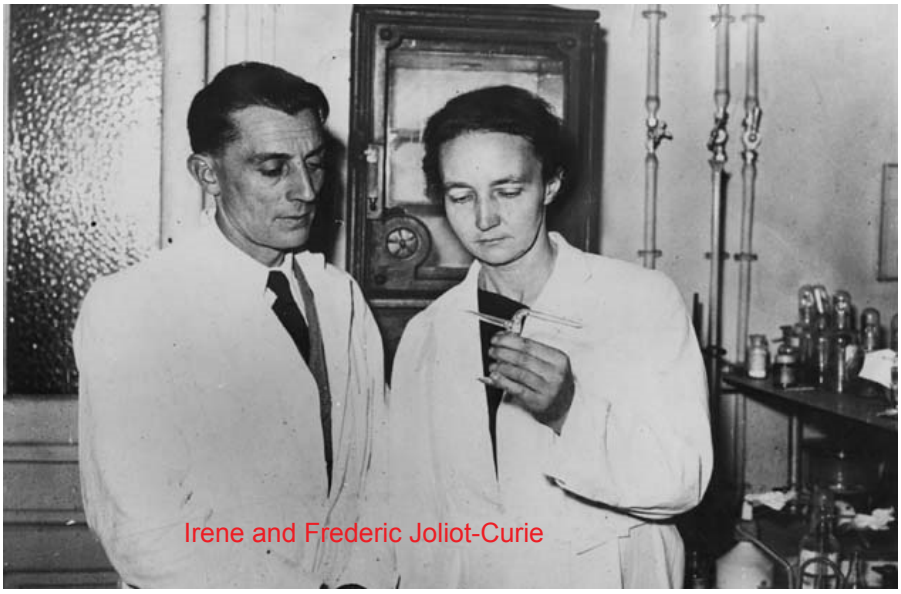


Irene and Frederic Joliot-Curie

Chapter 4

Artificial Radioactive Isotopes

The Discovery

It was mentioned earlier that Irene Joliot-Curie and her husband Frederic Joliot-Curie was awarded the Nobel prize in Chemistry in 1935. The prize was given "in recognition of their synthesis of new radioactive elements" .

Let us see more into the details of this experiment.

Irène and Frederic Joliot-Curie had a large supply of polonium, after Irene`s parents. The polonium emitted alpha particles which they used to bombard different elements. In 1933 they used alpha particles and bombarded an aluminum plate. When they removed the α–particle source, it appeared that the aluminum plate emitted radiation with a half-life of approximately 3 minutes. The explanation was that the bombardment had resulted in a nuclear reaction. The α-particle penetrated the aluminum nucleus and changed it into phosphorus by emitting a neutron. The new phosphorus isotope was radioactive and was responsible for the observed radiation. Its designation is P-30.

This nuclear reaction may be written as follows:

$^{27}Al + \alpha \Rightarrow {}^{30}P + n$ or like this $^{27}_{13}Al + {}^{4}_{2}He \Rightarrow {}^{30}_{15}P + {}^{1}_{0}n$

Decay of P-30 is: $^{30}_{15}P \Rightarrow {}^{30}_{14}Si + \text{positron}$ Energy 3.24 MeV

The neutron emitted can be observed as long as the bombardment takes place, but disappears immediately when the α-source is removed. However, the phosphorus isotope is radioactive. The decay mode is positron emission as shown above. The half-life is 2.5 minutes.

Irene and Fredric Joliot-Curie used their alpha bombardment technique on some other elements and found that it was possible to transform an element into another, with a higher number of protons in its nucleus. They announced this breakthrough to the Academy of Sciences in January of 1934 – and received the Nobel prize the year after. It can be mentioned that Marie Curie died July 1934 and was thus able to follow this exciting research almost up to the third Nobel prize in the family.

Radioactive isotopes are made!

Radioactive isotopes can be made by bombarding an element with a particle (α-particle, deuteron, proton, electron, neutron and even high energy x-rays). The neutron is the most efficient.

In general, the probability of a reaction depends on the energy of the particle – and it is measured in the so called "*reaction cross section*". It appears that neutrons with low energy (slow neutrons) in general have a larger cross section than fast neutrons.

In the mid 1930's, several laboratories had developed equipment, such as the accelerator called cyclotron, to bombard stable atoms with protons, deuterons and α-particles and found that new isotopes were formed. Some of these isotopes were radioactive.

Since protons, deuterons and α-particles are charged, they will mainly be scattered (because of Coulomb interaction) when approaching the positive nucleus of an element. Because of this, and since the techniques available for accelerating particles in the 1930-ties was rather poor, it was impossible (with the available energy) to transform elements with atomic number above 20 .

Use of neutrons – discovery of the efficiency of slow neutrons

A very efficient particle that can be used for bombardment is the neutron. This particle has no charge and will consequently not be influenced by the electric field around the atomic nucleus. The neutron readily penetrates the atomic nucleus with the result that new isotopes are formed.

In Rome, Italy, a small group of excellent scientists (Amaldi, d'Agostino, Pontecorvo, Rasetti, and Segré), headed by Enrico Fermi, used neutrons for bombardment. The neutron source consisted of beryllium powder and radon in a small glass bulb (the α-particles from radon hit beryllium with neutrons as a result). With this relatively strong neutron source they bombarded a number of heavy elements and observed numerous new radioactive isotopes. They assumed that they found both element 93 and 94 (see later).

During these experiments Fermi made an important observation. He found that when the source was surrounded by paraffin or water, the efficiency of the neutrons increased by up to 100 times. Consequently, when the neutrons were slowed down by collisions with hydrogen atoms, they become more effective. *Slow neutrons* yield a larger cross-section for many reactions than fast neutrons. With this technique they observed several hundreds new isotopes. These experiments from the 1930-ties earned Fermi the Noble prize in physics for 1938.

Enrico Fermi
(1901 – 1954)

Enrico Fermi was one of the top scientists of the 20th century. He discovered in 1926 the statistical laws, nowadays known as the «Fermi-Dirac-statistics». In 1927, Fermi became Professor of Theoretical Physics at the University of Rome (a post which he retained until 1938, when he – immediately after he received of the Nobel Prize – emigrated to America, primarily to escape Mussolini's fascist dictatorship).

In 1934 he presented his explanation of the β-decay (a paper that was first not accepted in Nature). He then carried out the work with neutrons that earned him the Nobel prize.

In USA he was in charge of the work that led to the first controlled nuclear chain reaction – the reactor on a squash court situated beneath Chicago's stadium.
Today we have several things with his name – such as element number 100 which is called Fermium, ”The Fermi National Accelerator Laboratory” and ”The Enrico Fermi US Presidential Award”, which was started in 1956.

Noble prize in physics 1938

To Enrico Fermi
"for his demonstrations of the existence of new radioactive elements produced by neutron irradiation, and for his related discovery of nuclear reactions brought about by slow neutrons"

Reactors are excellent sources of neutrons and are used for the production of radioactive isotopes needed for biomedical research and the treatment of disease. Also a number of accelerators are used for the formation of particular radioactive isotopes.

The number of artificial isotopes increased rapidly in the years after 1934. Thus, by 1937, approximately 200 isotopes were known, in 1949 the number was 650 and today more than 1,300 radioactive isotopes have been produced.

Fission

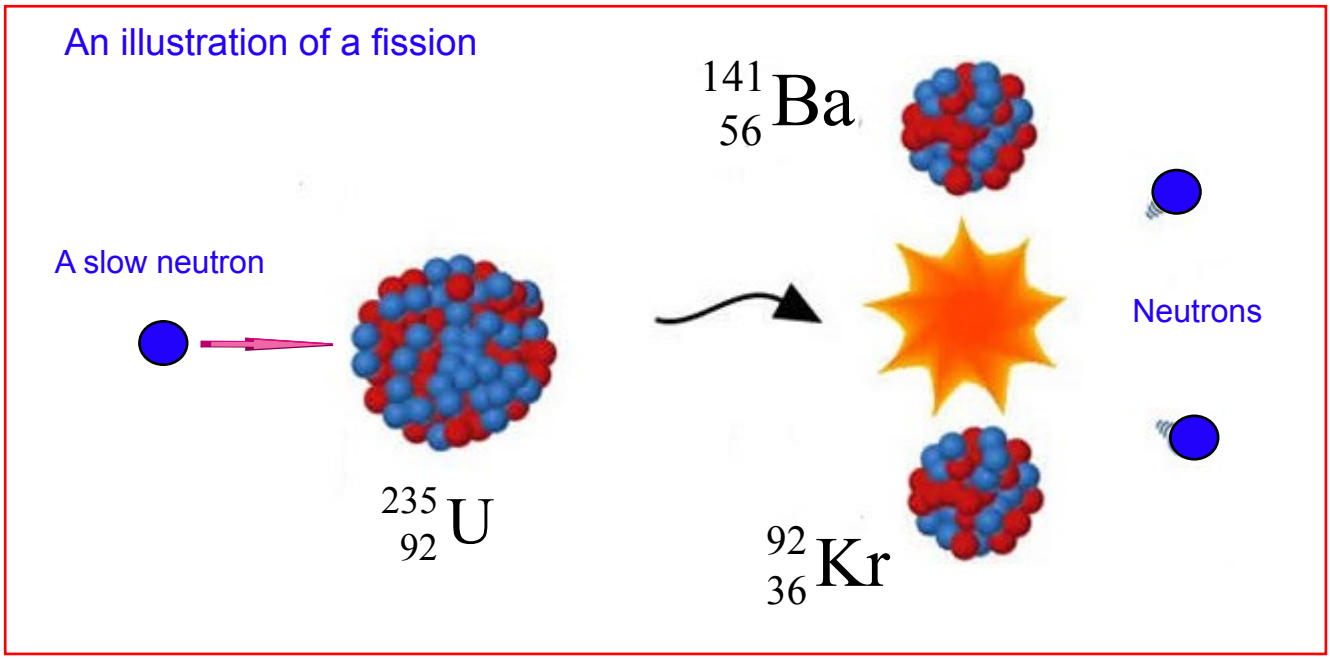
A short history

James Chadwick discovered the neutron in 1932, and this initiated a number of research projects. The goal was to make and identify the isotopes formed when neutrons penetrate various atomic nuclei. We mentioned above that Fermi used neutrons for this purpose. In 1934 he observed β-particles when he bombarded uranium. Fermi`s interpretation was that he had made a transuranic element – like 93. This interpretation was questioned by the German chemist **Ida Noddack.** In a famous paper, "*Über das Element 93*" published in "Zeitschrift fur Angewandte Chemie" in 1934. Noddack suggested a

number of possibilities such as "it is conceivable that the nucleus *breaks up into several large fragments,* which would of course be isotopes of known elements, but would not be neighbors of the irradiated element." In this way Ida Noddack suggested a nuclear fission – which was found a few years later. Since however, she presented no theoretical or chemical basis for this possibility, the paper was more or less ignored at that time.

Ida Noddack
(1896 – 1978)

At The Kaiser Wilhelm institute in Berlin **Otto Hahn, Lise Meitner** and **Fritz Strassmann** worked with uranium and neutron bombardment. This research resulted in the conclusion that uranium can be split in two large fragments when bombarded with neutrons (see illustration below).



Lise Meitner, was an important member of the small group in Berlin. She and Otto Hahn had a lot of discussions in the summer of 1938 – when the political situation in Germany became more and more difficult for Lise Meitner. She was of Jewish ancestry and had lost her Austrian citizenship after the Anschluss and was at great risk. Otto Hahn, Niels Bohr and Dutch physicists were able to help Lise Meitner to escape via the Dutch border and go to Sweden. Here she took up a post at Manne Siegbahn's laboratory.

Lise Meitner
(1878 – 1968)

Hahn and Strassmann continued the experiments in Berlin and showed in an experiment on the 17th of December 1938, that it **was not possible to separate barium from the compound formed** when uranium was hit by neutrons. On the 19th of December, Otto Hahn wrote a letter to Meitner about the latest results. He ended his letter with the sentence: *"Perhaps you can suggest some fantastic explanations".*

Lise Meitner travelled to Kungälv (just north of Göteborg) to spend the December holidays together with some of her family. Here she met her young nephew **Otto Frisch** who worked with Bohr. One day they sat on the trunk of a tree, discussing the barium mystery. Using the the nuclear model of Bohr (the so called liquid drop model) as a basis, they calculated that if a neutron penetrated the nucleus, it could set up oscillations that would split the atom - *fission was possible !*

Maybe it can be said that the atomic age started on a timber log north of Göteborg during the December holidays of 1938. Hahn and Strassmann published their experiments in the German Journal *Naturwissenschaften*. Meitner and Frisch published their theoretical calculations in the British Journal *Nature* and Bohr let the news explode at a conference in the United States in January 1939.
It was however Otto Hahn – and only him – that was awarded the Noble prize for this discovery.

Otto Hahn
(1879 – 1968)

Hahn started to work together with Lise Meitner already in 1907 – and this collaboration lasted for more than thirty years. They identified several radioactive isotopes.

Following the discovery of artificial radioactivity by Joliot-Curie and the use of neutrons by Fermi for atomic nuclear processes, Hahn, Meitner and later Fritz Strassmann collaborated on the project that resulted in the discovery of fission.

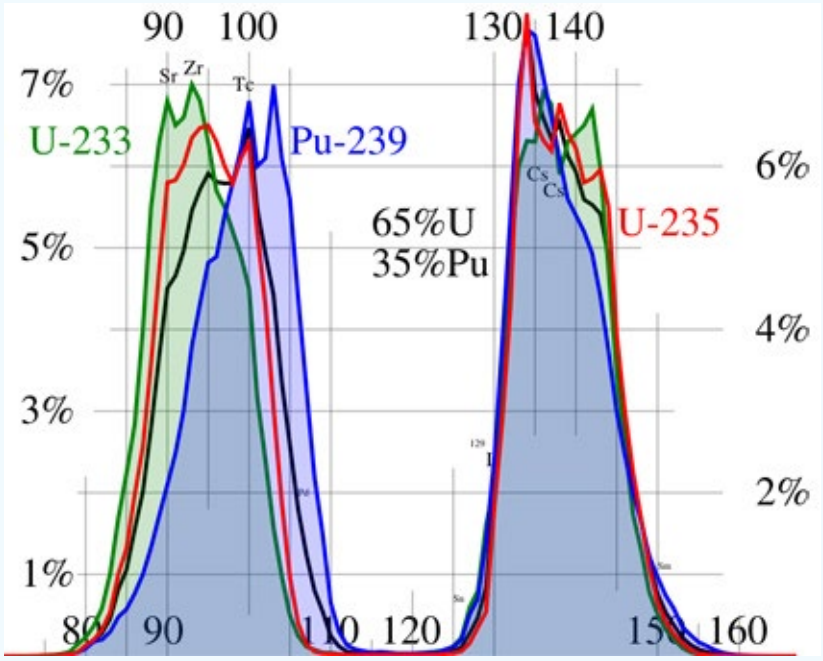
Otto Hahn have been considered as "the father of nuclear chemistry". He started early to work on the chemistry of radioactive isotopes. He became professor in 1912 and then head of The Radioactivity Department of Kaiser Wilhelm Institute for Chemistry in Berlin.

Nobel prize in chemistry 1944

To Otto Hahn
"for his discovery of the fission of heavy nuclei".

Isotopes formed by fission

The splitting of a heavy atomic nucleus, such as U-235, occurs because of intrinsic instabilities when a neutron enters. There are numerous pathways by which the nucleus emits energy and creates new products. The fission products formed are very unstable and undergo β-decay to form new products. More than 200 fission products are known. The products formed can be divided into two groups, one "heavy" group with an atomic weight of about 130 – 140 mass units and one "light" group with an atomic weight of 90 – 100 mass units. See illustration on next page.



Here is a figure that shows the yield of fission products for U–235 (red), U–233 (green), Pu–239 (blue) and a mixture of U and Pu (black). Two elements are formed in each fission. The two elements are not equal in mass. The average mass of the lighter element is between 90 and 100 atomic units and of the heavier element is between 130 and 140 atomic units. The distribution is shown by the graph. The vertical-axis indicate the yield.

A large amount of energy is released in the fission process. Most of the energy is released directly during the process of fission but a small amount is released at a later stage by those fission products that are radioactive. Most fission products have short half-lives. However, unstable daughter products have longer and longer half-lives.

From an environmental point of view, Cs-137 and Sr-90 are the most important fission products of U-235. They have a half-life of about 30 years, which is important with regard to storage and disposal of these products. The fission process leads to three different types of radioactive isotopes: *fission products, transuranic elements, and activation products*.

Table 4.1: Some important fission products

The Light Group			**The Heavy Group**		
Isotope	Symbol	$t_{1/2}$	Isotope	Symbol	$t_{1/2}$
Krypton-85	Kr-85	10.7 yr	Tellurium-129m	Te-129m	33.6 d
Strontium-89	Sr-89	50.5 d	Iodine-131	I-131	8.04 d
Strontium-90	Sr-90	29.1 yr	Xenon-133	Xe-133	5.3 d
Yttrium-91	Y-91	58.5 d	Cesium-137	Cs-137	30.0 yr
Zirconium-95	Zr-95	64 d	Barium-140	Ba-140	12.7 d
Technecium-99	Tc-99	213,000 yr	Praseodymium-143	Pr-143	13.6 d
Ruthenium-103	Ru-103	39.3 d	Neodymium-147	Nd-147	11.0 d
Ruthenium-106	Ru-106	368 d	Promethium-147	Pm-147	2.6 yr

1. Fission products

There are a large number of fission products, and some of the most important ones are given in the table above. In the first period after fission occurs isotopes with short half-lives dominate, i.e. Zr-95 and I-131. Later, Sr-90 and Cs-137 are predominant.

2. Transuranic elements

Transuranics are elements with an atomic number larger than 92 (uranium). Most transuranic elements are made in accelerators when heavy atoms such as uranium are bombarded with neutrons, deuterons or – other small charged particles. Thus plutonium was first produced and isolated in 1940, by Glenn T. Seaborg, Edwin McMillan and colleagues by deuteron bombardment of uranium in the 60-inch cyclotron at the University of California, Berkeley. They shared the Nobel prize in chemistry in 1951 for the discovery of the transuranium elements. Plutonium is also formed when U-238 absorbs a neutron and subsequently emits two β-particles. In a reactor this process can produce large amounts of plutonium.

McMillan and Seaborg

In addition, it can be noted that the transuranic elements are usually α-particle emitters, whereas the fission products are β-particle emitters. This implies that when the transuranics come into the body, through inhalation or ingestion, they deposit all their energy within the body. Thus, transuranic elements may be a health concern.

3. Activation products

The third type of radioactive isotopes which may be produced in combination with reactors and nuclear weapons, are activation products. These radioactive isotopes are formed when stable isotopes are bombarded by neutrons. Co-60 is an activation product formed when Co-59 absorbs a neutron. Likewise, Cs-134 is formed from Cs-133 by neutron capture. The materials around a reactor or a nuclear bomb explosion can be made radioactive by neutron capture.

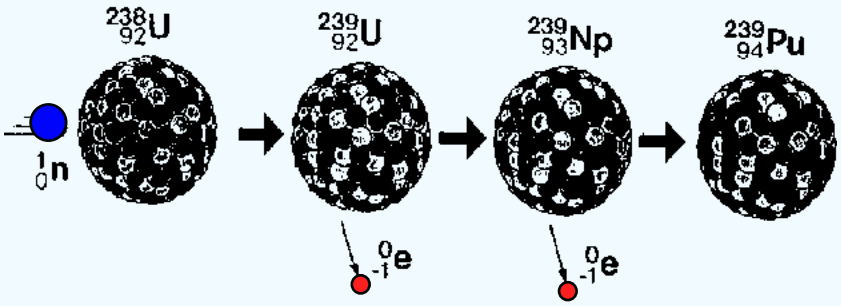
If a nuclear bomb detonates in the atmosphere, large numbers of neutrons will be released. They can then react with stable atoms such as N-14. In this way the radioactive isotopes H-3 and C-14 are formed.

If the detonation takes place just above the ground, large amounts of materials (earth, rocks, building materials, etc.) will be activated and vaporized in the “fireball” which is formed. Such nuclear tests yield large amounts of radioactive fallout.

The use of neutrons to form new radioactive elements can also be used in research to attain new and valuable information. This will be mentioned under "activation analysis".

Plutonium

Few radioactive isotopes have attracted more interest than plutonium – a tranuranic element with atomic number 94. Several plutonium isotopes exist. Thus, we have Pu-238 (half-life 87.7 years), Pu-239 (24,400 years), Pu-240 (6,570 years) and Pu-241 (14 years). Pu-239 is the best known and its environmental impact has been heavily debated during the last decades. It is formed in a reactor (see the illustration below) after neutron bombardment of U-238. Pu-239 sits in the spot light because it is not only a cost effective fuel used in fission reactors but also, it is a key ingredient in nuclear weapons.



Pu-239 production starts with a neutron absorption by U-238. U-239 and subsequently Np-239 are unstable, and both emit β-particles. The final product is Pu-239

With regard to the environment, Pu-239 is the most important of the plutonium isotopes. It emits an α-particle with an energy of 5.15 MeV. In air, these α-particles have a range of a few cm, whereas, in tissue, the range is less than one mm.
The following important conclusion can be made from this: *plutonium has a minor influence on a person's health when it is outside the body since the emitted α-particles will not enter the body. On the other hand, if plutonium enters the body all the emitted α-particles will deposit their energy within the body.*

Large amounts of plutonium have been released to the atmosphere due to atmospheric nuclear tests. It has been estimated, that for the period 1945 to 1974, approximately 400,000 Ci or $1.5 \cdot 10^{16}$ Bq Pu-239 were released – corresponding to 6.5 tons. As a consequence, plutonium is now found in nature. The fallout of plutonium is approximately as fast as that for strontium (Sr-90). The ratio between plutonium and strontium fallout has been rather constant since the large weapon tests ended in 1963. Calculations show that the total fallout on the Northern hemisphere is approximately 50 Bq per square meter.

Plutonium entering the body via the food chain represents a small radiation problem since only 30 ppm is absorbed in the blood from the intestine. However, the plutonium which enters the body via inhalation (such as those attached to dust particles in the air) presents a more serious problem.

Activation Analysis

The activation of certain materials by neutron irradiation is an elegant analytical method for identifying chemical species. When a compound is irradiated with neutrons, many elements are activated and become radioactive. The radioactivity can be measured easily and the properties of the radiation can be used to identify an element. Thus, it is possible to observe with excellent sensitivity about 70 elements. In some cases, multi-element analysis at the ppm level for up to 30 elements can be accomplished with a sample weighing less than a gram.

It is a type of analysis which do not destroy the sample and it is used by archaeologists to obtain important information about old coins, pieces of ceramic pots, and other relics.

Napoleon

Criminologists use activation analyses in the solution of criminal cases. One example that has been heavily debated is the activation analysis which showed that the hair of Napoleon contained arsenic (work in the 1960-ties). This suggested the possibility that Napoleon was murdered. However, it is possible that the arsenic could have come from his environment. At that time, arsenic was used in wall coverings and could have been picked up by touch or given off into the atmosphere.

New experiments carried out in 2007 seem to prove that more than 97% of the arsenic found in the hair of Napoleon is in inorganic form, which is consistent with a chronic intoxication by the most toxic inorganic arsenic species.

Rocks from the moon

It is also of interest to mention that when the composition of the moon was determined, the analysis was assisted by neutron activation. Rocks, brought back to Earth by the astronauts, were bombarded by neutrons, forming radioactive products. The subsequent radioactive emissions were then used to identify elements in the moon rocks.

Women in nuclear chemistry

For all those that have followed this short glimpse of history into the field of radioactivity and the formation of radioactive isotopes may have noticed that a surprisingly large fraction of the scientists are women. Remember this research took place between 1900 and 1940. In the foregoing we have tried to mention their work – and we would like to end this chapter by lining them up again. The women are: Marie Curie, Irene Joliot-Curie, Ida Noddack and Lise Meitner.


Mare Curie


Irene Joliot-Curie


Ida Noddack


Lise Meitner

These four ladies were forerunners in nuclear chemistry and led us into the atomic age.

One of Marie Curies students was the Norwegian Ellen Gleditch. She came to Paris in 1907 and joined the Curie - group. She later introduced nuclear chemistry to Norway. Furthermore, she was very much involved in the purchase of four grams of radium – that was the foundation of the Norwegian Radium Hospital in 1932.


Ellen Gleditsch
(1879-1968)


Ellen Gleditch in a photo from 1912. She had just obtained the degree " Licenciée ès Sciences".