# Chapter 4. Radioactivity and the Environment

# 4.1 Introduction

Radioactive particles have an impact on the environment in many ways. We have the direct effect on humans as ionization radiation, but also as a tool for studying the human body as well as treatment of humans within medicine.

There is also empirical evidence for between cosmic rays (high energy protons) and the climate on Earth.



Showers of cosmic rays interacting with the atmosphere.

In this section we will first discuss radioactivity and then the interaction of cosmic rays with solar activity and how that influences our climate.

#### Radioactivity

Thomson discovered the electron in 1897 (but its mass was still unknown) and proposed a model of the build-up of the atom, like a plum-pudding.



Thomson's "Plum pudding"-model of the atom

Rutherford, Geiger and Marsden investigated the so called Plum pudding model of



the atom and found that the positive charge (and thus the mass) of the atom is concentrated to a small region, a nucleus, of the atom. The size of the nucleus is fund to be around 1/10,000 of the atom.

Looking at the density, matter has a density around 1 g/cm<sup>3</sup>. The density of the Sun is somewhat higher, whereas stellar objects as white dwarfs have a density of  $10^4$  g/cm<sup>3</sup>. The density of so called neutron stars is higher and is found between  $10^6$  to  $10^{14}$  g/cm<sup>3</sup>, and the density of the nucleus is found

around  $10^{14}$  g/cm<sup>3</sup>. The largest densities are found in black holes with a density of  $10^{17}$  g/cm<sup>3</sup>.

# 4.2 Radioactive decay

The nucleus consists of protons and neutrons, so called nucleons and their total *mass number* is *A* 

The number of protons in the nucleus is given by the *atomic number* Z The number of neutrons in the nucleus is given N

A = Z + N

Chemical identical agents but with different masses, emit radioactive.

These substances with the same chemical properties are called *isotopes*. They have different *atomic masses* (different A) but the same *atomic number* (the same number of protons Z).

Nuclides with the same N but with different Z are called isotones, while nuclides with the same A are called *isobars*. Below is shown the so called nuclide map:



Nuclide map with Z as a function of N and below we have a detail of the nuclide map:



Here, the black section are nuclei that are stable and do not decay. Of the around 2000 known nuclides, there are only 280 stable, while the rest are *radioactive*, i.e. they decay spontaneously. In nature, there are only a few naturally radioactive, while the rest are produced by man and are called *induced* radioactive.

# Natural radioactivity

Becquerel discovered the radioactive radiation in 1896, and that it can consist of three different components,

- α-particles (helium nuclei)
- β- particles (electrons or positrons)
- γ- particles (photons)

In rare cases neutrons and larger nucleus fragments can be found. The number of decays from a radioactive substance declines exponentially. The probability that the nucleus shall decay in the time dt (dt is so small that dt << 1) is dt. If there are N radioactive nuclei at time t, the following number of nuclei is decaying (that is why we have a minus sign)

-dN/dt = N; This differential equation has the solution  $N = N_0 e^{-\lambda t}$ 

**Example.** Calculate the number of atoms in 1.00 gram of <sup>226</sup>Ra! Avogadro's number is 6.022 x  $10^{23}$  /g, why the number of nuclei is:  $N = (1/226) \times 6.026 \times 10^{23} = 2.67 \times 10^{21}$ 

 $N_0$  is the number of nuclei at time t=0 and  $\lambda$  is the decay constant, different for different nuclei. This is due to the interaction between the nucleus and the *inner electrons* of the atom, which is different for different nuclides. That is why it does not depend on the state of the atoms, if they consist of a gas, a liquid or a solid. The decay equation is shown in the graph below:



Radioactive decay with the number of nuclei as a function of time.

The so called **half-life**  $T_{1/2}$  is the time that takes for the number of nuclei to decay to half of that number, i.e. N<sub>0</sub> till N<sub>0</sub>/2, shown in the figure.

Let us study the half-life. When N = N<sub>0</sub>/2 we have t =  $t_{1/2}$  and thus N<sub>0</sub>/2 = N<sub>0</sub> e<sup>- $\lambda$  T1/2</sup>, or 2 = e<sup> $\lambda$  T1/2</sup>. We take the logarithm of both sides and get In2 =  $\lambda$ T<sub>1/2</sub>.

### The half-life is $T_{\frac{1}{2}} = \ln 2/\lambda$ .

#### Example

How many nuclei remain after 3T<sub>1/2</sub>?

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### Solution

After  $t = T_{1/2}$  we have N<sub>0</sub>/2 nuclei left. After  $t = 2T_{1/2}$  we have  $(N_0/2)/2 = N_0/4$  and after  $t = 3T_{1/2}$  there remains  $(N_0/4)/2 = N_0/8$ 

#### Example

How many nuclei remain after  $nT_{1/2}$ , where n is an integer? **Solution** After t =  $T_{1/2}$  we have N<sub>0</sub>/2 nuclei left. After t =  $2T_{1/2}$  we have  $(N_0/2)/2 = N_0/2^2$ . Thus we have after n half-lives N<sub>0</sub>/2<sup>n</sup> left.

In some cases we also talk about the life-time  $\tau = 1/\lambda$ . Then we can write the radioactive decay as

 $N = N_0 e^{-t/\tau}$ 

Thus, we have  $t = \tau$  when the activity is 1/e of the starting activity.

Note: The decay law can also be written using  $T_{1/2}$ :  $N = N_0 2^{-t/T_{1/2}}$ 

### $\alpha$ -particle decay



Looking at the diagram above, we see that in the  $\alpha$ -particle decay we start with Z and N and end up with Z-2 and N-2, i.e. a He-nucleus has left the original nucleus.

- $\alpha$  particles are helium nuclei
- $\alpha$  particles only decay where nuclei have A>200

•  $\alpha$ -particle kinetic energy has a specific value (or a few values if the daughter nucleus can exist in different energy levels). Normally, the energy is within the 4-10 MeV region, as can be seen in the table below

Isotope	Kα(MeV)	T <sub>1/2</sub>
<sup>232</sup> Th	4.01	1.4x10 <sup>10</sup> y

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<sup>238</sup> U	4.19	4.9x10 <sup>9</sup> yr
<sup>230</sup> U	5.89	20.8 days
<sup>220</sup> Rn	6.29	56 s
<sup>216</sup> Rn	8.05	45 μs
<sup>212</sup> Po	8.78	0.30 μs

One also notices that some heavy elements decay fast, such as radon, Rn, while others have decays of the order of  $10^{10}$  years.

Generally, the reaction formula for  $\alpha$ -particle decay can be written  ${}^{A}{}_{Z}X \rightarrow {}^{A-4}{}_{Z-2}Y + {}^{4}{}_{2}He$ As an example we can study the uranium isotope s  ${}^{238}$ U that will decay in the following way

 ${}^{238}_{92}U \rightarrow {}^{234}_{90}Th + {}^{4}_{2}He$ 

(Half-life of <sup>238</sup> U is 4.9x10<sup>9</sup> years)

# β-decay

In the  $\beta$ -decay an electron (or a positron) is released. If you measure its kinetic energy, one obtains a continuous spectrum although one would expect just one single value. However, the decay is shown in the figure below:



In order to fulfil the laws about the conservation of energy and momentum, one has to make one more assumption, namely that there must be still one more particle emitted together with the electron, the so called the neutrino (electron neutrino) ve. Pauli suggested the existence of the neutrino around 1930 to explain the  $\beta$ -decay. It is extremely difficult, but not impossible to detect. In a famous experiment from 1953 Reines and Cowan succeeded in doing this by using the neutrino flow from a reactor. The probability (or cross section) for a neutrino reaction to take place is extremely small, of the order of 10<sup>-20</sup> barn, giving a free mean distance of many thousands of light years in water. The unit 1 barn = 10<sup>-28</sup> m<sup>2</sup>.

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 $^{-}$  decay to the left and  $\beta$   $^{+}$  decay to the

right The reaction formula for the β-decay car

The reaction formula for the  $\beta$ -decay can generally be written  $A_Z X \rightarrow A_{Z+1} Y + e$ - + anti- $n_e$  for the  $\beta$ - -decay  $A_Z X' \rightarrow A_{Z-1} Y' + e$ + +  $n_e$  for the  $\beta$ + -decay

For certain nuclei there is another process present, so called electron capture that is a kind of  $\beta$ -decay. The nucleus captures a K-electron (inner shell). In this case just only one neutrino is ejected, but it is followed by X-ray radiation from the electronic transition when the temporary free place of the K-shell is filled.

# γ-decay

At  $\gamma$ -decay, electromagnetic radiation, or a photons are emitted. The number of nucleons does not change in this process, why the reaction formula can be written

$$A_Z X^* \rightarrow A_Z X + \gamma$$



The  $\gamma$ -decay goes from an excited state to a lower energy state. Z does not change. So, the nucleus goes from an excited state (denoted \*) to a state of lower energy. The spectrum is discrete why the emitted radiation only has just one energy (or a few energies), or one can say that the nucleus energy is ruled by the laws of quantum physics.

# 4.3 Activity and dose. Biological effects regarding dose.

The **activity (decay rate)** *R*, is defined as the number of decays per time unit, and can be written

$$R = - dN/dt = N_0 e^{-\lambda t} = R_0 e^{-\lambda t}$$

That also can be written as

 $R = \lambda N$ 

The activity is measured in the unit 1 Bq (Becquerel) = 1 decay/s. Earlier the 1 Cu (Curie) =  $3.7 \ 10^7$  decay/s was used.

### Example

Two radioactive samples with the same number of nuclei, one contains  $^{230}$  U (T<sub>1/2</sub> 56s) and one with  $^{212}$  Po (T<sub>1/2</sub> 0.30 µs). Compare the activities.

### Solution

 $R_{Po} = \lambda_{Po}N_{Po}$  and  $R_U = \lambda_U N_U$ , where  $N_{Po} = N_U$ . We have T  $_{2} = \ln 2/\lambda$ . This gives  $R_{Po}/R_U = t_U/t_{Po} = 56/(0.3 \times 10^{-6}) = 1.9 \times 10^8$ . Thus, the activity of the Po sample is much higher!

**Absorbed dose**: Absorbed radiation in energy per mass unit. S.I.-unit: Gy (gray) = J/kg. Older unit: rad, 1 rad= 0.01Gy.

**Dose equivalent**: Absorbed dose normalised with a **quality factor** that grades the specific danger of the radiation. S.I.-unit: Sv(Sievert). Older unit: rem, 1 rem=0.01 Sv.

### Quality factors ( $w_{R}$ ) for different types of ionizing radiation

X-ray and $\gamma$ -radiation:	1
$\beta$ -radiation (electrons):	1
Protons:	5
Fast neutrons:	20
$\alpha$ -particles:	20
Heavy ions:	20

We see that the heavier the particle, the higher the quality factor. However, for neutrons the quality factor depends on the neutron kinetic energy, E (in MeV), according to:

 $w_R = 5 + 17 \exp(-(\ln(2E)^2/6))$  where E>0.5 MeV.

### Example

Calculate the quality factor for slow neutrons with kinetic energy 0.01 MeV. **Solution** Applying the formula above we have  $w_R = 5 + 17 \exp((\ln(2x0.01)^2/6)) = 5 + 0.001 = 5$ .

# Example

Calculate the quality factor for fast neutrons with kinetic energy 0.6 MeV. **Solution** Applying the formula above we have  $w_{-} = 5 \pm 17 \exp((\ln(2x0.6)^{2}/6) = 5 \pm 17x1$ 

Applying the formula above we have  $w_R = 5 + 17 \exp((\ln(2x0.6)^2/6) = 5 + 17x1.005 = 22$ .

#### **Biological effects of ionization radiation**

lonization radiation can in the human body, create ions, i.e. remove electrons from the atomic shells. This gives rise to free radicals that can create chemical changes in the cells of the body. Ionization can result in permanent changes or damages of the matter that has been irradiated, for example in the DNA of the cells.

#### Direct biological effects of radiation doses

Dose (Sv)	damages/symptoms on humans
< 0.25	No visible effects
	Chromosome changes in rare cases
0.25 -1.0	Temporary changes in the blood
1.0 -2.0	Radiation illness. Illness. Reduction of white blood cells.
	Normally the patient recovers after a couple of months
2.0 -6.0	After a few weeks fever, bleeding and infections occur, depending on reduced immune defence
	At a dose of 3 Sv half of the patients die within a month
>6.0	Few people survive

#### Examples

The Hiroshima bomb in 1945 resulted in a maximum dose around 10 Sv on the ground and resulted in around 100,000 dead within four months.

In the so called Tokaimura accident in 1999, three people were injured by ionization radiation. Two died after 3 respectively 7 months. Their doses were estimated to be 3, 10 and 17 Sv.

In 2006 a Russian x-spy got a large dose of radiation through <sup>210</sup> Po and died shortly after (<u>http://news.bbc.co.uk/2/hi/uk\_news/6180682.stm</u>).

In 2005 <u>IAEA</u> (International Atomic Energy Agency) reported about the Tjernobyl accident at 1986. They claim that <50 direct deaths occurred due to the radiation damage and gave an estimate of the number of expected deaths in cancer to be around 4,000. Around 600,000 people have been exposed to the ionization radiation.

To compare with deaths concerning other energy sources such as carbon, that stands for 2/3 of the Chinese energy consumption. In the Chinese carbon mines more than 2600 workers diseased through accidents in 2009.

### Distribution of 137 Cs in Sweden caused by the Tjernobyl accident.



From SSI Stockholm 1986. Nowadays "Strålsäkerhetsmyndigheten", SSM.

#### Distribution of different radiation sources effective dose (USA 1997).

Radon	55%
Medical X-ray	11%
Natural internal	10%
Cosmic rays	8%
Terrestrial	8%
Medicine	4%
Commercial products	3%
Other	<1%

There is a large variation of these estimated mean values depending on radon concentration.

People living at high altitudes get larger radiation doses due to cosmic radiation. Flying also increases the annual exposure of ionization radiation.

#### **Natural radiation**

We can divide the natural radiation in three groups, radiation due to cosmic origin, radiation from the ground and buildings and radioactivity within the human body. All this together will give humans in Sweden a dose of 1 mSv/year. This is just about  $\frac{1}{4}$  of the total yearly dose.

### Radioactivity from cosmos

The cosmic radiation from space, galactic radiation and ionizing radiation from the Sun, mainly is due to photons, protons, alpha-particles, electrons and a small fraction of heavier charged particles. Secondary radiation is created in the atmosphere when these particles are absorbed. The radiation intensity is higher at large altitudes. The radiation is doubled each 1500 m. The contribution at the surface of the Earth is around 0.3 mSv/year.

# Radioactivity from the ground and buildings

The dose we get from the ground varies very much depending on where you live in Sweden. The average dose is 0.5 mSv/year but at some spots the dose can be 4 times higher. The origin of this radiation comes from long-lived isotopes with half-lives around 10<sup>9</sup> years, once created in stars or supernovae, which holds for <sup>40</sup>K, <sup>232</sup>Th, <sup>235</sup>U and <sup>238</sup>U. All these substances give rise to series of daughter products. When <sup>238</sup>U decays we get <sup>226</sup>Ra, <sup>222</sup>Rn and <sup>210</sup>Pb, which are import nuclei regarding radiation protection. Especially radium (<sup>226</sup>Ra) and radon (<sup>222</sup>Rn) are abundant in drinking water as well as sea water (around 4 mBq/liter), in provisions and food and in the ground (40 mBq/g). Radium and potassium can be stored in the body.

# Radioactive substances in the human body

The dominant radioactive substance in man is <sup>40</sup>K and <sup>14</sup>C. Since the life-time of <sup>40</sup>K is long,  $T_{1/2} = 1.28 \times 10^9$  years, it was already here when Earth was created. <sup>14</sup>C that is described below, has a half-life of 5730 years is continuously created in our atmosphere. The dose we get from <sup>40</sup>K and <sup>14</sup>C is relatively small, around 0.2 mSv per person and year, since the relative concentrations in the body is small and since carbon and potassium only consist of 18% and 1.5% of the elemental composition of the human body.

### Distribution of different radiation sources effective dose in Sweden 1997

Radon	45%
Medical X-ray & treatments	35%
Natural background	19%
Other	1%

#### Normal Background Radiation in Sweden

The normal background from radioactive radiation in Sweden it is  $\sim$  4 mSv/year (whereas  $\sim$  3 mSv from radon indoor)

# 4.4 Isotopes used in environmental science. C-14 method



Within environmental science and archaeology, the isotope <sup>14</sup>C is used, having a suitable half-life of 5730 years. This isotope is produced in the atmosphere where <sup>14</sup>N-nuclei are hit by thermal neutrons and then the following reaction occurs

$$^{14}N + n \rightarrow {}^{14}C + H$$

The production of this isotope is supposed to have been constant during a long time and that an equilibrium has occurred where the number of created <sup>14</sup>C-atoms = number decays. The figure below describes how the creation of <sup>14</sup> C takes place.

# **Creation of C-14 in the atmosphere**

The decay of C-14 takes place as follows

$$^{14}C \rightarrow ^{14}N + \beta^- + \overline{\nu}$$

All living organisms consist of carbon and are thus slightly radioactive. As long as the organism is alive, the activity remains constant, but when the organism dies, the activity declines according to ( $T_{1/2}$  = 5730 years):

$$R = R_0 \cdot e - (ln2)t/5730$$

Thus, by measuring the activity R and comparing it with the corresponding activity of living material today, we can determine the age of the sample. During a long period, the concentration of 14-C has been constant. However, around 1963, after the large series of nuclear bomb tests had been performed, the concentration rose about 100%, the so called "bomb pulse". The same holds for the radioactive tritium.

So, 14-C functions like a radioactive **clock for organic material**. When an organism dies, the intake of carbon stops and 14-C decays; the clock starts.

### Measurements of the age of organic samples by the <sup>14</sup>C method

The concentration of <sup>14</sup>C in a sample can be done in two ways:

1. Detecting and counting the decay of <sup>14</sup>C. Around **1** g is needed. For living organisms the **activity is 13.6 decays/(g minute)** for carbon.



One often uses a so called proportional counter, shown in the figure from the Swedish museum: "Naturhistoriska Riksmuseet":





One can also directly measure the number of <sup>14</sup>C in a sample comparing it with the number of <sup>12</sup>C atoms. This is done in a massseparating accelerator. In this case only around 1 mg of the sample is needed. A rather large mass separator at Uppsala University is shown in the figure. The sample is injected at the green area to the left in gas phase and is ionized in order to enter the accelerators. The molecular ions enter an analyzing magnet (upper right) that is set for the carbon masses. The ions finally reach a particle detector (below).

The relative mass fraction  ${}^{14}C/{}^{12}C$  is measured at rather high accuracy.

The advantages with this instrument are that a small amount of material is need and that the measurements are very precise. However, the instrument is very large.

#### Example

A sample from a fossil plant is heated to transform into a gas. One measures the  $\beta$ -decay in a proportional counter and obtains 3.2 decays/(g minute). Determine the age of the sample.

### Solution

Since  $N = N_0 e^{-t/\tau}$ 

We obtain the activity  $R = dN/dt = -N_0 e^{-t/\tau}/\tau$ 

Thus  $R(t)/R(t=0) = e^{-t/\tau} / 1 \rightarrow e^{-t/\tau} = 3.2/13.6$ . Taking the logarithm of both sides:

 $t = \tau \ln(13.6/3.2) = (T_{1/2}/\ln 2)x \ln(13.6/3.2) = (5730/\ln 2) x \ln(13.6/3.2)years = 11,961 years or around 12,000 years.$ 

#### Tritium <sup>3</sup>H

The hydrogen isotope <sup>3</sup>H, tritium, decays through  $\beta$ -decay to <sup>3</sup>He with a half-life of 12.38 years. Tritium is both a cosmogenic and anthropogenic nuclide. Tritium is created naturally in the atmosphere by spallation, i.e. breaking molecular nitrogen and oxygen in reactions with high-energy cosmic protons.

During the bomb tests around 1960 the activity was increased, the so called "bomb pulse".

Tritium is the most important isotope for making environmental studies of lakes, seas and ground water.

The concentration relation <sup>3</sup>H/ <sup>3</sup>He can for example be used to measure the "age" of ground water, i.e. how long it has been isolated from the atmosphere. It is also a great way to detect surface water penetration in a ground water reservoir.

#### Beryllium <sup>10</sup>Be

The beryllium isotope <sup>10</sup>Be decays through  $\beta$ -decay to <sup>10</sup>B with a half-life of 1.5x10<sup>6</sup> years.

One can say that it is a cosmogenic nuclide, which like tritium is created naturally in the atmosphere through spallation i.e. breaking 14-nitrogen and 16-oxygen in reactions with cosmic radiation. Secundary cosmic radiation at the surface of the Earth,

in the form of fast neutrons and myons (the myon is an electron-like elementary particle) also gives rise to the production of <sup>10</sup>Be in surface materials.

<sup>10</sup>Be is an important isotope for geological investigations, but also in environmental studies. It can be used to determine sediment formation velocities in marine environments.

By studying <sup>10</sup>Be-variations in drilling cores from the Greenland ice or from deep sea bottoms it has been possible to observe geomagnetic reversals (The magnetic North pole has changed from a magnetic South pole and vice versa during time.

### Aluminium <sup>26</sup>Al

The aluminum isotope <sup>26</sup>Al decays through  $\beta$ + -decay to (positron decay) to <sup>26</sup>Mg with a half-life of 0.7 x10<sup>6</sup> years.

It is a cosmogenic nuclide, which is created naturally in the atmosphere through spallation the rare gas <sup>40</sup>Ar. However, this is done a a small extent. Instead, secondary cosmic radiation at the surface of the Earth, in the form of fast neutrons and myons has a larger contribution to the <sup>26</sup>Al production in surface materials. <sup>26</sup>Al is therefore used as <sup>10</sup>Be in geological investigations.

<sup>26</sup>Al is also created in stellar processes, for instance in super novae. In order to detect these processes one studies the characteristic  $\gamma$ -rays at 1809 keV that is created in the decay process. One uses  $\gamma$ -ray detectors I satellites to clearify the stellar processes (nuclear astrophysics).

#### Xenon <sup>133</sup>Xe, <sup>135</sup>Xe for nuclear bomb test detection

At nuclear bomb tests, large amounts of isotopes of xenon are created. Since xenon is a rare gas, it does not easily react with the surroundings. It can unchanged leak through the ground after a bomb test. If it does not directly be pushed out at the explosion it will after a few days leak into the open air and finally be dragged into an atmospheric low pressure. The most interesting isotopes have half-life's between nine hours and eleven days.

There are xenon sensors all over the World due to the CTBT-treaty (Comprehensive Nuclear-Test Ban Treaty). The sensors consist of both seismic detectors and radioactivity-detectors. In Stockholm (FOI) there is a sensor, SAUNA II placed in Kista. It is an automatic system for detection of the radioactive isotopes of xenon, <sup>133</sup>Xe, <sup>135</sup>Xe, and <sup>131</sup>Xe. <u>SAUNA</u> collects a sample through 12 hours by filtering 15 m<sup>3</sup> of air. The air is cleaned from water and carbon oxide and then passes through piles of active carbon. Here, the xenon gets stuck. While this sample is analysed a new sample is inserted. Thus, the process is continuous.

As a last step the ratios between the concentration of the xenon isotopes is determined and the resulted spectrum is sent to CTBTO via satellites.

# 4.5 Radon

Radon is one of the rare gases, like helium, neon, argon. It has as the other rare gases filled shells (electrons 2, 8, 18, 32, 18, 8). It is radioactive. There are several isotopes, where <sup>222</sup> Ra is the most common in Nature. This isotope is created by the

decay of  $^{226}$  Ra (radium). The full decay chain is shown below. It all starts with  $^{238}$  U and ends with  $^{206}$  Pb that is stabile:

<sup>238</sup> U (4.5x10<sup>9</sup> yr)  $\rightarrow$  <sup>234</sup> Th (24 days) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>234</sup> Pa (1.2 s) +  $\beta$  +  $\gamma$   $\rightarrow$  <sup>234</sup> U (247,000 yr) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>230</sup> Th (80,000 yr) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>226</sup> Ra (1,622 yr) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>222</sup> Rn (3.8 days) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>218</sup> Po (3.0 s) +  $\alpha$  +  $\beta$   $\rightarrow$  <sup>214</sup> Pb (26.8 s) +  $\alpha$  +  $\beta$   $\rightarrow$  <sup>214</sup> Bi (19.7 s) +  $\alpha$  +  $\beta$   $\rightarrow$  <sup>214</sup> Po (0.16 ms) +  $\alpha$   $\rightarrow$  <sup>210</sup> Pb (22 yr) +  $\beta$  +  $\gamma$   $\rightarrow$  <sup>210</sup> Bi (5 months) +  $\alpha$  +  $\beta$   $\rightarrow$  <sup>210</sup> Po (138 months) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>206</sup> Pb (stabile)

The radioactive gas radon can cause radioactive pollution. It easily spread and gives rise to solid decay products, so called radon daughters. We have as shown by the chain above that we have all types of decay, such as  $\alpha$ -,  $\beta$ - and  $\gamma$ -decay.

The different radon isotopes <sup>220</sup>Rn, <sup>222</sup>Rn and <sup>226</sup>Rn were earlier called *toron*, *radon* and *aktinon*. It all depended on the constituencies of the sample. Rather soon radon was set as the name of the elements.

# Threshold values of radon gas in houses

One of the major sources for radon in the air in houses is air coming from the ground. Already ground air can contain 10 kBq/m<sup>3</sup>. Some building material such as blue concrete (Swedish "blåbetong") can emit radon to the indoor air since it contains minerals with uranium.

Normally in Sweden, one has tried to remove the radon by a large air flow. A better way of reducing the radon concentration is to look at the point sources of emission from the ground and how to hinder the radon to get indoors by introducing an air flow under the house. Radon exposure in combination with smoking has been found to be very dangerous. Earlier WHO set the threshold value for radon gas to be 1000 Bq/m<sup>3</sup>. Nowadays this threshold has been lowered to 100 Bq/m<sup>3</sup>. In Sweden, however, **the limit is 200 Bq/m<sup>3</sup>** due to local factors such as the natural radon content of the ground. In Sweden, Socialstyrelsen and Boverket, have given regulations how to make precautions in order to reduce the radon concentrations.

# Threshold values of water containing radon

In Sweden, water containing radon is often found in drilled wells. In a normal well that has been done by digging, outdoor removal of radon is done through air. The so called "Livsmedelsverket" in Sweden is responsible for threshold values in water and ahs set the limit to 1000 Bq/liter, whereas 100 Bq/liter is a limit without any complains.

# Measuring the radon content in humans

Is it possible to measure the radon content in humans? By looking at the long lived radon daughter <sup>210</sup>Pb that can be found in the skeleton of humans, it is possible to measure the radon content. By studying the  $\gamma$ -ray decay from excited <sup>210</sup> Bi to <sup>210</sup> Pb at 47.1 keV, we can measure the radon content. This is shown in the energy diagram below, just like in atoms:





# <sup>220</sup> Rn important daughters

Both <sup>212</sup>Bi and <sup>212</sup> Po are daughters of <sup>220</sup> Rn via the so called thorium decay chain. The thorium chain has radon 220 Rn as a link. However, this radon-isotope, called toron, is too short-lived to be able to spread in the environment. Toron's half-life is only  $T_{1/2}$  = 56 s. Below is shown the thorium decay chain:

<sup>232</sup> Th (14x10<sup>9</sup> yr)  $\rightarrow$  <sup>228</sup> Ra (5.8 yr) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>228</sup> Ac (6 h) +  $\beta$  +  $\gamma$   $\rightarrow$  <sup>228</sup> Th (1.9 yr) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>224</sup> Ra (37 days) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>220</sup> Rn (58 s) +  $\alpha$  +  $\gamma$   $\rightarrow$  <sup>216</sup> Po (0.15 s) +  $\alpha$   $\rightarrow$  <sup>212</sup> Pb (1 h) +  $\beta$   $\rightarrow$  <sup>212</sup> Bi (1 h) +  $\beta$   $\rightarrow$  <sup>212</sup> Po (0.3 µs) +  $\alpha$   $\rightarrow$  <sup>208</sup> Pb (stabile)

We also have a branch from <sup>212</sup> Bi to <sup>208</sup> Pb: <sup>212</sup> Bi (1 h) +  $\alpha \rightarrow$  <sup>208</sup> Th (3 min) +  $\beta \rightarrow$  <sup>208</sup> Pb (stabile)

# 4.6 Nuclear bomb tests. Effects on the environment

Since 1945 a large number of nuclear bomb tests have been performed under the surface of the Earth as well as in the atmosphere. Only USA has done at least 1054 different tests. China, the Soviet Union and France have performed an unknown



number of tests. Lately, India and Pakistan have done some underground nuclear bomb tests. This has caused a significant rise in background radiation for many people around the World. It is on the North semi-sphere where most nuclear bomb tests have been performed.

In the North and in Russia, the "Samer" have been influenced through their diet. However, the people on the Marshall Islands have been under the highest influence. Both USA and France have done

tests in the South Sea. How the local people in Russia and Chine we do know very little about.

I the USA, the "National Cancer Institute" as found that the largest health risk on the American population has been in the form of thyroid gland cancer. It is due to the nuclear tests in Nevada and New Mexico.

We can still measure how the activity from the atmospheric tests during 1960-1970 is circulating in the upper atmosphere and comes down to Earth through rain.

China was the last country that performed atmospheric nuclear bomb tests and ended in 1979. Leakage from the underground experiments can nowadays easily be detected.

There is now an international organization (CTBTO) having measuring sites all over the World to detect bomb tests, so one can see that the agreements still hold. See section "Xenon <sup>133</sup>Xe, <sup>135</sup>Xe for nuclear bomb test detection" above.

# 4.7 Nuclear energy. Nuclear plants. Fusion energy. Transmutation

### 4.7.1 Nuclear energy. Introduction

#### Politics and science--Why fission or fusion?

The world needs energy sources, since global energy consumption is about 0.3 Q per year (Q =1x10<sup>21</sup>J).

Let us make a simple calculation: The World population  $x \ 2 \ kW$ /person  $x \ 1 \ year = 0.3 \ Q$ ).

If one looks at the World reserves:

\* Fossil fuels ~ 100 Q (mostly coal)

```
* <sup>235</sup> U ~ 3 Q
```

\* Breeder reactor ( $^{238}$  U) ~ 300 Q

(A breeder reactor uses burnt fuel)

No source is ideal when we consider requirements for:

- environment and safety
- economy
- society

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Nuclear reactor based on fission energy

Nuclear test reactor based on fusion energy

The Einstein mass-energy relation:  $E = mc^2$ 

The nuclear mass, that can be measured with high accuracy is given in **atomic** mass units defined by the mass of 1/12 of the <sup>12</sup>C-atom and has the value  $1 \text{ u} = 1.66056 \text{ x} 10^{-27} \text{ kg}$ 

Perhaps it can be worth noticing that  $uc^2 = 931.494$  MeV and in the same way we get The energy of one proto:  $m_pc^2 = 938,3$  MeV, of one neutron:  $m_nc^2 = 939,6$  MeV and of one electron:  $m_ec^2 = 0.511$  MeV

The nuclear mass, M, is less than the sum of the masses of the protons and the neutrons. This mass-defect  $\Delta m$  corresponds to an energy  $\Delta E_{be}$  (>0), the nuclear binding energy  $\Delta E_{be} = \Sigma (mc^2) - Mc2$  (=  $\Delta mc^2$ )

If one adds energy to the nucleus, one can split it into its parts. If the electrons binding energies of the atom can be neglected, the binding energy  $E_{be}$  for a nucleus with Z protons and (A-Z) neutrons be written

 $E_{be} = Z(M_H - m_e)c^2 + (A-Z)m_nc^2 - (M-Zm_e)c^2$ 

M is the ATOMIC MASS and  $m_{\mbox{\tiny e}}$  the mass of the electron. The electron masses vanish in the expression, why

 $E_{be} = ZM_{H}c^{2} + (A-Z)m_{n}c^{2} - Mc^{2}$ 



The binding energy/nucleon then becomes  $\epsilon$  =  $E_{be}/A$ ,  $\epsilon$  depends on A according to the figure.

We observe from the diagram that one can obtain kinetic energy from rest energy if 1) two light nuclei add to form a heavier nucleus, in a process called **fusion** or

2) a heavy nucleus is split into two lighter nuclei, a process called **fission** 

### 4.7.2 Fission

If a nucleus with mass number A and atomic number Z decays into two equal parts (a possible decay, but not the most common) the difference in rest energies (fore-after) becomes

 $\mathsf{A}(\epsilon_{\mathsf{A}/2} - \epsilon_{\mathsf{A}}) > 0$ 

With A = 238 and  $\varepsilon_{A/2}$  = 8,5 MeV/nucleon and  $\varepsilon_A$  = 7,6 MeV/nucleon respectively, we get 238(8,5-7,6) MeV/nucleus = more than 200 MeV/nucleus!

In order to achieve fission, we have to make the nucleus to become unstable. This can be done by sending neutrons to hit the nucleus. If one studies normal fission elements like <sup>235</sup>U and <sup>238</sup>U, we can look at the diagram below, showing the probability or



the cross section, for fission to occur. From the diagram we see that it for <sup>235</sup>U is enough with slow (thermal) neutrons for the reaction to start. For <sup>238</sup>U on the other hand, neutrons with kinetic energies in the MeV-region are needed.

As we have mentioned, normally two equally large fission fragments

are not created at nuclear reactions. An example is shown below where <sup>235</sup>U captures a neutron:







In the figure above, we see different possibilities for fission when a high-energy proton (1 GeV) falls upon a uranium nucleus. Highest probabilities are observed around mass numbers 100 and 140f we look at the figure to the left. Since there are more neutrons created in the nuclear reaction, all these secondary neutrons can react with another nucleus, thus creating still more neutrons, why a nuclear chain reaction can follow. The energy release is due to the kinetic energy of the participating daughter nuclei.



Since there are more neutrons created in the nuclear reaction, all these secondary neutrons can react with another nucleus, thus creating still more neutrons, why a **nuclear chain reaction** can follow. The energy release is due to the kinetic energy of the participating daughter nuclei.

The mean value of created neutrons after each reaction, the so called v -factor, which for <sup>235</sup>U is v = 2,44 and for <sup>238</sup>U is v = 2,89. It is very important, in order to control a nuclear reaction, to use a moderator that controls the reaction. The moderator controls the number of slow neutrons making the cross section large enough for a reaction to take place as in the case of <sup>235</sup>U.

- So, critical mass fission can be controlled by letting the neutrons be absorbed. En kritisk-massa fission kan alltså kontrolleras genom att man låter neutroner absorberas. Some of the neutrons are delayed and are being emitted by the daughter nuclei. These delayed neutrons allow the nuclear reactor to be controlled.
- Control rods steers the absorption of neutrons in order to maintain a controlled nuclear reaction.



A nuclear reactor where the energy is transformed to boiling water that is used for electricity production. In the detailed picture we see the control rods and fuel rods.

The most common method to take care of the heat from the reactor is to use a heat exchanger.

- In the *boiling water reactors* one let the water boil and transform to steam that can be connected to a turbine connected to generators for electricity production.
- In the pressure water reactors one lets the steam under high pressure circulate from the reactor to an external heat exchanger, where one lets the steam run a turbine.

Boiling reactors are simpler constructed than pressure water reactors, but the possibility that the steam can become contaminated by radiation is higher than for the pressure water reactors. Their two-step design hinders the water to be contaminated.



The nuclear power is used very differently in various countries. In the diagram below we see how much the nuclear reactors are used for electric power production (2000).



We can also compare the energy content of different fuels. It shows that the energy content differ many orders of magnitude for the same amount of fuel.

Fuel energy content			Daily fuel demand for a 1000 MW power plant		
Material	Amount	Energiy(J)	Material	Amount	
Carbon Oil Natural gas Petrol Uranium fission Uranium fusion	1 kg 1 ℓ 1 m <sup>3</sup> 1 ℓ 1 kg 1 kg	3x10 <sup>7</sup> 4x10 <sup>7</sup> 4x10 <sup>7</sup> 3x10 <sup>9</sup> 10 <sup>14</sup> 2x10 <sup>14</sup>	Carbon Oil Natural gas Uranium	8x10 <sup>6</sup> kg 1 train/day 6000 m <sup>3</sup> 1 tanker/week 7x10 <sup>4</sup> m <sup>3</sup> 3 kg	

#### Fusion

We have for long realized that one can obtain energy by fusion and make two light nuclei collide in order to create a larger nucleus.

Let us look at the Energy requirements in a modern society.

The yearly consumption of energy per capita is 60 000 kWt in Sweden, which corresponds to:

- 0.7 gram D&T fusion fuel
- 6 m<sup>3</sup> oil
- 27 000 kg biomass = 20 000 m<sup>2</sup> land

Let us look at economy, safety, environmental effects and when it is possible to have fusion plants.

#### Economy

- The fuel is available everywhere in great amounts
- Low cost for land
- High investment for the plant available in 30-50 years

#### Safety

• No uncontrolled reactions can occur. Inga

There is radioactive waste - Tritium gas ( $T_{1/2}$  = 12.38 years) decays to <sup>3</sup>He

#### Environmental

- No greenhouse gases are produced
- Waste will not be a burden for generations to come
- No requirements for large land areas

### Solar fusion

The solar fusion reaction is based on the figure below:

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Here two light hydrogen nuclei form a helium nucleus, and by doing so we obtain lots of energy. However, we have to overcome the repulsive Columb barrier in order to get the positive hydrogen nuclei close enough to let the attractive nuclear force be dominant, since it works on small distances. It is easy to estimate how large the Coulomb barrier is for two colliding hydrogen nuclei.

The Sun radiates isotropicly the energy 3,9 10<sup>26</sup> J/s The fusion in the centre of the Sun is a several step process making hydrogen convert to helium. In the first step two protons (hydrogen nuclei) collide, creating deuterium (heavy hydrogen nucleus) and at the same time we get one positron (positive electron) and an electron-neutrino as below (the Q-vlue is excess energy):

$${}^{l}H^{+} + {}^{l}H^{+} \rightarrow {}^{2}H^{+} + e^{+} + v_{e}$$
 (Q = 0,42 MeV)

Let us summarize the solar total *p*-*p*-cycle:

$$2({}^{1}H^{+} + {}^{1}H^{+}) \rightarrow {}^{4}He^{2+} + 2(e^{+} + n_{e}) + 2\gamma$$

If we add four electron masses on each side one can instead use atomic masses in the calculations and determine the O-value of the process.

$$Q = \Delta mc^2 = 4(1,007825) \text{ uc}^2 - 4,002603 \text{ uc}^2 = 0,028697 \text{ uc}^2 = 26,7 \text{ MeV}$$

Both neutrino particles carry about 0.5 MeV of this energy from the Sun. When all hydrogen has burnt out in the Sun, in about  $5x10^9$  years, the Sun's mass is not large enough to start a helium cycle and will end its days as a star as a Red Giant.

#### **Fusion reactor**

Elements with mass numbers larger than 56, are supposed to be created in Super Novae explosions. One has tried to copy the Sun's elegant fusion reaction at Earth, but it it hindered by the process slow behavior Tempting alternative fusion reactions that could work would be:

$^{2}\text{H}^{+} + ^{2}\text{H}^{+} \rightarrow ^{3}\text{He}^{2+} + n$	(Q = 3,27 MeV)
$^{2}H^{+} + ^{2}H^{+} \rightarrow ^{3}H^{+} + ^{1}H^{+}$	(Q = 4,03 MeV)
$^{2}\text{H}^{+} + ^{3}\text{H}^{+} \rightarrow ^{4}\text{He}^{2+} + \text{n}$	(Q = 17,59 MeV)

There are three criteria to have to be fulfilled for a successful Earthbound fusion reactor:

- High particle density of a neutral plasma, n
- High plasma temperature, T
- Long confinement time  $(\tau)$  of the plasma

One can show that a fusion reactor using the deuterium-tritium reaction has to fulfill the so called Lawson criterion:  $n\tau > 10^{20} \text{ s/m}^3$ .



Magnetc confinement in a so called Tokomak where ITER is the name of the latest research plant that is supposed to be in action at 2016.

Run your own Tokomak Reactor. The simulation needs Java.



ITER is the next step towards fusion power.

• It will demonstrate the scientific performance reaching 500 MW fusion power.

- Collaboration between EU + Japan + Ryssa + Canada + USA + China + South Korea
- Decision was taken to build ITER 2004
- Start 2015
- 4 billion Euro invested

### 4.8 Radiation shield

When you visit a hospital to have an X-ray picture taken, one has to avoid unnecessary exposure to X-rays. Personnel are often sitting in another, shielded room to avoid the ionizing radiation, which most often is X-rays (Röntgen).

### Absorption of X-rays or $\gamma$ -rays by matter

The absorption of X-rays in matter decreases exponentially with the thickness **x** of the absorbing layer and the mass absorption coefficient  $\mu_m$ . The remaining intensity *I***(x)** after a passage through a layer with thickness **x** can be described by the following formula for absorption:

$$I(x) = I(0) e^{-(\mu} m^{\rho)x} = I(0) e^{-\mu x}$$

We can define the linear absorption coefficient  $\mu = \mu_m \rho$  with the dimension cm<sup>-1</sup>.

Just as in the case of radioactive decay one can here define the half-value thickness (layer), which just is the thickness of the shield to reduce the radiation intensity to half of the incoming intensity,  $x_{\frac{1}{2}}$ .

$$I(0)/2 = I(0) e^{-\mu x l/2}$$
 gives  $ln 2 = \mu x_{l/2}$  why

Half value thickness  $x_{1/2} = \ln 2/\mu$ 

#### Absorption parameter $\mu$ for 50 keV X-rays

Element	$\mu = \mu_m \rho(cm^{-1})$	
Air Aluminum Lead	0.0 0.21 1.22	

#### Example

Calculate how thick layer (in mm) of lead is needed to stop 99% of incident 50 keV X-rays.

### Solution

Only (1-0.99)I(0) of the intensity remains, why the formula for absorption gives  $(1-0.99)I(0) = I(0) \exp(-\mu x)$  which means that

0.01 = exp - 1.22x where we obtain x in cm. We take the logarithm and get: ln(1/0.01) = 1.22x and x = 4.605/1.22 cm = 3.77 cm = **38 mm**.

# Half-value layer in medicine

The same laws of absorption of X-rays hold when we look at medicine and X-rays of the human body. Some linear attenuation coefficients for photon energies of diagnostic X-ray beam are:

 $\begin{array}{l} \mu_{air} = 0.000112 \ cm^{-1} \\ \mu_{bone} = 0.48 \ cm^{-1} \\ \mu_{muscle} = 0.180 \ cm^{-1} \\ \mu_{blood} = 0.178 \ cm^{-1} \end{array}$ 

Thus, the differences are very small for the soft tissue for the human body. This and the fact that the X-ray films usually allow contrasts of the order of 2 % limits the possibilities of traditional X-ray imaging. This can be illustrated by the following calculations.

### Example

A 1 cm rib and a 1 cm air filled volume in the lung is easily detectable:  $I_{bone}(1 \text{ cm}) / I_{air}(1 \text{ cm}) = e^{-0.48} / e^{-0} = 1.63$ , which means 63 % contrast.

# Example

A blood filled vessel embedded in a soft tissue, the muscle, of the same dimension is

not visible:

 $I_{blood}(1 \text{ cm}) / I_{muscle}(1 \text{ cm}) = e^{-0.178} / e^{-0.180} = 1.002$ , and the contrast will be only 0.2 %.

# 4.9 Nuclear waste

In Sweden 10 nuclear reactors are in operation, covering around 45% of the Swedish electricity production. Besides energy production, nuclear waste is produced. It has



to be handled in a way as to protect humans and the environment.

This has to be taken care of in a short time aspect as well as for the long-term. This is the task for SKB (<u>http://skb.se/</u>)



(Svensk Kärnbränslehantering AB), Swedish Nuclear Fuel and Waste Management Company, owned by the nuclear-power companies Nuclear power plants have been producing electricity in Sweden for more than 30 years. All Swedes have been using electricity and we all have a common responsibility for the future for disposing of the waste. In the picture above the specially designed vessel Sigyn transports the waste to its final destination.

Research has been made on the storage of the long-lived nuclear waste. Preparations of the encapsulation and final disposal of spent nuclear fuel are on-going. Waste has to be isolated for at least 100,000 years, if we do not invent a method to convert long-lived isotopes to short-lived (Transmutation).

### Repository for final disposal



Around 20 years ago, SKB started locating a final repository for disposal of the spent nuclear fuel.

Now, in March 2011, SKB will submit an application to the Swedish Radiation Safety Authority (SSM) and to the Environmental Court in order to create a final repository in Forsmark.

### The activity of the nuclear waste determines the handling of the material

It is not only spent nuclear fuel SKB deals with. Replaced reactor materials as well as nuclear waste from hospitals and industry where SKB focuses on.

#### **Operational waste**

So called Operational waste is about 85 per cent of all nuclear waste, mostly low and intermediate-level waste (isolation around 500 years). Low-level waste needs no radiation shielding. It can thus be transported and stored in ordinary containers.





#### Spent nuclear fuel

Spent nuclear fuel has to be taken care of by radiation-shielded and cooling methods and storage and isolated for at least 100,000 years. Several decimeters of steel or

several meters of water is used to protect against radiation. There is a national and international consensus that geological final disposal is the best solution.

#### SKB:s method of final disposal

SKB uses a special method of final disposal of the spent nuclear fuel. The method is based on three protective barriers.

- 1) The spent nuclear fuel must first be encapsulated in copper.
- 2) The impermeable copper canisters are then placed in crystalline basement rock at a depth of about 500 metres, embedded in bentonite clay.
- 3) After disposal the tunnels and rock caverns are sealed.



#### **Final disposal**

The tunnels will be around 250 metres long, and positioned about 40 metres apart. In the base of the tunnels there will be disposal holes about six metres apart. The copper canisters are to be be placed in disposal holes and embedded in a bentonite-clay buffer. When all the spent nuclear fuel has been deposited in the crystalline basement, the tunnels and shafts will be filled in with swelling clay.

#### Bentonite clay protects canisters

The impermeable copper canister fully contains the spent fuel. The bentonite buffer protects the canister against corrosive attack and rock movements. If a fracture occurs in a canister, the bentonite clay buffer and undamaged parts of the canister will prevent water from penetrating into the canister. The buffer will also prevent the leakage of radioactive substances from the canister. The rock provides a natural environment in which the function of the technical barriers is maintained over a very long period. The rock, coupled with the great depth of the deposit, effectively isolates the spent fuel from human beings and the environment.

#### Problems with copper canisters?



Researchers at KTH have found evidence that the copper canisters may corrode far too rapidly. The estimated service life of 100,000 years for the canisters may as short as less than 1,000 years. The research is based on studies of copper coins which ended up at the bottom of the sea when the ship 'Wasa' sunk, and stayed there for 333 years. The environment for terminal nuclear storage and the bottom of the sea where Wasa lay are quite similar to each other.

It is perhaps necessary to use 1 m of copper metal in order to last 100,000 years. In addition, there is another disturbing factor. The copper capsules are heated up by the radioactive waste inside them, and warm copper corrodes even faster.

### Swedish uranium mining

Sweden has uranium content over the mean if we look worldwide. Only theoretically, we could have our own production of nuclear material. Uranium has so far not been accepted in Sweden for mining, although mining companies are working in Sweden.

In Sweden, the uranium mostly is found in slate (alunskiffer). The richest findings are located between the lakes Vänern and Vättern, but also in the North.

Landscape	uranium g/1000kg	Total uranium (10 <sup>6</sup> kg)
Västergötland		
(between the lakes Vänern and Vättern):	200-300	1400
Östergötland	100-200	1600
Jämtland (Northern Sweden)	150-250 1	0 000
Skåne (Southern Sweden)	50-150 1	8 000

If we summarize, we find that the total amount of uranium in slate is around  $30x10^9$  kg that is relatively easy to mine and extract.

SGU, "Sveriges Geologiska Undersökning", (http://www.sgu.se) has a missing to in-



vestigate the geology of Sweden and one of its task is to neasure the uranium content. This is also done by measuring applying sensitive gspectrometers flying at a height around 50 m navigating by GPS in the North-South or East-West directions along lines around 500 m over land. The flight speed was 250 km/h. The map shows the measured uranium surface content. In the ground water one measures the radioactivity and the following limits are given:

Radon 222 > 1000 Bq/l Not usable Uranium > 15 μg/l Just usable

#### Problems with nuclear power

- Criticality
- Rest heat
- Used fuel
- Plutonium problem

Let us first discuss the problems with criticality. Reactors are based on a self-going chain reaction and we rely on delayed neutrons. As a consequence, the fuel has to be well characterized.



• Problems have occurred with reactor excursion (rushing) in certain types of reactors such as Tjernobyl. The Swedish or Western reactors based on the Light Water Reactor type have so called negative well designed negative feed-back mechanisms that will stop the reactions if problems occur.



 There are also Rest heat problems that can cause melting of the core in a closed reactor (TMI).

Solid fuel needs an intensive cooling, but there are no good passive cooling systems. Economy pushes the reactor constructions to give a high power.

- Nuclear power produces long-lived waste, most of all in the form of Actinides, which consists of the 15 elements between <sup>89</sup>Ac (actinum) and <sup>103</sup>Lr (lawrencium) in the Periodic Table. The most long-lived isotope, <sup>247</sup>Cm (curium) has a half-life of 1.6x10<sup>7</sup> years.
- Plutonium, <sub>94</sub>Pu in the waste is a potential security risk for producing nuclear bombs, which was discovered in 1941 when <sup>238</sup>U was bombarded by deuterons.

#### Radio-toxicity of used nuclear waste

The following nuclei are involved in the burning of fuel in reactors based on uranium:

Nucleus	gram/1000 kg fuel	T½ (years)
<sup>235</sup> U	7 400	7, 037 x 10 <sup>8</sup>
<sup>234</sup> U	200	2, 454 x 10⁵
<sup>238</sup> U	945 000	4, 468 x 10 <sup>9</sup>
<sup>237</sup> Neptunium	440	2,14 x 10 <sup>6</sup>
<sup>238</sup> Plutonium	150	88
<sup>239</sup> Plutonium	4 700	24 100
<sup>240</sup> Plutonium	2 000	6 570
<sup>241</sup> Plutonium	1 000	14,4
<sup>242</sup> Plutonium	660	376 000
<sup>243</sup> Americium	100	7 370
<sup>244</sup> Curium	30	18

The table of nuclei, their half-life and concentration can be described by the following figure. The curve representing the effect of the TRU (trans-uranium) is shown:





#### So, the question is:

Do we have to wait 10<sup>5</sup> years to reach an acceptable level of radioactivity? Are there methods to speed up the decay or converting the responsible nuclei to other types of nuclei?

The answer to this could be *transmutation*.

### 4.10 Transmutation of nuclear waste

What is transmutation?

Transmutation was first demonstrated 1919 by Rutherford where <sup>14</sup>N is transferred to <sup>17</sup>O by a reaction with  $\alpha$ -particles from a radioactive sample (He-nuclei): <sup>14</sup>N +  $\alpha \rightarrow$  <sup>17</sup>O + p

The first *accelerator driven* transmutation was made in 1930 by Cockroft and Walton:  $^{7}Li + p \rightarrow {}^{4}He + \alpha$ 

So, this is a nuclear process where an element is transferred (by transmutation) to another element by nuclear reactions.

Can we make the nuclear power reactors more safe and be able to "clean" nuclear waste ?

A new type of reactor construction, an under-critical reactor can improve the safety as well as reducing the radioactivity of the nuclear waste. It can clean nuclear waste also if we include plutonium and improve the nuclear cycle.

#### Transmutation by neutron capture

As an example we can take transmutation of technetium, <sup>99</sup>Tc. This isotope (<sup>99</sup>Tc) can be transferred to <sup>100</sup>Tc that in turn will decay to stable Ru isotopes. The long-lived 99Tc ( $T_{1/2}$  = 2.1x10<sup>5</sup> years) will convert to short-lived Tc isotopes that can be converted to stabile Ru isotopes.



#### **Transmutation of trans-uranium**

Transmutation can be made through neutron capture by fast neutrons and fission. In the picture we see the <sup>237</sup>Np nucleus capture a neutron and by fission we get more neutrons and new fission fragments. We also see the cross section (probability) for <sup>237</sup> Np capture and fission around 10-100 eV. (Thermal neutrons have energies around 0.01 eV).







See also the Department of physics at KTH (<u>http://www.physics.kth.se</u>) where the different groups of <u>Nuclear Safety</u>, <u>Reactor Physics</u>, <u>Reactor Technology</u> are present.



# Incitaments for transmutation

- Radio-toxicity of used Light Water Reactor fuel is dominated by TRU, trans uranium.
- Fission products (FP) only contributes only until <sup>90</sup>Sr and <sup>137</sup>Cs decay (T<sub>1/2</sub> 28 respectively 30 years.
- The equilibrium radio-toxicity of uranium in Nature is ~ 20 mSv/g
- According to the diagram, It takes more than 300 000 years to reach the normal natural background

Pu can be transmuted (by fission) in fast reactors, but unfortunately it is problems with Am. Only 5% of Am can be loaded in a reactor. Excess of Am and Cm has to be "burned" in specially designed reactors that need under-critical systems.

### A "Swedish" scenario for transmutation of Pu and Am (Gudowski, KTH)



We see that from a reactor of 30 GWh of 300 000 kg spent fuel from a nuclear power plant we can transform it to 12+4 kg TRU ( $T_{1/2}$  300 000 years) and 14 000 kg FP (with  $T_{1/2}$  < 30 years).

The Department of Physics (Gudowski, Wallenius etc) together with units in Europe are developing a "better" nuclear power, the Generation IV power plants with lower toxicity and waste as can be described below:

Early Prototyp Reactors	e 🗖	MARKEY, L		Controling 1	and a second sec		
		ommercial Pow Reactors	er 🛛	Advanced LWRs		Near-Term Deployment	Generation IV
Shippingport     Dresden, Ferr     Magnox	NI			- ABIR	7	Brolutionary Designs Offering Improved Economics	<ul> <li>Highly Economical</li> <li>Enhanced Salety</li> <li>Minimal Waste</li> </ul>
	- L - ( - Y	NR PWR, BW XNDU VERIRBMK	R	- Systemat - AP600 - EPR	-		- Proineration Resistant

Comparing of cost of electricity (COE) in the unit mills/kWh (1 mill =1/1000 of the U.S. dollar = m\$).



Here ADS stands for Accelerator Driven System (for transmutation), and MOX stands for mixed uranium-plutonium dioxide.

ADS can create a better acceptance for nuclear energy according to nuclear scientists. The nuclear reactors will have

- Better on-line security in operation
- Drastically changes the time dimension of the waste
- · Can solve the problem with the production of plutonium
- Takes over the evolutionary energy production from today's reactors
- Can be constructed to safe circulation of plutonium

(Gudowski, KTH)

# 4.11 Radioactive particles and climate change

First let us discuss what factors are responsible for climate change variations. We can list at least six factors (J. Kristjansson, University of Norway):

- 1. Variation in Earth's orbit around the Sun
- 2. Solar activity
- 3. Volcanic activity
- 4. Internal oscillations
- 5. Anthropogenic greenhouse gases
- 6. Anthropogenic gases

In this section we will discuss the question about solar activity and cosmic rays.



The climate of the Earth is strongly influenced by solar activity. Changes in solar activity has a direct correlation between cloud formations, temperature changes etc on the Earth.

Cosmic rays from super novae outburst have an impact on the solar emission if the cosmic rays are intense enough.

The effect of cosmic rays in the creation of clouds that will in turn will affect the climate.

In the figure we see that the solar radiation heats the Earth and causes heat transport towards the poles, the colder parts of the Earth.



Recently VR, the Swedish Research council had a seminar on the climate system and the solar changes. Link to Seminar on climate change. This seminar shows a strong link between solar radiation and climate changes.

However, direct solar irradiance measurements have only been going on for 30 years by satellite detection.



The solar emission shows a cycle of around 11 years. For the moment the solar activity is low as can be seen to the

left of the figure, whereas the activity was high around the year 2002. We also observe that the variations are less than 0.1%.

The sunspot activity is also higher when the intensity is high, why studies of the number of sunspots can be used as a tool for indirect solar intensity measurements. This has been done for longer periods as is shown below:



We observe that today's activity is the lowest in around 100 years. Also here we observe that the sunspots give peaks within around 11 years and that it has been going on in the same way since around the year 1750, when observations were steady.

Now the question we ask, is if the climate changes are larger when the Sun spot activity is high or low.

#### **Cosmic accelerators**

An important discovery is the outburst of supernovae, the origin of cosmic rays. The main content of the outburst are protons, generated by the supernovae. These pro-



ton generators or *cosmic accelerators* can last for about 300 000 years. The pictures show oscillations of super novae.

Super novae oscillations; Producers of high energy protons.



High energy protons travel into our solar system. They have to pass the solar magnetic field caused by the solar wind. Only 50% of the protons enter the region where the Earth is situated. If the solar activity is high, the solar wind is larger and the number of incoming protons will be reduced. So, if we look at low sunspot rates we would have an increase in protons when sunspots are few, just as it is for now.

The picture shows high and low energy protons enter the solar system and the Heliosphere around the Sun.

Recent research shows that the Sun moderates 10% of the incoming cosmic rays or protons.

What happens when the cosmic rays enter the Earth atmosphere?



Here is shown a simulation where protons enter the atmosphere at 20 km above the ground. The high energy protons produce a shower of particles on its way downwards. There might be millions of secondary particles produced on the proton travel on its way to the surface of the Earth. New isotopes are produced such as like <sup>14</sup>C and <sup>10</sup>Be. Ionization also happens along the protons pathway, which is important for the chemistry of the atmosphere. For example, close to the ground, around 140 muons pass through our bodies per second. (Muon,  $\mu^{-}$ , charge -1e, mass 106 MeV/c<sup>2</sup>, T<sub>1/2</sub> = 2.2 µs).



The figure shows the variation of the cosmic ray flux during the last 12 000 years. The cosmic ray flux is lower in the positive y-axis direction. During the 1000 last years we have the so called *Little Ice Age*. The CO<sub>2</sub> content has been constant, around 280 ppm, during the last 12 000 years, according to ice core studies. During the last decades, however,

there has been an increase in the atmospheric CO<sub>2</sub> content as we have pointed out in Environmental Science I (SK182N).

# Coherence between rain and cosmic rays

There are other means to study the influence of cosmic rays and the environment on Earth. One example is to study the rain or monsoon over various areas on Earth. A measure to study this has been made in Oman where one examined caves with stalactites ("droppstenar") that grew depending on the water content of the air.

One has measured the <sup>14</sup>C content that is directly correlated to the intensity of the cosmic rays, since they are responsible for the <sup>14</sup>C production (Paragraph 4.4) in the atmosphere. One also measured the water content by studying the growth of the

stalactites. In the picture below we see the result in Oman between the years 7900 to 8300 years ago.



The correlation between the cosmic ray intensity = <sup>14</sup>C creation and the growth of the stalactites (water content = monsoon) is astonishingly strong even on small timescales like in the figure.(H. Stensmark, National Space Institute, Denmark)

In the seminar H. S. also looks at the last 1000 years and compares available tem-



perature data with cosmic ray data and finds a nice correlation also in this case. The blue line is the global temperature measurements the last millennium with uncertainties and the red line corresponds to the isotopic change corresponding to the cosmic ray change or its activity.

How can now stellar particles like protons have an influence on the Earth's temperature? It seems that solar activity and climate are closely related. Researchers believe that the incoming particles change the Earths cloudiness. Since, if one changes the clouds, one changes the energy that reaches the surface of the Earth. The cooling of the surface by cloud formation is a cooling of the order of 30 W/m<sup>2</sup>. In the figure below we see the correlation between the cloudiness (blue) and the incoming cosmic rays (red) as measured by a neutron monitor. The cloudiness has been measured by satellites for some 30 years.



Also here, cloud formation and cosmic ray activity are closely correlated to each other. The solar activity alone is so small it could hardly be the explanation. However, if it affects cloud formation, then we have a parameter that could be responsible for climate changes.



So we have to look for a mechanism that is responsible for the climate change that is correlated to solar activity and/or cosmic rays.

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This question is hard directly to answer. It must however, be cloud formation that is a kind of amplifier of solar or cosmic ray activity.

Therefore, we have to discuss cloud formation and see if we have the solution to the problem there.

#### **Cloud formation**

When clouds are formed, a cloud droplet has to be produced around a cloud condensation nucleus (CCN) as shown to the left of the figure below. But how do we pro-



duce these CCNs? It is believed that the CCN are created through a process where trace gases in the atmosphere, like H<sub>2</sub>SO<sub>4</sub> as shown. Then there has to be a channel leading to ultrafine condensation nuclei (UCN). Then they grow to CN and CCNs.



The process is not fully understood where the aerosol sizes grow from around 1 nm to 10  $\mu$ m for the droplet. It has been proposed that it is the cosmic rays and the ionization that is responsible for the creation of UCNs that form the CNs and the CCNs.

Experiments have been going on where large chambers with trace gases,  $O_3$ ,  $SO_2$  and water vapour are inserted that are relevant for the atmosphere. Cosmic rays are being simulated by using radioactive sources emitting  $\gamma$ -rays causing ionisation of molecules. Detection with mass, aerosol and ion spectrometers was used in the experiments. Small particles like the ones in the atmosphere were to the UCNs pacessary for cloud formation

found correspond to the UCNs necessary for cloud formation.

Studies have been performed where the cosmic radiation leading to ionization in the



atmosphere has been compared with the water content of clouds over the oceans. The red curve in figure shows the ionization and the produced by cosmic rays and the water content of the clouds. The water content curve follows the ionization, but is shifted around 10 days. The ionization change is of the order of 10% and the liquid water content around 6%. One believes that an increase in ionization could give rise to a rather high decrease in water content of the clouds.

So, as a conclusion:

- Variations in cosmic ray intensity affects the climate of Earth
- Evidence suggests that clouds play an important role
- It involves ions and aerosols formation