

Uranium enrichment technologies

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Summary

The enrichment of uranium has taken place since World War II, initiated by the quest for the atomic bomb to win the war. To enrich uranium means to separate the two main isotopes that make up natural uranium (U-235 and U-238). The details of the possible enrichment processes are generally classified because highly enriched uranium may be used for weapons production, but the principles are well known.

The gaseous diffusion method developed during the war constitutes the first generation of uranium enrichment techniques and has been used both for enrichment to weapons grade uranium and also for production of low-enriched uranium for nuclear power plants. This technique has high energy consumption and takes up a lot of space and is therefore being phased out in favour of gas centrifuges. *Gas centrifuges* take up less space and use only about one tenth of the energy of the gaseous diffusion method. Gas centrifuges are harder to build technically, but once they are built they can more easily be used to produce highly enriched uranium, as well as low-enriched uranium, for weapons or other purposes. The gas centrifuge technology is spread around the world and has even been for sale on the black market. *Laser isotope separation methods* are about to become the third generation of commercial scale uranium enrichment technologies. These methods are researched by many countries, and have been developed to a great extent. The energy consumption of these processes is about the same as for gas centrifuges, but the facilities require much less space than the centrifuge plants. These are high-precision technologies, and making the lasers and other necessary equipment has posed significant technical challenges. It appears difficult to expand from laboratory scale to large scale facilities, and most of the research has been terminated. The only still active laser process is the SILEX process which is about to go commercial in a few years.

Another technique which has been used on a large scale is *the aerodynamic technique* used in South Africa. This technique was used both for production of low-enriched and highly enriched uranium, but it is not regarded as commercially competitive because of its high energy consumption. Also *the mass spectrometer method* was used in a relatively large scale in the United States during World War II, but this process is too slow and consumes too much energy to be commercially competitive as well. The mass spectrometer method is relatively easy to perform, and could therefore be used in a non-commercial setting as a slow but reliable option for enrichment. This could also be said about *the thermal diffusion method* which was used to produce feed for the mass spectrometer method in the United States during World War II. The thermal diffusion method could not be used for high-level enrichment. *Plasma processes* have not been applied on a large scale yet. *The chemical enrichment processes* developed in France and Japan have been utilised in pilot plants, but not yet in full-scale facilities.

Norwegian summary

Urananrikning fant først sted i forbindelse med utviklingen av kjernevåpen under andre verdenskrig. Anrikning av uran innebærer at en skjelner mellom de to viktigste isotopene i naturlig uran (U-235 og U-238). Detaljene i de mulige anrikningsprosessene er generelt gradert fordi høyanriket uran kan brukes i kjernevåpen, men prinsippene er velkjente.

Gassdiffusjonsmetoden som ble utviklet under andre verdenskrig, utgjør første generasjon urananrikningsteknologi og har vært anvendt både til å lage våpenmaterialer og til å lage lavanriket uran til kjernekraftverk. Denne metoden krever mye energi og store anlegg og blir derfor faset ut til fordel for gassentrifuger. *Gassentrifuger* er mindre plasskrevende og krever bare om lag en tidel så mye energi som gassdiffusjonsmetoden. Sentrifugene er teknisk krevende å lage, men når de først er på plass, er de enklere å benytte enn diffusjonsutstyr til produksjon av både lavanriket og høyanriket uran. Gassentrifugeteknologien er i bruk mange steder i verden og har også vært tilgjengelig på det svarte markedet. *Laserisotopseparasjonsmetoder* er i ferd med å bli tredje generasjon av kommersielle urananrikningsteknologier. Det er blitt forsket på disse metodene i en rekke land, og en god del utviklingsarbeid har funnet sted. Energibehovet er omtrent som for gassentrifugemetoden, men anleggene er vesentlig mindre plasskrevende. Lasermetodene krever avansert teknologi med høy presisjon, og det har vist seg svært utfordrende å lage både laserne og annet nødvendig utstyr. Det har vært vanskelig å gå fra laboratorieskala til industriell skala, og mye av forskningen har blitt avsluttet. Den eneste aktive lasermetoden er den såkalte SILEX-prosessen som forventes å bli kommersielt tilgjengelig i løpet av noen få år.

En annen metode som har vært anvendt i stor skala, er *aerodynamisk urananrikning*, som Sør-Afrika benyttet seg av. Der ble metoden brukt både til lavanriket og høyanriket uran, men den anses ikke kommersielt konkurransedyktig fordi den er svært energikrevende. Under andre verdenskrig ble også *massespektrometermetoden* anvendt i relativt stor skala i USA, men denne metoden er for langsom og for energikrevende til å være kommersielt konkurransedyktig. Metoden er relativt enkel og kan derfor tenkes anvendt som en langsom, men pålitelig, mulighet i ikke-kommersielle situasjoner. Det samme kan sies om metoden med *termisk diffusjon* som ble brukt til å produsere utgangsmaterialet for de amerikanske massespektrometrene under andre verdenskrig. Termisk diffusjon kan ikke anvendes til å lage høyanriket uran. *Plasmaprosesser* har ennå ikke blitt anvendt i stor skala. *Kjemiske anrikningsmetoder* har blitt utviklet i Frankrike og Japan. De har vært demonstrert i pilotanlegg, men ikke i industriell skala.

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1 Introduction

Uranium enrichment is the key to making uranium-based nuclear weapons. It is also a process that is important in the nuclear power industry. In the race for the atomic bomb during World War II, the United States pursued several methods for enriching uranium all at once. Also Germany and the Soviet Union worked on developing enrichment methods at this time. Some of these methods proved inefficient and have been discarded. Others, where the most prominent examples are the gaseous diffusion method and the gas centrifuge method, have been further developed and made into commercial processes used today for enriching uranium for commercial nuclear power plants. The enrichment of uranium is considered the most technically and industrially demanding part of any nuclear program (either civilian or military).

Today most enrichment plants produce low-enriched uranium, but even now almost all enrichment plants can be modified to produce highly enriched uranium.[1] This report gives an overview of the most common enrichment technologies and addresses some of the technical and economical difficulties associated with them as well as when and where these technologies have been in use. All of these techniques have been researched by more than one country, but not all of them have made it to actual production. Several other techniques exist either on the drawing board or have been tried to some extent, but this report covers only the techniques considered most promising for future use.

To understand the techniques discussed in this report, some basic knowledge of physics and chemistry is needed. Some of this information is provided in Appendix A.

2 Uranium enrichment

Like all elements, uranium consists of different *isotopes* (see Section A.1); the two that are the most abundant and relevant, are known as *U-235* and *U-238*. Natural uranium consists of approximately 99.3 % *U-238* and 0.7 % *U-235*. When uranium is to be used as fuel for a power plant or in a bomb, it is the less abundant isotope *U-235* that is needed because it is this isotope that is most likely to *fission* (see Section A.2) when it absorbs one neutron and releases several neutrons in each fission process. *U-235* is able to sustain a *chain reaction* if enough of the isotope is present. The challenge is to find a way to extract this isotope from the natural uranium; this process is called *enrichment*. There are several ways of doing this, and some of the most important techniques are discussed in this report. It is customary to speak of three different levels of enrichment. *Low-enriched uranium (LEU)* contains less than 20 % *U-235*; the fuel used in power plants is usually enriched to about 3-5 %. *Highly enriched uranium (HEU)* contains more than 20 % *U-235*; uranium enriched to this level is used in some specialised reactors. If the uranium is enriched beyond about 90 %, it may also be referred to as *weapons grade uranium*.

2.1 How enrichment processes work

The different methods for uranium enrichment all have in common that they make use of the different properties of the isotopes (either physical or chemical) to physically separate U-235 from U-238. No enrichment technology is perfect so the products will never be 100 % U-235 in one place and 100 % U-238 in another; there will always be two new mixtures, one which is enriched in U-235, and one which is correspondingly depleted in U-235. Because of its physical properties, *depleted uranium* (DU) is commonly used in conventional armour-piercing munitions,¹ but it is in fact a waste product from the nuclear industry.

The physical property that most enrichment technologies make use of, either directly or indirectly, is the *mass difference* between the isotopes. Heavier particles move differently from lighter particles when experiencing the same external influence, and even though the mass difference between the two isotopes of uranium is very small, it is possible to exploit this to separate them from each other. In the *gas centrifuge* method, gas molecules containing U-238 will concentrate closer to the centrifuge wall than the molecules containing U-235. The *gaseous diffusion* method exploits the fact that heavier molecules with a certain energy move slower than lighter molecules with the same energy and therefore will diffuse more slowly through a semi-permeable membrane. The *mass spectrograph* separates the isotopes because particles with a different charge-to-mass ratio will move differently in a magnetic field. Even chemical properties of a molecule may be affected by the mass of the atoms it consists of.

Much of the information below is from Reference [2].

The separation elements in an enrichment facility and also the full facility may be described in a general way. In the following discussion some common factors of the various enrichment facilities will be described, and the elements may be considered as black boxes (it does not matter which enrichment technique is used). The *feed*, the form of natural uranium or uranium enriched to a lower degree than what is desired, is entering at one end, and the *product* (the enriched uranium) and the *tails* or *waste* (the depleted uranium) emerge at the other end. This terminology is also used for the entire facility. The *separation factor* of an element is the ratio of the relative amount of U-235 in the product to the relative amount in the tails, and is therefore a measure of the efficiency. Separation factors are just above 1 for each step in the most common enrichment methods such as gaseous diffusion and gas centrifuges.

Enrichment methods with low separation factors have been made effective by organizing the elements into *stages* and *cascades*. A stage is comprised of one or more elements that are organized in parallel; this means that they all receive their input from the same source and contribute to the same output product. A cascade consists of a number of stages organized in a series; this means that the product of one stage is used as feed for the next and that the waste may be returned to the previous stage or earlier (known as *reflux*). The cascade is called *ideal* if the

¹ Uranium has a very high density. Furthermore, it is pyrophoric, that is, it ignites and burns as it penetrates the armour.

arrangement is such that streams of uranium of different enrichments are never mixed. This minimizes the ratio between energy input and product gained from the cascade. Ideal cascades also have the shortest *equilibrium time* (described below) for a given product enrichment. The downside of an ideal cascade is that all the stages must be of different size and use a different amount of power, so construction of an ideal cascade is not economical. In practice a compromise between what is feasible to construct and idealization is used.

It is also possible to use *batch recycling* to enhance the level of enrichment of the uranium, this means that enriched material is run through the enrichment facility one or more times extra to increase the enrichment level. Which properties of the enrichment facility make this a good or a bad idea will be mentioned below. Yet another way of increasing the enrichment level of the product is to use *stretching* [1]. When stretching is used there is a large reflux and the product is extracted at a very low rate. This gives a high percentage of U-235 in the product and a very low percentage of U-235 in the tails at the expense of efficiency and production rates. Batch recycling or stretching are methods which may be used to enrich to a higher degree than originally intended for the enrichment facility. If a country already has a plant for enriching uranium for use in nuclear power reactors and subsequently decides to produce HEU, then these methods could be used instead of rebuilding the plant or building a new one.

Some factors that are general and therefore suitable for comparing different enrichment technologies are the *equilibrium time*, the *inventory*, and the *kilogram separative work units* (SWU^2). When a cascade is started, all the stages are filled with material of the same isotopic composition; therefore it takes a while before the product reaches the desired enrichment. Until this point is reached, the cascade is operating in total reflux (no product or tails are extracted). The equilibrium time is a measure of the time it takes from the initial start-up until any product can be extracted. To use batch recycling to gain a higher level of enrichment, the whole cascade must be emptied, refilled and started up again, so the equilibrium time is an important factor in determining whether this is suitable. The inventory (the amount of uranium it takes to fill the cascade) is of course also important in this discussion. To reach weapons grade uranium from LEU by batch recycling, the batch will have to be recycled a few times. Batch recycling can be wasteful in uranium because some material will be lost in each recycling.

The kilogram separative work units that a facility can produce per year depend on the feed flow (*the throughput*) and the separation factor in each stage. The unit is useful for comparing different technologies when there are many stages and a small separation factor per stage, for example in the case of centrifuges and gaseous diffusion. If the percentage of U-235 in the feed, the product and the tails are all set, then the number of SWUs produced by the facility is directly related to the amounts of product, feed and tails. For instance, in order to produce 25 kg of 93 % U-235, 5000 SWU and 5050 kg of natural uranium feed is needed if the tails contain 0.3 % U-235 [1]. In commercial processes, the costs are generally directly related to the number of SWUs, and so this has come to be a commonly used parameter for describing enrichment facilities.

² The kilogram separative work unit is abbreviated kg SWU or just SWU [1].

When enriching uranium, one important aspect is *criticality*, the amount of enriched uranium may become large enough to sustain a nuclear chain reaction (see Section A.2). For low-level enrichment, criticality is not a problem because the concentration of U-235 will not be high enough. When producing HEU, the issue is present; it is important not to reach critical mass or critical density. If the inventory is large, this problem is more prominent than if the inventory is small, either way pressure must be controlled as to not reach critical density.

The most common uranium compound utilised in enrichment facilities is *uranium hexafluoride* (UF₆) gas (see Section A.3). This gas is highly corrosive; therefore all the parts of machines that come into contact with the gas must be corrosion resistant. Uranium hexafluoride is used in the gaseous diffusion method, the gas centrifuge method, thermal diffusion, aerodynamic processes and molecular laser isotope separation. Uranium hexafluoride gas is a good molecule for uranium isotope separation because the molecule is simple, and the mass difference between the uranium atoms is the only source of mass difference between the molecules (fluoride has only one stable isotope). Uranium hexafluoride is also preferred because of the extensive experiences accumulated by working with it (it has been used for the enrichment of uranium since the early 1940s). Other techniques employ uranium in the form of *uranium tetrachloride* (UCl₄) or in its pure metallic form.

3 Gaseous diffusion

3.1 The main principle

This method indirectly uses the different masses of the isotopes. If a container is filled with gas and the walls are *semi-permeable* membranes, then the molecules in the gas will have a small probability of penetrating the walls. These walls will be referred to as *barriers* throughout this chapter. Molecules with a comparatively high speed will encounter the membranes more frequently than the slower ones and therefore have a larger probability of penetration (see Section A.4). And since the lighter molecules on average move faster than the heavier ones, the gas on the other side of the wall will be enriched in the lighter molecules. In principle this process is very simple, but it is not very efficient because the mass difference between the molecules is so small. To make the diffusion go faster, a pressure difference is set across the membrane.

The gaseous diffusion technology is relatively simple, but not very effective. The quality that can make gaseous diffusion interesting to a new participant in the enrichment industry, is that the technology has been proven to work on an industrial scale. The gaseous diffusion method employs uranium in the form of uranium hexafluoride gas (see Section A.3).

3.2 The enrichment plant

Each stage in the gaseous diffusion process increases the fraction of U-235 by very little (the separation factor is about 1.004), but contains a large amount of gas. This makes the process less flexible. Batch recycling is not a good option for increasing the level of enrichment, but stretching

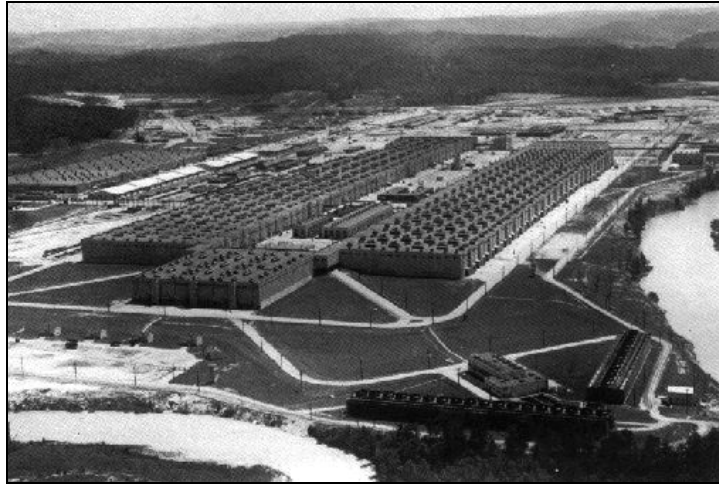


Figure 3.1 The gaseous diffusion plant built at Oak Ridge, Tennessee, in the United States during World War II. Photo courtesy of the United States Department of Energy.

the cascade may be an alternative. If stretching is to be used in a gaseous diffusion plant, then precautions to prevent the possibility of a criticality accident are necessary to take. This also goes for batch recycling. More than 3500 stages is needed to produce weapons-grade uranium from natural feed, and there is an equilibrium time of more than a year [1;3].

3.2.1 The separation stage

The main components of a single gaseous diffusion stage are [4]:

- A large cylindrical vessel called a *diffuser* or a *converter* that contains the barrier;
- A *compressor* used to compress the gas to the pressures needed for it to flow through the barrier;
- An *electric motor* to power the compressor;
- A *heat exchanger* to remove the heat of compression;
- *Piping* and *valves* for stage and inter-stage connections and process control.

The key parts in the gaseous diffusion method are the barriers and the compressors. This technology seems to be relatively simple to develop. It was done by the United States in the 1940s, and reportedly every country that has tried since has been able to do it [5]. When the gaseous diffusion method was first developed, the corrosive nature of uranium hexafluoride was one of the major obstacles[1].

There must not be any leakage in the system letting uranium hexafluoride gas out or air in. Metals such as aluminium and nickel are used for the surfaces that are exposed to the gas, for example piping and compressors, because they are corrosion resistant. Since there is little enrichment in just one stage of a gaseous diffusion plant, the cascades are long. Also the equipment tends to be rather large, so a gaseous diffusion plant takes up a lot of space. When the gaseous diffusion plant at Oak Ridge was built in the early 1940s (see Figure 3.1), it was the largest industrial building in the world [6]. Support facilities for a gaseous diffusion plant would include a large electrical power distribution system, cooling towers, a fluorination facility, a steam plant, a barrier production plant, and a plant to produce dry air and nitrogen. [4]

In a typical stage the feed enters at a pressure of one-third or one-half atmosphere. Only half of the gas is allowed to diffuse through the barriers; this gas will have a lower pressure, and it is sent through a heat exchanger to be cooled. Then the gas is compressed and mixed with depleted gas from a higher stage. The cooling is necessary because the compression will substantially increase the temperature of the gas. This energy-demanding process is the main source of the high power consumption of the gaseous diffusion process. Modern compressors are similar to those used in jet aircraft engines. The compressor rotor rotates at high speed and must handle the corrosive uranium hexafluoride gas at high temperatures in continuous operation for many years. [3]

3.2.2 The diffusion barrier

The barrier must meet some requirements:

- The size of the pores must be very small, but not so small that the diffusion rate gets too low, and they have to be of a uniform size. The diameter should be much smaller than the average distance travelled by the molecules between collisions with other molecules (determined by the pressure of the gas), but still large enough that the molecules are able to penetrate the barrier at a reasonable rate. About 25 nm would be appropriate if the gas is kept at 80 °C and a pressure of about one half of an atmosphere [3].
- The barrier must be thin enough for sufficient permeability, but must be thick enough to withstand the pressure difference across it. The material of the membranes must be bonded under high temperatures and pressures into thin sheets, and these sheets must be able to withstand a pressure difference of 0.3 to 0.5 kg/cm² for a long time (years). This can be achieved by using many layers of different porosities and strengths [3]. The diffusion barrier is typically just 5 mm thick [6].
- It must be highly corrosion resistant, so that the holes are not plugged [3]. Typical materials are nickel and aluminium oxide [4].

The actual methods for producing barriers are kept classified by the countries that use the technique. Japanese researchers have experimented with nickel, aluminium and teflon barriers. The United States uses sintered³ nickel powders while in the French Tricastin plant the barriers are ceramic. The efficiency of the barriers is about 1 SWU/y per square meter of barrier. [3] There has been success in using barrier tubes made from anodized⁴ aluminium which is usually considered too fragile for use in an industrial enrichment plant [1].

To increase the efficiency of the method, the surface of contact between the gas and the barrier should be as large as possible; this can be done by shaping the barriers as thin tubes. The barrier tubes must be thin to provide a large contact surface with the gas, but not so tight that it hinders the gas flow. It is assumed that the tubes are about 2 m long and 1 cm in diameter. This means that 160 000 such tubes are needed in a stage with the capacity of 12 000 SWU/y. [3]

³ Sintering is a process in which a powder is compacted and heated to a point below its melting point; the particles then attach to each other without melting together.

⁴ The thickness of the natural oxide layer on the surface is increased; this makes the metal more corrosion resistant.

3.2.3 Energy consumption

The gaseous diffusion method consumes a lot of energy, about 2500 kWh/SWU. The energy consumption was more than ten times this level when the process was first developed, but it decreased by a factor of ten in about ten years of further development. One method for dealing with the expenses arising from the high energy consumption, which is inevitable in the gaseous diffusion method, is to use the plant only in periods of lower electricity prices. This has been done in France. [1]

3.3 When and where has the technique been used?

The gaseous diffusion method was first used in the United States during World War II, but has since been used in the United Kingdom, France, China, Russia and Argentina as well. Since gaseous diffusion is an out-dated and highly power-consuming process, it is not in wide use anymore. Today only the United States and France use gaseous diffusion on any significant scale [7].

4 The gas centrifuge method

4.1 The main principle

This method employs uranium in the form of uranium hexafluoride gas (see Section A.3) and utilizes the mass difference between the isotopes. It is common knowledge that when the only influence on an object is gravity, it will fall toward the ground. Also if there are several objects, like grains of sand for instance, in a container they will arrange themselves so that there will be larger density closest to the ground (the smaller grains will be on the bottom and the larger ones that cannot be packed as compactly will be on top). This also applies to molecules in a liquid or a gas. Just like the grains of sand, the molecules of uranium hexafluoride gas made with the heavier uranium isotope (U-238) will tend to move toward the bottom, and the lighter molecules will tend to move toward the top because all the molecules take up the same amount of space, and the ones with the higher mass will therefore also have the higher density. This process is very slow, and since there is so little U-235 in natural uranium, just filling uranium hexafluoride in a container and waiting for the molecules to separate into two layers will be very ineffective. Instead it has proved efficient to construct a fake gravitational field that is much stronger than the earth's field by putting the gas in a container and spinning it very fast. The molecules will then be exposed to the same *centrifugal force* that we feel when we are driving around a corner, and by spinning the centrifuge faster, we can make the force stronger, and the separation of the isotopes will happen quicker. If we use a cylindrical container, we could then in the ideal case extract the enriched gas from around the centre (the rotation axis) and the depleted gas from near the cylinder wall. But because the molecules in a gas also perform a random motion (see Section A.4), and all of the gas will be pressed close to the wall, separating the two isotopes is not so simple, and they cannot be separated completely.

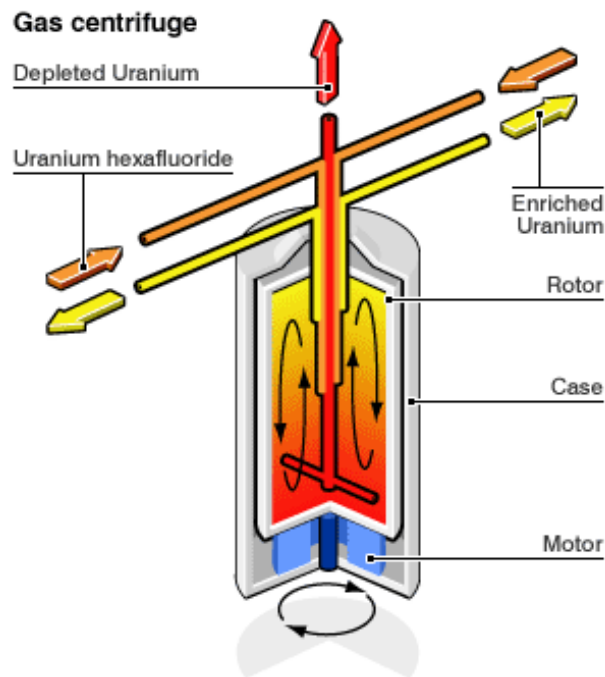


Figure 4.1 Sketch of a gas centrifuge. One can see the pattern of the motion of the gas inside the rotor tube. Here the lower part of the centrifuge is warmer than the upper part so that the enriched gas will settle in the upper part of the centrifuge. Reprinted with permission from BBC News.

To increase the separation factor, a relatively slow *counter current* is set up in the centrifuge. This is often set up by the same principle that makes it possible to fly a hot air balloon: hot gas rises. If the temperature is higher in one end of the centrifuge than in the other, the gas in the hot end will move toward the rotational axis since this is “up” in this “gravitational field” while colder gas from elsewhere in the centrifuge fills the void, and the gas in the cold end will move toward the wall since this corresponds to “down.” In this way a convection current flow pattern will emerge (see Figure 4.1). As can be seen from the figure, the gas closest to the rotational axis will flow toward the cold end, and the gas closest to the centrifuge wall will flow toward the hot end. Since the molecules containing the U-235 isotope will be closer to the centre than the molecules containing the U-238 isotope on average, there will be an accumulation of molecules containing the light isotope in the cold end, and the gas here will be enriched in U-235. The same flow pattern can be supported mechanically by use of scoops. Instead of (just) making use of the temperature difference, the gas is scooped toward the centre in one end and toward the wall in the other end.

4.2 Technical challenges

4.2.1 Building the centrifuge

From the above discussion, it is obvious that the longer the centrifuge tube and the higher the rotation velocity, the greater the separation factor. But there are some problems related to increasing the velocity and the length of the tube that have to be overcome. There will be vibrations and flexing of the rotor tube as the rotation velocity increases, and this limits the length

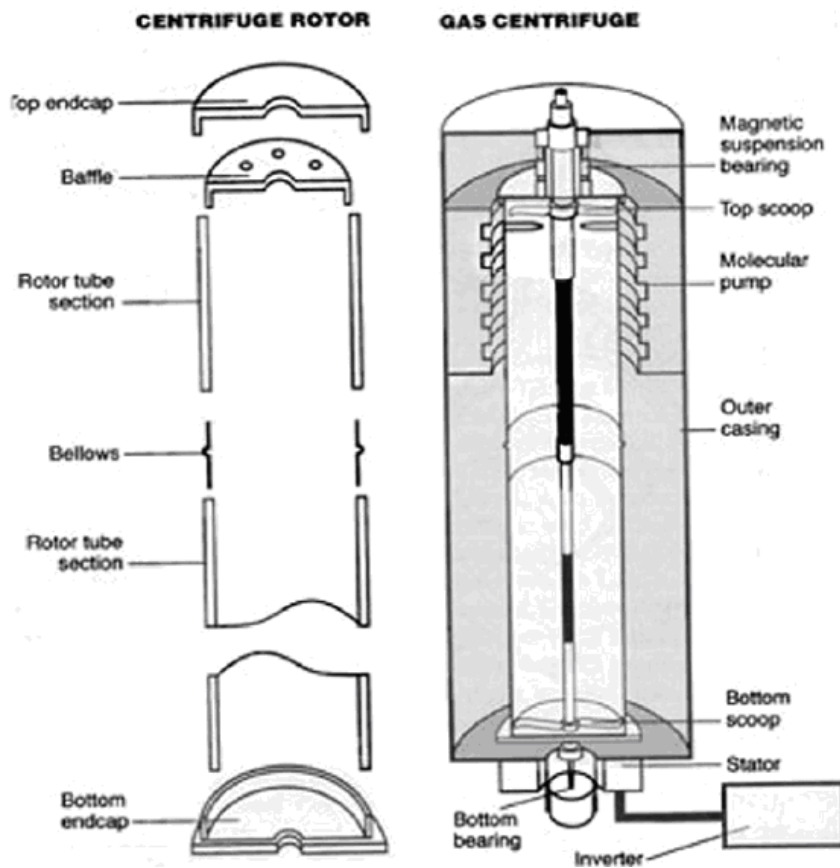


Figure 4.2 Sketch of the different parts of a centrifuge. Copyright © 1992 by the Bulletin of the Atomic Scientists, Chicago, IL 60637. Reproduced by permission of the Bulletin of the Atomic Scientists: The Magazine of Global Security, Science, and Survival.

of the tube since it is impossible in practice to balance the tube perfectly. A long thin rotor will have certain characteristic vibrations, so flexion at the corresponding “resonant” speeds may cause the rotor or the bearings to break. Therefore passing these “resonant” speeds will be difficult. Centrifuges which rotate faster than these resonant speeds, and therefore have to go through them in the acceleration process, are called *supercritical*. Centrifuges which rotate slower than the first resonance speed are referred to as *subcritical*, and are not subject to the same amount of stresses and strains as the supercritical centrifuges, but are also less efficient. The rotor tube must be made from a high-strength material, and the vibrations can also be reduced by reducing the friction force on the rotor. The following will be a discussion of the different components that are needed to build a centrifuge, and the technical challenges connected to them. One obvious challenge is the corrosiveness of uranium hexafluoride. The components that come in direct contact with the gas must be made of or lined with corrosion resistant materials.

As can be seen from Figure 4.2, the centrifuge mainly consists of a rotating tube which contains the gas. This tube is suspended inside a casing and driven by a motor. The main components of a centrifuge are:

- A rotor tube
- Bellows
- Top and bottom end caps

These are welded to the rotor tube to contain the uranium hexafluoride. The top cap supports

part of the upper bearing and the bottom cap carries the rotating part of the bottom bearing. [8]

- *An outer casing*

- *A vacuum system*

There must be a pumping system to create and maintain vacuum between the rotor and the casing.

- *Suspension and bearings*

The suspension system keeps the centrifuge in place.

- *Scoops*

The scoops are small tubes which extract enriched and depleted gas, respectively, in either end of the rotor interior, and they are relatively easy to make. The scoops may be stationary while the centrifuge rotates around them. They help amplify the convection flow pattern in the centrifuge and create a pressure difference which makes it possible to extract the product and tails without using compressors. [1;8]

- *Motor system*

- *Pipes*

Various piping systems are necessary to transport gas of different enrichment into and out of the centrifuge.

4.2.1.1 Rotor

The maximum speed of the rotor depends on the physical strength of the tube material, and since most of the stresses on the tube come from its own weight, the tube material must have a high strength to weight ratio [9]. Suitable rotor materials include alloys of aluminium (alloys containing zinc, magnesium and copper are the strongest [9]) or titanium, maraging steel⁵ or composites reinforced by certain glass, aramid or carbon fibres. With maraging steel, the maximum rotor wall speed is approximately 500 m/s, while carbon fibre-resin composites allow speeds in the range of 700–800 m/s. The aluminium alloys used in early centrifuges permit maximum speeds of about 350 m/s. Maraging steel is much more difficult to work than aluminium. Currently, manufacturing carbon fibre-resin composite machines is beyond the technical capability of states of particular proliferation concern. [10]

The load on the bearings of a spinning centrifuge tube can be reduced by keeping its weight as low as practically possible, requiring that the walls of the rotor be as thin as possible, typically a millimetre or so. [9]

4.2.1.2 Bellows

A low-tech solution to the problem of flexural vibrations with long rotors is to break up the length of the rotor with short, thin flexible joints called bellows, located so as to absorb the energy of the vibrations at the resonant speeds. While this method is still used in relatively short aluminium and maraging steel centrifuges, longer, faster carbon fibre-resin composite machines instead use

⁵ Maraging steel is an alloy of iron containing less carbon than other steels, but a large amount of nickel. High strength, low weight and corrosion resistance are some of the properties making maraging steel suitable as a rotor material.

special drives that can accelerate the rotor rapidly through the resonances before the amplitude of the vibrations grows sufficiently large to damage the machine. This technology is quite sophisticated. [10]

The bellows act as a spring, and therefore they make it possible for the rotors to pass the resonance frequencies without being torn apart. The subcritical centrifuges have short rotor tubes without bellows [1]. Making the bellows out of maraging steel is one of the difficulties in manufacturing a centrifuge.

4.2.1.3 Outer casing

The rotor is suspended inside an outer casing, and the space between the rotor and the casing is evacuated to reduce friction as much as possible. The casing must therefore be air-tight. Furthermore, the casing also serves the purpose of containing the parts of the centrifuge in case the centrifuge breaks down, so it must be made of a high-strength material (the rotor moves with a very high velocity).

4.2.1.4 Suspension and bearings

The suspension system must be capable of damping some amount of vibration because it is impossible to balance the rotor perfectly to avoid all vibrations [4]. Achieving such precision is becoming easier as modern, numerically controlled machine tools continue to improve [9].

The top bearings can be made out of ring magnets. This way there does not have to be contact between the rotor and the suspension on the one hand, and the pipes that carry the uranium hexafluoride into the rotor on the other hand. The bottom bearing may be a needle with an oil damper to absorb some of the vibrations. These can be designed such that the rotor acts as a self-stabilizing spinning top. Manufacturing the top and bottom bearings have constituted some of the greatest challenges to countries trying to build centrifuges. [1]

4.2.1.5 Motor system

AC motors⁶ are used, and they provide a centrifuge speed that is proportional to the frequency of the supplied current. [8] The power supply must accept alternating current input at the 50 Hz or 60 Hz line frequency available from the electric power grid and provide an AC output at a much higher frequency (typically 600 Hz or more). The high-frequency output from the frequency changer is fed to the high-speed gas centrifuge drive motors. The centrifuge power supplies must operate at high efficiency, and provide precise control for the output frequency. [4]

4.2.2 Energy consumption and space requirements

The electrical consumption of a gas centrifuge facility is much less than that of a gaseous diffusion plant. An energy need around 100–300 kWh/SWU was claimed in 1983 [11], but in 2008 it was claimed to be as low as 50 kWh/SWU [7]. Consequently, a centrifuge plant will not

⁶ That is, motors that are powered by alternating current (AC).

have the easily identified electrical and cooling systems typically required by a gaseous diffusion plant.

A typical centrifuge facility appears to have a capacity of 10–20 SWU/m², and a facility capable of producing one bomb per year would then require about 600 m² of floor space [12].

4.2.3 Feasibility for production of HEU

The throughput of a single centrifuge is small, which leads to rather small separative capacities for typical proliferator centrifuges. The separation factor of a centrifuge is about 1.5. To be able to produce only one weapon per year, several thousand centrifuges would be required. [4]

Centrifuges have a small equilibrium time and a small in-process inventory. The inventory is small because the more gas there is in a centrifuge and the faster it spins, the higher the pressure near the wall will be. And uranium hexafluoride may then desublime or liquify. Centrifuges are flexible when it comes to changing the cascade design (parallel to series) [12]. This makes it easier to change the level of enrichment of the product, and the small equilibrium time and inventory make batch recycling more feasible. The centrifuge process is economic on a smaller scale, under 2 million SWU per year, which also enables staged development of larger plants. [7]

4.3 When and where has the technique been used?

Today, the gas centrifuge method is taking over for the gaseous diffusion method as the most important method for uranium enrichment. Plants are being built in all parts of the world, and in almost all countries that enrich uranium.

The development of centrifuge technology for the separation of isotopes started in the mid 1930s in Germany. The first known separation of uranium isotopes using a gas centrifuge was performed in 1941, and the research was conducted by Professor Jesse W Beams and his colleagues at the University of Virginia. [1;13]

The Soviet Union started developing the gas centrifuge in 1946. The work was conducted in part by captured German scientists; one of them was Gernot Zippe. In 1956, the Germans were allowed to leave the Soviet Union. Zippe received permission from the Soviet Minister of Atomic Energy to work freely on centrifuges in the West in 1958. Zippe contributed to the development of advanced Western centrifuges and the formation of the uranium enrichment company Urenco. The first industrial-scale plant in the Soviet Union was built in 1959. [1]

China started research and development of gas centrifuges in 1958, and Chinese officials have stated that the country may build a gas centrifuge enrichment plant to replace their gaseous diffusion plants. Russia has agreed to provide China with a gas centrifuge plant. [1]

In Europe, the company Urenco has gas centrifuge facilities in Germany, the Netherlands and the United Kingdom. Urenco's present German enrichment plant has been in operation since 1985.

India also masters the technique of centrifuge separation of isotopes [14], and it is believed that research on gas centrifuges has taken place in India since the early 1970s or even earlier [1].

The Pakistani scientist Abdul Quadeer Khan was the long-time head of the Pakistani Engineering Research Laboratories which worked on uranium enrichment after he had worked until 1976 for a subcontractor to Urenco. He is assumed to have brought with him to Pakistan designs and assembly instructions for centrifuges and lists of suppliers used by Urenco. The information was used in Pakistan's nuclear programme, and has later been sold along with hardware to other countries. Khan built a whole network trading in enrichment and nuclear bomb technology. Khan has admitted selling to Libya, Iran and North Korea, but there are suspicions that he has also sold his services to Iraq, Syria and other countries. [15]

Iraq pursued centrifuge technology in the 1980s. Valuable mechanical design information of an oil centrifuge were unclassified and available to the Iraqis, and this is what they started working on. The fact that key parts are moving at great speeds makes centrifuge technology difficult for inexperienced engineers. In 1988 Iraqi centrifuge experts approached the German firm H&H Metalform GmbH, for help. H&H was well connected to European centrifuge experts, and the company became Iraq's most important contact for assistance in obtaining expertise and manufacturing equipment from abroad. Iraq received designs for Zippe centrifuges in 1988 through this contact. Iraq was able to import raw materials, motors and frequency converters from Europe in sufficient quantities for research and development. [1]

Israel may have pursued gas centrifuge technology at least as a research activity, and may also have expanded on this work [1]. Japan also uses gas centrifuges for uranium production [16].

Urenco received a licence to build a plant in New Mexico, USA, in 2006. This plant was expected to be on line in 2009 [17]. The United States does not have centrifuge enrichment on an industrial scale yet, but USEC⁷ began construction of a plant in Ohio in 2007. Commercial plant operation is expected in 2010 [18].

5 Thermal diffusion

5.1 The main principle

The thermal diffusion process is based on the same principle as the counter current flow pattern in the gas centrifuges, and employs uranium in the form of uranium hexafluoride (see Section A.3). In this process a thin container of gaseous or liquid uranium hexafluoride is oriented vertically (see Figure 5.1). One side is then heated and the other side is cooled, and this gives rise to a thermal convection current with an upward flow along the hot surface and a downward flow along the cold surface. Under these conditions, the lighter U-235 gas molecules will diffuse toward the hot surface, and the heavier U-238 gas molecules will diffuse toward the cold surface.

⁷ United States Enrichment Corporation

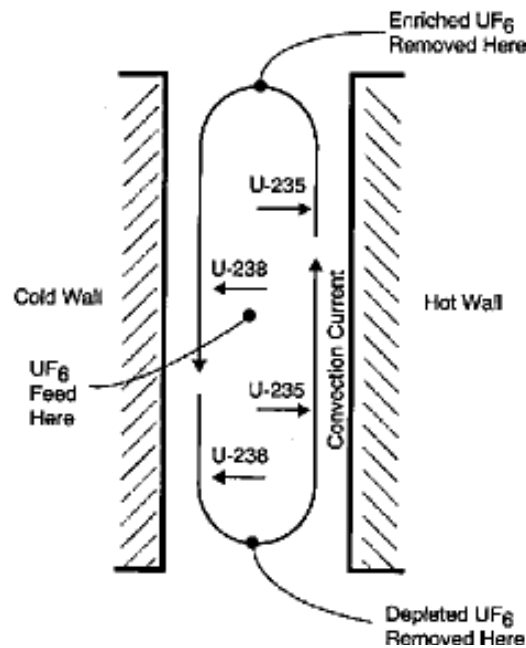


Figure 5.1 Cross section of the film of uranium hexafluoride used in the thermal diffusion method. Illustration courtesy of the United States Department of Energy.

The result of these motions is that the U-235 molecules concentrate at the top of the film, and the heavier U-238 molecules concentrate at the bottom. [4]

5.2 When and where has the technique been used?

The thermal diffusion method was one of many methods for enrichment of uranium pursued by the United States during World War II. It was not found to be a very good method then, and there is no sign of it having been used since. The process has a low capital cost, but high heating requirements. It is about 140 times less energy efficient than the gaseous diffusion process [4].

The United States used thermal diffusion to provide slightly enriched feed material for the electromagnetic separation on the calutrons (see Section 6.1). The film was an annular region between two cylindrical pipes. The inner pipe was heated by steam, while the outer was cooled by water. The uranium hexafluoride was kept under pressure to stay in liquid form. The process was very slow, and only low enrichments were gained even in tubes as high as about 15 m. Still, some 2000 such tubes were constructed to provide feed of 0.89 % U-235 for the calutrons. [19]

6 Electromagnetic processes

The electromagnetic processes for the separation of uranium isotopes all have in common that they make use of uranium *ions*⁸. Ions of different mass but equal charge will behave differently in a uniform *magnetic field*. The first process, known as EMIS, is relatively simple in theory, and the main challenge is that it is relatively slow. The two other methods are plasma processes. A

⁸ Ions have a different numbers of electrons than they have protons; this means that they have a net electric charge.

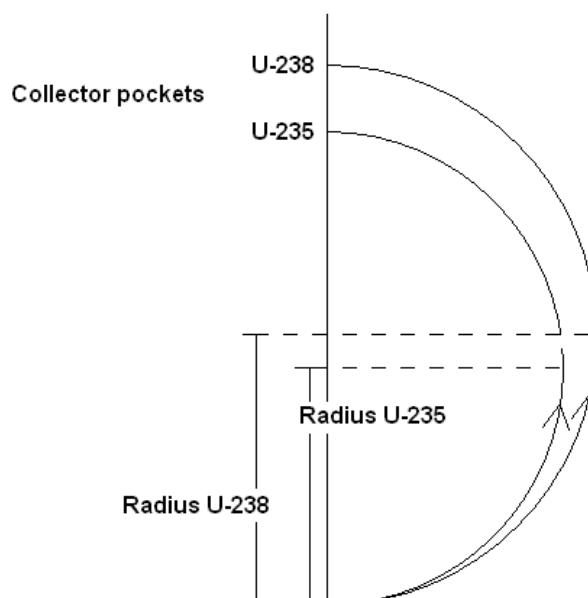


Figure 6.1 Simple drawing of a mass spectrometer. The particles enter in the bottom with the same kinetic energy and the same electric charge, and the beam will then split in two because of the different masses of the isotopes. The magnetic field points into the paper.

plasma is a gas consisting of electrically charged particles. The plasma processes are all fairly complicated theoretically because the physics of plasmas is rather complicated.

6.1 Mass spectrometer: EMIS (calutron)

6.1.1 The main principle

In a uniform magnetic field, a charged particle with a velocity component orthogonal to the field will follow a *circular* or *helical path*. If it has no velocity component along the field, the trajectory will be circular in the plane orthogonal to the magnetic field; this is the case in the mass spectrometer. The radius of the circular path is determined by the strength of the field, the electric charge, the kinetic energy of the particle and the particle's mass. A beam of uranium ions with equal charge and kinetic energy being sent into such a uniform magnetic field will split into two beams because of the different masses of the isotopes. The beams will form circles, and the one containing U-238 will have a larger radius than the one containing U-235. This is illustrated in Figure 6.1. The isotopes can be collected for example after half a rotation in the field. The mass spectrometer method is often referred to as EMIS (electromagnetic isotope separation), and the spectrometer is referred to as a *calutron*, which is what the American machine developed at the University of California in the early 1940s was called [20].

6.1.2 Equipment and efficiency

In principle, a mass spectrometer consists of a *large magnet* that can provide a uniform magnetic field at a sufficient strength, a *vacuum pump*, an *ion source* and *collectors*. The ions of the two uranium isotopes U-235 and U-238 differ in mass by 1.27 %, and they will have radii that differ by only 0.64 %. [20]

The uranium ions can be made from uranium tetrachloride (“green salt”). Solid uranium tetrachloride is electrically heated to produce uranium tetrachloride vapour. The vapour molecules are bombarded with electrons, and this knocks loose U^+ ions. The ions are then accelerated over an electric potential to high speeds and follow circular paths in the magnetic field. Because they are accelerated over the same potential they acquire the same kinetic energy. [4]

The EMIS process has a very high separation factor (somewhere between 30 and as high as 80 000), but a very low throughput. It also has a very high energy consumption (3800 kWh/SWU for the early machines, but this value may be lower today). [20]

One limitation is the ion beam intensity because the charges on the ion beam particles will repel each other. To avoid this problem, the ions can be allowed to collide with a background gas so that electrons are released and neutralize the repulsion, but if the gas is too dense and the collisions became too frequent it would disturb the ion beam too much. The typical currents are a few hundred milliamperes, and at this rate it would take more than 400 years to collect a critical mass. [20]

Another limitation is the fact that only a small amount of the feed material can be converted into product in one run. Only about 50 % of the uranium tetrachloride feed is converted to uranium ions, and only about 50 % of the ions are actually collected in the end. This is a problem if valuable partially enriched uranium is used as feed, and the insides of the vacuum chamber have to be cleaned so the material can be recovered. This is a time-consuming process which decreases the efficiency. [4]

Note that EMIS is an isotope separation method which can be used also to separate the isotopes of plutonium [20].

6.1.3 When and where has the technique been used?

The method was used, among others, by the United States during World War II to enrich uranium for their uranium bomb, and the machines were called “calutrons”. Because of the low throughput, over 1000 calutrons were used to help produce uranium for the first American bomb. This cost roughly a billion dollars over three years [20]. The enrichment was done in two stages; in the first stage natural or slightly enriched uranium was used to increase the enrichment to 12–20 %; and in the second stage the product from the first stage was further enriched to weapons grade. EMIS could be used to produce limited amounts of weapons-grade uranium, and it may be especially attractive for further enrichment of partially enriched material. The United States, the United Kingdom, France, the Soviet Union and China all tried this enrichment method. It was also pursued by the Iraqi uranium enrichment program, and this happened after the process had been discarded by other countries. The process is both energy demanding and labour intensive, and it is not economically competitive with other technologies. [4]

The United States declassified much of its EMIS technology after World War II, and this is well documented in open literature. Also the basic scientific and technical problems are not too hard to master, the feed material is relatively easy to produce and handle, and if LEU is used as fuel the productivity increases substantially. Also, when Iraq decided to use EMIS in the early 1980s, the computational software and the main equipment were not on any of the international export control lists, and the prototypes for the main equipment could be designed and manufactured in the country. Another advantage is that the EMIS technology does not require moving parts at high speeds but rather static equipment which makes it easier to use for engineers without special experience. [1]

6.2 Ion cyclotron resonance

6.2.1 The main principle

This process employs the same principle as used in the mass spectrometer, only this time the particles also have a velocity component along the magnetic field. The motion of the charged particles becomes a helix with its axis along the magnetic field. The frequency of the rotation is determined by the strength of the magnetic field and the mass and charge of the particles, and is called the cyclotron frequency. If the uranium ions have the same charge, then the U-235 ions will have a larger cyclotron frequency than the U-238 ions. If the ions are subjected to an *oscillating electric field* in the plane orthogonal to the magnetic field, which has the same frequency as the *cyclotron frequency*⁹ of the U-235 ions, then their energy will increase (the ions are *excited*) and the radius of the helix for these ions will increase while the radius of the U-238 ions remains about the same. A typical unexcited ion will have a helix radius of the order of a millimetre, but excited U-235 ion can be made to move in circles several centimetres in diameter [20]. The ions with the larger radius in their orbit can be collected by plates parallel to the helix axis while most of the ions which still have a small radius can pass without being caught by the plates.

6.2.2 How it is done

The ions can be produced from a feed plate of solid uranium. The atoms are vaporized by bombarding the plate with energetic ions in a process called sputtering. A microwave antenna located in front of the plate, energizes free electrons which collide with neutral uranium atoms in the vapour sputtering off the plate. This in turn displaces electrons from the uranium atoms and produces a plasma of U-235 and U-238 ions. The ions then move in their helical paths toward the collector inside a cylindrical vacuum chamber. The magnetic field is produced by a *super conducting magnet* located around the outside of the chamber. [4]

The strength of the magnetic field is about 2 T which gives the U-235 ions a cyclotron frequency of 130 kHz. The frequency of the oscillating electric field must be stable to an accuracy of 1 % or better to distinguish U-235 from U-238. It is difficult to produce a magnetic field which is uniform to better than 1 % over a large area. Modern magnet technology can achieve this goal

⁹ The number of rotations in the helix done by the ions per second.

only with relatively sophisticated and expensive solenoid designs. Because plasmas consist of charged particles, they shield electric fields. The characteristic shielding length of uranium plasma is a few micrometres. This means in practice that even though the electric field can be applied in different ways, it must be applied indirectly. [20]

To separate the two isotopes properly, the ions must be sent through several revolutions. The influence of the electric field on the U-238 ions will average to zero while the U-235 ions will get a substantially larger radius. There should be about 80 cyclotron periods before the ions reach the collector. If the separation tube is 1 m long this means that the axial velocity of the ions cannot be more than 1 600 m/s which corresponds to a temperature of about 75 000 K.¹⁰ This again means that the temperature of the plasma has an upper limit for the method to work. When the ions collide, they will be thrown out of phase with the electric field and loose energy. This puts a limit on the density of the plasma; the ion density must be below 10^6 ions per m^3 . This means that with a beam diameter of about a metre, using natural uranium as feed and assuming that all the U-235 ions are collected, then about 70 kg of U-235 could be collected per year. [20]

The process needs large amounts of liquid helium¹¹ to maintain the magnets in their superconducting state [20].

The process is a batch process, and has to be repeated to produce HEU from natural uranium feed. The enrichment factor is high, and the energy consumption of the process is quite low, about 330 kWh/SWU. [1;4]

6.2.3 When and where has the technique been used?

The only countries known to have carried out serious experimental programs are the United States and France. The technique became a part of the Advanced Isotope Separation research and development program of the American Department of Energy in 1976, but development was stopped in 1982. The French developed their own version, and this program ended around 1990. [4]

6.3 The plasma centrifuge method

The information in this section is mainly from “Uranium Enrichment and Nuclear Weapon Proliferation” [20].

In the plasma centrifuge method, a plasma is set into fast rotations. As in a gas centrifuge, the heavier isotopes will gather closer to the wall of the centrifuge than the lighter isotopes. A main difference between the gas centrifuge and the plasma centrifuge is that the plasma container does not have to spin like the rotor in the gas centrifuge. This is a feature of the plasma centrifuge method which makes it easier to operate at a high rotational velocity. The bearings will not wear

¹⁰ The Kelvin temperature scale has the same unit size as the Celsius scale, but it starts at absolute zero. This means that water freezes at 273 K.

¹¹ Liquid helium has a temperature of 4.2 K (-268.9 °Celsius) at atmospheric pressure.

out and the walls do not have to withstand the same amount of tension as in the rotating gas centrifuge. But as the rotational speed increases in the plasma centrifuge, energy consumption and problems with collisions between ions and neutral particles increase.

The plasma is made to rotate by creating an electric discharge perpendicular to a strong magnetic field. As in the EMIS and the ion cyclotron resonance processes, the magnetic field makes the charged particles move in circles. By adding a *radial electric field* all the particles in the plasma (independent of whether the charge is negative or positive) will rotate in the same direction and very high speeds can be attained. In the rotating plasma, the different isotopes will have a velocity relative to each other. In collisions between ions, the heavier isotopes tend to gain speed and the lighter to slow down. This increases the velocity difference between the isotopes and enhances the separation effect.

The energy consumption of the plasma centrifuge method has been said to be about ten times that for gaseous diffusion, and even though this probably can be reduced, it may still be larger than that of mechanical centrifuges. One suggestion for reducing the energy consumption is to use a partially ionized plasma, then the rotation must be transferred to the neutral gas by collisions.

The separation factor of a plasma centrifuge is not too great, but in some designs it may be possible to enhance it by setting up a counter current flow pattern as in the gas centrifuges.

Several countries have researched plasma centrifuges, also for separation of isotopes of other elements than uranium. There are a number of variations in the design, but no one has been used for enrichment of uranium to any degree worth mentioning.

7 Aerodynamic processes

There are two aerodynamic processes that are developed to a degree that makes them possible to use: The *separation nozzle* process developed in Germany and the *vortex tube* separation process developed in South Africa. They both rely on centrifugal forces to separate the isotopes, but unlike in the gas centrifuge process there are no spinning parts.

A stream of uranium hexafluoride gas is sent into a curved wall which bends it. During this bending, centrifugal forces press the heavier gas molecules closer to the wall than the lighter molecules. Then the stream is separated in one heavier and one lighter fraction.

The centrifugal forces are greater the larger the speed of the gas stream. To make use of this, the uranium hexafluoride is diluted by a light *carrier gas* such as hydrogen or helium. The velocity of sound is higher in a lighter gas than in a heavy gas, and this means that the gas can travel faster without energy being lost in sonic shock effects. Also the light carrier gas makes it easier for the isotopes of uranium to separate.

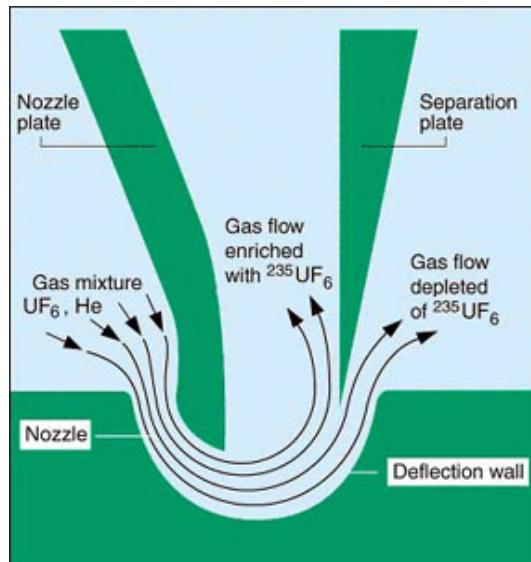


Figure 7.1 Schematic view of a separation nozzle. The smaller the nozzle is, the less turbulence there will be in the gas flow. The sketch is reprinted here with permission from Informationskreis KernEnergie, Berlin (www.kernenergie.de).

Both of the aerodynamic processes described below have a high energy consumption because of the high proportion of carrier gas relative to uranium hexafluoride. The processes are not considered economically competitive because of this high energy consumption.

A complicating feature of the aerodynamic processes is the handling of hydrogen. If the temperature is too high, hydrogen will react with uranium hexafluoride, so the temperature must be kept under control. [1;4]

7.1 The separation nozzle process

The separation nozzle process, also known as the Becker nozzle process after its inventor, was developed in Germany and has been used both there and in Brazil, but both countries have since abandoned this activity [1]. Gaseous uranium hexafluoride diluted by hydrogen or helium is compressed and directed along a curved wall at high speed, and the heavier molecules gather closer to the wall. The centrifugal forces on the gas in the nozzle can exceed those in a centrifuge by more than a thousand times [21]. Then the stream is split by a knife edge, and the heavy and the light fraction is withdrawn separately [4] (see Figure 7.1).

The smaller the physical dimension of the separation nozzle, the more economic it is, and the radius of curvature may be as small as 10 μm . Production of the nozzles is done by photo etching of metal foils and then stacking them. A stage typically consists of a vertical *cylindrical vessel* containing the separation elements, a cross piece for *gas distribution*, a *gas cooler* to remove the heat of compression and a *centrifugal compressor* driven by an electric motor. [4]

The energy costs for a separation nozzle plant is about 3600 kWh/SWU, and originate mostly in the re-compression of the depleted and enriched gas streams. The separation factor of this method is about 1.015 which is lower than for gas centrifuges but higher than for gaseous diffusion [1;22].

7.2 The vortex tube process

The process developed in South Africa can be characterized as an advanced vortex tube or a stationary-wall centrifuge. In this process, uranium hexafluoride gas is diluted by hydrogen gas and then compressed. The gas mixture enters tangentially through nozzles or holes at speeds close to the speed of sound into a tube shaped like a cone [4]. The gas spirals down the tube and at the end of the tube the heavier gas exits through holes in the side while the lighter gas exits straight out the end [1]. The vortex tube has a relatively high separation factor of between 1.025 and 1.035 [23;24].

The South Africans developed a technique called *Helikon* to cascade the elements, and avoid difficulties in piping. A typical Helikon module consists of a large *cylindrical steel vessel* that houses a separation element assembly, two *axial-flow compressors* (one mounted on each end) and two water-cooled *heat exchangers*. [4] A module with the capacity of 10 000 SWU per year would be about 3.6 m in diameter and 10 m long.

South Africa used the vortex tube method to produce both reactor fuel and HEU for their weapons program [25]. The Y-plant at Valindaba produced hundreds of kilograms of HEU [4]. This plant was the pilot enrichment facility designed to produce weapons-grade uranium, and it started operation in 1977. The first enriched uranium was withdrawn in 1978. In 1988 production started at another plant located next to the Y-Plant. This plant was semi-commercial and employed the Helikon technique, and it produced 3.25 % enriched uranium for a power reactor. The plant could be used to produce HEU by batch recycling or stretching, but was not intended for this.

The energy consumption of the vortex tube process is about 4000 kWh/SWU, which is high compared to other techniques. Because of the high costs of this process, South Africa also investigated both gas centrifuge and MLIS processes (see below). [1]

8 Laser isotope separation

Laser isotope separation makes use of the quantum mechanical properties of atoms and molecules described in Section 10A.5. Lasers are light sources that can provide an intense beam of light with a very precise wavelength or energy of the photons. If one can produce a laser capable of emitting light at the exact frequency that is needed to separate the two isotopes of uranium, it will be a very efficient way of enriching.

Although the laser isotope separation (LIS) techniques are relatively simple to understand, they are considered technically difficult, and it has been assumed that only technically advanced countries would be able to master them. Much relies on laser and optical devices. The laser industry is in rapid development on its own, and it is therefore not unlikely that the technical obstacles will decrease as time passes. LIS can of course be used on other chemical elements than uranium, so the technology is interesting also for other purposes than uranium enrichment. It is

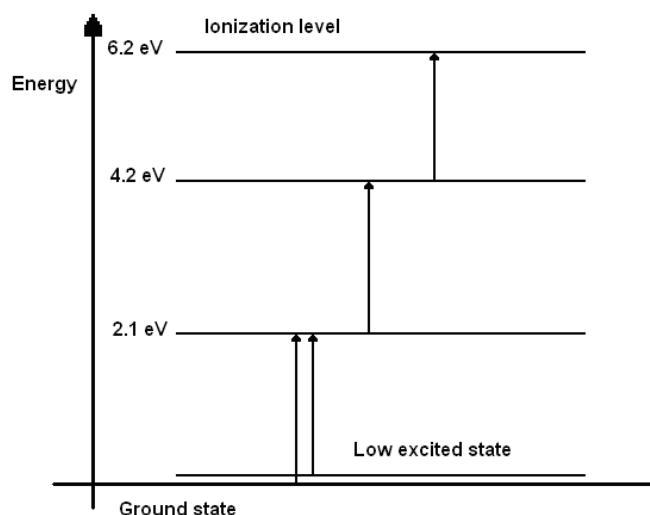


Figure 8.1 Ionization of the uranium atom is done in three steps of about equal size. Each arrow in the diagram represents one laser that has to be present for the AVLIS process to work. The energy levels shown are quoted from [26].

therefore possible to “practise” the use of LIS technology and LIS equipment without using uranium just by tuning the lasers to a different frequency.

Unlike many other isotope separation methods, LIS can be used to separate the isotopes of plutonium [26].

8.1 Atomic vapour laser isotope separation (AVLIS)

8.1.1 The main principle

The atomic vapour laser isotope separation (AVLIS) process consists of a laser system and a separation system. Uranium in its pure metallic form is melted and heated in a crucible by an electron beam to the point where enough of the atoms evaporate. The evaporated atoms move radially away from the ingot. Warm liquid uranium is very corrosive and must not come into contact with the support structure. It is also important that the heat does not disappear, and that the atoms can move relatively freely; therefore an extensive mechanism is needed to create vacuum and contain the heat. The liquid uranium is in one end of an evacuated chamber, and laser beams are cutting through the other end. As the uranium gas moves away from its source, the atoms collide and most of their internal energy is converted to kinetic energy. When the atoms reach the radiation zone (the laser beams), about 40 % of them are in their lowest energy state (the ground state). [26]

When the atoms reach the radiation zone, the U-235 atoms which are in the ground state will be excited two times and then ionized (see Figure 8.1). The ionization energy for the uranium atom is about 6.2 eV.¹² In theory, just one laser with the exact energy could be used to ionize the atom,

¹² The electron volt (eV) is a small physical unit for energy, appropriate on atomic levels. It is the same amount of energy as 1.602×10^{-19} joules.

but in practice it is necessary to do the ionization in at least two steps. Three steps of about equal size have been used to take advantage of the readily accessible and well-established technology of rhodamine dye lasers (see Appendix B for more information on lasers). The ions are then collected on charged plates oriented parallel to the radial direction, while most of the neutral atoms continue outwards and are collected on plates orthogonal to their direction of motion. It is impossible to avoid collecting some neutral atoms on the collector plates for the ions because of the random motion of the molecules in a gas (see Section A.4). This and other problems decrease the efficiency such that the maximum separation factor will be about 15. In practice, a separation factor of between five and ten is more likely, and this is far greater than the separation factor of processes that are already in use, such as gaseous diffusion and gas centrifuges where the separation factors are just above 1. [26]

An AVLIS module consists of the crucible holding the liquid uranium and the collector plates, and it is encapsulated in a vacuum system. All laser sources and mirrors are kept outside the volume containing the uranium steam. Each module can be typically 1 m high and 3 m long. The laser light is reflected many times by mirrors and may have a path length of more than 100 m. [26]

Since only about 40 % of the U-235 atoms are in their ground state, only these 40 % can be excited, and this lowers the efficiency of the process. But an additional about 30 % are in a low excited state and can be included by adding another laser with a slightly lower energy which excites these atoms into a state with the same energy as the atoms in the ground state are excited to (cf. Figure 8.1). In this way, 70 % of the U-235 atoms may be ionized instead of only 40 %. The four lasers that are needed all have slightly different colours corresponding to the slightly different energies required, but all are in the red-to-orange region of the spectrum. [26]

8.1.2 Advantages and problems with the AVLIS process

One of the advantages of the AVLIS process is the high separation factor. This suggests that producing highly enriched uranium would be easier with this process than with many other processes. With a separation factor of ten, only three times of batch recycling will be needed to reach a product of 97 % U-235. But there are some obstacles. When the fraction of U-235 in the feed is increased, a higher laser intensity is needed to maintain the same separation factor because there are more atoms present which need to be excited. When the feed contains more than 50 % U-235, it will be better to tune the lasers to ionize the U-238 atoms. Then the intensity of the lasers will not have to increase as the enrichment level becomes larger, but can instead decrease. If the steam is too dense, it will be more difficult to collect all the atoms, and some will be lost. On the other hand, if the density of the steam is too low, the atoms will not have the opportunity to collide often enough to reach the ground state before they enter the radiation zone. [26]

Also only about 50 % of the atoms reach the radiation zone with the laser beams after being evaporated, the remaining atoms are collected on the walls of the chamber and must be recycled. This makes the process less efficient. [26]

The AVLIS enrichment process will generate a small amount of waste and consume only about as much energy as the centrifuge process, 100–200 kWh/SWU [4;23]. The process does not require as much space as a centrifuge plant, however [27].

8.2 Molecular laser isotope separation (MLIS)

8.2.1 The main principle

The energy differences between the vibrational states of a molecule are smaller than the differences between the electronic states of an atom, so for the dissociation of molecules infrared light must be used instead of visible light. *Photo dissociation* of a molecule (see Section A.5) can be done in one, two or more steps. When two steps are used in the molecular laser isotope separation (MLIS) process for enrichment of uranium, the first photon is in the infrared section of the electromagnetic spectrum. This is the one that selects the desired molecule by exciting the vibrational states of the molecules containing U-235 and not U-238. Then a photon of higher energy, in the ultraviolet end of the spectrum, breaks the bond completely. Molecules which are already excited have a larger probability of absorbing the photon and be dissociated than those in the ground state, and in this way the selectivity of the first step is conserved. But it is not impossible also for the unexcited molecules to be dissociated by the UV photons, and this makes the process less effective. In this two-step process the excitation and the dissociation must occur on a time scale that is short compared to the lifetime¹³ of the intermediate vibrational state. Alternatively, several infrared photons of the same frequency may be used to dissociate the molecules and still be isotopically selective. [28]

The MLIS process uses uranium hexafluoride gas because of its chemical qualities¹⁴, and because its handling is well known from its use in the gaseous diffusion method and the gas centrifuge method. Other molecules could also be used. The idea is to break the bond between the uranium atom and one of the fluorine atoms in the molecules containing U-235 so that uranium pentafluoride and a free fluorine atom are formed. Uranium pentafluoride enriched in U-235 will then precipitate¹⁵ as a white powder and can subsequently be collected [4].

As in the AVLIS process, it is the ground state that has to be occupied initially if the laser is going to be able to excite the molecules. The molecules in the uranium hexafluoride gas collide so often that almost none of them occupy the ground state. The molecules can be forced into the ground state by cooling, but before room temperature is reached uranium hexafluoride will precipitate. One way of getting around this is by super-cooling the gas. Super-cooling means cooling below the sublimation point without crystallization taking place. This is a very unstable state, and if just a small crystal is present or the gas is disturbed, then crystallization will occur. Gaseous uranium hexafluoride mixed with a carrier gas and a scavenger gas is expanded through a nozzle at large speed, this cools the gas down to low temperatures without the uranium

¹³ The lifetime is the average time it takes from a molecule is excited until it deexcites by getting rid of its excitation energy.

¹⁴ One being the possibility to convert it into uranium pentafluoride by photo dissociation.

¹⁵ Form solid particles.

hexafluoride precipitating. Suitable carriers are hydrogen or a noble gas. A scavenger gas (such as methane) is used to capture the fluorine atoms that are released as a result of the dissociation of the uranium hexafluoride molecules containing U-235. [4]

By this process, 95 % of the molecules will descend to the ground state, and only one laser is needed to excite most of the molecules (unlike in the AVLIS process where two lasers were needed and only 60 % of the atoms were excited). [26]

The energy consumption of the MLIS process is estimated at about 235 kWh/SWU which is slightly higher than for the AVLIS process. The use of uranium hexafluoride and low power consumption are some of the advantages of the MLIS process [1;4].

8.2.2 Potential for HEU production

It was a concern when developing the process that the uranium pentafluoride and the uranium hexafluoride molecules might exchange fluorine atoms before being separated, but this turned out not to be a big problem [28]. Also as the enrichment in the feed increases, this problem will decrease because there will be fewer U-238 atoms present to change places with the U-235 atoms. This makes the MLIS process more suitable for cascading than the AVLIS process. Also removing the uranium pentafluoride and converting it back to uranium hexafluoride does not pose great difficulties. And the amount of U-235 can always be made to match the lasers, independent of the enrichment level, because the uranium hexafluoride gas is diluted by a carrier gas. All of this implies that using the MLIS process for HEU production may be possible in relatively small plants.

8.2.3 Separation of isotopes by laser excitation (SILEX)

The SILEX process was developed in Australia by the company Silex Systems Limited (Silex). It is an MLIS process using uranium hexafluoride as feed [29]. Many details of the process are classified, and it is difficult to distinguish SILEX from the general MLIS technique described above. The technique uses a pulsed CO₂ laser of wavelength 10.8 μm which is converted to about 16 μm by use of a parahydrogen Raman converter [30]. The 50 Hz pulse rate of the lasers is not enough to produce significant enrichment; it must be increased at least ten times [30]. Both the product and the tails are removed mechanically, and the separator system is cascaded [29].

SILEX has advantages to other processes for uranium enrichment such as low power consumption, simple and practical separation modules and versatility. [31] Because of this, the SILEX method is suggested as the third generation of uranium enrichment, where gaseous diffusion and gas centrifuges constitute the first and second generations, respectively. The separation factor is classified, but announced by the company Silex to be somewhere between 2 and 20 whereas it is 1.3 for gas centrifuges and 1.004 for gaseous diffusion [31].

8.3 Laser assisted processes

The information in this section is from “Uranium Enrichment and Nuclear Weapon Proliferation” [26].

The laser’s ability to excite only one kind of molecule can be utilized in several different ways in addition to those mentioned above. One example is that they may raise the temperature of one component of a gas mixture so rapidly that the rest of the mixture is not heated notably during the process. This could be done to uranium hexafluoride molecules by use of an infra red laser similar to the one used in the MLIS process. When the molecules are excited into a higher vibrational state, and the molecules collide with others, much of the vibrational energy can be converted into kinetic energy, which makes the molecules move and diffuse faster. This is a quality that can be used to enhance the effect of some of the methods which use uranium hexafluoride.

One example is the thermal diffusion process. If only uranium-235 hexafluoride molecules are heated, then fewer stages are needed to achieve the same separation of U-235 and U-238 than without the use of lasers. Another method is to combine it with an aerodynamic process.

Another proposed method involves letting a stream of a gas mixture of uranium hexafluoride and argon be irradiated by an infrared laser. At the end of the tube, there are very cold plates (below -150 °C) on which the gas molecules will condense if they collide with it. Because the molecules containing U-235 will have a higher velocity, they are more likely to collide with the plates. When the tube is evacuated, the plates can be heated to free the uranium hexafluoride in gaseous form again and the gas can be sent on to the next stage in the process.

8.4 When and where have the LIS techniques been used?

More than 20 countries are known to have done research on laser isotope separation techniques.¹⁶ France, the United Kingdom and the United States have developed LIS programs that could move beyond the lab into the pre-industrial phase and ultimately into commercial production. In August 2002, it was announced by the dissident group National Council of Resistance of Iran that Iran had started an LIS program and developed a laser enrichment facility. This enrichment program was not detected by the International Atomic Energy Agency (IAEA), but had supposedly only enriched milligrams of uranium. In September 2004, IAEA discovered South Korean experiments; in 2000, scientists at the Laboratory for Quantum Optics at the Korea Atomic Energy Research Institute had separated about 0.2 g of U-235 enriched to levels between 10 % and 77 %. It seems that LIS methods are hard to upgrade to commercial scale, but scientists would not need a commercial-scale plant to enrich enough uranium for a single nuclear weapon if they had one or two years in which to do it. [32]

¹⁶ This includes Argentina, Australia, Brazil, China, France, Germany, India, Iran, Iraq, Israel, Italy, Japan, the Netherlands, Pakistan, Romania, Russia, South Africa, South Korea, Spain, Sweden, Switzerland, the United Kingdom, the United States and Yugoslavia.



Figure 8.2 Laser corridor during AVLIS operation. Over 2000 optical elements convey these green and yellow beams. Each wall in the dye laser corridor has two dye laser chains, which amplify the light for later use in the separator tank. Special filters clean the air in the laser enclosures, making it 1000 times cleaner than that in a room with standard air conditioning. Photo courtesy of the United States Department of Energy.



Figure 8.3 Photograph showing some of the approximately 4500 optical elements used in the AVLIS process. These high-quality optical elements reflect light approximately 100 times more efficiently than a bathroom mirror. Optical elements convey light through the laser system, and some also regulate the size and shape of the laser beams. Photo courtesy of the United States Department of Energy.

The challenges posed by the AVLIS technique are mainly technical. The complex laser systems (cf. Figure 8.2 and Figure 8.3) and the fact that uranium vapour is very corrosive, lead to technical difficulties that make the process hard to apply on a commercial scale. In June 1999, the United States was on the verge of commercialization when it was decided to cancel its AVLIS program. About two billion dollars had then been spent on the program. Faced with stiff

competition from gas centrifuges, large-scale LIS processes have not been developed by any of the countries who have sought this path for uranium enrichment. [32] The only potential large-scale LIS project seems to be the one conducted by the Australian company Silex (cf. Section 8.2.3).

Silex Systems Limited (Silex) was established in 1988. The principle was demonstrated to work in 1994, and in 1996 Silex reached an agreement for the development and licensing of SILEX technology with the United States Enrichment Corporation (USEC). In 2000, the process was demonstrated at prototype level, and in 2001 the technology was classified by the United States and Australian governments. The classification of the process has been interpreted by some to mean that the process can be used to produce highly enriched uranium for weapons use, but in 2007, it was claimed that the SILEX process was not mature enough to enrich significant amounts of uranium to the levels required in nuclear weapons [30]. USEC withdrew from the project in 2003, but in May 2006, General Electric Company signed an exclusive commercialisation and license agreement for the SILEX uranium enrichment technology. [33]

9 Chemical separation methods

Two chemical processes have been developed to a point where they could be considered possible to use for the enrichment of uranium. The CHEMEX process was developed in France, and an ion exchange method has been developed in Japan. The chemical approaches to the separation of uranium isotopes are attractive because of their low energy consumption; there is no need for powerful compressors or pumps [34]. Furthermore, these processes are based on standard chemical engineering technology [4], which makes them favourable.

Both processes are based on the fact that U-235 is more likely than U-238 to be in a higher oxidation state (see Section A.6) [4]. This means that if natural uranium is present in different chemical states or compounds, then there will be an accumulation of U-235 in the higher oxidation states. If it is possible to separate the states (for example if they are different types of molecules that do not mix very well) then the compound which is enriched in U-235 can be extracted. For the separation effect to be the greatest, the two compounds used should have uranium in as different oxidation states as possible, and the temperature should be as low as possible [34]. Unfortunately these are criteria which also make the process go slower and lead to the need of good catalysts¹⁷.

A separation stage consists of mixing of the two compounds of uranium for a long enough time that chemical equilibrium may be established. This can be done by letting the two substances flow in opposite directions in a column. The main difficulty of chemical enrichment methods is the reflux problem. When the substances emerge at the ends of the column, some can be extracted as product and tail, but much has to be sent back into the column to maintain the counter current flow pattern. Before they can be sent back into the column, the enriched substance has to be

¹⁷ A catalyst is a substance which facilitates a chemical process without being consumed itself.

transformed into the form of the depleted substance and vice versa. This is a non-reversible process and the main source of power consumption in the chemical enrichment processes. Even a small material loss in the reflux processes can reduce the efficiency of the plant significantly. [34]

9.1 The French process (CHEMEX)

In the French process, two different compounds including uranium are dissolved in different liquids. These liquids are immiscible¹⁸ in order to make possible the later separation of the compounds; one is in an aqueous phase, and one is in an organic phase. [22]

The two liquids flow through each other in a column. The liquids are agitated so that the organic one forms small droplets to increase the reaction surface; this is done either by the use of an external mechanical device or a series of reciprocating discs. Limiting factors are the rate of the chemical reaction and the rate of diffusion in and out of the oil droplets for the two chemical species. The chemical reaction rate can be sped up by changing the uranium compounds or by the use of catalysts and raising the temperature. Some compounds from the group known as “crown ethers” have good qualities for uranium enrichment because they both extract the uranium from one liquid phase to another and enhance the separation effect. The diffusion rate of the uranium compounds in and out of the oil droplet can be made higher by decreasing the size of the droplet. But there seems to be a practical limit at a diameter of a few millimetres, and this makes the process slower than the Japanese process discussed below (Section 9.2). [34]

The French enrichment facility is proposed to be constructed of two identical columns connected in a way that reduces the equilibrium time. The standard dimensions of the exchange column in the CHEMEX process is 1.6 m in diameter and 35 m high [22], and the capacity is 3000–5000 SWU per year. The concentration of uranium is greater in this process than in the Japanese process. The inventory is about one order of magnitude larger than that for a gaseous diffusion plant of about the same capacity. There is also a need for large volumes of chemicals. Some of them, such as the crown ethers, can be expensive. [34]

At the ends of the column, the two uranium compounds have to be transformed into each other, in order to have reflux. The chemical converters must be very efficient; they are the main sources of energy consumption in this process. [22]

The uranium compounds, the catalysts and the reflux reactions are kept secret. The components needed in the process are made from plastic materials because of corrosion problems. [34]

The estimated energy consumption is about 600 kWh/SWU [23].

9.2 The Japanese process (ACEP)

The Japanese ion exchange process known as the Asahi Chemical Exchange Process (ACEP) has been under development since 1972. It is based on a solid uranium adsorbent which has been

¹⁸ Immiscible liquids do not mix. An example is oil and water.

developed by Asahi Chemical; this selectively adsorbs¹⁹ hexavalent uranium. The breakthrough of this process was the discovery of a uranium adsorber of high reaction speed. [22] The adsorber is a resin²⁰ in which the individual particles have the forms of spheres with a porous surface to increase the contact surface. It has the function of holding one of the two uranium compounds on its surface [34].

First the resin is charged with an oxidizing agent. A solution containing a uranium compound in which uranium is in the 4+ oxidation state is introduced. Then the uranium is oxidized to the 6+ state and replaces the oxidizing agent on the resin. This process will leave a sharp boundary between the part of the column where the resins are occupied by the oxidizing agent and uranium, this boundary will move slowly down the column when more uranium solution is added at the top of the column. After some time a new solution is added at the top of the column, this one containing a reducing agent. This will act on the uranium absorbed on the resin and reduce it back to the 4+ oxidation state and then replace it on the resin. The 4+ uranium goes into the solution and continues down the column with it. The 4+ uranium will then come into contact with the 6+ uranium on the resins further down the column, and the isotopic change will take place. There will eventually be a higher concentration of U-235 in the compound adsorbed on the resin than in the solution. The portion of the column where the isotope exchange takes place is well defined because also the reducing agent forms a sharp boundary with the uranium compound. [34]

The process is carried out by the use of four columns; the actual ion-exchange process takes place in the first three columns which are connected in series, and the recharging of the agents is done in the fourth. Each column should be a few meters high. Very little energy is needed in the ion-exchange columns unless the temperature has to be raised. The main energy consumption comes from recharging the oxidizing and reducing agents and the pumping of the recycled solution. [34] The energy consumption of the process is estimated to be about 150 kWh/SWU [23]. Because of good exchange kinetics, the ACEP process requires smaller volumes than the CHEMEX process [22].

To find suitable resins, oxidizing and reducing agents, processes for recharging these as well as a suitable catalyst for the exchange process has taken many years. [34]

9.3 A third process

There is also a third process that has been studied by several countries. It appears to be attractive, but there are problems related to developing it past the laboratory stage. The problems seem to be mainly large energy costs and losses of material in the recycling of the compounds. The development of this process seems to be far behind the two processes discussed above. It makes use of NOUF₆ dissolved in dry hydrogen fluoride and UF₆ dissolved in freon or some other saturated fluorocarbon in the liquid phases. It is the execution without large energy costs and

¹⁹ When something is adsorbed it forms a layer on the surface of the adsorber.

²⁰ A resin is a substance that acts as an ion exchanger. It holds ions in its structure and is therefore able to hold ions with the opposite charge to its surface. When new ions are adsorbed, old ones are released.

losses of material of the reaction that recycles NOUF_6 which seems to be the biggest challenge. [34]

9.4 When and where has the technique been used?

Chemical exchange processes are under study in several countries, but there are still no full-scale chemical enrichment plants in the world [4]. The only countries that have attempted this technique to any extent worth mentioning seem to be France, Japan and Iraq. The French process is offered on sale to countries that do not pursue other enrichment paths [5]. It is believed that North Korea may have done research on chemical enrichment using knowledge obtained in Japan [35;36].

Iraq decided to concentrate on chemical enrichment methods for the production of LEU to be used as fuel for their EMIS program when their gaseous diffusion program was not progressing well in the late 1980s [1].

10 Brief summary and comments

Even though a number of methods for the enrichment of uranium have been investigated and developed over the years, only very few have been applied on a large scale, and presently the gas centrifuge method appears to be the only commercially viable method. It is possible that other techniques, such as for example laser isotope separation, may be successfully used also on an industrial scale in the future.

One should be aware that some of the techniques that cannot be used for large-scale enrichment, for example the mass spectrometer method, may still be of use to small-scale bomb makers. It is therefore important to maintain export control regimes capable of preventing equipment for all kinds of uranium enrichment from falling into the wrong hands.

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Appendix A Some relevant physics and chemistry

A.1 Isotopes

An atomic nucleus contains two types of particles: *protons* which have a positive electric charge, and *neutrons* which have no electric charge. Around the nucleus there are electrons which have the same charge as the protons only with opposite sign; they are negatively charged. In a neutral atom there is therefore the same number of electrons around the nucleus as there are protons inside the nucleus. It is mainly the number of electrons that determine the chemical properties of an atom, and therefore different atoms which have the same number of protons (and therefore also electrons) constitute the same *chemical element*. So when a chemical element is defined, nothing is said about the number of neutrons. The number of neutrons in a nucleus is typically about the same as the number of protons, but it may vary a little, and atoms of the same chemical element, but with a different number of neutrons, are known as different *isotopes* of that element.

The main difference between two isotopes is their masses; the *mass number* of an atom indicates the total number of protons and neutrons. When discussing isotopes, the mass number is generally provided along with the chemical symbol to specify which isotope we are referring to. Uranium is the heaviest naturally occurring element, and there are two important isotopes U-238 and U-235, where U-238 contains three more neutrons than U-235. In naturally occurring uranium, U-238 amounts to approximately 99.3 % and U-235 only 0.7 %.

A.2 Nuclear fission and chain reaction

A nuclear power plant and a bomb are based on the same principle, the principle of *fission* of an atomic nucleus. Some large nuclei are unstable and can sometimes split into two smaller nuclei of roughly equal size while emitting a few independent neutrons; this process is called spontaneous nuclear fission. These new nuclei and neutrons will have a total mass that is smaller than the mass of the original nucleus. This *mass defect* is compensated by an energy release in accordance with Einstein's famous equation $E = mc^2$ where c is the velocity of light (300 000 km/s). This is the energy that is utilised in a power plant or a bomb. The uranium isotopes have a low probability of spontaneous fission²¹ (if spontaneous fissions occurred readily, it would not be possible to use uranium in a reactor). When U-235 captures an extra neutron, it may fission and release two or three new neutrons. If these new neutrons are captured by another nucleus and subsequently lead to a new fission, a *chain reaction* is started. In a nuclear reactor such chain reactions are controlled to give an approximately constant release of energy, whereas in a bomb the chain reaction is allowed to run wild. For a chain reaction to be sustained, on average at least one of the freed neutrons from each fission has to lead to a new fission. For this to occur there has to be a certain amount of uranium present, and this minimum amount is known as the *critical mass*; if the

²¹ The partial half lives due to spontaneous fission for U-238 and U-235 are 9×10^{15} y and 1.2×10^{17} y, respectively [37].

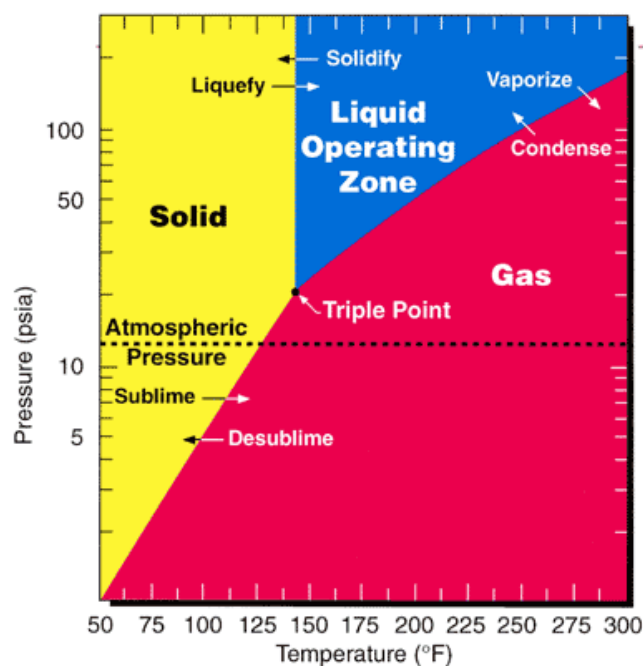


Figure A.1 Phase diagram of uranium hexafluoride (copied from <http://web.ead.anl.gov/uranium/guide/prodhand/sld022.cfm>).

mass is greater it is called *supercritical*. Alternatively, it is possible to compress a smaller mass to bring the atoms closer together and thereby reach a *critical density*. Since there is so little U-235 in natural uranium, it is necessary to enrich the uranium in this isotope for the material to be able to sustain a chain reaction. Otherwise the critical mass or the critical density would be too high to reach in practice.

A.3 Uranium hexafluoride

UF₆, *uranium hexafluoride*, also referred to as “hex,” is a substance with good properties for uranium isotope separation. Each molecule consists of only one uranium atom and six fluorine atoms, and because fluorine only has one stable isotope, F-19, there are only two possible compositions of the molecule, each with a specific molecular mass dependent of the uranium isotope. The mass numbers of the molecules containing the different isotopes of uranium are 352 and 349, respectively, so the mass difference is three units which amount to only 0.85 %. Because the mass difference comes directly from the different masses of the uranium isotopes, uranium hexafluoride is a good choice for separation of these isotopes, but because the relative mass difference between the molecules is so small, the enrichment will be less effective than if the mass difference had been greater.

At room temperature and atmospheric pressure, uranium hexafluoride is a crystal, but at 56.5°C it turns directly into a gas without first becoming a liquid (it sublimates) (see the phase diagram in Figure A.1). The molecule is organized with the uranium atom in the centre and the six fluorine atoms spaced symmetrically around it, giving the molecule three perpendicular symmetry axes. This makes it possible to make theoretical calculations of the vibrational energy states which are utilised in the laser isotope separation methods. UF₆ is highly reactive and reacts violently with

water and organic compounds. UF_6 is also highly corrosive, and nickel or aluminium alloys are needed to handle the gas. [2]

Uranium hexafluoride is used in gaseous form in centrifuges, in the gaseous diffusion method, in the aerodynamic processes and in molecular laser isotope separation. It is used in liquid form in the thermal diffusion process. To obtain the liquid form of uranium hexafluoride, the pressure must be raised to just over atmospheric pressure, as can be seen from the phase diagram.

A.4 Statistical description of the molecules in a gas

A gas consists of a very large number of molecules that are moving around all the time at different speeds and exchanging energy through collisions with each other. Because there are so many molecules in a gas, it is impossible to describe the properties of the gas by considering each molecule separately; instead it has been found fruitful to use a *statistical description*. This means that while we cannot determine the exact speed or the exact energy of each molecule, we can determine the average or the *most probable* values. The actual values for each molecule will generally not be the same as this most probable value. Because of collisions between the molecules, energy will be exchanged between them and the energy of individual molecules will thus vary.

If the gas is in *thermal equilibrium* (in practice this means that the temperature and the pressure are stable), all molecules have the same kinetic energy on average. The kinetic energy of a molecule is given by $E = \frac{1}{2}mv^2$ where m is the mass of the molecule and v is its velocity. This means that molecules with smaller mass must have a larger velocity to match the kinetic energy of the heavier molecules. But this is only on average; the heavier molecules may have higher velocities at some times, and the lighter molecules may have lower velocities.

A.5 Quantum mechanics

On the atomic and molecular level nature is not smooth. For instance a system (like an atom or a molecule) cannot have a continuous energy spectrum, but can only take distinct, discrete energy levels. Transitions between these discrete states occur with emission or absorption of a quantum of energy; these transitions are called quantum leaps and are the smallest changes possible. This quantum of energy carries the exact energy that separates the two states and is usually in the form of a photon. Photons are the particles that light and other electromagnetic waves such as microwaves, infra red light, visible light, ultraviolet light and x-rays consist of. A light ray is a bundle of these particles, and each of them carry the energy $E = h\nu$ where h is a scaling constant and ν is the frequency of the light. This means that microwaves, x-rays, blue light and red light all consist of photons with different energies. Electromagnetic waves with photons of high energy have high frequencies (and correspondingly short wavelengths). By sending photons with the exactly right energy (this means electromagnetic waves with the exactly right frequency) at an atom or a molecule, it can be *excited* (go to a given state with a higher energy).

In an atom, the electrons around the nucleus occupy discrete energy levels. By adding energy to an atom, an electron can be excited into a higher energy state. But for this to occur, the energy which is added has to match the energy difference between the old and the new energy levels. Electrons can also be ejected completely if enough energy is added and this will happen easier if the electron is already excited. When an electron is ejected the atom becomes an *ion*. The energy that is added to ionize the atom does not have to be specific as long as it is large enough; spare energy will just become kinetic energy for the electron (energy that gives it a velocity). An ionized atom has a net electric charge which can be exploited if we want to influence its motion. The energy levels of the electrons in an atom are determined by the nucleus of the atom. Therefore there will be slight differences in the energy levels of different isotopes of the same chemical element. By choosing photons of the exactly right energy, it is possible to excite and ionize only one isotope in a mixture.

In molecules, the energy levels are mainly determined by the chemical bonds between the atoms. When two or more atoms are bound together, they may vibrate relative to each other, leading to discrete vibrational modes with distinct energies. When a molecule is excited, there is a change from one vibrational mode to another, and the energy added to excite it has to match the energy gap between the modes exactly. If more energy is added to the system, the chemical bond can be completely broken, and new chemical properties are created. If the energy is added through a photon, this is called *photo dissociation* of the molecule. As mentioned above, the energy levels of the vibrational modes of a molecule are determined by the atoms involved in the chemical bond. There can be slight differences between the energy levels of a certain chemical bond depending on the isotopes of the chemical elements involved. Therefore it is possible to excite and dissociate only molecules containing a certain isotope if the photons can be tuned accurately enough.

A.6 Oxidation states

The oxidation state gives the degree of oxidation of an atom in a chemical compound. It is equal to the hypothetical charge that an atom would have if all bonds to other elements were completely ionic (complete exchange of electrons). An increase in oxidation state is known as oxidation and a decrease in oxidation state is called reduction. The algebraic sum of the oxidation numbers of all the atoms in a compound is always equal to the net electric charge of the compound. The term oxidation state is often used interchangeably with the terms oxidation number, even though the actual definitions are slightly different, the values are usually the same. A given chemical element may have different oxidation states in different compounds; this is the origin of the term redox reaction. A redox reaction is shorthand for reduction-oxidation reaction and is a chemical reaction where the oxidation states of the atoms involved are changed.

Substances that are oxidative are known as oxidizing agents, oxidants or oxidizers, in a redox reaction the oxidizing agent will be reduced. Substances that are reductive are known as reducing agents, reductants or reducers, the reducing agent will be oxidized in a chemical reaction.

Uranium can be found in oxidation states +2, +3, +4, +5 and +6 [38].

Appendix B Lasers

This appendix discusses in more detail the laser requirements necessary for laser isotope separation.

In the atomic vapour laser isotope separation (AVLIS) technique (Section 8.1), to be able to excite only one isotope, the lasers must be tuned with an accuracy in wavelength of one part in 100 000. This is possible with modern lasers. Especially a type of lasers called dye lasers, which use organic dyes, may be used. High-power green or yellow light from copper vapour lasers (cf. Figure 8.2) is fired into streams of special dyes, which convert the laser energy into beams tuned to the exact colour absorbed by U-235 [39]. Four slightly different beams are needed (cf. Figure 8.1); it is not possible to distinguish between these beams with the human eye. The light that causes the desired transitions has a red-to-orange colour and can be made by use of a common dye called rhodamine 6G. Because excited atoms will decay back down into a lower state, the lasers must be intense enough to re-excite the ones that decay. The lowest acceptable power density is on the order of tens of kilowatts per square centimetre. Because the uranium atoms move through the radiation zone, the laser pulses must come often enough so that all the atoms may be hit by them. The problem sufficiently high frequency pulses can be solved by using several lasers that are synchronised such that the total frequency will be high enough. [26]

In the molecular laser isotope separation (MLIS) technique (Section 8.2), the required wavelength is 16 μm , and there are several possible lasers. Powerful infrared lasers which use CO or CO₂ are available in many frequencies, but the infrared frequency that is needed to dissociate the uranium hexafluoride molecule is harder to make [26].

The candidate which seemed to be the best alternative when the technique was developed, uses Raman scattering from parahydrogen to increase the wavelength of the 10 μm light from carbon dioxide. Raman scattering refers to an interaction of photons with molecules in which the scattered photons have different energy than the incident photons; the change is determined by the molecules' rotational or vibrational energy levels. [28] Parahydrogen is a state of the hydrogen molecule (H₂) in which the two atoms have antiparallel spins.

Another option is to use a free electron laser. Free electron lasers are widely tuneable and used for many purposes other than isotope separation, so there is a lot of activity in that area. In a free electron laser, the electrons are accelerated to relativistic speeds and pass through a transverse magnetic field giving them a sinusoidal path. This acceleration results in the release of photons. Today, a free-electron laser requires the use of an electron accelerator with its associated shielding, as accelerated electrons are a radiation hazard. The accelerator requires a high voltage supply. Usually, the electron beam must be maintained in a vacuum which requires the use of numerous pumps along the beam path. Free electron lasers can achieve very high peak powers. Their tuneability makes them desirable in several disciplines. [40] Other types of lasers were also suggested at the time the process was developed [28].

After the molecules have been excited by the infrared laser, an ultraviolet laser is used to dissociate them completely. There is a larger probability for the excited molecules to be dissociated by the ultraviolet photons than for the unexcited ones. The difference is not very large, and this affects the efficiency of the method and implies that the ultraviolet laser must be powerful [28].

A way to avoid the difficulties of finding a suitable laser is to use a molecule which fits with a laser that is already developed. This has not been given much attention because of the preference for uranium hexafluoride, but it has been proven possible. A successful demonstration has been given [41] of uranium-isotope selective dissociation of a molecule with a CO₂ laser, but the molecule was very complex, and the work was ended because this approach was considered unlikely to be competitive with approaches based on uranium hexafluoride or AVLIS.