Abstract

Uranium enrichment is a process, that was first tested in the early 1940s in the United states for production of fissionable material for the atomic bomb. Since then technology and manufacturing has improved and newer methods became available. In this seminar we will explore some of the basic concepts of enriching natural uranium to the degree where it can be used in nuclear power plants or in nuclear weapons. We will also introduce mass uranium enrichment and view some basic ideal cascade shapes.
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1 Isotope Separation

Natural uranium is a mixture of 0.711% $^{235}U$ and 92.89% of $^{238}U$. The enrichment process enriches the $^{235}U$ content in natural uranium to the desired percentage. Low-enriched uranium which is typically used in nuclear reactors has 3-4% percent of $^{235}U$, while the highly enriched uranium has more than 50% of $^{235}U$ and is typically used in nuclear weapons.

Uranium is made from uraninite, which is a mixture of $UO_2$, $UO_3$, oxides of lead, thorium and rare earth elements. Uraninite is calcined to evaporate some impurities then agglomerated and crushed. $U_3O_8$ is then put in a kiln with hydrogen:

$$U_3O_8 + 2H_2 \rightarrow 3UO_2 + 2H_2O, \quad \Delta H = -109kJ/mole \quad (1)$$

$$UO_3 + H_2 \rightarrow UO_2 + H_2O \quad \Delta H = -109kJ/mole \quad (2)$$

The uranium di-oxide is then treated with hydrogen fluoride in another kiln.

$$UO_2 + 4HF \rightarrow UF_4 + 2H_2O. \quad \Delta H = -176kJ/mole \quad (3)$$

Finally the tetrafluoride is fed into a fluidized bed reactor with fluorine to produce the uranium hexafluoride ($^{238}UF_6$, $^{235}UF_6$) that is used in the separation process.

$$UF_4 + F_2 \rightarrow UF_6 \quad (4)$$

After the separation of molecules the $UF_6$ is vaporized in autoclaves with steam and reacted with hydrogen at 700°C:

$$UF_6 + 2H_2O + H_2 \rightarrow U_3O_8 + 6HF \quad (5)$$

The final product is the so-called yellow cake, which is the basic raw material for nuclear fuel fabrication. For more information see [1].

Uranium hexafluoride is used because it has great storage properties. It can be used as a solid, liquid or gas, with minimum variations in pressure or temperature. It is usually stored as a solid, when in use it can be turned into liquid which is ideal for pumping. For the actual separation process it is used as a gas (see figure 4).

1.1 Theory

1.1.1 Separative work unit

Enrichment of $^{235}U$ in $UF_6$ is a painstaking process. Since both isotopes $^{235}U$ and $^{238}U$ have the same number of protons and electrons, chemical separation methods can not be applied and we have to use special techniques to separate the $^{235}U$ from $^{238}U$, which differ in mass by 2%.

$$M_f = M_p + M_t \quad (6)$$

We can also note the amount $^{235}U$ is

$$x_f M_f = x_p M_p + x_t M_t \quad (7)$$
where $x_f, x_p, x_t$ are the concentrations of $^{235}U$ inside the mixture.

$$M_f = \left(\frac{x_p - x_t}{x_f - x_t}\right) M_p$$  \hspace{1cm} (8)

From this equation we may calculate the amount of natural Uranium that is needed in the process of separation for the expected amount of enriched uranium. For nuclear reactor Krško we require about 50 tons of uranium at $x_p = 0.043$. We must therefore provide 430 tons of natural uranium, assuming $x_t = 0.0024, x_f = 0.00711$.

Since the process of separation costs energy we devise a special unit called *separative work unit* (SWU). The concept is based on the laws of thermodynamics, where we have to separate atoms, that were previously mixed. Since the ordered state is lower in entropy, additional work must be added to the process of separation. SWU is calculated from the *value function*:

$$V(x) = (1 - 2x)ln\left(\frac{1 - x}{x}\right)$$  \hspace{1cm} (9)

Figure 2: Value function diagram

The *value function*, see Figure 2, is dimensionless. In terms of SWU we write:

$$SWU = M_p V(x_p) + M_t V(x_t) - M_f V(x_f)$$  \hspace{1cm} (10)

We can also use $M_f - M_p = M_t$. Thus we eliminate the mass of tails:

$$SWU = M_p [V(x_p) - V(x_t)] - M_f [V(x_f) - V(x_t)]$$  \hspace{1cm} (11)

We can now show the optimal tails enrichment considering the price of SWU and the cost of residual feed material. Figure 3 was calculated for LEU ($x_f = 0.00711, x_p = 0.043, 50 t$) and is using a cost of $160 per SWU (February 22, 2010) [2] and the price of yellowcake is $41.75/pound (February 22, 2010) [2].

Figure 3: The cost of SWU (blue) and the cost of of feed needed (black) as a function of tails enrichment

The rate at which enrichment facilities can produce enriched uranium is defined per unit time $SWU/yr$. For example in 2008 the US government had the ability to produce 11300 kSWU/yr, while France has 10600 kSWU/yr. To facilitate a nuclear power plant that outputs 1300 MW we must produce
120 kSWU/yr. The current energy cost for SWU in the US is 50-60 kWh. The industrial tail enrichment is calculated cost wise, depending on the price of natural uranium and electricity [3].

1.2 Separation factor

The separation factor is the ability of a process to separate isotopes [4]. It is defined as

$$
\alpha^2 = \left( \frac{x_p}{1-x_p} \right) / \left( \frac{x_t}{1-x_t} \right)
$$

(12)

Where the numerator is the molar ratio of the product stream and the denominator is the molar ratio of the tails stream. We can express the product and the tails stream $x_p$ and $x_t$ in terms of:

$$
\alpha R = \frac{x_p}{1-x_p}, \quad \frac{1}{\alpha} R = \frac{x_t}{1-x_t}
$$

(13)

where $R = x_f/(1-x_f)$. The two streams do not divide equally. A factor between 0 and 1, also referred to as the cut $\theta$, splits the input stream into $\theta$ and $1-\theta$ respectively. The gas flow $X_p$, which is defined as number of moles per unit of time [1/s] is then stated as

$$
\theta X_p + (1-\theta)X_t = X_f
$$

(14)

Equations 13 can be rewritten in terms of $x_p$ and $x_t$ and inserted into 14, which yields

$$
\frac{\theta \alpha R}{1+\alpha R} + \frac{(1-\theta)R/\alpha}{(1+R/\alpha)} = \frac{R}{1+R}
$$

(15)

The cut $\theta$ is therefore a function of $R$ and $\alpha$:

$$
\theta = \frac{(\alpha R + 1)}{(\alpha + 1)(R + 1)}
$$

(16)

2 Enrichment processes

Researchers have developed at least a dozen of different techniques for the separation of isotopes, but only a few have been shown to be cost effective. All methods are based on gas separation. Since uranium has a boiling point of 4131 °C, we use the $UF_6$ gas. Figure 4 represents the phase diagram of $UF_6$. Fluorine is a mono-isotopic element, so the mass differences in the gas molecules arise entirely from the differences in the mass of the $U$ isotopes.

Fluorine is also very reactive with water and creates hydrofluoric acid, so minimum moisture must be maintained inside the separation devices.
2.1 Gaseous Diffusion

The first enrichment process created was the gaseous diffusion. It works on the principle that on average the lighter molecules travel faster than the heavier ones. The molecules hit a porous barrier which has holes big enough to permit their passing, yet too small to permit a mass-flow of gas. The lighter molecules will hit the walls more than heavier ones. If there is a pressure difference between the two sides of the barrier, on the other side of the barrier the gas will be slightly enriched with the lighter molecules[3].

This process is the oldest and is extremely energy inefficient. The gas must be re-compressed after each stage of the process, which leads to compression heating. It must then be cooled before entering the next diffuser. A plant capacity of 10 million SWU/yr requires about 2700 MW of electrical power, which is 15 times more than a plant with gas centrifuges.

The number of collisions is proportional to the average velocity of the molecules:

$$\alpha^2 \propto \frac{v_1}{v_2}$$

(17)

At a constant temperature gas molecules have on average the same energy:

$$W = \frac{1}{2}mv_{\text{avg}}^2.$$  

(18)

From the ideal gas law, we write

$$MpV/RT = m.$$  

(19)

Where $p$ is pressure [Pa], $V$ is volume [m$^3$], $M$ is the molar mass, $R$ is the gas constant which is 8.314472 J-K$^{-1}$-mol$^{-1}$ and $T$ is temperature [K].

$$v_{\text{avg}}^2 = \frac{2WRT}{pVM}$$  

(20)

The ideal separation factor is proportional to the square root of the molecular weights:

$$\alpha^2 = \sqrt{\frac{M_2}{M_1}} = \sqrt{\frac{(19*6+238)}{(19*6+235)}} = 1.0043$$  

(21)
The enrichment technology is moving away from gaseous diffusion. See table 1 for more details.

### 2.2 AVLIS and MLIS separation

LIS is an acronym which stands for laser isotope separation. The AV and M represent atomic vapor and molecule. The absorption lines of $^{235}\text{U}$ and $^{238}\text{U}$ slightly differ due to hyperfine structure of the isotopes. The difference of energies of excited states differ to some degree because the atoms and molecules have some dependence to their atomic masses. The resonance absorption is shown in Figure 6.

AVLIS uses tunable lasers, which can be set so that only $^{235}\text{U}$ absorbs the photons and undergoes photoionization. Photoionization is the process where an incident photon ejects one or more electrons. We are then left with ionized $^{235}\text{U}$ which we can extract with an electric field produced by a collector [7].

The MLIS method is energy more efficient, due to the fact that it uses $\text{UF}_6$ which requires much lower temperatures to vaporize. Molecules are irradiated with multiple lasers, causing $\text{UF}_6$ to undergo photolysis. The $\text{UF}_6$ is separated into $\text{UF}_5$ and $\text{F}$. The uranium pentfluoride atoms form a solid, which is separated from the gas by a cyclone separator. $\text{UF}_5$ must be converted back to $\text{UF}_6$ for further processing [8].

The AVLIS method is still in research and will be commercially available in the near future. The MLIS on the other hand has been abandoned by the US, France, UK, Germany and South Africa, however Japan still has a small scale program in operation.
2.3 Gas Centrifuge

The gas centrifuge method is based on a common procedure used in biology and medicine. A fast centrifuge creates a strong centrifugal force, which pulls the heavier molecules to the edge of the centrifuge. The gas is then heated to create a convection cycle. The lighter uranium molecules are extracted at the bottom of the centrifuge while the heavier molecules are extracted at the top of the centrifuge (see Figure 7). The capacity of a single centrifuge is much smaller than that of a single difuser, but the capability to separate isotopes is much greater. These centrifuges are about 4 to 5 meters tall (some even up to 12 meters), 20 cm in diameter and rotate at about 50000-70000 rpm. The centrifugal force is about $10^4 - 10^5$ stronger than the gravitational force: $a = \omega^2 r = 1.4 \times 10^5 \text{ m/s}^2$, compared to the $g = 9.81 \text{ m/s}^2$.

The separation factor is then calculated from ideal gas equation [10]:

$$pV = nRT \quad (22)$$
$$dp = -a \rho dr \quad (23)$$

Pressure can be expressed from the ideal gas equation in the form of $p = RT \rho / M$.

$$\frac{dp}{p} = -\frac{M g}{RT} dr, \quad p = p_0 e^{-M \omega^2 r^2 / 2RT} = p_0 e^{-M v^2 / 2RT} \quad (24)$$

With the substitution of $g = \omega^2 r$, where $r$ is the radius of the centrifuge. Separation at radius $r$ is then:

$$\alpha^2 = e^{(M_2 - M_1) v^2 / RT} \approx 1 + \frac{(M_2 - M_1) v^2}{2RT} \quad (25)$$

The rotor is suspended in vacuum to allow minimal friction and resistance. Special features also include magnetic bearings at the top of centrifuge for stabilization. On the bottom of the centrifuge there is a needle-like bearing that the rotor sits on. The UF$_6$ is fed in at a pressure of 1 mbar and a typical centrifuge can hold only a few grams of inventory at a time.

Method comparison

Usage of the most common methods is shown in table 1. It is obvious that diffusion process is being abandoned due to the high energy costs [1].

<table>
<thead>
<tr>
<th>Method</th>
<th>kWh/SWU</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffusion</td>
<td>2000-3000</td>
</tr>
<tr>
<td>Centrifuge</td>
<td>100-300</td>
</tr>
<tr>
<td>Laser</td>
<td>100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Supply source:</th>
<th>2007</th>
<th>2017</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffusion</td>
<td>25%</td>
<td>0</td>
</tr>
<tr>
<td>Centrifuge</td>
<td>65%</td>
<td>93%</td>
</tr>
<tr>
<td>Laser</td>
<td>0</td>
<td>3%</td>
</tr>
<tr>
<td>HEU ex weapons</td>
<td>10%</td>
<td>4%</td>
</tr>
</tbody>
</table>

3 Cascades

There are two basic types of hook-ups, first being the serial connections. The output of one centrifuge is connected directly to the input of the second centrifuge. This type of connection is obviously not efficient,
since the enrichment factor is 1.3 and a lot of uranium is lost with waste. For a cascade of 10 centrifuges which are connected serially and an input of 10,000 kg at 0.711% enrichment, we get 8.81% enrichment and a total mass of 2.66 kg. Clearly not the best way to go.

In the parallel case the outputs of 10 centrifuges are connected and each one has its own feed. Again for a cascade of 10 centrifuges with an input of 10,000 kg and an enrichment of 0.711%, we get in the product stream an enrichment of 0.92% and a total mass of 4.362 kg. Which is basically the same as feeding the same input stream into 20 centrifuges [4].

![Figure 8: Two basic types of connections](image)

It is obvious that we waste too much uranium in the process. Therefore an ideal cascade was designed, see Figure 9. In this case, we made a simple approximation. Equation 16 is rewritten for $\alpha \sim 1$

$$\theta = \frac{1}{\alpha + 1}, \quad 1 - \theta = \frac{\alpha}{\alpha + 1}$$

(26)

$\theta$ is no longer a function of input concentration. In a cascade like this the waste from one centrifuge is fed back into the previous centrifuge to be recycled. The following diagram was constructed with the assumption given by equation 26.

![Figure 9: Ideal symmetrical cascade](image)

We start off at the product side and reverse the procedure of enrichment. The left most centrifuge splits the feed stream:

$$\theta F = 1 \rightarrow \frac{1}{1 + \alpha} F = 1 \rightarrow F = 1 + \alpha$$

(27)

$$(\theta - 1)F = T \rightarrow \frac{\alpha}{\alpha + 1} \alpha + 1 = T = \alpha$$

(28)

The procedure is then repeated until we arrive to the input stream. The reverse process is then again used from the tails side of the ideal cascade. The enrichment factor is then $\alpha ^ N$, where $N$ is the number of stages.

Cascades of this sort are not usually symmetrical meaning that the product branch is longer than the waste branch. Designing cascades is an art form itself and cascades are usually custom designed to fit each customer’s needs.

The question now is: How many stages do we need on each side? We now introduce stages, since we can have more than one centrifuge in each stage, meaning they are connected in parallel. For the product side we write

$$\frac{x_f}{1 - x_f} \alpha^N = \frac{x_p}{1 - x_p} \rightarrow \alpha^N = \frac{x_p}{1 - x_p} \frac{x_f}{1 - x_f} \rightarrow N_P = \frac{ln \left( \frac{x_p}{1 - x_p} / \frac{x_f}{1 - x_f} \right)}{ln(\alpha)}$$

(29)
and for the tails side:

\[
N_T = \frac{\ln\left(\frac{x_f \alpha}{1 - x_f} - \frac{x_t}{1 - x_t}\right)}{\ln(\alpha)} - 1 \tag{30}
\]

4 Numerical calculations

We start of the process with natural enriched uranium \(w_f = 0.00711\), and would like it to be enriched to LEU \(w_p = 0.043\), with tail concentrations of \(w_t = 0.0028\). The early gaseous centrifuges had an enrichment factor \(\alpha\) of about 1.2. Using equations 29 and 30, we get the number of series on the product and the tails sides [4]:

\[
N_P = 9, \quad N_T = 5
\]

Where the main feed series has been subtracted from the number of tails series.

The next question is how much uranium do we need, to produce 50 kg of HEU (93%). For this we use equation 8. We need a feed of 11038 kg of natural enriched uranium. The same amount would produce 1153 kg of LEU.

Separative work units needed to produce 50 kg of HEU can be calculated from equation 11 is 10382 SWU. To produce 1025 kg of LEU, we need 7096 SWU. It is now obvious that if we started to enrich from LEU to HEU, we would only need 3285 SWU.

A standard centrifuge can produce approximately 2.5 SWU/year (number varies). Producing the previous amount of LEU in a year would require 2800 centrifuges, or three years with about a 900 centrifuges.

4.1 Cascade Shape

In an ideal cascade, we would continuously feed material into the cascade. We know the number of stages needed to achieve the desired enrichment, but how many centrifuges are there in each stage?

Since designing and building centrifuges is an expensive process, they are usually identical to one another and built on a mass scale. Therefore all centrifuges in an “ideal” cascade operate best when they have the same input flow.

The number of centrifuges is a function of \(\theta\), since every centrifuge will output a product flow of \(\theta L_{in} = L_p\) and a waste flow of \((\theta - 1) L_{in} = L_t\). Yet \(\theta\) is also a function of \(w_{in}\) and \(\alpha\), see equation 16.

We now write

\[
N^i L_{in}^i = N^{i-1} L_{p}^{i-1} + N^{i+1} L_{p}^{i+1} \tag{31}
\]

Where \(N^i\) is the number of centrifuges in the \(i\) stage, \(L_{in}^i\) is the input flow, \(L_p^i\) is the output product flow and \(L_t^i\) is the tails flow. We then write \((1 - \theta^i)L_{in}^i = L_t^i\) and \(L_{in}^i = L_p^i/\theta^i\) and get

\[
L_{t}^{i+1} = \frac{1 - \theta^{i+1}}{\theta^{i+1}} L_{p}^{i+1} \tag{32}
\]

We then rewrite equation 31 and use equation 32, to get:

\[
N^i L_{in}^i = N^{i-1} L_{p}^{i-1} + \left(\frac{1 - \theta^{i+1}}{\theta^{i+1}}\right) N^{i+1} L_{p}^{i+1} \tag{33}
\]

Finally we substitute \(L_{in}^i = L_p^i/\theta^i\) and get

\[
L_{t}^i = (1 - \theta^i)L_{in}^i \quad N^i \frac{L_{p}^i}{\theta^i} = N^{i-1} L_{p}^{i-1} + \left(\frac{1 - \theta^{i+1}}{\theta^{i+1}}\right) N^{i+1} L_{p}^{i+1} \tag{34}
\]

We can now build a tridiagonal matrix \(A\) and a vector of flows, called \(\vec{L}\):

\[
A \ast \vec{L} = \vec{b} \tag{35}
\]
and solve it for

$$\vec{L} = A^{-1}\vec{b}$$  \hspace{1cm} (36)$$

Where $\vec{b}$ is a vector of zeros, except on the input stage. We can now observe flow chart for different cascade configurations. In the ideal cascade shape, the flow should be equally distributed over all the centrifuges, yet we still do not know the ideal shape.

Equation 34 can also be rewritten to solve the problem of how many centrifuges we actually need in each series since we have a condition that all flows must be same. This also means that the product flow of the last stage must be the same as previous flows and write:

$$0 = L_p^{i-1}N^{i-1} - \frac{L_p^i}{\theta}N^i + \frac{(1 - \theta^{i+1})}{\theta^{i+1}}L_p^{i+1}N^{i+1}$$  \hspace{1cm} (37)$$

Where $L_p$ is calculated from equation 8, except is defined per unit of time.

$$B*\vec{N} = \vec{b}$$  \hspace{1cm} (38)$$

Where $B$ is again a tridiagonal matrix and $\vec{N}$ is the number of centrifuges in each stage. We solve equation 38:

$$\vec{N} = B^{-1}\vec{b}$$

Figure 10 was calculated for production of 1025 kg of LEU per year and an enrichment of 4.3%, a waste of 0.28% and an average of 83 kg per centrifuge per year and an initial feed of 11038 kg.

This all adds up to 2633 centrifuges. The flow diagram from equation 36 would be a constant, yet is has some error due to rounding of numbers of centrifuges. In order to achieve HEU enrichment, we would have to pass the material about 3 times through this cascade and feed it 10 times the material, which one produces.

If we wanted to produce the HEU directly we would need only 3642 centrifuges in 30 stages, setup correctly.
5 Conclusion

Uranium enrichment is a growing industry. New and better methods are still in development and nuclear energy is still the cleanest $CO_2$ wise. Many countries are even now building new nuclear power plants and LEU consumption is going to increase. Unfortunately, enrichment plants can also be converted easily to produce weapons-grade enrichment. There are now 9 countries that posses nuclear bombs and the number is also increasing which is a growing concern.

References


