# Chapter 10

# ISOTOPIC SEPARATION AND ENRICHMENT

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# 10.1 INTRODUCTION

There is a need in nuclear applications to separate the light isotopes of the elements such as deuterium  $_1D^2$  from hydrogen, Li<sup>6</sup> from lithium, B<sup>10</sup> from boron as well as the heavy isotopes such as U<sup>235</sup> from natural uranium and Pu<sup>239</sup> from a mixture of plutonium isotopes. Large commercial enrichment plants are in operation in France, Germany, The Netherlands, UK, USA, and Russia, with smaller plants in Japan, South Africa, Brazil, Argentina, Pakistan, Democratic People Republic of Korea (DPRK) and Iran. Four major suppliers of enrichment services are: the Unites States Department of Energy (USDOE), Eurodif in France, Urenco, a British, Dutch, and German group, and Techsnabexport in Russia.

Canada and India, which both rely on heavy water nuclear power plants for electricity, make the most heavy water. Other countries with heavy water production facilities include Argentina, Iran, Romania, and Russia.

The world has used just three very inefficient nuclear enrichment processes: gaseous diffusion, centrifugation, and Electro Magnetic Isotope Separation (EMIS.) EMIS enrichment is an inefficient process: It would cost \$81,000 to enrich a single pound of uranium. Gaseous diffusion is not all that much better. This 1940s vintage energy intensive enrichment process currently accounts for 25 percent of all enriched uranium and takes more than 1,400 stages to enrich uranium to a useful level. The centrifuge technology is taking over the enrichment industry. The United States Enrichment Company (USEC) is building the American Centrifuge plant in Ohio. This project is facing constant delays and budget overruns and is expected to reach full production in 2012. There is a new enrichment facility under construction in New Mexico by the European firm Urenco, which will not be running at full capacity until 2017.

The separation of the light isotopes occurs in nature in closed bodies of water such as the Great Salt Lake in Utah, USA, and the Dead Sea in the Middle East. Evaporation of water leads into an enrichment into heavy water: D<sub>2</sub>O through the process of fractional distillation, because it has a higher boiling point than ordinary water H<sub>2</sub>O. Electrolysis can also be used to produce heavy water since the dissociation of light water proceeds faster than that for heavy water leading to its enrichment in the electrolytic solution. Heavy water is currently used as coolant and moderator for a reactor design using natural uranium as a fuel: the Heavy Water Reactor (HWR) design.

The need for separating the two isotopes of uranium: U<sup>235</sup> and U<sup>238</sup> arose initially as part of the Manhattan project for the manufacture of the atomic bomb. Later, it became the basis for using light water as a coolant and moderator for propulsion naval reactors as well as land-based reactors. In propulsion reactors there was a need to increase the enrichment from the natural abundance of 0.72 percent in U<sup>235</sup> to 97.3 percent as a way of providing enough reactivity to overcome the xenon poisoning and the reactor dead time arising in these systems, increasing the

time between refueling to about ten years, and even for the whole operational time of the core. For nuclear warheads, the enrichment is increased to 93.5 percent. For land-based reactors, the U<sup>235</sup> is raised to the 3-5 percent range to achieve criticality with light water. Interestingly, this is the range of enrichment that occurred earlier in the history of the Earth at the natural reactors at the Oklo uranium mine site. For space reactors an enrichment of 97.3 percent is envisioned in view of reducing the launch weight.

Isotopic separation is also encountered in the separation of the Li<sup>6</sup> isotope from lithium for thermonuclear weapons and future peaceful fusion applications. For the reactors using natural uranium as a fuel, separating heavy water from ordinary water becomes a replacement for the enrichment of uranium. However, it is easier to isotopically separate the light elements compared with the heavy ones.

The most important separation methods for the heavy elements as well as the light elements are described.

# 10.2 STABLE ISOTOPES APPLICATIONS

The stable isotopes produced by isotopic separation are used worldwide in both medical and industrial applications.

Three segments of the medical field are served:

# 1. DIAGNOSTICS AND IMAGING

Nuclear diagnostic imaging has an important role in the identification and management of conditions such as heart disease, brain disorder, lung and kidney functions and a broad range of cancers. The high sensitivity and specificity of nuclear diagnostic imaging techniques offer the important advantages of being able to identify diseases at an early stage, to track disease progression, to allow for accurate disease staging and to provide predictive information about likely success of alternative therapy options.

The two most important techniques are Gamma Imaging and Positron Emission Tomography (PET).

### **Gamma Imaging**

There are some 8,500 nuclear medicine departments in the world using gamma cameras to detect diseases of various organs including heart, brain, bone, lung and the thyroid. A total of some 20,000 gamma cameras are in use. Gamma imaging uses the stable precursors for Gallium<sup>67</sup> such as Zinc<sup>68</sup> and Zinc<sup>67</sup>, Indium<sup>111</sup> (Cadmium<sup>112</sup>) and Iodine<sup>123</sup> (Xenon<sup>124</sup>).

#### **Positron Emission Tomography (PET)**

There are about 200 PET centers in the world operating a total of some 300 PET cameras. They are used mainly for the diagnosis and staging of cancer. Use of PET is growing as a result of the recognition of clinical benefits from PET.

# 2. RADIATION THERAPY

# Brachytherapy

Brachytherapy is the procedure of using temporary irradiation very close to the area of disease, in particular cancer and stenosis. Iridium<sup>191</sup> is produced for Iridium<sup>192</sup> sources used in remotely controlled after loaders. A modern development pursued by many companies is Seeds Implantation: implanting radioactive sources ("seeds") in tumors, in particular prostate cancer. This minimizes the side effects known from surgery and/or external radiation. More than 300,000 new cases of prostate cancer are diagnosed every year in the USA and the EU alone, and patients are increasingly being treated with seeds implantation. The radioactive source most often used in the seeds is Iodine<sup>125</sup> (made from Xenon<sup>124</sup>).

Restenosis is the re-narrowing of a coronary artery after angioplasty treatment. This happens in about 30 percent of the patients within a few months after treatment, a percentage which can be dramatically reduced by restenosis treatment: the irradiation through brachytherapy of the area unblocked by angioplasty treatment. The treatment is yet far from standard procedure. Many tests are being conducted, including tests to determine what radioactive isotopes and thus what precursors are most suitable such as Tungsten<sup>186</sup>, which is a precursor for Rhenium<sup>188</sup>, a possible candidate for restenosis treatment.

A new development is to deplete Titanium<sup>46</sup>. Natural Titanium is a suitable biocompatible material, which is being used to encapsulate radioactive seeds for brachytherapy, in particular for prostate cancer. Instead of the complicated process of sealing radioactive sources into Titanium capsules, it would be much easier to seal the stable precursors into Titanium capsules before irradiating the precursors. However, this would make it necessary to deplete the Titanium<sup>46</sup> isotope in order to eliminate the adverse radioactivity of activated Titanium<sup>46</sup> (Scandium<sup>46</sup>).

#### 3. PAIN RELIEF

Palliative care of pain arising from secondary metastasis derived from spread of breast, prostate and lung cancers is under development. Growth is rather slow due to the problem of acceptability and the conservatism amongst doctors preferring traditional medicines like morphine.

# 10.3 INDUSTRIAL ISOTOPES

# **DEPLETED Zinc<sup>64</sup> (DZO)**

Depleted zinc<sup>64</sup> is used in the nuclear industry. The addition of natural zinc to nuclear reactor cooling water inhibits corrosion and the subsequent generation of radioactive Cobalt<sup>60</sup>, thus acting to reduce worker radiation exposure. Natural zinc contains 48 percent zinc<sup>64</sup> and as the cooling water is subjected to continual neutron bombardment this isotope is activated to radioactive zinc<sup>65</sup>. This isotope is a strong radiation emitter with a long half life and thus contributes greatly to the storage time and hazard of waste cooling water.

The use of depleted zinc<sup>64</sup> compounds enable the maximum benefits of zinc injection to be reaped without the attendant radio nuclides consequences.

Nuclear plants use Depleted Zinc Oxide (DZO) pellets or powder for BWR's, and Depleted Zinc Acetate (DZA) for PWR's. Using centrifuge technology concentrations can be enriched to exceed 99 percent or depleted below 1 percent.

The USA's EPRI (Electric Power Research Institute) recommends the use of depleted zinc. The added zinc reduces the amount of radioactive cobalt<sup>60</sup> formed because of the irradiation of natural cobalt in the construction materials of the reactor. <sup>60</sup>Co is a major contributor to radiation build up in the cooling systems and therefore causes elevated dose rates to personnel.

# TRANSMUTATION OF LONG-LIVED RADIO-ISOTOPES IN SPENT NUCLEAR FUEL

Depleted molybdenum<sup>95</sup> is used in an experimental process of transmutation of the long lived radio isotopes, which are present in spent nuclear fuel. Americium in the form of AmO<sub>2</sub> is embedded in a matrix of molybdenum metal, processed to cermet pellets and irradiated in a nuclear reactor. The americium is then transmuted into other, much shorter living radio isotopes. The molybdenum has to be depleted in the isotope Mo<sup>95</sup> in order to reduce its radiative capture cross section for neutrons.

# NON-DESTRUCTIVE TESTING (NDT) SOURCES

NDT using gamma cameras is an important and growing application of radioactivity. The majority of the cameras use an iridium<sup>192</sup> sources, although selenium<sup>75</sup> sources are a promising growth sector.

Iridium<sup>192</sup> and selenium<sup>75</sup> use the stable precursors iridium<sup>191</sup> and selenium<sup>74</sup> respectively.

#### SEMICONDUCTOR APPLICATIONS

Increased miniaturization of chips might require ultra pure silicon, germanium and gallium material to build new generation semiconductors. Research in this field is ongoing.

# **10.4 CHEMICAL EXCHANGE**

This technique makes use of different chemical reaction rates between isotopes. It works best for light elements where the reaction rate differences are large. Practical plants use reactions that allow the two reactants to be in different phases such as gas/liquid, solid/liquid, and immiscible liquids. This permits convenient separation of the enriched and depleted materials, and allows continuous countercurrent operation. By also including temperature differences between the phases, the separation factor can be considerably enhanced. This is the most important process today for producing heavy water, for which it is by far the most energy-efficient method. Chemical exchange techniques for uranium have also been developed by Japan and France, but have not been used for production work. Chemical exchange is also used for Li<sup>6</sup> enrichment.

#### HEAVY WATER, HDO AND D2O

Heavy water HDO and D<sub>2</sub>O, differs from the widely abundant light water H<sub>2</sub>O in the presence of the deuteron nuclide D, rather than the hydrogen nuclide H, in the water molecule. Ordinary water is a mixture of its heavy and light forms. Heavy water has a density of 1.1 [gm/cm<sup>3</sup>], compared with 1.0 [gm/cm<sup>3</sup>] for light water at room temperature, which explain its name. In nature, deuterium occurs with an abundance that varies between 130-160 parts per million or ppm in hydrogen, with an average of 0.015 in atomic percent or 150 ppm. The ratio varies according to location, seasonal temperatures and meteorological factors. Because of its rarity, it occurs in the form of HDO, or the mono-deuterated molecule.

Heavy water is used as coolant and neutron moderator in nuclear reactors using natural uranium as fuel, such as the Canadian Uranium Deuterium CANDU type (Fig. 1).





Figure 1. Darlington 935 MWe CANDU Heavy Water Reactor (top), and reactor complex at Ontario, Canada.

One cubic meter or one metric tonne is needed per MWe of installed capacity in the HWR design. Its excellent moderating properties, high scattering cross section for neutrons, and low absorption cross section makes it a unique moderator in this case. Heavy water must be replaced in these systems because of leakage, in the form of steam, from the pressurized water circuits. Building volumes are sealed around the coolant circuit, and the D<sub>2</sub>O vapor, which leaks into them, is condensed and extracted for both economic reasons, and to avoid leakage of the tritium gas.

Tritium, an isotope of hydrogen that is beta radioactive with a half-life of 12.34 years, is produced from deuterium through neutron capture in the reaction:

$$_{1}D^{2} + _{0}n^{1} \rightarrow _{1}T^{3} + \gamma$$
 (1)

where  $\gamma$  is a gamma photon.

Tritium can be extracted for radioactive isotopes applications such as activation of Liquid Crystal Displays (LCDs), and future use in fusion reactors or thermonuclear weapons through the DT fusion reaction:

$$_{1}D^{2} + _{1}T^{3} \rightarrow _{0}n^{1} + _{2}He^{4} + 17.6 \text{ MeV}.$$
 (2)

In future fusion reactors, deuterium can fuse with other deuterium nuclei producing energy in the process through the reaction:

$$_{1}D^{2} + _{1}D^{2} \rightarrow _{0}n^{1} + _{2}He^{4}$$
, (neutron branch) (3)

$$_{1}D^{2} + _{1}D^{2} \rightarrow _{1}H^{1} + _{1}T^{3}, \quad \text{(proton branch)}$$
 (4)

This reaction, which branches with equal probability, is the ultimate use of hydrogen as deuterium in the oceans, and constitutes a practically unlimited future supply of fusion energy on Earth.



Figure 2. Heavy water ice sinks in light water because of its higher density. Light water ice would float because of its lower density. Source: Nova.

#### **ELECTROLYSIS**

To separate heavy water from light water, electrolysis, distillation of water or of liquid hydrogen and a chemical exchange method, have been used commercially. The concentration of heavy water in ordinary water is about 0.01 percent. Its concentration must be increased to 99.76 percent, which is essentially pure heavy water.

A multistage electrolysis process can concentrate heavy water, since light water can be dissociated preferentially by an electric current gradually concentrating the heavy water. To make the water conduct electricity potash lye, which is actually potassium hydroxide is added which makes the water very caustic with a pH value of 14 compared with the neutral value of 7.

Germany used this process in the Second World War in an aborted attempt under the leadership of their leading scientist Werner Heizenberg at building a heavy water nuclear reactor.

The process used abundant electrical power in a plant at Vemork, Norway, operated by Norsk Hydro.

The Norsk Hydro company used hydroelectric power to manufacture ammonia, the basis of fertilizer and explosives, both vital to the German war effort. To make chemical fertilizer, the company needed hydrogen, and hydrogen could be made from water electrolysis.

The Vemork plant is shown in Fig. 3. Norwegian commandos blew up the plant, but the Germans had it working again within three months. Then the USA Eighth Air Force bombed Vemork, but the heavy water plant survived unscathed. Eventually, in 1944, members of the resistance learned that the entire production plant and 15 tons of partially purified heavy water were to be shipped to Germany. A commando attack destroyed the plant and sunk the Hydro ferry shipment of heavy water destined for Germany at the cost of 14 local Norwegian and 4 German victims in the 400 meters deep icy waters of Lake Tinn. Some half filled barrels floated and eventually found their way to Germany. The Germans needed about five tonnes of heavy water for a single reactor; when at least ten reactors would have been needed for weapon effort. but only half a ton of very diluted heavy water was shipped. The heavy water was destined for a civilian experimental program, not a large scale military one. This explains why it was not heavily guarded. Nevertheless, the raid slowed down and eventually stopped any German effort at building any self-sustained fission chain reaction, even though a subcritical system using natural uranium and heavy water was eventually built. The German nuclear bomb had inspired much fear but was no more than a mirage. There was no German equivalent of the massive USA Manhattan Project.

In nature, successive evaporation leads to concentration of heavy water in isolated water bodies such as the Dead Sea in the Middle East and the Great Salt Lake in Utah, USA. The concentration process is due to the lower boiling point of light water relative to Heavy Water.

Water containing normal hydrogen is more easily disassociated into hydrogen and oxygen gases by an electric current than water containing deuterium. This allows the isotopes to be separated. In the USA, at the Savannah River Site (SRS) a now dismantled heavy water plant used the hydrogen sulfide-water exchange process to partially enrich heavy water. Deuterium was further concentrated by fractional distillation, and then by electrolysis. The moderator rework unit at SRS used fractional distillation to re-enrich reactor moderator that had become depleted in deuterium.



Figure 3. Norsk Hydro heavy water electrolysis production plant at Vemork, Norway.



Figure 4. Norsk Hydro 18 electrolysis cells at Vemork. Source: Nova.

# FRACTIONAL DISTILLATION

Water molecules containing deuterium atoms vaporize at a higher temperature than those without deuterium, so the boiling point of heavy water is slightly higher than that of normal water. Water vapor above a mixture of normal and heavy water will be slightly depleted in deuterium as a result, while the liquid will be slightly enriched. Enrichment results from successively boiling off and removing vapor containing normal hydrogen.



Figure 5. Savannah River heavy water plant's 130 foot tall distillation towers.

Industrially, a distillation process called the GS Dual-temperature Hydrogen Sulfide exchange method is used to produce heavy water. K. Geib and J. S. Spevack invented the process. However GS here stands for Girdler-Sulfide, where Girdler is the name of the company, which designed the first such plant in the USA. It uses a process whose rate depends on the different masses of the molecules involved.

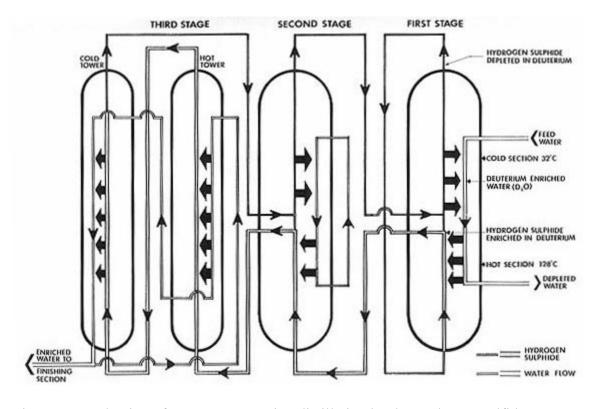


Figure 6. Production of Heavy Water using distillation by the Hydrogen sulfide process.

Since they possess the same number of electrons, the hydrogen and the deuterium isotopes are thought to be chemically identical. However, they do undergo chemical reactions at different rates according to their atomic masses.

The difference in the reaction rates increases with the difference between the isotopic masses. This is most pronounced in the case of hydrogen and deuterium, since the latter has twice the mass of the former. Deuterium is exchanged between streams of water and hydrogen sulfide gas, as shown in Fig. 6. An Ontario, Canada heavy water production plant is shown in Fig. 7. The Arak, Iran heavy water plant is shown under construction in Figs. 8, 9.



Figure 7. Ontario, Canada, Heavy Water production plant.



Figure 8. Arak, Iran heavy water plant under construction.



Figure 9. Arak, Iran heavy water production plant showing distillation towers, February 2005.

The water trickles through a series of perforated plates in a tower, while the gas flows upward through the perforations.

In this case a reversible reaction occurs:

$$H_2O + HDS \Leftrightarrow HDO + H_2S$$
 (5)

The equilibrium constant K for this reaction in terms of the concentrations, can be written as:

$$K = \frac{[HDO][H_2S]}{[H_2O][HDS]} = \frac{\frac{[HDO]}{[H_2O]}}{\frac{[HDS]}{[[H_2S]}}$$
(6)

If H and D were the same chemically, the equilibrium constant for the reaction would be equal to unity. In fact K is not equal to unity, and is furthermore temperature dependent:

$$K = 2.37$$
 at 25 °C,  
 $K = 1.84$  at 128 °C.

It becomes clear from the above equation that the concentration of HDO in H<sub>2</sub>O is greater than the concentration of HDS in H<sub>2</sub>S. Furthermore, the relative concentration of HDS in H<sub>2</sub>S increases with increasing temperature. This makes it possible to separate D from H.

At 32 <sup>o</sup>C, equilibrium favors the concentration of deuterium in water. However, at around 128 <sup>o</sup>C, the equilibrium favors the concentration of deuterium in the hydrogen sulfide.

The separation tower is divided into an upper cold section, where the concentration of deuterium in water is increased. This is then used as feed for the lower hot section, where the exchange leads to a further enrichment, this time in the hydrogen sulfide stream. The enriched gas from the hot section is fed to a second stage for further concentration. In the third stage, deuterium from the return stream of water is transferred to hydrogen sulfide in a cold tower, and enriched gas is transferred to water in the hot tower. Only a few stages are needed to produce heavy water at about 20 to 30 w/o D<sub>2</sub>O. Water is then fed to a vacuum distillation system to almost pure deuterium at a concentration of 99.75 percent, as needed by the CANDU reactor concept.

At the Bruce plant in Canada, the first stage towers are large structures. The first stage consists of 3 towers in parallel with a height of 90 meters and a diameter of 9 meters. Large amounts of power are needed: 300 MWth to produce 400 metric tonnes of heavy water per year.

Other possible processes would involve exchange between hydrogen and ammonia as gases, or amino methane and hydrogen gas. These processes would reduce the power requirements, with smaller size plants, and avoid corrosion problems typical of the hydrogen sulfide process. Exchange between water and hydrogen gas is possible if a suitable catalyst can be identified.

A common feature of both heavy water and light water reactors is the radiolysis or the decomposition of water under the effect of gamma radiation into hydrogen ions and free hydroxyl radicals:

$$\gamma + D_2O \rightarrow D + OD$$
 (7)

these can recombine back into heavy water in the first way as:

$$D + OD \rightarrow D_2O$$
 (8)

or in another way in pairs into deuterium gas and deuterium peroxide, when the specific ionization is high, as:

$$D + D \rightarrow D_2 \tag{9}$$

$$OD + OD \rightarrow D_2O_2 \tag{10}$$

The deuterium peroxide would then decompose leading to the release of substantial amounts of deuterium gas D<sub>2</sub>, and oxygen O<sub>2</sub>. Arrangements must be made for the removal of the deuterium gas since it constitutes an explosion hazard in the presence of oxygen. Oxygen itself must also be removed since it constitutes a corrosion hazard, by injection of hydrogen gas into the coolant to reduce the amount of oxygen, in both heavy water and light water cooled reactors.

Deuterium can also be produced using distillation from liquid hydrogen.

# 10.5 LITHIUM ENRICHMENT

Deuterium separated from heavy water is combined with enriched Li<sup>6</sup> to make ceramic-

like lithium<sup>6</sup> deuteride, Li<sup>6</sup>D parts for the secondary stages of thermonuclear weapons. A mixture of deuterium and tritium gases is injected into the pit of the primary fission stage of modern USA thermo nuclear weapons to boost the nuclear explosive yield by the effect of the 14.06 MeV neutrons from the fusion DT reaction.

The USA, UK, France, China and Israel and believed of having facilities for Li<sup>6</sup> separation in quantities sufficient for the manufacture and maintenance of boosted fission and thermonuclear weapons. Russia exploded a device using lithium deuteride powder Li<sup>6</sup>D before the USA did, even though the so-called sloika or layer cake design was not a true thermonuclear device scalable to any desired yield. The USA is reported to have stopped stockpiling Li<sup>6</sup> around 1963.



Figure 10. The Y-12 plant at Oak Ridge, Tennessee.

The Oak Ridge Y-12 Plant, which also included the magnetic separation process of  $U^{235}$  using Calutrons, began the initial effort to develop lithium isotope separation processes in 1950. Three processes were explored: the COLEX, ELEX, and OREX processes.

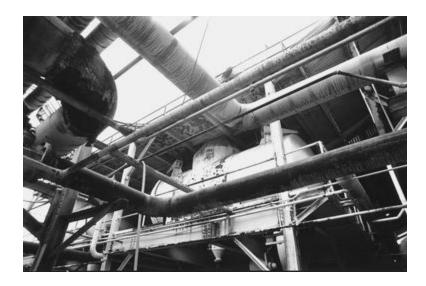


Figure 11. Lithium Column Exchange, COLEX process enrichment equipment. Isotopes of lithium are partially separated when transferring between an aqueous solution of lithium hydroxide and a lithium-mercury amalgam. The isotope Li<sup>6</sup> has a greater affinity for mercury than does the isotope Li<sup>7</sup>. A lithium and mercury amalgam is first prepared using natural lithium. The amalgam is then agitated in a natural lithium hydroxide solution. The rare Li<sup>6</sup> isotope concentrates in the amalgam and the more common Li<sup>7</sup> isotope migrates to the hydroxide solution. A counter-current flow of amalgam and hydroxide passes through a cascade of stages until the desired enrichment in Li<sup>6</sup> is reached.



Figure 12. Lithium solution tank used in the COLEX process. The Li<sup>6</sup> can be separated from the amalgam and the tailings fraction of Li<sup>7</sup> is electrolyzed from the aqueous solution of lithium hydroxide for further use.



Figure 13. Mercury flasks used in the Li<sup>6</sup> enrichment process.

The first successful laboratory separation was achieved with the ELEX process-an electrically driven chemical exchange process similar to that used in chlor alkali plants for the manufacture of chlorine gas and sodium hydroxide. The ELEX pilot plant was built at Y-12 in 1951. This plant was cleaned out and dismantled by 1959.

The mercury exchange process involves an amalgam of lithium and mercury which is made via electrolysis. Then the mercury lithium amalgam is mixed with an aqueous lithium hydroxide (LiOH) solution. Li<sup>6</sup> has a slightly higher affinity to mercury than Li<sup>7</sup>, so Li<sup>7</sup> diffuses out of the amalgam more quickly than Li<sup>6</sup>, leaving the amalgam with increased, or enriched, concentration of Li<sup>6</sup>. The Li<sup>6</sup> can be separated from the amalgam. The tailings fraction of Li<sup>7</sup> is electrolyzed from the aqueous solution of lithium hydroxide. Afterwards, the lithium hydroxide can be reused.

The OREX pilot plant was built by the Chemical Technology Division of Oak Ridge National Laboratory (ORNL) in 1952 and was operated in a sort of inter-plant competition with the Y-12 COLEX process to determine the better process. Both plants were operated by the Union Carbide Nuclear Corporation.

In the OREX process, an organic solution of lithium was exchanged with a solution of lithium in mercury or an amalgam in pulse columns using Propylene-di-amine (PDA) as the organic phase. The COLEX process won the competition, and the OREX pilot plant was subsequently dismantled between 1957 and 1959.

As a side note, an amalgam of mercury and silver was used for dental filling of decayed teeth. Due to the toxicity of the leaching of mercury, possibly leading to immune system responses, this approach has been abandoned in favor of ceramic fillings and existing amalgam fillings are being regularly replaced by the dentists at the request of their patients.

The COLEX process, an acronym for Column Exchange, is based on the fact that isotopes of lithium are partially separated when transferring between an aqueous solution of lithium hydroxide and a lithium-mercury amalgam. The isotope Li<sup>6</sup> has a greater affinity for mercury than does the isotope Li<sup>7</sup>. A lithium and mercury amalgam is first prepared using natural lithium. The amalgam is then agitated in a natural lithium hydroxide solution. The rare

Li<sup>6</sup> isotope concentrates in the amalgam and the more common Li<sup>7</sup> isotope migrates to the hydroxide solution. A counter current flow of amalgam and hydroxide passes through a cascade of stages until the desired enrichment in Li<sup>6</sup> is reached. The Li<sup>6</sup> can be separated from the amalgam and the tailings fraction of Li<sup>7</sup> is electrolyzed from the aqueous solution of lithium hydroxide for further use. The mercury is recovered and is mixed with a fresh supply for reuse.

Some anecdotal accounts are that the lithium medication on pharmacies shelves used for the treatment of schizophrenia is primarily of the purified Li<sup>7</sup> isotope.

A hoax may have been perpetrated about what was for a while called "red mercury" during the tenure of President Boris Yeltsin in Russia. Red mercury could have been the lithium mercury amalgam. It was attributed some unfounded capability of building so called "ballotechnic" weapons, possibly pure fusion weapons. A ballotechnic would refer to an explosive material that, when ignited, would generate enough energy, temperatures and pressures without exploding to compress a DT fusion charge without the need for a primary fission device. Claims were made that it is the Hg<sub>2</sub>Sb<sub>2</sub>O<sub>7</sub> compound.

The material was offered for sale to black market operatives attracted to its purported characteristics. Some who fell to the hoax were bilked out of their money, including some governments as well as the USA government who was curious about the claims and purchased a sample for analysis in government laboratories. The other face of the hoax, it is reported, was that it was a sting operation by the Russian authorities used to catch nuclear materials black market operatives and dispatch them to linger in Siberian prisons.

The COLEX process supplied most of the enriched lithium needed for the USA weapons complex. The Atomic Energy Commission, AEC built two large COLEX facilities, called Alpha-4 and Alpha-5 at the Y-12 Plant. Alpha-4 operated from January 1955 until 1963. The unit was placed on standby until it was dismantled in the late 1980s. Alpha-5 began operating in 1955. It was shut down in 1959 and restarted in 1963 for a six-month campaign. The Y-12 Plant engineers dismantled and disposed of the Alpha-5 COLEX process equipment in 1965 and 1966. Site contractors operated an open-air mercury receiving operation, where mercury flasks were emptied into a pipe leading to the COLEX plants. They used a furnace in a shed to roast sludges, wastes and other materials for mercury recovery.

Lithium enrichment has created a considerable amount of materials in inventory. The Department of Energy, DOE stores the lithium enrichment tailings, depleted in the Li<sup>6</sup> isotope, at the Portsmouth, Ohio enrichment plant and the K-25 Site at Oak Ridge, Tennessee. K-25 also stores a stockpile of unprocessed lithium. The Y-12 and K-25 sites both store the DOE's stockpile of enriched lithium.

The COLEX process employed approximately 24 million pounds of mercury. Most of the mercury used in the COLEX and ELEX processes was returned to the General Services Administration, GSA once it was no longer needed. However, a great deal of mercury was lost in wastes, spills, and through evaporation. A mercury-nitric acid purification system utilized in the COLEX process between 1955 and 1960 was the source of the major mercury bearing waste stream at Y-12. This system discharged a diluted, neutralized acid waste containing mercuric nitrate to the East Fork Poplar Creek. Mercury vapor from the plant was exhausted to the environment by the building ventilation system. Mercury from spills also contaminated basement sumps which were pumped through three concrete sedimentation tanks into the storm sewer and from there were pumped directly into the East Fork Poplar Creek. The DOE believes that small amounts of residual mercury are still present in the Y-12 plant sewers. Inorganic

mercury compounds of the type released at Y-12 plant were not initially believed to be toxic unless inhaled. It was not until 1970 that scientists discovered the biological methylation of inorganic mercury in the environment, which raised concerns over mercury discharges to surface water.

Approximately two million pounds of mercury used in the lithium enrichment processes have not been accounted for. Approximately 730,000 pounds or about 4,000 gallons of this material is believed to have been lost in waste streams, evaporation, and spills. A study done in 1983 estimated that evaporation during maintenance operations, seepage from pumps and other equipment, the venting of mercury vapors, and the smelting of mercury-contaminated scrap released 51,300 pounds of mercury into the air. The COLEX process discharged 239,000 pounds of mercury to the East Fork Poplar Creek in the process waste stream, some of which exists in sediments at the bottom of the New Hope Pond. The DOE believes that these waste discharges are also the source of some of the mercury contamination in Watts Barr Lake, Poplar Creek and the Clinch River. However, these bodies of water are also downstream from a commercial chlor-alkali plant. Residual mercury contamination at Y-12 includes sludges and mercury residue in building sewers and drain systems. A 1983 study also estimated that approximately 425,000 pounds of mercury were lost to the soil in eight accidental spills at the Y-12 Plant.

# 10.6 BORON ISOTOPE SEPARATION

Boron<sup>10</sup> is a powerful neutron absorber with many uses such as control rods, neutron detectors, boron cancer therapy and autocatalytic nuclear devices applications. The B<sup>10</sup> production process uses a dimethyl ether-boron trifluoride complex. The complex is fed into a distillation system. When the complex is boiled, part of the vapor phase breaks down into boron trifluoride and dimethyl ether. Boron trifluoride vapor molecules containing lighter B<sup>10</sup> atoms recombine into the liquid phase more rapidly than molecules containing the heavier B<sup>11</sup> isotope. As a result, the heavier isotope is concentrated in the vapor phase and the lighter isotope in the liquid phase.

To supply B<sup>10</sup>, the AEC built a plant in Model City, New York, near Niagara Falls. The plant operated from September 1954 until 1958, when the AEC placed it on standby. The Model City plant was rehabilitated in mid 1964 and restarted. First, the restarted plant converted the remaining inventory of B<sup>10</sup> from potassium fluoborate, KBF<sub>4</sub> to elemental boron to meet immediate weapon and reactor program demands. The plant continued to produce B<sup>10</sup>, until it was placed on standby again in March 1971. Since that time, the USA government has relied on commercial nuclear industry suppliers to convert its inventory of enriched B<sup>10</sup> to a powder form, and to supply additional B<sup>10</sup>.

#### 10.7 FISSILE AND FISSIONABLE FUELS

Nuclides that fission upon capture of a neutron in such a way that the added neutron itself is sufficient to overcome the fission barrier, even if it had zero kinetic energy, and induce fission are designated as fissile nuclei. Examples of fissile nuclei are:

$$U^{233}$$
,  $U^{235}$ ,  $Np^{237}$ ,  $Pu^{239}$  and  $Pu^{241}$ .

These are nuclei of even atomic number Z and odd mass number A.

Nuclides that fission with neutrons possessing a significant amount of kinetic energy or fast neutrons, are designated as fissionable nuclei. Examples of these even A isotopes are:

$$U^{232}$$
,  $U^{234}$ ,  $U^{236}$ ,  $U^{238}$ ,  $Th^{232}$ ,

as well as the fissile nuclei such as  $U^{235}$ .

It is possible to "breed" fissile nuclei from otherwise fissionable nuclei though the process of neutron capture. In this case a nucleus that is incapable of fissioning with "slow" neutrons can instead be transmuted into one that can.

Breeding can be achieved with fast neutrons with the fissionable isotope  $U^{238}$  to produce the fissile isotope  $Pu^{239}$  through the reactions:

$${}_{0}n^{1} + {}_{92}U^{238} \rightarrow {}_{92}U^{239} + \gamma$$

$${}_{92}U^{239} \rightarrow {}_{93}Np^{239} + {}_{-1}e^{0} + \nu^{*} + \gamma$$

$${}_{93}Np^{239} \rightarrow {}_{94}Pu^{239} + {}_{-1}e^{0} + \nu^{*} + \gamma$$

$${}_{0}n^{1} + {}_{92}U^{238} \rightarrow {}_{94}Pu^{239} + {}_{-1}e^{0} + 2 \nu^{*} + 3\gamma$$

$$(11)$$

Breeding can also be achieved with slow neutrons with the fissionable  $Th^{232}$  isotope to produce the fissile  $U^{233}$  isotope through the reactions:

$${}_{0}n^{1} + {}_{90}Th^{232} \rightarrow {}_{90}Th^{233} + \gamma$$

$${}_{90}Th^{233} \rightarrow {}_{91}Pa^{233} + {}_{1}e^{0} + \nu^{*} + \gamma$$

$${}_{91}Pa^{233} \rightarrow {}_{92}U^{233} + {}_{1}e^{0} + \nu^{*} + \gamma$$

$${}_{0}n^{1} + {}_{90}Th^{232} \rightarrow {}_{92}U^{233} + 2{}_{1}e^{0} + 2\nu^{*} + 3\gamma$$

$$(12)$$

# **10.8 URANIUM MINING**

Uranium is mined in different locations in the USA and Canada in open pit or underground mines. Two particularly rich mining districts exist in New Mexico and in Colorado. Underground mining poses a health hazard to the miners in the process of inhalation of the radon<sup>222</sup> gas, and its solid daughters in mine dust. Radon<sup>222</sup> is itself a daughter nuclide in the uranium<sup>238</sup> decay chain. Adequate mine ventilation is necessary to ensure the safety of the mining process.

The Grants Mineral Belt uranium was discovered at Poison Canyon by sheepherder Paddy Martinez in 1951. His sheep were dying and he thought it was caused by radioactivity in uranium. The fact is that the area had abundant selenium-bearing loco weed in the area, hence the name: Poison Canyon. Selenium occurs in conjunction of uranium, and loco weed absorbed

it, and becomes a marker for the location of uranium ore.



Figure 14. Haysrack Mountain in the uranium mining district of New Mexico.



Figure 15. McArthur River Cameco underground uranium mine in Northern Saskatchewan, Canada.



Figure 16. Jackpile open pit uranium mine, Grants, New Mexico. The Grants Mineral Belt is the world's second largest historic uranium producer despite production cessation after the collapse in uranium prices and the collapse of the USA's uranium industry in the 1980's following the 1979 Three Mile Island accident.



Figure 17. Rossing open pit mine, Namibia.

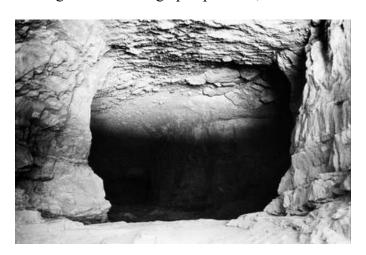


Figure 18. Underground uranium mine, Grants, New Mexico.

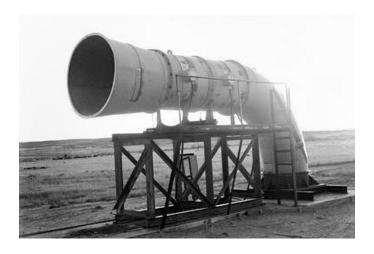


Figure 19. Radon gas ventilation shaft at the Ambrosia Lake uranium mine, Grants, New Mexico.



Figure 20. Reclaimed open pit and underground Poison Canyon uranium mine site, Ambrosia Lake area north of Grants, New Mexico. Host rock is sandstone from the late Jurassic Morrison Formation, 140-150 million years before present. Source: Michael S. Fulp.



Figure 21. Uranium mineralization in petrified tree logs exposed by a flash flood is sought by geologists and prospectors to locate uranium ore bodies. Petrified log at Poison Canyon, Ambrosia Lake, New Mexico. The yellow-green secondary uranium minerals appear on the weathered surface. Source: Michael S. Fulp.

# 10.9 URANIUM CONVERSION AND REFINING

Uranium leaves the mine as the concentrate of a stable oxide known as  $U_3O_8$  or as a peroxide. A typical commercial ore contains approximately 0.3 percent in  $U_3O_8$ . The uranium undergoes a milling and extraction process solvent extraction leaching process concentrating  $U_3O_8$  which is now designated as the yellow cake with a 65 to 85 percent content of  $U_3O_8$ .

The yellow cake is dissolved in nitric acid yielding a uranyl nitrate UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> liquid solution. This is purified with a selective solvent extraction process. The purified solution is treated with ammonium hydroxide yielding a precipitate of ammonium diuranate (NH<sub>4</sub>)<sub>2</sub>U<sub>2</sub>O<sub>7</sub>. The different forms of these compounds are shown in Fig. 24.

Upon calcinations and reduction with hydrogen, the UO<sub>2</sub> brown oxide is obtained. Ingots of the uranium metal can now be obtained by reduction with magnesium or calcium in electrical induction furnaces (Fig. 25). These can be alloyed with different elements, extruded or machined for use as a natural uranium metallic reactor fuel (Fig. 26).

For the production of enriched fuel, hydro fluorination with hydrofluoric acid HF of the brown oxide yields uranium tetrafluoride UF<sub>4</sub> powder. In a fluorination step treatment in a fluidized bed with fluorine gas F<sub>2</sub>, one obtains uranium hexafluoride UF<sub>6</sub> as a feed to the enrichment process.

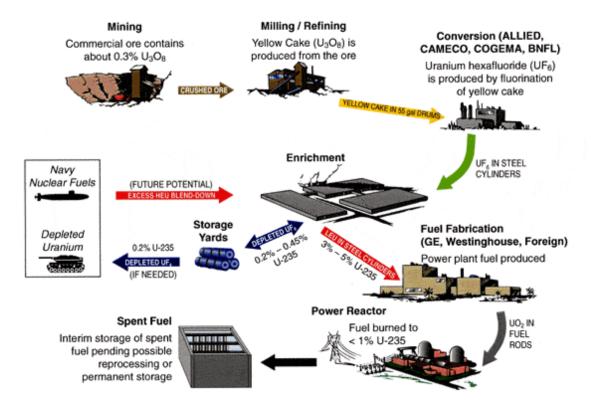


Figure 22. The uranium mining and conversion cycle.

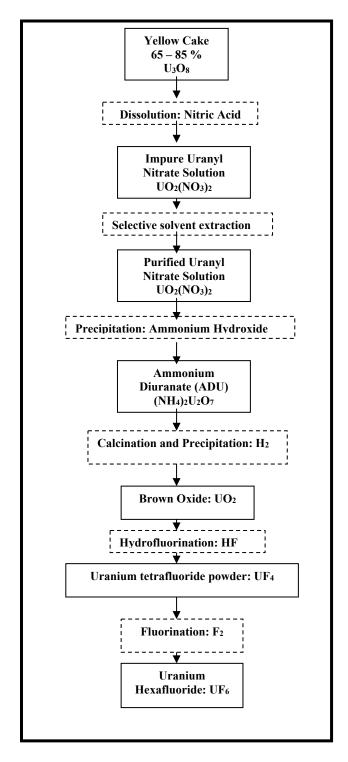


Figure 23. The uranium refining process.



Figure 24. Uranium mill at Key Lake, Northern Saskatchewan, Canada. Source: Cameco.



Figure 25. Uranium refinery at Blind River, Ontario, Canada. Source: Cameco.

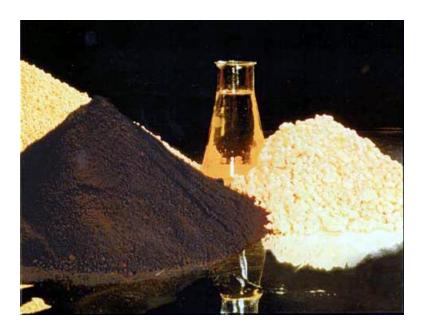


Figure 26. Yellow cake, U<sub>3</sub>O<sub>8</sub>, Uranyl Nitrate liquid UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>, Ammonium Diuranate, (NH<sub>4</sub>)<sub>2</sub>U<sub>2</sub>O<sub>7</sub>, and uranium dioxide UO<sub>2</sub>.



Figure 27. Separated yellow cake, U<sub>3</sub>O<sub>8</sub>.

Uranium hexafluoride: UF<sub>6</sub>, commonly referred to as "hex" is stored in cylinders made of the Monel alloy, nickel or steel, shown in Fig. 27. At room temperature it forms crystals that look like rock salt (Fig. 28).



Figure 28. Induction furnaces for the magnesium reduction of uranium metal.



Figure 29. Billets of depleted uranium weighing 1,100 lbs each.



Figure 30. Inspection of UF<sub>6</sub> storage cylinders.



Figure 31. UF<sub>6</sub> crystals in a glass vial.

After initial refining, which may involve the production of uranyl nitrate, uranium trioxide is reduced in a kiln by hydrogen to uranium dioxide: UO<sub>2</sub>. This is then reacted in another kiln with hydrogen fluoride HF to form uranium tetrafluoride UF<sub>4</sub>, a powder. The tetrafluoride is then fed into a fluidized bed reactor with gaseous fluorine to produce UF<sub>6</sub>. Removal of impurities takes place at each step.

An alternative wet process involves making the UF<sub>4</sub> from UO<sub>2</sub> by a wet process, using aqueous HF.



Figure 32. Enriched U<sup>235</sup> buttons.

Uranium hexafluoride, UF<sub>6</sub>, particularly if moist, is highly corrosive. When warm, it is a gas, suitable for use in the enrichment process. At lower temperature and under moderate pressure, the UF<sub>6</sub> can be liquefied. The liquid is run into specially designed steel shipping cylinders which are thick walled and weigh over 15 metric tonnes when full. As it cools, the liquid UF<sub>6</sub> within the cylinder becomes a white crystalline solid (Fig. 28) and is shipped in this form.

The siting, environmental and security management of a conversion plant is subject to the regulations that are in effect for any chemical processing plant involving fluorine-based

chemicals.

The enriched uranium undergoes a metallurgical process in which it is turned into metal buttons (Fig. 29), which could be used as a metal or converted to the fuel form desired such as uranium dioxide, UO<sub>2</sub>.

# 10.10 NUCLEAR FUEL CYCLES

Three types of fuel cycles are considered for fission power plants, depending on whether the fuel is recycled and on the type of reactor used for electrical generation as shown in Fig. 30.

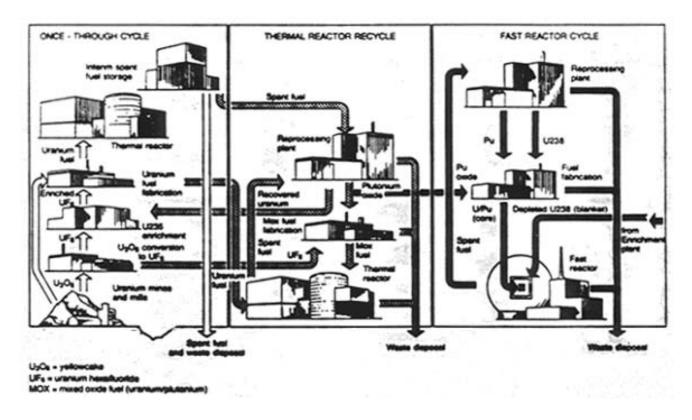


Figure 33. The once through, thermal reactor recycle and the fast reactor fuel cycles.

In the *once-through fuel cycle*, the spent fuel is not reprocessed but kept in storage in water pools at the reactor sites, until it is eventually reprocessed or stored as waste with a contingent of the ability of being able to retrieve if ever needed. This is the fuel cycle currently adopted in most currently operating reactors. It entails a waste of resources since the fuel is disposed for primarily structural and metallurgical reasons, not for the depletion of the fuel in it. The abbreviated overall reactions are:

$${}_{0}\mathbf{n}^{1} + {}_{92}\mathbf{U}^{238} \rightarrow {}_{92}\mathbf{U}^{239} + \gamma$$

$${}_{92}\mathbf{U}^{239} \rightarrow {}_{93}\mathbf{N}\mathbf{p}^{239} + {}_{-1}\mathbf{e}^{0} + \nu^{*} + \gamma$$

$${}_{93}\mathbf{N}\mathbf{p}^{239} \rightarrow {}_{94}\mathbf{P}\mathbf{u}^{239} + {}_{-1}\mathbf{e}^{0} + \nu^{*} + \gamma$$

$${}_{94}\mathbf{P}\mathbf{u}^{239} \rightarrow {}_{92}\mathbf{U}^{235} + {}_{2}\mathbf{H}\mathbf{e}^{4} + \gamma$$

$$(13)$$

$${}_{0}\mathbf{n}^{1} + {}_{92}\mathbf{U}^{238} \rightarrow {}_{92}\mathbf{U}^{235} + {}_{2}\mathbf{H}\mathbf{e}^{4} + 2{}_{-1}\mathbf{e}^{0} + 2 \ \nu^{*} + 4\gamma$$

This shows that the produced  $Pu^{239}$  decays into  $U^{235}$ . The mined  $U^{238}$  is ultimately converted into  $U^{235}$  in this cycle, in addition to the generated fission products and alpha particles.

In the *thermal reactor recycle*, the spent fuel is reprocessed and the uranium and plutonium isotopes are separated from the fission products. These are recycled in new fuel elements in the form of the Mixed Oxide fuel: MOX composed of UO<sub>2</sub> and PuO<sub>2</sub>. It is also possible to recycle only the uranium and to store the plutonium, or vice versa. This fuel cycle entails a better use of resources, and the disposal is simplified to the volume of the fission products, instead of the whole fuel elements.

In the *fast reactor breeder* cycle, the uranium and plutonium in the spent fuel are separated. They are then refabricated into new fuel elements. In fast breeder reactors, a blanket of depleted U<sup>238</sup> intercepting the otherwise leaking neutrons and breeding plutonium, surrounds a central core of uranium and plutonium fuel. Breeder reactors can produce more fuel than they consume, hence the name: breeder.

# 10.11 FUEL BURNUP AND CONSUMPTION RATES

For a reactor operating at a power of P<sub>th</sub> thermal megawatts (MWth), and assuming a recoverable energy per fission event of 190 MeV/fission, excluding the unavailable 10 MeV of energy carried by the antineutrinos, one can write for the fission rate dF/dt:

$$\frac{dF}{dt} = P_{th} [MWth] .10^{6} \left[ \frac{Watt}{MWth} \right] \left[ \frac{(Joules/sec)}{Watt} \right] . \frac{1}{190} \left[ \frac{fission}{MeV} \right] . \frac{1}{1.6 \times 10^{-13}} \left[ \frac{MeV}{Joule} \right] .86,400 \left[ \frac{sec}{day} \right]$$

$$= 2.84 \times 10^{21} P_{th} \left[ \frac{fissions}{day} \right]$$
(14)

Using Avogadro's law we can express the number of fissions N in g grams of U-235:

$$N = \frac{g A_{v}}{M} \tag{15}$$

where A<sub>v</sub> is Avogadro's number,

M is the molecular weight of  $U^{235} = 235$  amu,

g is the mass in grams,

we can convert the fission rate into the burnup rate, BR, where:

$$BR = \frac{dF}{dt} \cdot \frac{g}{N}$$

$$= 2.84 \times 10^{21} P_{th} \left[ \frac{fissions}{day} \right] \cdot \frac{M}{A_{v}} \left[ \frac{gm U^{235}}{fissions} \right]$$

$$= 2.84 \times 10^{21} P_{th} \frac{235}{0.6 \times 10^{24}}$$

$$= 1.112 P_{th} \left[ \frac{gm U^{235}}{day} \right]$$
(16)

Thus, a reactor operating at a power of 3,000 MWth, will have a burnup rate of about  $3,336 \text{ gms } U^{235}/\text{day}$ .

Actually, not all the U<sup>235</sup> nuclei present in the reactor undergo fission, some will absorb the neutrons through the radiative capture process, where a neutron is absorbed and a gamma photon is emitted.

If the ratio of radiative captures to fission reactions is given by:

$$b = \frac{\sigma_{\gamma}}{\sigma_{f}} \tag{17}$$

where b = 0.169 for  $U^{235}$ , the total number of absorptions per fission is given by:

$$(1+b) = \frac{\sigma_f + \sigma_{\gamma}}{\sigma_f} = \frac{\sigma_a}{\sigma_f}$$
 (18)

where  $\sigma_{\gamma}$  is the microscopic radiative capture cross section [barns],

 $\sigma_f$  is the microscopic fission cross section [barns],

 $\sigma_a$  is the microscopic absorption cross section [barns].

Consequently, the consumption rate, CR is larger than the burnup rate by the factor (1+b):

$$CR = 1.112 (1+b) P_{th} [gm U^{235}/day].$$
 (19)

Substituting the value of b for  $U^{235}$ , we find that for thermal neutrons fissions,  $U^{235}$  is consumed at the rate of:

$$CR = 1.112 (1+0.169) P_{th} [gm U^{235}/day],$$
  
= 1.299  $P_{th} [gm U^{235}/day].$  (20)

Thus 1.299 grams of U<sup>235</sup> per day are consumed per one MWth of reactor power.

# 10.12 MASS BALANCE IN THE ENRICHMENT PROCESS

An important consideration in the fuel cycle is for a utility to contract for a given amount of feed material to obtain a certain amount of enriched fuel needed for the operation of the plant (Fig. 34). The yearly mass CR of U<sup>235</sup> consumption from Eqn. 19 is given by:

$$CR = 365 \times 1.112 (1+b) P_{th} \left[ \frac{gm U^{235}}{year} \right]$$
 (21)

The amount of the natural uranium consumed per year is:

$$M_{\rm f} = \frac{\rm CR}{\rm x_{\rm f}} = \frac{365[1.112 (1+b) P_{\rm th}]}{\rm x_{\rm f}} \quad \left[\frac{\rm gm \ nat \ U}{\rm year}\right]$$
 (22)

where  $x_f\!\!=\!\!0.72$  percent is the enrichment of  $U^{235}$  in natural uranium.

This amount of natural uranium would be contracted-for with a uranium mine for a natural uranium reactor such as the CANDU heavy-water design.

For reactors using enriched fuel such as research reactors, light water reactors, gas cooled reactors and fast neutron breeder reactors, it is interesting to estimate the amount of natural uranium feed-material  $M_f$  that should be contracted for with a uranium mine for power plant operating at a thermal power  $P_{th}$ .



Figure 34. Mass balance in an enrichment plant.

Notice that the product mass being  $M_p$ , the amount of  $U^{235}$  in the product is consequently equal to:  $x_pM_p$ . The product material in this case is given by:

$$M_{p} = \frac{CR}{x_{p}} = \frac{365[1.112 (1+b) P_{th}]}{x_{p}} [\frac{gm enr U}{year}]$$

Assuming that there are no losses in the enrichment process, a mass balance for the feed material as uranium entering and exiting the plant can be written as:

$$M_{f} = M_{p} + M_{T} \tag{23}$$

Similarly, a mass balance can be conducted for the U<sup>235</sup> isotope as:

$$X_{f}.M_{f} = X_{p}.M_{p} + X_{T}M_{T}$$
 (24)

where  $x_i$  stands for the enrichment for the feed (f) as natural uranium, product (p) enriched fuel, and depleted uranium tailings (T).

The two equations 23 and 24 can be solved for the two unknowns  $M_f$  and  $M_T$  through the variables elimination method.

Substituting for the tails mass from the first equation as:

$$M_{T} = M_{f} - M_{p}$$

into the second equation, we get:

$$\mathbf{x}_{\mathrm{f}}.\ \mathbf{M}_{\mathrm{f}} = \mathbf{x}_{\mathrm{p}}.\ \mathbf{M}_{\mathrm{p}} + \mathbf{x}_{\mathrm{T}}\ [\mathbf{M}_{\mathrm{f}}\ - \mathbf{M}_{\mathrm{p}}]$$

$$= \mathbf{x}_{\mathrm{p}}.\ \mathbf{M}_{\mathrm{p}} + \mathbf{x}_{\mathrm{T}}.\mathbf{M}_{\mathrm{f}}\ - \mathbf{x}_{\mathrm{T}}.\ \mathbf{M}_{\mathrm{p}}$$

Rearranging, we get:

$$[x_f - x_T]. M_f = [x_p - x_T]. M_p,$$
 (25)

From this we can write for the needed mass of feed material in terms of the enrichments of the product, the feed and the tails:

$$M_{f} = \frac{[x_{p} - x_{T}]}{[x_{f} - x_{T}]} M_{p}$$
 (26)

The enrichment of natural uranium feed is  $x_f = 0.72$  percent. The enrichment of the tails,  $x_T$ , is normally at 0.2 percent in weight.

#### **EXAMPLE**

An executive at an electrical utility company needs to order uranium fuel from a mine. The utility operates a single 1000 MWe power plant of the CANDU type using natural uranium, and operating at an overall thermal efficiency of 33.33 percent. What is the yearly amount of:

- a. U<sup>235</sup> burned up by the reactor?
- b. U<sup>235</sup> consumed by the reactor?
- c. Natural uranium that the executive has to contract with the mine as feed to his nuclear unit?

$$P_{th} = \frac{P_e}{\eta_{th}} = \frac{1,000MWe}{1/3} = 3,000MWth$$

BR= 1.112 
$$P_{th} \left[ \frac{gm \ U^{235}}{day} \right]$$

CR = 365×1.112 (1+b) 
$$P_{th} \left[ \frac{gm \ U^{235}}{year} \right]$$
  
= 365×1.112 (1+0.169) 3,000  $\left[ \frac{gm \ U^{235}}{year} \right] \frac{1}{10^6} \frac{mt}{gm}$   
= 1.423  $\frac{mtU^{235}}{year}$ 

$$M_{\rm f} = \frac{\text{CR}}{\text{x}_{\rm f}} = \frac{365[1.112 \text{ (1+b) } P_{\rm th}]}{\text{x}_{\rm f}} \left[ \frac{\text{gm nat } U}{\text{year}} \right]$$
$$= \frac{1.423}{0.72/100} = 197.63 \frac{\text{mt nat} U}{\text{year}}$$

#### **EXAMPLE**

An executive at an electrical utility company needs to order uranium fuel from a mine. This utility operates a single 1,000 MWe Pressurized Water Reactor PWR power plant operating at an overall thermal efficiency of 33.33 percent.

The fuel needs to be enriched to the 5 w/o level in  $U^{235}$ .

Consider that the enrichment plant generates tailings at the  $0.2\ \text{w/o}$  in  $U^{235}$  level.

Calculate the yearly amount of natural uranium metal in metric tonnes that the executive has to contract with the mine as feed to the enrichment plant, then to his nuclear unit.

CR = 365×1.112 (1+b) 
$$P_{th} \left[ \frac{\text{gm U}^{235}}{\text{year}} \right]$$
  
= 365×1.112 (1+0.169) 3,000  $\left[ \frac{\text{gm U}^{235}}{\text{year}} \right] \frac{1}{10^6} \frac{mt}{gm}$   
= 1.423  $\frac{mtU^{235}}{year}$ 

This is the same amount of  $U^{235}$  as the Candu reactor to produce the same amount of power.

$$M_p = \frac{\text{CR}}{x_p} = \frac{365[1.112 \text{ (1+b) P}_{th}]}{x_p} \left[ \frac{\text{gm enr U}}{\text{year}} \right]$$
$$= \frac{1.423}{5/100} = 28.46 \frac{\text{mt enr U}}{\text{year}}$$

This is a smaller amount than the amount fed into the Candu option, hence not requiring online refueling.

$$M_{f} = \frac{[x_{p} - x_{T}]}{[x_{f} - x_{T}]}.M_{p}$$

$$= \frac{5 - 0.2}{0.72 - 0.2}28.46 = 262.70 \frac{mt \, nat U}{year}$$

This is a larger amount of  $U^{235}$  than the Candu reactor because of the amount of  $U^{235}$  left in the tailings.

# 10.13 SEPARATIVE WORK UNIT (SWU)

The capacity of uranium enrichment plants is measured in terms of the Separative Work Units or SWUs. The SWU is a complex unit which is a function of the amount of uranium processed and the degree to which it is enriched, or the extent of increase in the concentration of the U<sup>235</sup> isotope relative to the remainder, and the level of depletion of the remainder. The unit is specifically expressed as "Kilogram Separative Work Unit," and it measures the quantity of separative work performed to enrich a given amount of uranium a certain amount. It is thus indicative of energy used in enrichment when feed and product quantities are expressed in kilograms. The unit "Metric tonnes SWU" is also used.

As an example, to produce one kilogram of uranium enriched to 3 percent  $U^{235}$  requires 3.8 SWU if the plant is operated at a tails assay of 0.25 percent, or 5.0 SWU if the tails assay is 0.15 percent, thereby requiring only 5.1 kg instead of 6.0 kg of natural uranium feed.

About 100,000-120,000 SWU is required to enrich the annual fuel loading for a typical 1,000 MWe light water reactor. Enrichment costs are substantially related to electrical energy used. The gaseous diffusion process consumes about 2,500 kWhr or 9,000 MJ per SWU, while

modern gas centrifuge plants require only about 50 kWhr or 180 MJ per SWU.

Enrichment accounts for almost half of the cost of nuclear fuel and about 5 percent of the total cost of the electricity generated. It also accounts for the main greenhouse gas impact from the nuclear fuel cycle if the electricity used for enrichment is generated from coal. However, it still only amounts to 0.1 percent of the carbon dioxide from equivalent coal fired electricity generation if modern gas centrifuge plants are used, or up to 3 percent in a worst case situation.

# 10.14 THERMAL DIFFUSION

The kinetic theory of gases predicts the differences in the rates of diffusion of gases of different molecular weights. In this process, a liquid compound rises as it heats, falls as it cools, and tends to separate into its lighter and heavier components as it cycles around a column.

If there is a temperature gradient in a mixed gas, there is a tendency for one type of molecule to concentrate in the cold region and the other in the hot region. This tendency depends on the molecular weights as well as on the forces between the molecules. If the gas is a mixture of two isotopes, the heavier isotope may accumulate at the hot region or the cold region or not at all, depending on the nature of the intermolecular forces. The direction of separation may reverse as the temperature or relative concentration is changed.

H. Clausius and G. Dickel in 1938, in Germany, built a vertical tube containing a heated wire stretched along the axis of the tube and producing a temperature difference of about 600 °C between the axis and the periphery. The heavy isotopes became concentrated near the cool outer wall. In addition, the cool gas on the outside tended to sink while the hot gas at the exit tended to rise. Thermal convection thus set up a counter current flow, and thermal diffusion caused the preferential flow of the heavy molecules outward across the interface between the two currents.

Applied to uranium enrichment, a gaseous uranium compound such as uranium hexafluoride is circulated in an annular region between two vertical pipes kept at different temperatures. The lighter molecules of  $U^{235}F_6$  and  $U^{234}F_6$  end to get more concentrated near the hot surface where they are carried upwards by the convection current. An exchange occurs with the current moving downwards along the cold surface producing a fractionation process. After a state of equilibrium is reached, the gas near the upper end contains more of the light molecules than near the lower end.

A system of two concentric tubes of 2 mm separation, 3 cm in diameter, and 150 cms long, would produce a difference of 40 percent in the concentration of the rare isotopes between its ends, and about 1 gm/day could be drawn from the upper end without upsetting the equilibrium condition.

To produce large amounts of enriched  $U^{235}$ , a large number of these units must be used in parallel and in series. To produce 1 gm/day of 90 percent enriched  $U^{235}$  would require about 1,000 tubes.

In the thermal diffusion rack shown in Fig. 32, steam circulates through an inner pipe and cooling water through an outer pipe surrounding it. This caused U<sup>235</sup> to diffuse inward and circulate upward. The S-50 thermal diffusion plant shown in Fig. 33 was built during the Manhattan project at Oak Ridge Tennessee. The resulting slightly enriched material could be later fed to other enrichment devices such as the magnetic separation Calutrons.

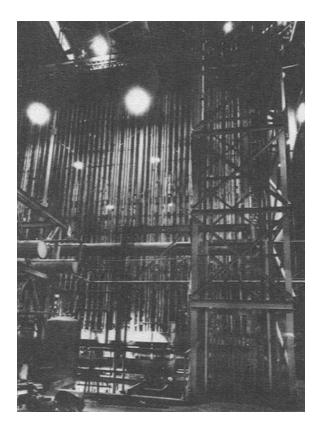


Figure 35. Liquid Thermal Diffusion Rack.

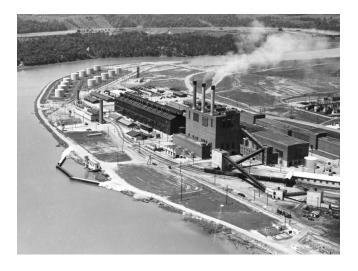


Figure 36. The S-50 Thermal Diffusion Plant, Oak Ridge, Tennessee.

# 10.15 ELECTROMAGNETIC SEPARATION

Magnetic separation depends on the principle of the mass spectrometer. A mixture of the ions that are being separated is generated at an ion source. Upon emerging from the source, the

ions are accelerated through a potential difference V, maintained by an electric field. The positive ions carrying a charge q, acquire a kinetic energy equal to qV:

$$qV = \frac{1}{2}M_{235}v_{235}^2$$
, for the lighter U<sup>235</sup> isotope, (27)

and:

$$qV = \frac{1}{2}M_{238}v_{238}^2$$
, for the heavier U<sup>238</sup> isotope. (28)

The velocities acquired by the isotopes depend on their masses as:

$$\mathbf{v}_{235} = \left(\frac{2qV}{M_{235}}\right)^{1/2} \tag{29}$$

and:

$$\mathbf{v}_{238} = \left(\frac{2qV}{M_{238}}\right)^{1/2} \tag{30}$$

Lorentz equation governs the behavior of a charged particle in an electromagnetic field as:

$$\overline{F} = q\overline{E} + q\,\overline{\mathbf{v}}\mathbf{x}\overline{B}$$

 $\overline{v}$  is the velocity vector

 $\overline{\mathbf{B}}$  is the magnetic field vector

where:

 $\overline{E}$  is the electric field vector

x denotes the cross vector product

For a magnetic field presence only without an electric field:

$$\overline{F} = q \, \overline{\mathbf{v}} \mathbf{x} \overline{B}$$

This suggests that a charged particle injected in a magnetic field is subjected to a force that is perpendicular to both the magnetic field vector as well as the particle's velocity vector, resulting in a rotational motion around the magnetic field lines. Equating the centrifugal force to the magnetic force, using the magnitudes of the vectors, we get:

$$F = q vB = \frac{Mv^2}{R}$$

If the magnetic field B, generated by cylindrical coils in which a current is circulated, is perpendicular to the plane of the page, the ions will be forced to move in a circular path defined by the centrifugal force relationship:

$$Bqv_{235} = M_{235} \frac{v_{235}^2}{R_{235}}$$
, for the lighter U<sup>235</sup> isotope, (31)

and:

$$Bqv_{238} = M_{238} \frac{v_{238}^2}{R_{238}}$$
, for the heavier U<sup>238</sup> isotope, (32)

where  $R_{235}$  and  $R_{238}$  are the circular radii of the path taken. We can write expressions for the ensuing radii as:

$$R_{235} = \frac{M_{235} V_{235}}{Bq} = \frac{M_{235}}{Bq} \left(\frac{2qV}{M_{235}}\right)^{\frac{1}{2}} = \frac{\left(2qVM_{235}\right)^{\frac{1}{2}}}{Bq},$$
 (32)

and:

$$R_{238} = \frac{M_{238} V_{238}}{Bq} = \frac{M_{238}}{Bq} \left(\frac{2qV}{M_{238}}\right)^{\frac{1}{2}} = \frac{\left(2qVM_{238}\right)^{\frac{1}{2}}}{Bq}.$$
 (33)

There is a difference in the radii:

$$\delta = R_{238} - R_{235} = \frac{\left(2qVM_{238}\right)^{1/2}}{Bq} - \frac{\left(2qVM_{235}\right)^{1/2}}{Bq}$$
$$= \frac{\left(\frac{2V}{q}\right)^{1/2}}{B} (M_{238}^{1/2} - M_{235}^{1/2})$$

The ratio of the radii can also be written as:

$$\frac{R_{235}}{R_{238}} = \sqrt{\frac{M_{235}}{M_{238}}} \tag{34}$$

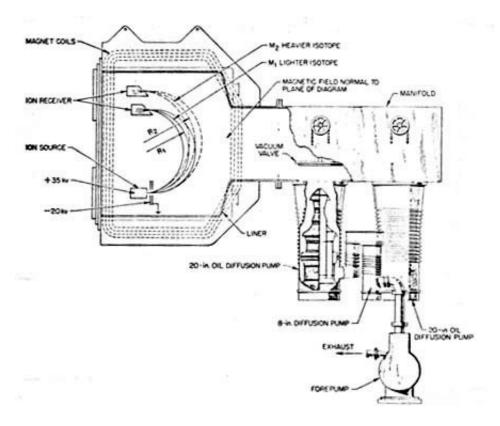


Figure 37. An 180-degrees magnetic separation device, showing the magnets and the vacuum system.

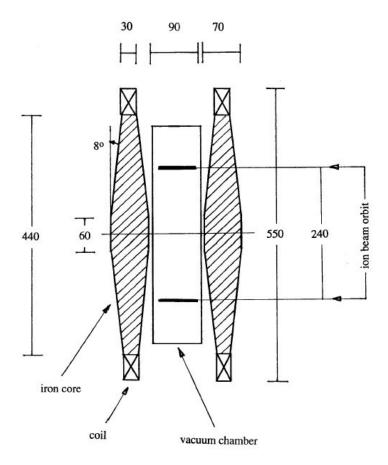


Figure 38. Two magnets surrounding a single collection chamber in an alpha track 255 degrees Calutron with a non-uniform 1/r magnetic field.

This shows that the isotopes of different masses will move in trajectories with different radii. These radii are proportional to the square root of the masses. Collectors can be placed at these radii to collect the different nuclei. These devices can generate only gram quantities of isotopes. That is why other methods like gaseous diffusion and centrifugation had to be developed.

In the magnetic separators or Calutrons (for Califiornia cyclotrons), uranium chloride UCl<sub>4</sub> is reported to have been used as a feed material. It oxidizes when exposed to air, creating a processing problem.

Figure 34 is a diagram of a beta Calutron separator, including the ion source, accelerating system, receiver, vacuum system and semicircular focusing ion paths in the magnetic field. In more advanced Calutrons, the magnets with an iron core are designed to generate a non uniform magnetic field with a radial dependence:

$$B(r) \alpha_{\ell} \frac{1}{r}$$

resulting in a uniform magnetic field in the space between them where the vacuum chambers and collection system are emplaced.

## 10.16 GASEOUS DIFFUSION

Commercial uranium enrichment was first carried out by the diffusion process in the USA. It has since been used in Russia, UK, France, China and Argentina as well. Today only the USA and France use the process on any significant scale. The remaining large USEC plant in the USA was originally developed for weapons programs and has a capacity of some 8 million SWU per year. At Tricastin, in Southern France, a more modern diffusion plant with a capacity of 10.8 million kg SWU per year has been operating since 1979. This plant can produce enough 3.7 percent enriched uranium a year to fuel some ninety 1,000 MWe nuclear reactors.

The gaseous diffusion process accounts for about 40 percent of world enrichment capacity. However, though they have proved durable and reliable, most gaseous diffusion plants are now nearing the end of their design life and the focus is on centrifuge enrichment technology which seems to be replacing them.

The diffusion process involves forcing uranium hexafluoride gas under pressure through a series of porous membranes or diaphragms. As  $U^{235}F_6$  molecules are lighter than the  $U^{238}F_6$  molecules they move faster and have a slightly better chance of passing through the pores in the membrane, possibly manufactured from sintered powdered nickel. The UF<sub>6</sub> which diffuses through the membrane is thus slightly enriched, while the gas which did not pass through is depleted in  $U^{235}$ .

This process is repeated many times in a series of diffusion stages called a cascade. Each stage consists of a compressor, a diffuser and a heat exchanger to remove the heat of compression. The enriched UF<sub>6</sub> product is withdrawn from one end of the cascade and the depleted UF<sub>6</sub> is removed at the other end. The gas must be processed through some 1,400 stages to obtain a product with a concentration of 3 percent to 4 percent U<sup>235</sup>.

It is not useful to try to isolate the isotopes of uranium in metallic form since uranium would rapidly oxidize. In the gaseous diffusion process for uranium enrichment, the gaseous uranium hexafluoride of the mixture of  $U^{234}$ ,  $U^{235}$  and  $U^{238}$  is used:

$$U^{234}F_6 + U^{235}F_6 + U^{238}F_6$$
.

Fluorine is selected since it occurs in nature with 100 percent atomic abundance in the isotope  $F^{19}$ . The lighter molecules flow slightly faster than the heavier molecules across the wall of a porous tube barrier. The portion of the gas passing through the barrier is slightly richer in  $U^{235}$ . The small fraction of  $U^{234}$  ends up with the  $U^{235}$  stream.

Uranium hexafluoride has a vapor pressure of 1 atmosphere at a temperature of 56 degrees C.

The separation factor, also known as the enrichment or fractionation factor of a process, is the ratio of the relative concentration of the desired isotope after processing to its relative concentration before reprocessing. If before and after processing the number of atoms of isotopes 1 and 2 are defined as:

 $b_1$  = number of atoms of isotope 1 per gram of the isotope mixture before processing,

 $b_2$  = number of atoms of isotope 2 per gram of the isotope mixture before processing,  $a_1$  = number of atoms of isotope 1 per gram of the isotope mixture after processing,

 $a_2$  = number of atoms of isotope 2 per gram of the isotope mixture after processing,

then, we can define the separation factor r as:

$$r = \frac{\left(\frac{a_1}{a_2}\right)}{\left(\frac{b_1}{b_2}\right)}.$$
(35)

This definition applies to one stage of a separation plant or to an entire plant consisting of many stages. One is interested usually in the either the *single stage* separation factor, or the *overall* separation factor of the whole process. For a single stage, r is usually slightly larger than unity, and we define the *enrichment factor* as:

$$\varepsilon = r - 1 = \frac{\left(\frac{a_1}{a_2}\right)}{\left(\frac{b_1}{b_2}\right)} - 1 \tag{36}$$

In natural uranium the mass of isotope 1 is 235 and the mass of isotope 2 is 238, and:

$$\left(\frac{b_1}{b_2}\right) = 0.72 \text{ percent} = 0.0072 = \frac{1}{140}.$$
 (37)

If we want to enrich the uranium to 90 percent in the isotope 235,

$$\left(\frac{a_1}{a_2}\right) = 90 \text{ percent} = 0.90 = \frac{90}{100}.$$

Consequently, a process producing 90 percent enriched uranium would need to produce a value of r equal to:

$$r = \frac{\left(\frac{90}{100}\right)}{\left(\frac{1}{140}\right)} = 126.0$$

In nearly every process, a high separation factor is associated with a low yield. This requires an optimization of the process. A separation device with a separation factor of 2,

$$r=2=r=\frac{\left(\frac{a_1}{a_2}\right)}{\left(\frac{1}{140}\right)},$$

implies a value of:

$$\left(\frac{a_1}{a_2}\right) = \frac{1}{70}.$$

A yield of 1 gram per day is one that, starting from natural uranium, produces in one day material that is 1 gram of  $U^{235}$  mixed with 70 grams of  $U^{238}$ .



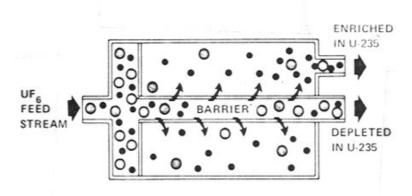


Figure 39. Gaseous diffuser enrichment cell.

The *holdup*, or total amount of material tied up in a separation plant may be very large in a plant consisting of many stages. In a plant with large holdup, a long time in the range of weeks or months is needed for steady operating conditions to be attained.

A mixture of two gases of different atomic weights can be partly separated by allowing some of it to diffuse through a porous barrier into an evacuated space as shown in Fig. 36. The molecules of the lighter gas will have a higher average speed than the heavier molecules for the same kinetic energy:

$$E = \frac{1}{2}M_{235}\mathbf{v}_{235}^2 = \frac{1}{2}M_{238}\mathbf{v}_{238}^2.$$
 (38)

The ratio of the velocities becomes:

$$\frac{\mathbf{v}_{235}}{\mathbf{v}_{238}} = \left(\frac{M_{238}}{M_{235}}\right)^{1/2} \tag{39}$$

The molecules of the lighter gas will diffuse through the barrier faster so that the gas, which has passed through the barrier (the *diffusate*), is enriched in the lighter isotope. The residual gas which has not passed through the barrier is impoverished, or depleted in the lighter isotope. For a mixture of two gases since the diffusion rates are proportional to the molecules velocities, the separation factor for the instantaneous diffusate, called the ideal separation factor  $\alpha$  is inversely proportional to the square root of the molecular weights, and is given by:

$$\alpha = \sqrt{\frac{M_{238}}{M_{235}}} \tag{40}$$

where M<sub>235</sub> is the molecular weight of the lighter gas, and M<sub>238</sub> is the molecular weight of the heavier one.

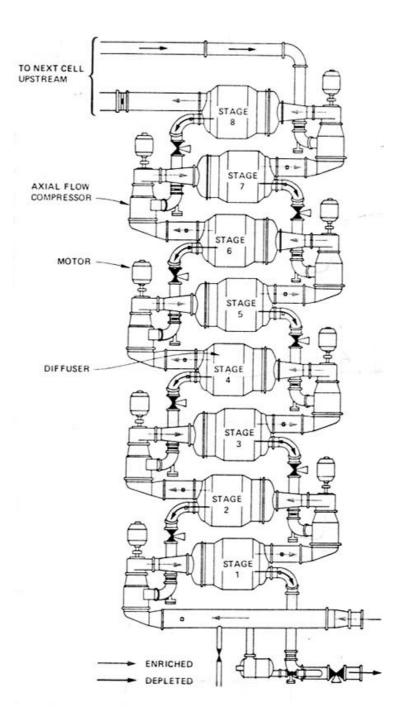


Figure 40. Gaseous diffusion cascade showing the cells, compressors, and the enriched and depleted streams.

Using the molecular weight of uranium hexafluoride,

$$\alpha = \sqrt{\frac{352}{349}} = 1.00429$$

If the fraction of the original gas that has diffused is small,

$$r = \alpha = 1.00429$$
.

If the fraction of the original gas that has diffused is appreciable, an expected diminution in separation is to be expected. If half the gas diffuses,

$$r - 1 = 0.69 (\alpha - 1)$$
.

For uranium hexafluoride, r = 1.003, a lower value than the ideal 1.004.

Because of back diffusion, of imperfect mixing on the high pressure side, and of imperfections in the barrier, a value in the range of 1.0014 can be expected in practice. For this reason, if one uses a cascade in which a reasonable overall enrichment factor is achieved per stage, then it turns out that about 4,000 stages are needed to obtain 99 percent enriched uranium, as shown in Fig. 37.

Thus, the theoretical separation factor is defined as the maximum increase in percent enrichment per stage and is given by:

$$r_{diffusion} = \left(\frac{m(U^{238}F_6)}{m(U^{235}F_6)}\right)^{\frac{1}{2}} - 1$$
$$= \left(\frac{352}{349}\right)^{\frac{1}{2}} - 1$$
$$= 0.00429$$

and the molecules speeds of the two types of UF<sub>6</sub> molecules differ by only about 0.43 percent.

Since uranium hexafluoride is highly corrosive, special alloys such as stainless steel or nickel plated components must be used. Figure 38 shows the compressors and diffuser cells.



Figure 41. Gaseous diffusion plant cells arrangement showing the compressors and diffusers for the cascade process.



Figure 42. The Tricastin Gaseous Diffusion plant, France can provide enough enriched fuel for 90 power plants. Four nuclear reactors are used to supply it with its electrical needs.



Figure 43. The K-25 Gaseous Diffusion Plant, Oak Ridge, Tennessee; no longer operational.



Figure 44. The Portsmouth, Ohio, gaseous diffusion enrichment plant, no longer operational.



Figure 45. The Paducah, Kentucky enrichment plant, only operating gaseous diffusion plant in the USA as of 2008.

# 10.17 CENTRIFUGATION PROCESS

## INTRODUCTION

The principle of centrifugation was first advanced by Linderman and Aston in 1919. At the University of Virginia in 1936, Beams separated the isotopes of chlorine using a gas centrifuge.

A centrifuge plant uses just 5 percent of the electricity needs of a gaseous diffusion plant

of comparable size.

The gas centrifuge process was first demonstrated using a gas centrifuge operating in a vacuum chamber in the 1940s during the Manhattan project in the USA and in Germany but was shelved in favor of the simpler diffusion process. It was then developed and brought on stream in the 1960s as the second generation enrichment technology. It is economic on a smaller scale at a performance of under 2 million Separative Work Unit per year (SWU/yr), which enables staged development of larger plants.

It has been deployed at a commercial level in Russia and in Europe by Urenco, an industrial group formed by British, German and Dutch companies. Russia¹s four plants at Seversk, Zelenogorsk, Angarsk and Novouralsk account for some 40 percent of the world capacity. Urenco operates enrichment plants in the UK, the Netherlands and Germany and is building one in the USA.

In Japan, the companies JNC and Japan Nuclear Fuel Limited (JNFL) operate small centrifuge plants. The capacity of JNFL's plant at Rokkasho was planned to be 1.5 million SWU/yr. China also has a small centrifuge plant imported from Russia at Lanzhou, with a capacity of about 0.5 million SWU/yr. Another small plant at Hanzhong is operating at 0.5 million SWU/yr. Brazil has a small plant with a capacity of 0.2 million SWU/yr. Pakistan has developed centrifuge enrichment technology, as well as North Korea. South Africa has an enrichment capability.

Iran became was a shareholder with French, Belgian, Spanish and Italian interests in Eurodif the gaseous diffusion group. Following the overthrow of the Shah of Iran by the Islamic Revolution, Iran was unceremonially thrown out of the consortium and since then it embarked on developing its own centrifuge technology as a matter of energy independence as well as national pride.

Both France and the USA are now considering centrifuge technology to replace their aging diffusion plants, not least because they are more economical to operate. No action has been taken in the USA in favor of the USA utilities purchasing highly enriched uranium from supposedly dismantled Russian nuclear warheads, and then blending it to a low enrichment to be used in the light water power plants under the "Megatons to Megawatts" program extending to 2009.

A license valid for 30 years for a plant in Piketon, Ohio including authorization to enrich uranium up to an assay level of 10 percent U<sup>235</sup> was issued to the United States Enrichment Corporation (USEC) which began construction of the American Centrifuge Plant in late May 2007. USEC is working toward beginning commercial plant operations in late 2009 and having approximately 11,500 machines deployed in 2012, which would provide about 3.8 million SWU/yr of production based on current estimates of machine output and plant availability.

As noted, a centrifuge plant requires as little as 50 kWhr/SWU power. For instance the Urenco plant at Capenhurst, UK has an input of 62.3 kWhr/SWU for the whole plant including infrastructure and capital works.

#### **HISTORY**

In the 1940s, the USA adopted gaseous diffusion as the most developed of the known enrichment techniques at the K-25 plant at Oak Ridge, Tennessee, which shipped enriched uranium starting in 1945. By 1941 Jesse W. Beams at the University of Virginia conducted the

first gas centrifuge experiments using the uranium isotopes. By 1956, two more gaseous diffusion plants were built at Portsmouth, Ohio and at Paducah, Kentucky. In 1976, responding to the needs of the civilian power industry, Porstmouth, Ohio was chosen to host a gas centrifuge enrichment plant using third generation machines designated as Set III with an annual performance of 200 SWUs per machine. Later the Advanced Gas Centrifuge (AGC) Set V design was developed with a capability of 600 SWUs.

At this time the promising Atomic Vapor Laser Enrichment Isotope Separation (AVLIS) method was developed. The AVLIS technology was chosen in 1985 as more versatile than the AGC and required less funding, being at an earlier stage of development. In 1992, the Energy Policy Act created the government Corporation: United States Enrichment Corporation (USEC) and transferred the uranium enrichment enterprise to it to be later privatized as a public corporation in July 1998. USEC invested \$100 million on AVLIS then dropped it in favor of a centrifuge design with 300 SWU of performance as a more economical option.

### **COMPONENTS**

The centrifuge design in the USA, Europe and Russia is based on one introduced by Gernot Zippe from Germany. In 1946, as a prisoner of war he started working on them in the USSR. He worked with Beams after his release in the USA from 1958 to 1960, when he returned to Europe.

The components of the Zippe design are a rotor, motor drive, casing, vacuum system, suspension system, and a column containing the feed, tails and product lines.

The casing surrounds the rotor and provides leak tightness to provide a vacuum and physical protection against the spinning rotor that rotates in a vacuum to minimize friction.

In advanced design centrifuges, magnetic bearings such as used in modern jet engines and Magnetically Levitated (Maglev) trains are used instead of ball bearings.

The rotor is a thin walled vertical cylinder that is spun by the drive motor. The suspension system holds the rotor upright within the casing.

The radial separation factor is proportional to the absolute mass difference between the two separated isotopes rather than the ratio of the molecular masses in the gaseous diffusion process.

In addition to the radial separation by the centrifugal force, there is also separation in the axial direction through thermal diffusion.

#### **OPERATION**

The UF<sub>6</sub> gas is introduced near the center of the rotor. It is acted upon by two simultaneous forces:

- 1. The centrifugal force induced by the rotation of the centrifuge.
- 2. An internal countercurrent circulation flow induced by an axial thermal gradient along the length of the rotor.

The thermal gradient is established by creating thermal non uniformities of temperature at the rotor end caps or along the rotor wall. The gas at the hotter lower end rises in the centrifugal

field moving radially inward. The gas at the colder end top cap sinks down and flows outwards. The countercurrent circulation created by thermal convection is superimposed on the radial centrifugal flow resulting in a relatively large assay difference between the bottom and top of the centrifuge, where the enriched product and depleted tailings are extracted at the top and bottom respectively.

In a gaseous diffusion plant about 1,000 stages are needed to reach the 4-5 percent U<sup>235</sup> enrichment needed for light water reactor designs. In contrast, these levels can be reached in about 10 stages in a centrifuge cascade. On the other hand, the throughput from a single centrifuge cascade is small and a commercial plant requires a large number of cascades, which are the basic building blocks of the plant.

## **THEORY**

When a mixture of the two molecules of  $U^{235}F_6$  and  $U^{238}F_6$  is rotated at high speed in a cylinder, the pressure distributions for the two molecules take slightly exponential gradients. The relative partial pressures at the cylinder axis and at its wall lead to a radial process separation factor. The pressure ration between the wall and the axis varies largely with the peripheral speed of the cylinder: at 300 m/s, the pressure ratio is 550:1, whilst at 500 m/s it rises to  $3.9 \times 10^7 : 1$ .

The radial separation factor at 27 °C and a peripheral speed of 300 m/s is about 1.055, compared with the theoretical process separation factor in the gaseous diffusion process of just 1.004. In practical centrifuges, the radial separation factor is further enhanced.

In the counter current centrifuge developed at the University of Virginia by Zippe in 1958, an internal recirculating flow sweeps the light molecule to the opposite end. This allows withdrawal of the two light and heavy molecular streams of the enriched and tails.

## **SEPARATION FACTOR**

Defining:

 $\alpha$  = the *enrichment factor* between the feed stream and the product stream,

 $1/\beta$  = the *depletion factor* between the feed stream and the depleted stream,

the maximum separation factor is described by the ratio:

$$r_{\text{max}} = \frac{\alpha}{\beta} = e^{\frac{1}{2}\Delta m v^2 \frac{1}{RT} \sqrt{2} \frac{\ell}{d}}$$

where:  $\Delta m$  is the difference in molecular weight between the isotopes,

R is the universal gas constant,

T is the absolute teperature in 
$${}^{\circ}K$$
,

 $\frac{\ell}{d}$  is the length to diameter ratio of the rotor,

v is the peripheral velocity, [m/s].

The theoretical separation factor is reduced by inefficiencies in the counter current flow and the feed and take off systems. It gives the discrete enrichment in one step of enrichment.

To obtain an estimate of the total number of centrifuge steps needed to obtain a certain level of enrichment, one uses the separative work concept. It depends on the enrichment and depletion of the isotopes, as well as on the mass of the gas handled. The Separative Work Unit (SWU) has the dimension of mass, since the enrichment and depletion factors are dimensionless. Enrichment services are provided in kg units of separative work.

### SEPARATIVE WORK POWER

The rate of performing separative work is termed separative power and has the units of [kg/s] or flow, which should not be confused with the product flow rate in the centrifuge. Dirac in 1941 derived the following expression for the maximum separative power of a centrifuge:

$$\delta U_{\text{max}} \approx \rho D \eta \left( \frac{1}{2} \Delta m \, \text{v}^2 \, \frac{1}{RT} \right)^2 \pi \, \frac{\ell}{2}$$

where: D is the back or self diffusion coefficient,

 $\rho$  is the gas density,

 $\Delta$ m is the molecular weight difference of the isotopes,

(42)

v is the peripheral rotor velocity,

R is the universal gas constant,

T is the gas temperature in degrees Kelvin,

 $\ell$  is the rotor length,

 $\eta$  is the circulation efficiency.

It must be noticed that the separative power is proportional to the centrifuge length, and to the fourth power of the peripheral velocity. It is independent of the rotor diameter, and favors operation at low temperatures.

A practical limit exists on lower temperatures to prevent the condensation of the UF<sub>6</sub> gas.

Two optimization factors are the centrifuge length and the peripheral velocity. The TC12 design used by Urenco is three times less in length than the Advanced Gas Centrifuge (AGC) Set V developed in the USA. The TC12 has an approximate annual performance of 40 SWU per machine, whereas the longer and faster Series V USA machine can provide 300 SWU per machine per year.

The maximum velocity achievable in a thin cylinder rotating around its axis is determined by the strength/density ratio of the material of construction. Each material possesses a typical bursting speed. It is also limited by the ability to attenuate harmonic flexural vibrations.

The maximum peripheral speed of a rotor is given as:

$$v = \sqrt{\frac{\sigma}{\rho}}$$
where: \sigma is the rotor tensile strength,
$$\rho \text{ is rotor density.}$$
(43)

A typical centrifuge can reach a rotational speed of 1,500 revolutions/ sec or 90,000 rpm, compared with 12-25 rpm in the spin drying cycle of washing machines. To reduce friction, a vacuum is created between the rotor and the stator. A magnetic bearing holds the top of the rotor steady in a levitated mode with just a needle contact at the bottom.

A pulsating magnetic field imparts angular momentum to the rotor and causes it to rotate. Various techniques are used to avoid destructive vibrations such as bellows allowing the controlled flexing of the rotor. Control of the rotational speed of the rotor should allow it quick passing through its natural frequency resonance speed during startup and shutdown.

Maraging-steel instead of high strength aluminum allows faster rotational speeds. A carbon fiber composite rotor as used in aircraft wings and wind turbine rotor designs stands out in that it can spin faster than other light weight and high strength materials.

Ultra high strength and super light alloys steels such as vanadium steel are promising candidates. Vanadium alloyed with titanium would even offer the best strength to weight ratio of any engineered material and is worthy of investigation.

Material	Density [gm/cm <sup>3</sup> ]	Maximum peripheral speed [m/s]	Tensile Strength [Newton/mm <sup>2</sup> ]	Tensile strength to density ratio
Carbon fiber composite	1.7	>800	2,000	1176.47
Glass fiber composite	2.0	600	700	350.00
Maraging steel, superhard alloy	8.0	525	2,250	281.25
High strength steel	8.0	455	1,700	212.50
Titanium	4.6	440	920	200.00
High strength aluminum alloy	2.8	425	520	185.71

Table 1. Candidate materials for centrifuge rotor tubes.

The length is determined by the materials choices as well as the dynamics of the rotor. Long small diameter and thin walled rotors can be rigid at only low rotational speed. As its length increases, the transverse bending frequency decreases up to the point of the frequency of angular rotation designated as the critical frequency. A subcritical centrifuge design such as the Zippe design would have a length that allows it to operate just below this critical frequency.

Beyond the critical speed, a supercritical machine experiences flexural vibrations. In this case the rotor vibrations are countered with precision balancing that reduces the vibration amplitude to a level within the confines of the centrifuge casing. This uses high quality manufacturing processes and fast computers to balance the rotor.

Circulation inefficiency resulting from back diffusion along the axial concentration gradient in the rotor limits the actual performance.

## **OPERATIONAL CHARACTERISTICS**

Like the diffusion process, the centrifuge process uses UF<sub>6</sub> gas as its feed and makes use of the slight difference in mass between U<sup>235</sup> and U<sup>238</sup>. The gas is fed into a series of tubes, each containing a rotor, one to two meters in length and 15-20 cm in diameter. When the rotors are spun rapidly, at 50,000 to 70,000 rpm, the heavier molecules with U<sup>238</sup> increase in concentration towards the cylinder's outer edge. There is a corresponding increase in concentration of U<sup>235</sup> molecules near the center. These concentration changes are enhanced by inducing the gas to circulate axially within the cylinder using the process of thermal diffusion. The vacuum contributes to thermal insulation.

The enriched gas forms part of the feed for the next stages while the depleted  $UF_6$  gas goes back to the previous stage. Eventually enriched and depleted uranium are drawn from the cascade at the desired assay.

To obtain efficient separation of the two isotopes, centrifuges rotate at very high supersonic speeds, with the outer wall of the spinning cylinder moving at between 400 and 500 meters/sec to give a million times the acceleration of gravity.

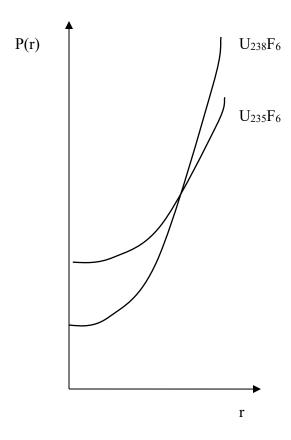


Figure 46. Radial pressure distribution in the centrifuge process.

Although the capacity of a single centrifuge is much smaller than that of a single diffusion stage, its capability to separate isotopes is much greater. Centrifuge stages normally consist of a large number of centrifuges in parallel. Such stages are then arranged in cascade similarly to those for diffusion. In the centrifuge process, however, the number of stages may only be 10 to 20, instead of a thousand or more for gaseous diffusion.

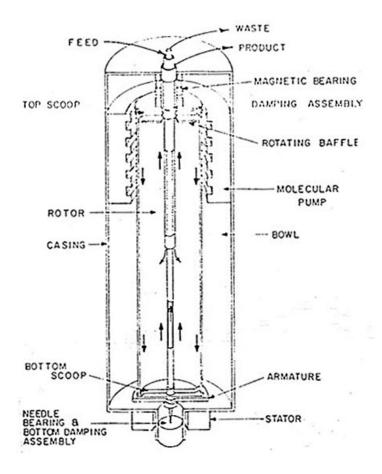


Figure 47. Single Centrifuge showing the flow of the depleted and the enriched streams. Input gas is released into the center. The centrifugal forces concentrate the heavier molecules near the edges. Heating the bottom of the centrifuge further causes the lighter gas to move through convection to the top while the heavier molecules would concentrate near the bottom. The top scoop collects the depleted stream, whilst the bottom scoop collects the enriched stream.

The centrifuge process uses less power than the gaseous diffusion process, but requires high strength components in the rotating parts on the centrifuge and more elaborate power inversion equipment. Magnetic bearings are used. High strength materials that would resist the extreme corrosive effect from the uranium hexafluoride including stainless steels and composites such as in aircraft components have been used in the rotors construction.

The great appeal of the centrifuge in the separation of the heavy isotopes is that the separation factor depends on the difference between the masses of the two isotopes:

$$\alpha_{centrifugation} \quad \alpha_{\ell} \quad \Delta m = (M_{238} - M_{235}) \tag{44}$$

not on the square root of the ratio of the masses as in diffusion methods:

$$\alpha_{diffusion} = \sqrt{\frac{M_{238}}{M_{235}}} \tag{45}$$

Figure 44 shows a cutout through a single centrifuge, the European and American centrifuge designs are shown in Fig. 45, assembly of Set III American centrifuges is shown in Fig. 46, and Fig. 47 shows a cascade of centrifuges.

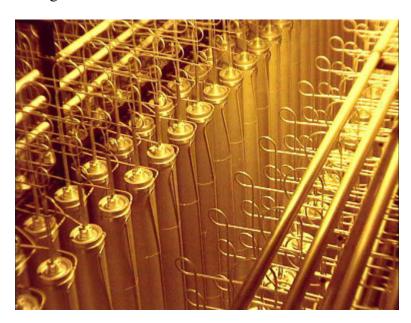




Figure 48. Centrifuge banks at the European Urenco plant (top), and the USA USEC demonstration plant (bottom).



Figure 49. Assembly of third generation Set III centrifuges with annual performance of 200 SWU/yr per machine.

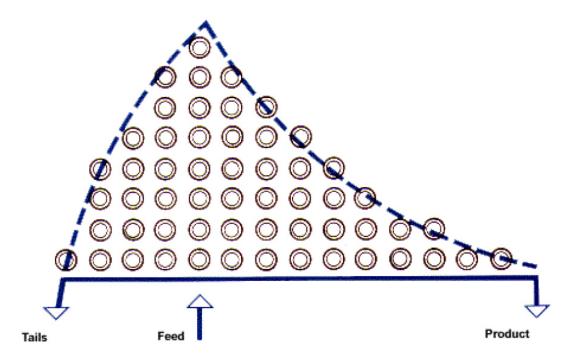


Figure 50. Serial and parallel connection in a centrifuge cascade configuration.

The theoretical separation factor is defined as the maximum increase in percent enrichment per stage and is given by:

$$r_{centrifuge} = \left(\frac{m(U^{238}F_6)}{m(U^{235}F_6)}\right) - 1$$

$$= \left(\frac{352}{349}\right) - 1 \qquad (46)$$

$$= 0.0086$$

Thus the molecules speeds of the two types of UF<sub>6</sub> molecules differ by only about 0.86 percent, double the amount in gaseous diffusion.

Half the stages required for enrichment to the same percentage as required by diffusion, makes the centrifuge process advantageous. However, this is associated with a large number of moving parts requiring maintenance and lubrication, in addition to constant inspection due to the stresses created by the high rotational speeds.

The centrifuge is a low output device measured in kg SWU/year. Many thousands of centrifuges are needed in a plant measured in the hundred of metric tonnes of SWU/year. The separation factor of a single centrifuge is inadequate to obtain the desired enrichment in one step, requiring the use of cascades of centrifuge operating in series and parallel arrays. The serial

centrifuges provide the multiplication of the separation effect, while the parallel components provide the magnitude of the separative work.

## PLANT DESIGN

The number of centrifuges in a cascade is governed by the separative work of the individual centrifuge, its cost, the cost of the cascade associated equipment, and the cost of the protection devices. A plant consists of many cascades grouped into operational units sharing the same process services. The whole plant must operate under high vacuum with the UF $_6$  pressures within the cascade pipe work of just a few hundred pascals.

Natural uranium as UF<sub>6</sub> is fed to the plant in its storage containers. They are steam heated to just below the triple point producing vapor through sublimation. The gas is passed through a pressure reducing manifold to the cascade manifold.

A cascade is bounded by feed, tailings, and product valves. The pumping action of the centrifuges causes the flow of the gas. The product and gas streams are withdrawn through desublimation in cold traps operated at  $-70~^{\circ}\text{C}$ . These are used in a batch mode: they are isolated when they are filled and heated to 55  $^{\circ}\text{C}$  and their contents passed to the tails and product containers.

The cascades need a medium frequency electric drive supply and a continuous supply of cooling water to remove the waste heat generated by inefficiencies in the motors and the frictional drag of the UF<sub>6</sub> gas on the static scoop arms used inside the rotor to remove the product and tails streams.

The cooling water is also used to create a thermal gradient along the length of the centrifuge to stimulate the counter current flow. This adds thermal diffusion to the centrifugal separation process.

Other components include steam heating, chilled water, venting systems, effluent scrubbing and UF<sub>6</sub> sampling systems.

## **DEVELOPMENT PROGRAMS**

One of the main objectives of centrifuge design is to maximize the length and peripheral velocities. This necessitates supporting studies of stress analysis, rotor dynamics, superconducting magnetic bearing development, separation theory, gas dynamics and heat transfer. Advances are pursued in the motor drive, composite materials, automated manufacturing techniques and digital instrumentation and control.

New developments include a rotating plasma centrifuge where a plasma is spun at high speed separating the two streams. A plasma as a conducting fluid is affected by a magnetic field, superimposing a magnetic filed configuration could lead to a centrifuge process combining the advantages of centrifugation and electromagnetic separation without the limitations of the mechanical moving parts of the conventional centrifuge.

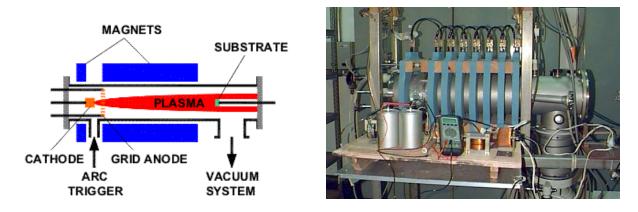


Figure 51. Plasma Centrifuge concept and experiment.

## 10.18 AERODYNAMIC NOZZLE AND VORTEX PROCESSES

Two aerodynamic processes were brought to demonstration stage. One is the jet nozzle process, with a demonstration plant built in Brazil, and the other the Helikon vortex tube process developed in South Africa. They depend on a high speed gas stream bearing the UF<sub>6</sub> being made to turn through a very small radius, causing a pressure gradient similar to that in a centrifuge. The light fraction can be extracted towards the centre and the heavy fraction on the outside. Thousands of stages are required to produce enriched product for a reactor. Both processes are energy intensive using over 3,000 kWhr/SWU.

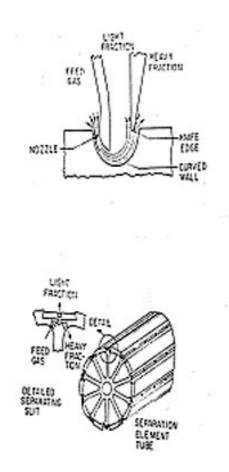


Figure 52. Isotopic Separation in the Nozzle Process.

The Becker nozzle separation process uses an aerodynamic process to separate the different isotopes. A mixture of UF<sub>6</sub> and an auxiliary gas such as hydrogen or helium, is forced to flow along a curved wall. The heavier molecules tend to remain closer to the wall than the lighter ones. The auxiliary gas aids in accelerating the uranium hexafluoride gas to allow separation of the molecules with the use of a knife's edge.

The separation nozzle approach provides enrichment per stage ratio that is 3 to 4 times that of the gaseous diffusion approach. It also has fewer moving parts than the gas centrifuge, minimizing wear and maintenance. The knife edge is however, subject to corrosion and must be adjusted continuously to provide the desired separation.

The separation nozzles are joined in a series of ten nozzles together in a tubular separation element. The inner channels provide the means for injecting the UF<sub>6</sub> and for the removal of the depleted gas. The enriched gas leaves radially and becomes the input to the next stage.

## 10.19 LASER ISOTOPE SEPARATION

Existing methods of enrichment cannot economically separate more than about 65 percent of the  $U^{235}$  isotope from natural uranium. The remaining 35 percent is left in the tails as depleted uranium. Thus one third of the useful  $U^{235}$  cannot be economically recovered by the existing

methods. Laser isotope separation may make that fraction available from the existing one half billion kilograms of tails.

Laser enrichment processes have been the focus of interest for some time. They are a possible third-generation technology promising lower energy inputs, lower capital costs and lower tails assays, hence significant economic advantages. None of these processes is yet ready for commercial use, though one is well advanced.

Development of the Atomic Vapor Laser Isotope Separation AVLIS, and the French SILVA, began in the 1970s. In 1985 the US Government backed it as the new technology to replace its gaseous diffusion plants as they reached the end of their economic lives early in the 21st century. However, after some USA \$2 billion in Research and Development (R&D), it was abandoned in the USA in favor of the SILEX molecular process. French work on SILVA has been stopped.

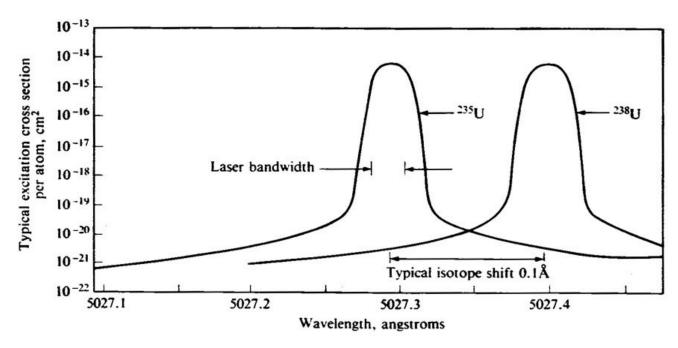


Figure 53. Excitation cross sections for the U<sup>235</sup> and U<sup>238</sup> isotopes at different laser wavelengths.

Atomic vapor processes work on the principle of photo-ionization, whereby a powerful laser is used to ionize particular atoms present in a vapor of uranium metal. An electron can be ejected from an atom by light of a certain frequency. The laser techniques for uranium use frequencies which are tuned to ionize a  $U^{235}$  atom but not a  $U^{238}$  atom. The positively-charged  $U^{235}$  ions are then attracted to a negatively-charged plate and collected. Atomic laser techniques may also separate plutonium isotopes.

In photochemistry, shining light into a mixture of chemicals can trigger chemical reactions. When a molecule absorbs light it rotates and vibrates faster, that is it is excited to a higher level. Photochemical reactions induced by lasers can be very specific in converting light energy into stored energy.

The wavelength and intensity of the absorbed light determine the excitation level of the reactants, and also determine the type of products the reaction will yield.

# **MOLECULAR SILEX PROCESS**

The main molecular processes which have been researched work on the principle of photo-dissociation of UF<sub>6</sub> to solid UF<sub>5</sub>, using tuned laser radiation. Any process using UF<sub>6</sub> fits more readily within the conventional fuel cycle than the atomic process.

The laser process under development on the world stage is Silex, an Australian development which is molecular and utilizes UF<sub>6</sub>. In 1996 USEC secured the rights to evaluate and develop Silex for uranium. It is also usable for silicon and other elements, but relinquished these in 2003. The Silex process is now at prototype stage with the Silex Company in Sydney, Australia and applications to silicon and zirconium are also being developed.

Laser photochemistry differs from classical photochemistry in major ways related from the extreme brightness of the laser within a narrow frequency band, or color. This brightness allows enough energy to be deposited in specific kinds of molecules so that all those molecules dissociate or break up into fragments, forming new products. Laser sources can be tuned to the frequencies required by a variety of molecules.

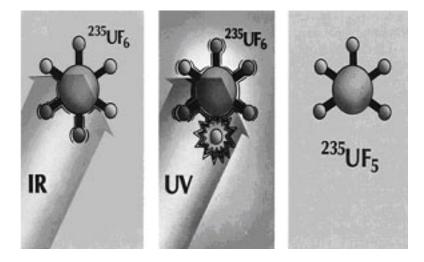


Figure 54. An infrared laser excites the U<sup>235</sup>F<sub>6</sub> molecule. The excited U<sup>235</sup>F<sub>6</sub> molecule loses one of its fluorine atoms as it absorbs an ultraviolet laser.

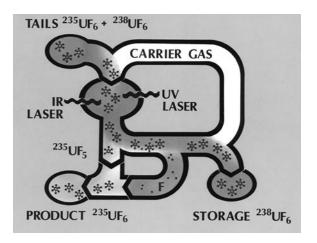


Figure 55. Schematic of the laser isotope enrichment process.

The purity of the wavelength in lasers provides the option of exciting only selected molecules. The molecules of  $U^{235}F_6$  and  $U^{238}F_6$  have identical chemical properties, but have different natural vibrational frequencies

Lasers can selectively be tuned to the frequency of a specific molecule with the result that only that molecule absorbs laser energy and undergoes a change.

Such selectivity is used to drive a chemical reaction in one isotope but not the other. An infrared long wave length laser excites the  $U^{235}F_6$  molecule. The excited  $U^{235}F_6$  molecule loses one of its fluorine atoms as it absorbs an ultraviolet or short wave length laser. The new  $U^{235}F_5$  molecule is in the form of a powder that can be easily separated from the gas, and collected by standard techniques.

A complete process can be devised where the removal of the U<sup>235</sup> isotope can be made to reach up to 100 percent. If applied to the existing tails, this would be a beneficial use of a now wasted resource. To avoid proliferation concerns, such a plant can be designed to enrich the uranium to a specified level suitable for nuclear reactor applications and no more. A plant could be designed to just enrich depleted uranium to the natural abundance of uranium for use for instance in heavy water reactors. It appears that its proliferation risk is lower than the other methods.

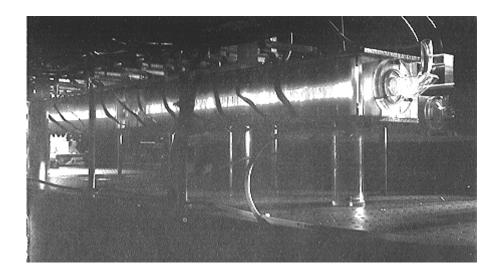


Figure 56. Krypton fluoride cable-fed rare gas halogen ultraviolet laser.

One of the candidate lasers is the Krypton fluoride rare gas halogen ultraviolet laser.

# ATOMIC VAPOR LASER ISOTOPE SEPARATION, AVLIS

In another approach to laser enrichment, molten uranium is used instead of uranium hexaflouride, where the uranium vapor is generated by an electron gun. The uranium vapor is expanded and irradiated with a laser light at a wavelength of 5,027.3 angstroms and a bandwidth of less than 0.1 angstroms. The  $U^{235}$  molecules are ionized and drawn off to a product collector, while the  $U^{238}$  passes off to the tails collector.



Figure 57. Green and yellow lasers beams dye corridor, AVLIS plant.

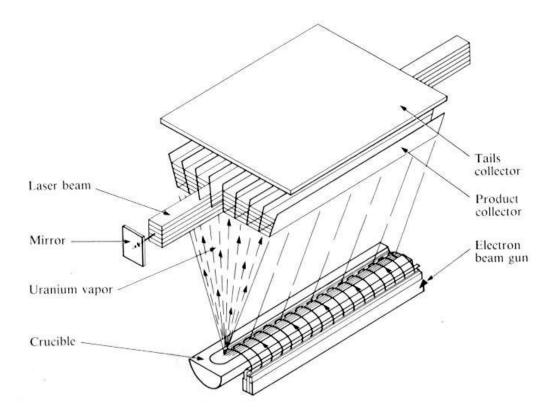


Figure 58. Atomic Vapor Laser Isotope Separation (AVLIS) Configuration.



Figure 59. The AVLIS pilot plant at the Lawrence Livermore National Laboratory, LLNL.

This technology makes use of the fact that isotopes of different masses absorb slightly different wavelengths of light; an indirect consequence of the nucleus mass difference. Precisely tuned lasers would excite only the isotope atoms desired in a stream of atomic vapor. The ionized atoms would then be separated from the neutral ones electromagnetically or by chemical reaction. AVLIS has not been used on an industrial scale yet. This technique promises to allow

high efficiency production of high-purity U<sup>235</sup> and Pu<sup>239</sup>, although its true useful is difficult to judge without an operating plant to observe. The AVLIS technology, if available, could make it possible for a country to produce substantial batches of weapon-grade uranium, neptunium or plutonium from commercial reactor fuel. The energy required for separation itself is very low, only enough to break the molecular bond or ionize the atom. Energy consumption is mostly determined by the efficiency of the laser used, which is generally on the order of 0.1 percent.

Due to its economic advantage, laser isotope separation may become the choice separation method for the twenty first century.

# 10.20 PLASMA ION CYCLOTRON FREQUENCY SEPARATION PROCESS

The Savannah River Plant (SRP) in the USA under contract with TRW developed the Plasma Separation Process (PSP) to recover the isotope  $U^{236}$  and to a lesser extent  $U^{234}$  and  $U^{238}$  from irradiated naval reactors fuel. All three build up during continued fuel recycle and irradiation.

The concentration of  $U^{236}$  may reach 35 percent. Left in highly enriched fuel the fissionable but non fissile isotopes absorb neutrons and result in significant deteriorated performance during reactor operation. Upon the removal of the fissionable non fissile isotopes the reactor efficiency increases and the need for additional Highly Enriched Uranium (HEU) decreases.

A variant of the process uses a plasma source and a uniform superconducting magnetic field. A microwave antenna excitation field feeds the ion cyclotron frequency that excites the isotope to be separated. Upon absorption of the resonance energy, its circular orbit around the magnetic field lines is increased leading to its collection on an array of plates.

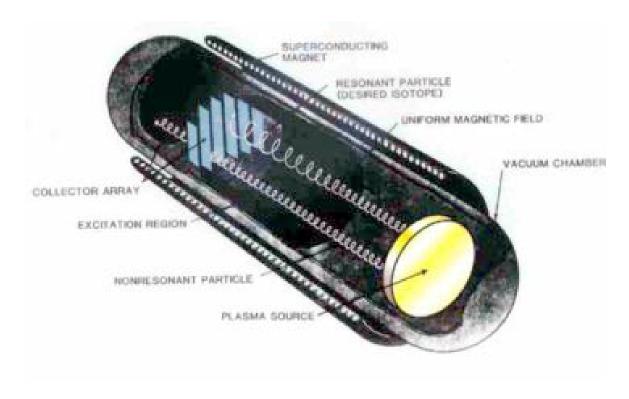


Figure 60. Schematic of plasma separation process using ion cyclotron frequency excitation.

Table 2. Percent isotopic content of recycled fuel.

	Without Plasma Separation Process	With Plasma Separation Process
$U^{234}$	1.6	1.2
$U^{235}$	49.0	68.7
$U^{236}$	35.0	25.8
$U^{238}$	14.4	4.3

## **10.21 CHEMICAL PROCESS**

A chemical process has been demonstrated to pilot plant stage but not used. The French Chemex process exploited a very slight difference in the two isotopes  $U^{235}$  and  $U^{238}$  propensity to change valency in oxidation/reduction, utilizing aqueous III valency and organic IV phases.

## 10.22 COUPLED LASER AND ELECTROMAGNETIC SEPARATION

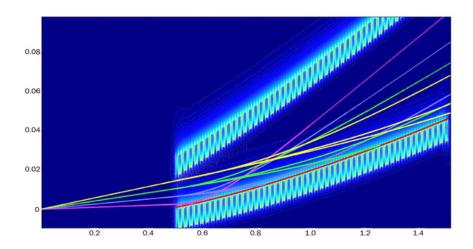


Figure 61. Simulated ion trajectories in the separation of the Li<sup>7</sup> and Li<sup>6</sup> isotopes. Lasers alter the magnetic moments of the different isotopes which would then follow different trajectories in applied magnetic fields. Source: Physics World.

A new methodology under study combines two techniques of isotopic separation: the electromagnetic separation or Calutron method and the laser isotope separation method. It is applied to the separation of the Li<sup>6</sup> and Li<sup>7</sup> isotopes. Lasers alter the magnetic moments of the different isotopes which would then follow different trajectories in applied magnetic fields.

Using lasers alone to separate isotopes with different nuclear magnetic moments has energy requirements that are prohibitive. The methods of enrichment using lasers are primarily based on ionization of the atoms requiring several different wavelengths and very high-power lasers. A subtler effect called optical pumping is suggested, whereby the absorption of laser light can change the magnetic moment of an ion, making it move in a specific direction in response to a magnetic field. Different isotopes are excited at slightly different frequencies, and lasers emit at only one, very precise frequency. This provides a very efficient way of changing the magnetic states of some isotopes but not others, and thus to separate the two.

The isotope Li<sup>7</sup> in the form of Li<sup>7</sup>OH is used as a buffering agent added to the boric acid used as a burnable poison in the Pressurized Water Reactors (PWRs) water coolant. Li<sup>7</sup> is also used in the resin used to purify the water coolant and control its chemistry. The isotope Li<sup>6</sup> is primarily thought-after in the isotopic separation process. Li<sup>6</sup> is used for the production, through neutron capture in a thermal neutrons spectrum, of tritium for maintaining the thermonuclear weapons stockpile. It would be used in future first generation fusion reactors using the DT fusion reaction. This is necessary since tritium decays with a half-life of 12.33 years into the He3 isotope and hence about 5 percent of it is lost every year and needs to be replenished to maintain the thermonuclear and the spiked (for yield control) weapons stockpile. Li<sup>7</sup> is thus a left-over or byproduct from the Li<sup>6</sup> production process using the COLEX process.

The introduction of the Th<sup>232</sup>-U<sup>233</sup> fuel cycle and the use of a lithium fluoride molten salt as a coolant would necessitate isotopic tailoring and the use of Li<sup>7</sup> as a salt component. In a thermal neutron spectrum, the naturally occurring Li<sup>6</sup> would produce undesirable tritium.

Technical issues exist in the choices of lasers as well as the magnets that would be needed for the process. Usually, electromagnets with copper wiring and iron cores would be used with the Calutron or magnetic separation process. Instead, permanent magnets using the rare earths alloying elements such as neodynium or lanthanum are suggested; and even superconducting magnets. A discerned difficulty is that he altered nuclear magnetic moment of an isotope has to be maintained via the optical pumping process during its movement in the applied magnetic field.

## 10.23 RE-ENRICHMENT OF RECYCLED URANIUM

In some countries spent fuel containing between 0.7-1.3 percent  $U^{235}$  is reprocessed to recover its uranium and plutonium, and to reduce the final volume of high-level wastes. The plutonium is normally recycled promptly into mixed oxide (MOX) fuel, by mixing it with depleted uranium.

Where uranium recovered from reprocessing spent nuclear fuel is to be re-used, it needs to be converted and re-enriched. This is complicated by the presence of impurities and two new isotopes in particular:  $U^{232}$  and  $U^{236}$ , which are formed by neutron capture in the reactor. Both decay much more rapidly than  $U^{235}$  and  $U^{238}$ , and Thallium<sup>208</sup>, one of the daughter products of  $U^{232}$  emits very strong 2.6 MeV gamma rays, which means that shielding is necessary in the plant.

U<sup>236</sup> is a neutron absorber which impedes the chain reaction, and means that a higher level of U<sup>235</sup> enrichment is required in the product to compensate for its presence. Being lighter, both the U<sup>232</sup> and U<sup>236</sup> isotopes tend to concentrate in the enriched rather than the depleted output, so reprocessed uranium which is re-enriched for fuel must be segregated from enriched fresh uranium.

Both the diffusion and centrifuge processes can be used for re-enrichment, though contamination issues prevent commercial application of the former. A laser process would theoretically be ideal as it would ignore all but the desired  $U^{235}$ , but this remains to be demonstrated with reprocessed feed.

# 10.24 UNITED STATES ENRICHMENT CORPORATION (USEC)

With plans underway for about 32 new reactors in the USA, a stable, domestic source of enriched uranium is vital. The USA's Nuclear Regulatory Commission (NRC) issued in April 2007 a construction and operating license for USEC Inc.'s American Centrifuge Plant in Piketon, Ohio. The license, which is good for 30 years, includes authorization to enrich uranium up to an assay level of 10 percent U<sup>235</sup>. USEC expects operates a Lead Cascade of centrifuge machines in the American Centrifuge Demonstration Facility. The Company was working toward beginning commercial plant operations and having approximately 11,500 machines deployed in 2012, which would provide about 3.8 million Separative Work Units (SWU) of production. USEC operates the only uranium enrichment facility in the USA: a gaseous diffusion plant in Paducah, Kentucky. The American Centrifuge Plant is expected to use 95 percent less electricity than a comparably sized gaseous diffusion plant.

## **EXERCISES**

- 1. An executive at an electrical utility company needs to order uranium fuel from a mine. The utility operates a single 1000 MWe power plant of the CANDU type using natural uranium, and operating at an overall thermal efficiency of 33 percent. What is the yearly amount of:
- a.  $U^{235}$  burned up by the reactor?
- b. U<sup>235</sup> consumed by the reactor?
- c. Natural uranium that the executive has to contract with the mine as feed to his nuclear unit?
- 2. An executive at an electrical utility company needs to order uranium fuel from a mine. The utility operates a single 1,000 MW(e) PWR power plant operating at an overall thermal efficiency of 33 percent. The fuel needs to be enriched to the 5 w/o in U<sup>235</sup> level. The enrichment plant generates tailings at the 0.2 w/o in U<sup>235</sup> level. What is the yearly amount of natural uranium that the executive has to contract with the mine as feed to his nuclear unit?
- 3. Compare the ratios and the difference in the separation radii in the electromagnetic separation method for the separation of the ions of the isotopes:
- a)  $U^{235}$  and  $U^{238}$ ,
- b) Li<sup>6</sup> and Li<sup>7</sup>.
- 4. Identify the level of U<sup>235</sup> enrichment in:
- a. Natural uranium,
- b. LWR: BWR and PWR, reactor fuel,
- c. Depleted uranium discharge from enrichment plant,
- d. Burnt-out discharged reactor fuel.

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## **APPENDIX**

#### CENTRIFUGE BEARINGS AND VIBRATION DAMPING

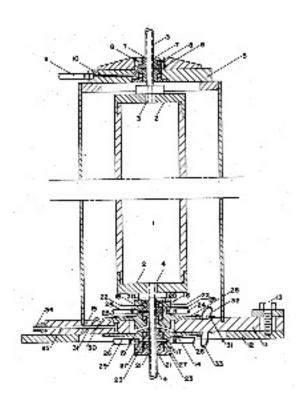


Figure A1. Initial centrifuge drawing in patent application by Jesse W. Beams and Leland B. Snoddy.

In a centrifuge there are certain speeds called the critical speeds at which the shaft is distorted or flexed out of the axis of rotation causing vibration forces which are transmitted to the bearings. The critical speed occurs when the speed of rotation of the device is equal to a natural mode of vibration of the rotating member and the supporting structure. They depend on the stiffness of the shaft and the inertia of the stationary and rotating parts of the device.

The vibrations must be damped through the design of the bearings, such as the use of dual bearings that are independently supported for the limited movement relative to each other. The bearings of the assembly are spaced axially of the shaft a distance different from the spacings of the nodes of the vibration waves at the critical speeds.

The rotating shafts are hollow so as to allow the introduction and withdrawal of the depleted and enriched streams. The bearings have an L shape and can be made of lead bronze with 20 percent Pb. The vibration absorbing rings could be made of neoprene. The bearing assemblies could be lubricated with oil or any suitable lubricant.