reagents; in fact, we are talking about micro-synthesis, which requires special equipment and methods of work.
- When working with large radioactivity, special safety measures are required.
- Sometimes it is necessary to take into account the effect of ionizing radiation on the course of the reaction (radiolysis of the starting compounds and reaction products).

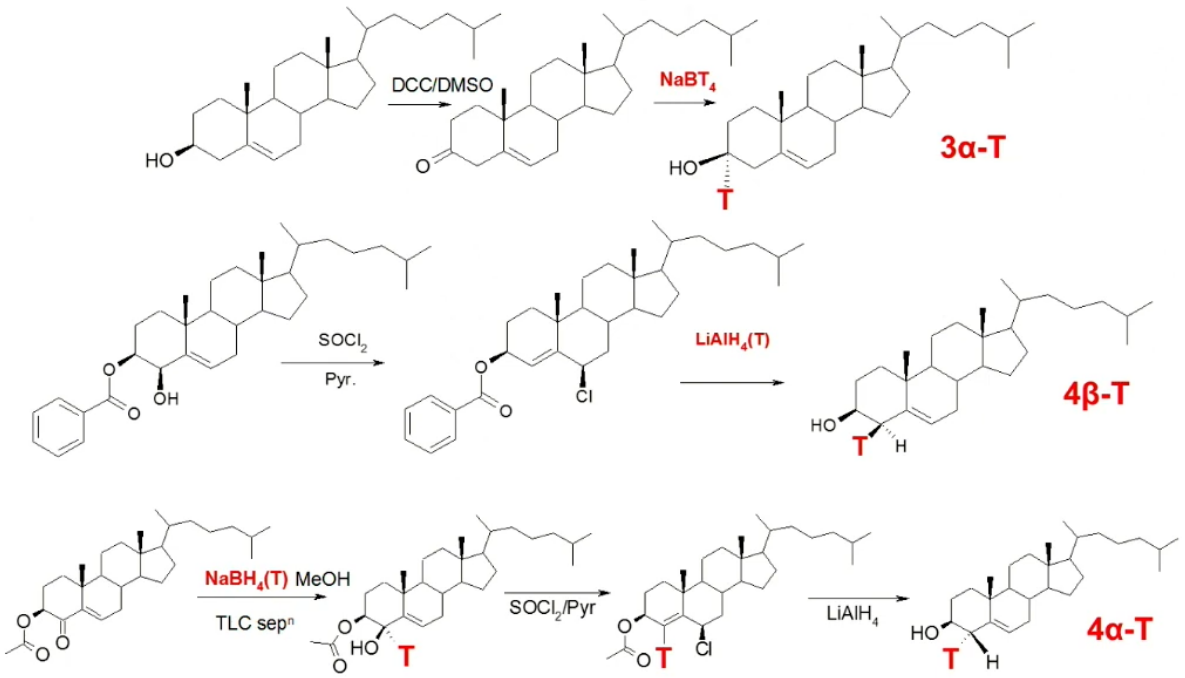


Figure 11.4. An example of **chemical synthesis**

Isotope exchange - the exchange of places between isotopes of atoms in different chemical or physical states or positions.

Isotope exchange is a chemical reaction in which the reactant and product chemical species are chemically identical but have different isotopic composition. In such a reaction the isotope distribution tends towards equilibrium as a result of transfers of isotopically different atoms or groups.

The whole variety of isotope exchange mechanisms can be divided into two groups:

1) Electronic transitions only (RedOx reactions) which proceed quickly and with low activation energy:

$$^{239}Pu^{3+} + ^{238}Pu^{4+} \rightleftarrows {}^{238}Pu^{3+} + ^{239}Pu^{4+} \quad (11.1)$$

$$^{99m}TcO_4^- + ^{99}TcO_4^{2-} \rightleftarrows {}^{99}TcO_4^- + ^{99m}TcO_4^{2-} \quad (11.2)$$

2) With the transfer of ions, atoms or groups of atoms. The exchange of isotopes in the latter case can proceed by **dissociative mechanism** :

$$^{123}I_2(gas) \leftrightarrow 2^{123}I \quad (11.3)$$

$$RI + ^{123}I \leftrightarrow R^{123}I + I \quad (11.4)$$

$$(11.5)$$

and by **associative mechanism**:

$$I_2(aq) + ^{125}I^- \leftrightarrow [^{125}II_2]^- \leftrightarrow ^{125}II + I^- \quad (11.6)$$

Biosynthesis of labeled compounds is transformation of radioactive substrates into more complex compounds in living organisms (in vivo) or by means of enzymatic reactions (in vitro).

^{14}C-Labeled Fructose from 14C-Labeled Glucose:

Reaction I (chemical):

$$Glucose + NaBH_4 \rightarrow Sorbitol \quad (11.7)$$

Reaction II (coupled enzyme):

$$Sorbitol + DPN^+ \xleftrightarrow[dehidrogenase]{sorbitol} Fructose + DPNH + H^+ \quad (11.8)$$

$$Pyruvate + DPNH + H^+ \xleftrightarrow[dehidrogenase]{lactile} Lactate + DPN^+ \quad (11.9)$$

Advantages of biosynthesis:

- obtaining biologically active compounds (in the required enantiomeric and diastereomeric form);
- no restrictions on the complexity of the chemical structure

Disadvantages of biosynthesis:

- high radiation sensitivity;
- impossibility of obtaining compounds with high total and specific radioactivity;
- the need to isolate the target product from a complex mixture of compounds

Self-induced exchange reaction with tritium gas (Wilzbach method).
A method for the random labeling of a compound with tritium by exposing it to isotopically pure tritium gas in a sealed container for several weeks. The exchange is promoted by the radiation.

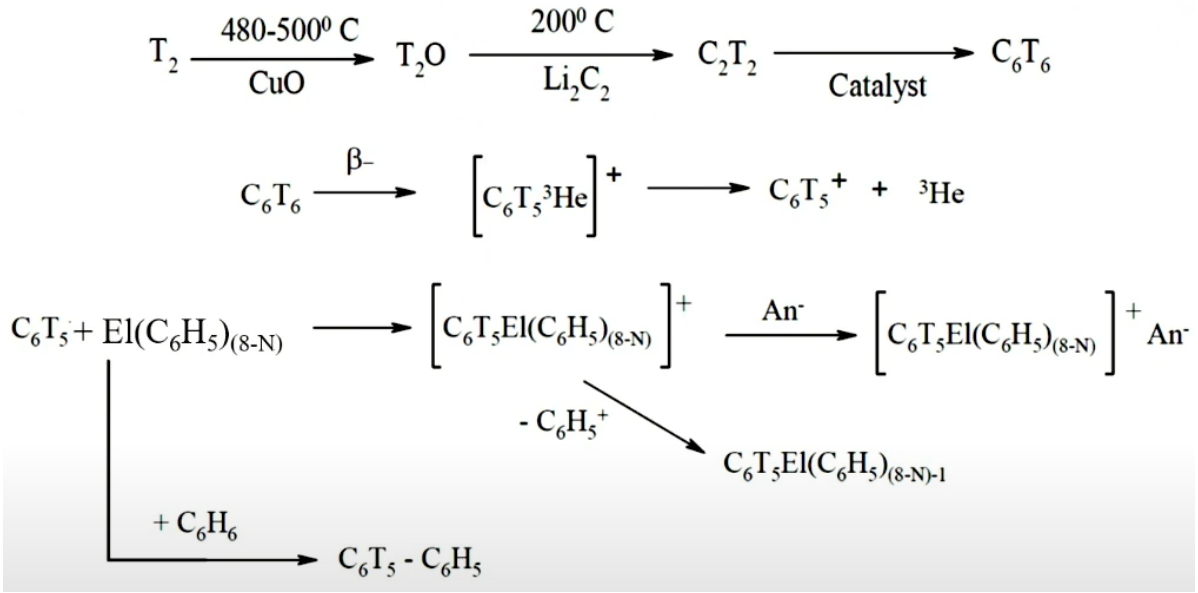


Figure 11.5. Nuclear chemical method for the generation of nucleogenic phenyl-cations and their use in organic synthesis

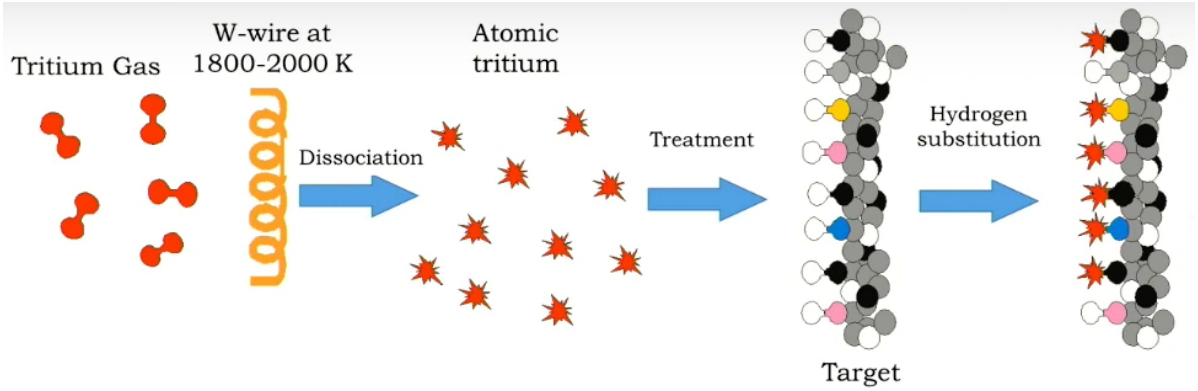


Figure 11.6. Formation of tritium atoms

Tritium atoms can substitute protium at any possible positions - so there occurs a competition of the three reactions (fig.11.7).

The list of compounds that can be activated by the means of tritium thermal activation technique is shown in fig.11.8. As can be seen from the table, the specific activities that are obtained due to tritium activation technique are rather high.

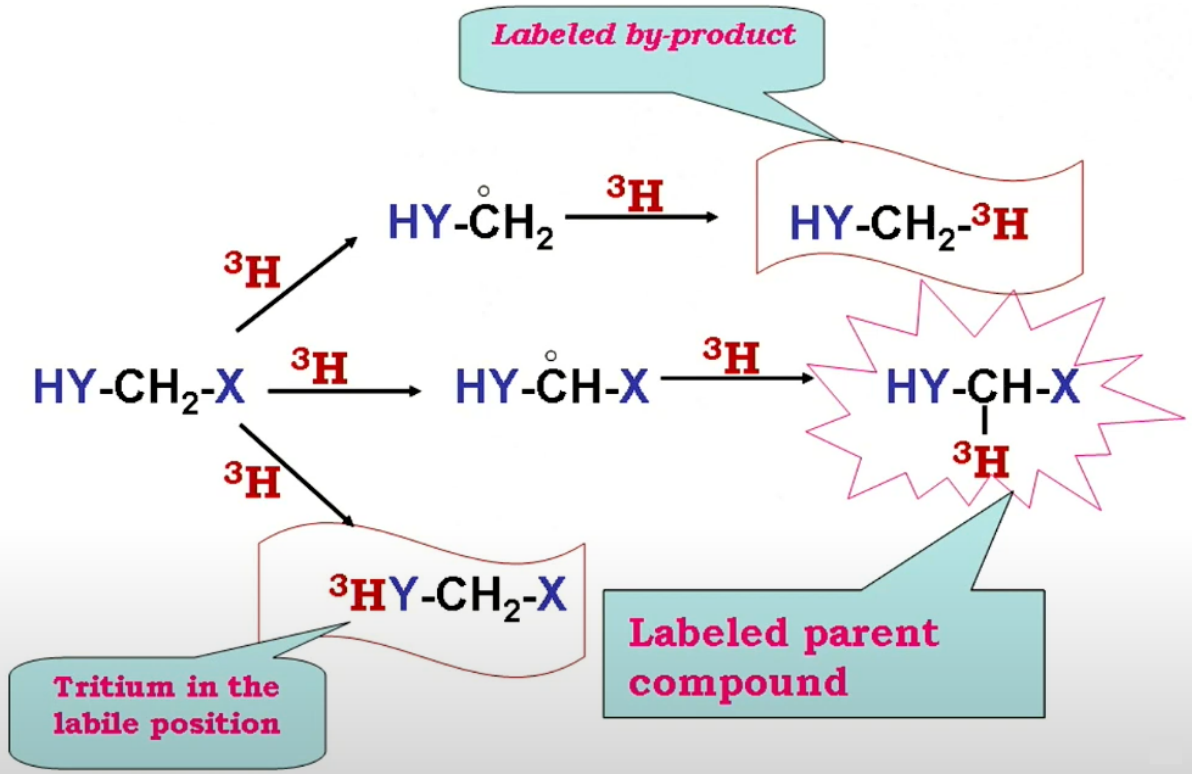


Figure 11.7. Reactions proceeded in the target

Target	Specific radioactivity
Lysozyme	1.71 TBq/mmol [1]
SDS	0.16 TBq/mmol [1]
DTAB	0.16 TBq/mmol [1]
Coal humic acids	0.55 TBq/g [2]
River humic acids	0.63 TBq/g
River fulvic acids	0.56 TBq/g
Nanodiamonds	2.6 TBq/g [3]
Radiochimica Acta 2012 [1], *2010* [2], *2014* [3]	

Figure 11.8. The list of compounds

Examples of application of tritium labeled compounds

- **Nanodiamonds distribution in water-organic liquid mixture**

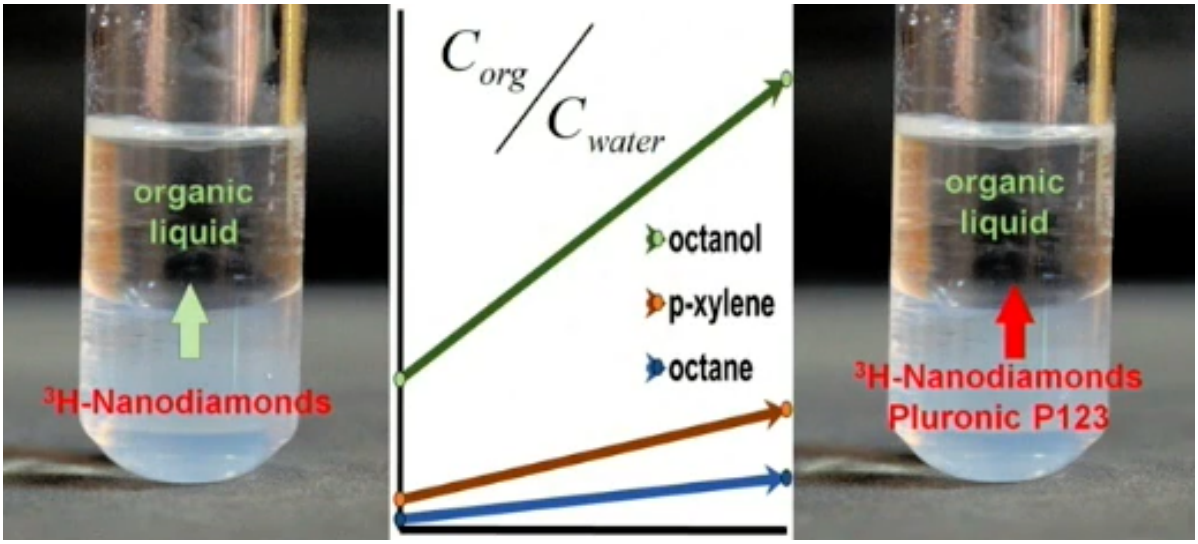


Figure 11.9. Nanodiamonds distribution in water-organic liquid mixture

- **Nanodiamonds uptake by wheat seedlings**

  We can analyze diamonds distribution in wheat seedlings to study how nanoparticles interact with plants by measuring the amount of labeled material in different parts of the plant.

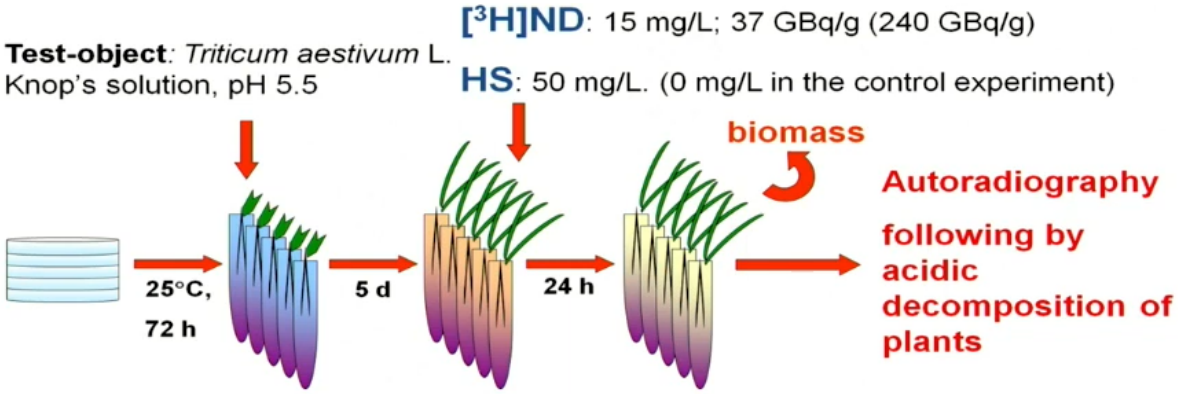


Figure 11.10. The study of nanodiamonds distribution in seedlings

- **Nanodiamonds interactions with cells MCF-7/R**

  Materials:

  – Tritium labeled Nanodiamonds (Sinta)
  – Tritium labeled Myramistin (drug)
  – Tritium labeled Nanodiamonds with adsorbed Myramistin

– Tritium labeled Myramistin adsorbed on nanodiamonds

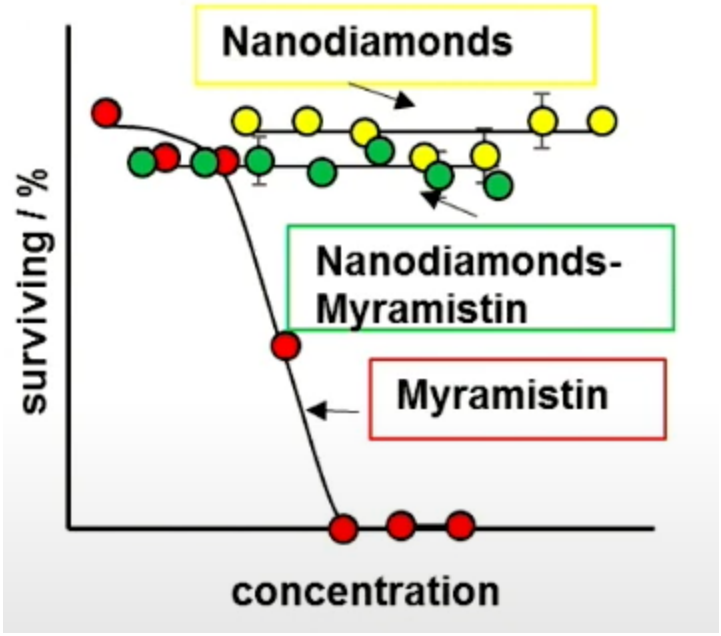
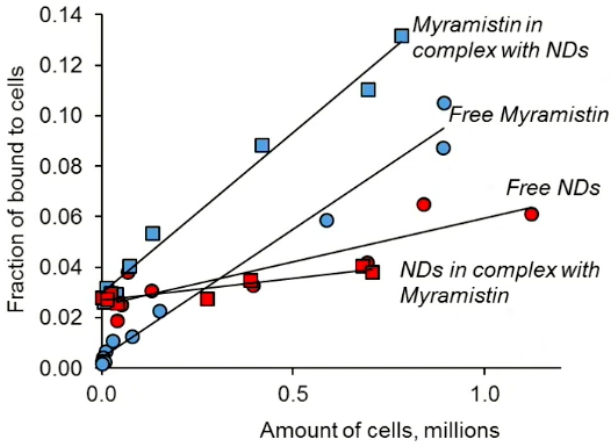


Figure 11.11. The toxicity rate of different materials

And here are some results on nanodiamonds binding with cancer cells. From fig.11.12 we come to a conclusion, that a complex drug+nanodiamond and a nanodiamond itself have significantly higher binding efficiency with cancer cells, therefore such type of drug-carrier has great prospects to be used in the development of drug delivery system.



Component	Binding efficiency, 1/million cells
Nanodiamonds	0.035±0.007
Nanodiamonds in complex with Myramistin	0.017±0.003
Myramistin	0.127±0.007
Myramistin in complex with nanodiamonds	0.102±0.004

Figure 11.12. Nanodiamonds binding efficiency with cancer cells

12. Lecture 12. The overview of the course.

The purpose of this lecture is to briefly summarize what we have studied so far.

- **Radioactivity** is a spontaneous process that is accompanied by the release of the energy and particles.

  Main types of radioactive decay: α, β, γ, spontaneous fission. The first three can be used in **nuclear medicine** for therapy and diagnostics. Spontaneous fission has applications in nuclear energy.

- Due to the radioactive decay there is not only the formation of some stable nucleus after the radioactive decay but it can be a prolonged chain of radioactive decays. There are natural chains and those formed by technogenic radionuclides that can be used for medical, scientific and other purposes.

- There are different types of interactions that occur between ionizing radiation and matter: **excitation**, **ionization** and **nuclear reactions**.

- We can only register ionizing radiation with special detectors. For each type of ionizing radiation different detection approaches are used: gas chambers, semiconductors, track detectors, etc.

- Protection from hazardous ionizing radiation is a matter of paramount importance. Choosing the appropriate **shielding material** we can prevent different types of radiation from entering a human body. People working with ionizing radiation rely on three pillars of radiation protection: **justification**, **ALARA**, **dose limits**.

- The main parameters describing a nuclear reaction are **cross section** which is basically the probability of interaction and the **energy effect** of the reaction.

- More than 10% of global energy production is nuclear and neutron induced nuclear reactions play crucial role in that. **Nuclear energy** production is a large chain of consequent steps: uranium mining, its purification, enrichment and conversion, production of nuclear fuel, production of electricity at nuclear power plants, storage of the spent nuclear fuel, re-processing (in closed nuclear fuel cycle) or disposal (in open nuclear fuel cycle). The re-processing processes aim not only to produce more nuclear fuel but also to decrease radiotoxicity of the radioactive waste. All in all, despite the common fallacy, nuclear energy turns out to be one of the safest and cleanest ways of energy production.

- The modern science in not pure chemistry or physics or whatever, but the co-operation of experts from many different branches of science.

ФИЗИЧЕСКИЙ
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