Chapter 6. Enrichment processes

I. Introduction

This chapter presents brief, quantitative descriptions of a number of uranium enrichment processes that have been developed or are undergoing serious research and development. There exist, in fact, literally hundreds of ideas for separating uranium isotopes, and those of other elements as well. New ideas are still being proposed at a steady rate, and patents for new methods are granted regularly in many countries.

However, only 10 of these techniques have shown sufficient promise to have either been developed for commercial or military use or to be the subjects of intense research and development efforts. It is the purpose of this chapter to describe these enrichment methods in enough detail to allow an assessment of the potential contribution each process might make to further nuclear weapon proliferation.

Most of the technical details of the processes are still secret, so it is necessary for the descriptions that follow to focus on those features which can be inferred from a basic physical understanding of the process. Such analyses can occasionally be misleading, but in the great majority of cases it is possible to derive useful information, especially for the purposes of policy analysis, without detailed engineering data. Since the policy problem is an important one, there seems to be no alternative to doing the best that can be done with the data which are available.
II. Gaseous diffusion

Basic principles

The basic physical principle underlying the gaseous diffusion method is the so-called ‘equipartition principle’ of statistical mechanics. This principle states that in a gas consisting of several types of molecules each type will have the same average energy of motion (kinetic energy). This equality of average energies is attained and preserved by the enormous number of collisions between molecules which are taking place at all times in the gas. These collisions ensure that any excess energy which may have been associated with one component will rapidly be shared equally with all the others. This equal sharing is called thermal equilibrium. The kinetic energy $KE$ of a molecule of mass $m$ is related to its velocity $v$ by the formula

$$KE = \frac{1}{2} mv^2$$

Therefore, molecules which have the same average kinetic energy will have average velocities which differ in inverse proportion to the square roots of their masses. Using the symbol $\langle \rangle$ to denote the average, the relationship can be written

$$\langle KE_1 \rangle = \langle KE_2 \rangle$$

$$\frac{1}{2}m_1\langle v_1^2 \rangle = \frac{1}{2}m_2\langle v_2^2 \rangle$$

or

$$\frac{\langle v_1 \rangle}{\langle v_2 \rangle} = \sqrt{\frac{m_2}{m_1}}$$

For uranium hexafluoride gas made up of $^{235}\text{UF}_6$ and $^{238}\text{UF}_6$ the respective molecular masses are 349 and 352, so the ratio of the velocities is 1.0043, with the lighter $^{235}\text{UF}_6$ molecules moving very slightly faster on the average. This last phrase is important, because the molecules move with a wide range of velocities, and the equipartition principle applies only to averages over large numbers of molecules.

The gaseous diffusion method of separation exploits this slight difference in average velocities by forcing the gas mixture to diffuse through a porous barrier under a pressure difference. The barrier is a thin wall of solid material containing many very small holes or passageways. The faster molecules will encounter the holes more often than the slower ones and will therefore be slightly more likely to pass through, causing the gas which emerges on the other side of the barrier to be enriched in the lighter isotope. This method is one of the oldest for separating isotopes and was first used by Aston in 1920 to partially separate isotopes of neon [11a].

The number of molecules of each type which emerge on the product side of the barrier is proportional to the rate of flow through the barrier of that type of molecule. This in turn depends on both the number of each type present on the feed side and their average velocities. So the ratio of
numbers emerging on the product side is

\[ R_p = \frac{N_{235}}{N_{238}} \frac{\langle v_{235} \rangle}{\langle v_{238} \rangle} = R_f \sqrt{\frac{M_{238}}{M_{235}}} \quad (6.1) \]

Therefore, the ratio of the average velocities is the ideal enrichment factor \( \alpha \), defined in chapter 5 (see equation 5.7, p. 104). The actual enrichment factor is less than this for two reasons. First, the concentration of the desired isotope on the feed side is not constant; it gradually decreases as the enriched mixture diffuses through the barrier. If the cut is assumed to be one-half, then this effect reduces the ideal enrichment gain \( \alpha - 1 \) by a factor of 0.69. An ideal cascade containing ideal separation elements would therefore have \( \alpha = 1.00297 \), corresponding to a stage separation factor \( q = 1.00595 \) (see chapter 5). This is further reduced by the separation 'efficiency', to be discussed below.

The diffusion barrier

The central problem in the use of gaseous diffusion is the manufacture of a suitable barrier material. The difficulty in making the barrier can be appreciated if one lists the properties it must possess.

1. The average diameter of the holes (pores) in the barrier must be much less than the average distance travelled between collisions (the 'mean free path') of a molecule. If this is not satisfied then a molecule is likely to suffer one or more collisions near the entrance to the pore and inside the diffusion channel. This would cause exchanges of energy with other molecules and tend to cancel out the slight velocity difference between light and heavy molecules. At the same time the holes must be large enough to allow the gas to pass through at a reasonable rate.

2. The barrier must be very thin so as to have an adequate permeability at reasonable pressure, but it cannot be so thin as to break under the necessary pressure difference across it. It is desirable to have this pressure difference as large as possible to increase the rate of flow through the barrier.

3. The barrier material must be highly resistant to corrosion by the very corrosive gas UF\(_6\). Any corrosion which occurs will cause plugging of the tiny holes in the barrier.

Requirements 1 and 2 can be made more quantitative by considering the actual properties of UF\(_6\) gas. A UF\(_6\) molecule has a diameter of about 0.7 nanometres (nm), and at a pressure of about one-half atmosphere and a temperature of 80°C the average separation between molecules is about 5 nm. The mean free path under these conditions is about 85 nm, so the average pore opening must be somewhat less than this, say about 25 nm. Using a rough geometrical argument it can be shown that such pore dimensions can be obtained by packing together spheres whose diameters are about 100 nm (0.1 \( \mu \)m). Figure 6.1 shows the relationship between these quantities drawn roughly to scale. Figure 6.2 shows an actual
Figure 6.1. Model diffusion barrier
This schematic diagram shows a barrier made of closely packed spheres 100 nm in diameter, giving average pore openings comparable to circles 25 nm in diameter. The average spacing between UF₆ molecules (the small dots below the barrier) is about 5 nm, and the mean free path of the UF₆ molecules is 85 nm.

Figure 6.2. A sintered nickel barrier
Photomicrograph of a piece of sintered nickel barrier cut parallel to its surface.

Source: Commissariat à l'Énergie Atomique, Paris, France.
photomicrograph of a piece of barrier made from nickel powder which has been sintered (i.e., packed together under high pressure and heated to a high temperature).

It is now easy to understand why a barrier is quite difficult to produce. The actual methods used by various countries are classified, but it is known that the United States uses sintered nickel powders [2n], while those in the new French Tricastin plant are “ceramic” [12].

Japanese researchers have experimented with nickel, aluminium and Teflon barriers [13a] and although the details of barrier manufacture are generally held to be secret, some publicly available Japanese patents are quite explicit in their descriptions of proposed methods [14].

Whatever the material, it must be bonded under high pressure and temperature into sheets only a few microns thick. These very thin sheets must be able to withstand pressure differentials of the order of 0.3 to 0.5 kg/cm² for many years without failure. Generally this requires a carefully designed supporting structure, and possibly even multilayer barriers composed of materials with different porosities and strengths [2o]. That this kind of reliability is indeed achievable can be seen in the record of US plants in which “barrier failures are too rare to justify maintaining separate records” [15a].

The barrier must be assembled in a way which will maximize its area of contact with the gas. In US gaseous diffusion stages this is done by manufacturing the barrier in the form of sheets of small tubes assembled in cylindrical tube bundles [2n].

The performance of a barrier depends not only on its own properties but also on the pressure and temperature of the gas which comes into contact with it. For example, as the pressure on the feed side increases, the mean free path gets smaller and the probability of collisions in or near the pores is increased. This reduces the efficiency of the barrier. It is also desirable to keep the pressure on the product side (back pressure) low in order to prevent too many molecules from diffusing backwards through the barrier. High temperatures would make the molecules move faster, thereby increasing the diffusion rate, but raising the temperature also forces the compressors to do more work for a given amount of gas. So the temperature must be kept as low as possible without allowing the gas to condense or solidify. Keeping the temperature low also reduces corrosion problems.

**A gaseous diffusion stage**

In the design of a separation stage all of these factors must be balanced in an optimum way. The operating pressures and temperatures, the properties of UF₆ gas, and the structure of the barrier all combine to produce a barrier efficiency $e_B$, which is a number somewhat less than 1. The actual separation gain is related to the ideal value by

$$ g = e_BG_{\text{ideal}} $$
The structure of the barrier also determines its ‘permeability’, or the rate of flow of gas through a given barrier area for a given pressure difference across it. Using data obtained from experiments with argon it is possible to estimate the dependence of permeability on pore size [13b]. Knowing the permeability of the barrier and its efficiency, it is then possible to compute how much separative power can be produced by a given area of barrier material. As an illustration assume that the barrier efficiency is about 0.7. Then the actual separation gain will be

\[ g = 0.7 \times 0.00595 = 0.0042 \]

which can be shown to lead to about 1 kg SWU per square metre of barrier per year. Therefore a plant such as the one at Tricastin, with a total capacity of 10.8 million SWU/yr, might have a total barrier area of \(10^7\) m\(^2\), or about 10 square kilometres. A single large stage with a capacity of about 12 000 SWU/yr will have a barrier area of roughly 1.2 hectares (3 acres).

The individual tubes which make up the barrier must be small enough to provide a large surface area for diffusion but large enough to permit easy flow of the process gas. Again, no information is available on the size of the tubes, but if it is assumed that the tubes are about 2 m long and 1 cm in diameter, then about 160 000 of them would be used in such a stage. This can be compared with some of the early US stages which contained several thousand tubes each [16].

Using the above value of \(g\) in table 5.1 (p. 113), it can be shown that an ideal gaseous diffusion cascade for the production of 3 per cent enriched product with a 0.2 per cent tails assay would have about 1 290 stages.

Table 5.1 can also be used to calculate the total cascade flow and the flow per stage. For a \(10^7\) SWU/yr gaseous diffusion plant the product flow \(P\) would be \(2.4 \times 10^6\) kg/yr, and the total cascade flow would be between \(4.6 \times 10^{12}\) and \(6.0 \times 10^{12}\) kg/yr. If the cascade were squared off, a typical large unit might carry a flow of \(5.5 \times 10^9\) kg/yr or 174 kg U/s. This implies a flow through a given stage of 250 kg UF\(_6\)/s, a value comparable to the rated capacities of the largest compressors at Tricastin (190 kg/s) [17a].

It should be emphasized once more that the numbers used here are only estimates designed to produce approximate values for stage and plant parameters. The precise values of numbers such as barrier efficiency are well-guarded secrets.

In a typical gaseous diffusion stage (see figure 6.3) feed enters the diffuser at a pressure of between one-third and one-half atmosphere [18a], and half of it is allowed to diffuse through the barrier tubes. The low pressure diffused gas is then cooled by passing it through a heat exchanger and drawn into a compressor where it is compressed and mixed with the depleted material from a higher stage. The precooling of the gas is necessary because the compression heats it substantially, and most of the compression energy must be rejected to maintain a steady operating temperature. This wasteful procedure accounts for the high energy
Figure 6.3. Gaseous diffusion stages

Three of the gaseous diffusion stages used at the French Tricastin facility shown connected in a symmetric cascade. In the stage at the right the three labelled components are: 1. diffuser, 2. heat exchanger, 3. compressor. Note that the product gas from the diffuser is cooled in the heat exchanger before it is compressed and sent to the next stage. This explains why the product and tails flows do not match those of figure 5.5.


consumption of gaseous diffusion plants, which is generally between 2 300 and 3 000 kilowatt hours for every SWU produced [2p].

The output of the compressor is sent forward to the next stage while the depleted half of the gas is drawn off through a separate port and sent back two stages where it is mixed with the diffuser output and recycled.

Modern compressors are of the axial flow type, similar to those used in jet aircraft engines (see figure 6.4). The compressor rotor rotates at high speeds and must handle large flows of corrosive UF₆ at relatively high temperatures for long periods (many years) of continuous operation. And because the entire UF₆ carrying system must be leak-proof, the compressor shaft bearings require special rotating seals and the use of nitrogen gas to isolate the UF₆ from the shaft lubricants [2q]. The giant compressors used at Tricastin have all been nickel-plated to prevent corrosion [17b].

The barrier and the compressors are the key components in a gaseous diffusion plant. All the rest is really just some high-quality plumbing. Detailed design and manufacturing data on the barrier and compressors are classified, but history has shown that all countries which have set out to build a gaseous diffusion plant have succeeded. This suggests that the secrets are probably less important in inhibiting the further spread of
gaseous diffusion technology than the enormous capital costs and industrial effort involved.

It remains to provide an estimate of the hold-up time in a gaseous diffusion stage. One way to do this is to use an estimated value for the 'specific hold-up', which is defined as the amount of uranium which must be kept in inventory to provide a separative power of 1 SWU/yr. One value which has been given is a specific hold-up of "not higher than 0.1 kg U/kg SWU/yr" [2r]. This can be used along with the value of $g = 0.0042$ in table 5.1 to show that $t_h = 6$ seconds. Given the uncertainty in this calculation it would be prudent to use 5–10 seconds as a range of possible values. Using this range the equilibrium time of a commercial cascade making 3 per cent product comes out to a minimum of 11 days if the lower value of $t_h$ is applied to an ideal cascade. A more reasonable estimate for a real cascade might be roughly three weeks.

The low single-stage enrichment factor and relatively long hold-up time combine to make gaseous diffusion a very large-scale, capital- and
energy-intensive process. To produce nuclear weapon-grade uranium from natural feed would require over 3 500 stages and an equilibrium time of at least a year. Production of the barrier, compressors, and piping would be major industrial undertakings, and the construction of the facility would put a heavy drain on the resources of most countries of the world. So, even though gaseous diffusion has been the major historical contributor to proliferation, it seems unlikely to continue to play this role in the future, especially in view of the capabilities of some of the other enrichment techniques under development or already in use.

III. The gas centrifuge

Basic principles

A good model for understanding the way in which a centrifuge separates a mixture of isotopes is to imagine a sample of gas in a room under the influence of gravity. Since each molecule is being pulled downwards, a certain amount of work must be done to lift the molecule to some height $h$. This work represents an increase in the molecule’s ‘potential’ energy, and this change is given simply by

$$\text{PE} = mgh$$

where $m$ is the molecule’s mass and $g$ is the acceleration due to gravity, equal to 9.8 m/s$^2$ at the Earth’s surface. If the temperature in the room is the same everywhere, molecules at all heights have the same average kinetic energy, but those near the ceiling have a higher potential energy than those near the floor. The same theory that predicts the equipartition of energy (see chapter 1) predicts that higher energies are less probable than lower ones. It predicts that the density of particles near the ceiling will be less than the density near the floor by the factor

$$\frac{N(h)}{N(0)} = \exp \left( \frac{-mgh}{RT} \right)$$

where $R$ is called the gas constant (8.3 joules per degree Kelvin), and $T$ is the temperature measured from absolute zero ($0\,\text{K} = -273^\circ\text{C}$). If two different species are present in the mixture, then an equation like 6.2 can be written for each one and the ratio taken:

$$\frac{N_1(h)}{N_1(0)}\frac{N_2(h)}{N_2(0)} = \exp \left[ \frac{-[(m_1 - m_2)gh]}{RT} \right]$$

If the factors on the left are slightly rearranged, the ratio can be seen to be equivalent to the ratio of the relative isotopic abundances at the heights $h$ and zero. So this equation can be rewritten as

$$\frac{R(h)}{R(0)} = \exp \left[ \frac{-(m_1 - m_2)gh}{RT} \right]$$
In this form it is seen to be nearly analogous to equation 6.1 for gaseous diffusion. Note that if subscript 1 refers to the lighter species, then equation 6.4 is consistent with the relative abundance of this species increasing with $h$. This calculation suggests that it should be possible in principle to separate isotopes of uranium by filling a room with UF$_6$, allowing it to come to thermal equilibrium, and then simply skimming off the top portion of the gas. Experiments similar to this were in fact performed over 80 years ago \[2s\]. However, this is not a practical process for uranium, since the effect is extremely small. For example, in a room 3 metres high at normal temperature the ideal separation gain for UF$_6$ would be only $3.4 \times 10^{-5}$, over 100 times smaller than for gaseous diffusion.

It was recognized very early that a rapidly rotating centrifuge could provide a much stronger force field and therefore increase the separation gain many times. In a centrifuge the acceleration of gravity is replaced by the 'centrifugal' acceleration, and the equation corresponding to 6.4 can be shown to be

$$\frac{R(r)}{R(0)} = \exp \left[ (m_1 - m_2) \left( \frac{\omega^2 r^2}{2RT} \right) \right] \quad (6.5)$$

where $r$ is the distance from the centre of the centrifuge, and $\omega$ is the angular velocity in radians per second. Notice that the sign of the exponential factor has changed, implying that the isotopic abundance of the lighter species increases towards the centre of the centrifuge. The wall of the centrifuge is then analogous to the floor in the previous example.

To see what equation 6.5 predicts for uranium enrichment consider a hypothetical centrifuge with a radius of 10 cm and an angular frequency of 800 revolutions per second ($\omega = 800 \times 2\pi = 5000 \text{ rad/s}$). The acceleration at the wall of this centrifuge is $2.5 \times 10^6 \text{ m/s}^2$ or more than 250 000 times as strong as gravity. Using $m_1 = 0.349 \text{ kg}$ and $m_2 = 0.352 \text{ kg}$ and assuming a temperature of 330 K ($57^\circ\text{C}$) the ratio of abundance between the centre and outer wall is found to be 1.147. The ideal value of $g = 0.147$ for this centrifuge can be seen to be over 16 times as large as the corresponding value for gaseous diffusion. Note also that the separation gain depends on the simple difference between the isotope masses, not on their ratio. This means that the advantage of the centrifuge over gaseous diffusion improves as the isotopic masses increase.

**Modern centrifuges**

All of these advantages were recognized by the early workers in isotope separation, and centrifuge research was actively pursued in initial efforts in both the USA and Germany during World War II (see p. 16). But very

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1. Actually the exponential quantity in equation 6.4 must be interpreted as a separation factor rather than an enrichment factor (see p. 95). Therefore equation 6.4 is analogous to the square of equation 6.1.
quickly a number of disadvantages were discovered, and it has taken many years of active research and development, and a number of important technological advances, to bring centrifuges into competition with gaseous diffusion. The following is a list of these problems and their solutions [19a].

1. The ideal separation factor derived above is actually the ratio of relative isotopic abundances at the centre of the centrifuge to that at the wall. To take full advantage of this difference, product would have to be extracted at the centre and tails at the wall; but the rapid rotation of the centrifuge causes virtually all the gas to concentrate near the wall. For example, in the above centrifuge the pressure of the gas at the wall can be shown to be 40 million times that at the centre [20a]. So it would be useless to attempt to extract any product, no matter how enriched, from near the centre. In fact all the separation must take place in a narrow annular region near the wall of the centrifuge, and this greatly reduces the separation effect unless a ‘countercurrent’ flow pattern is created.

The countercurrent flow is a form of internal reflux which causes a continual recirculation of the gas in the centrifuge, allowing a long-term exchange to take place between layers of differing isotopic concentrations [21] (see figure 6.5). The countercurrent flow is induced by aerodynamic interactions between the rotating gas and the bottom scoop. The lighter component tends to concentrate in the inner, rising layer and therefore at the top, while the heavier component concentrates at the outside and bottom. Feed is injected near the centre.

2. Normally when an object is rotated at high speeds it must be carefully balanced to prevent wobbling and vibration. At the rotational speeds necessary to get useful separation factors for UF₆ this problem becomes very severe. In addition to these difficulties there is a problem of ‘critical’ rotation frequencies at which the centrifuge can be set into a kind of resonant vibration which can grow to large amplitudes and destroy the centrifuge. Since it is desirable to operate the centrifuge above these critical frequencies if possible, the problem is to find ways both of damping these vibrations and of designing a centrifuge and bearings which can tolerate the stresses of passing through these frequencies as the centrifuge is accelerated.

These problems were solved in the late 1950s by the use of a simple oil-lubricated pivot and cup bearing at the bottom and a magnetic bearing at the top [19b]. In the latter there is no physical contact between the rotor shaft and housing, and consequently no friction. The magnetic bearing which holds the shaft in suspension is also designed for damping vibrations, as is the lower cup and pivot bearing (see figure 6.5). Friction is further reduced by enclosing the centrifuge rotor in a casing which is maintained at very high vacuums, probably between one-millionth and one ten-millionth of an atmosphere [22]. The casing must not only be leak-proof but must also be strong enough to contain the debris of a failed centrifuge.

3. The very high rotational speed of the centrifuge leads to severe mechanical stresses in the outer wall. For example, the tensile stresses in the wall of an aluminium centrifuge with a radius of 10 cm and a rotational
Figure 6.5. A modern gas centrifuge

The thin-walled rotor is driven by a small electromagnetic motor attached to the bottom of the casing. The top end of the rotor is held in a vertical position by a magnetic bearing and does not touch stationary components. Gas is fed into and withdrawn from the rotor through the stationary centre post, which holds three concentric tubes for the feed, the product and the waste. The stationary bottom scoop protrudes into the spinning gas and provides a mechanical means for driving the vertical flow of gas. The top scoop, which serves to remove the enriched product, is protected from direct interaction with the rotating gas by the baffle, which has holes allowing the enriched gas to be bled into the area near the scoop. The baffle is needed to keep the top scoop from imposing a vertical flow that would counteract the crucial one generated by the bottom scoop.

Table 6.1 Typical maximum peripheral speeds of thin-walled cylinders [20b]

<table>
<thead>
<tr>
<th>Material</th>
<th>Tensile strength $T$ (kg/cm²)</th>
<th>Density $\rho$ (g/cm³)</th>
<th>$T\rho$ ($\times 10^3$)</th>
<th>Approximate maximum peripheral speed (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium alloy</td>
<td>5 200</td>
<td>2.8</td>
<td>1.9</td>
<td>425</td>
</tr>
<tr>
<td>Titanium</td>
<td>9 200</td>
<td>4.6</td>
<td>2.0</td>
<td>440</td>
</tr>
<tr>
<td>High-strength steel</td>
<td>17 000</td>
<td>8.0</td>
<td>2.1</td>
<td>455</td>
</tr>
<tr>
<td>Maraging steel</td>
<td>22 500</td>
<td>8.0</td>
<td>2.8</td>
<td>525</td>
</tr>
<tr>
<td>Glass fibre/resin</td>
<td>7 000</td>
<td>1.9</td>
<td>3.7</td>
<td>600</td>
</tr>
<tr>
<td>Carbon fibre/resin</td>
<td>8 500</td>
<td>1.7</td>
<td>5.0</td>
<td>700</td>
</tr>
</tbody>
</table>

frequency of 800 rev/s will be 7 000 kg/cm², more than 1.3 times the tensile strength of aluminium (see table 6.1). In other words an aluminium centrifuge would have exploded long before these rotational speeds were achieved. This problem has been solved by the creation of new high-strength, lightweight materials, such as carbon or glass fibres, some of the properties of which are compared with older materials in table 6.1. It should be emphasized that the peripheral speeds in the last column of the table are the ‘bursting’ speeds of the materials. Actual centrifuges made of these materials would be run at lower speeds to allow an ample safety margin and to prevent excessive distortion (‘creep’) of the rotor material over time.

4. The separative power of a centrifuge is very sensitive to the detailed nature of the countercurrent flow pattern. Until the hydrodynamic equations governing this pattern were understood it was only possible to improve centrifuge performance by empirical, trial-and-error methods. In recent years substantial progress has been made in developing computer codes for solving these very complicated equations [2t]. This has made it possible to optimize centrifuge designs in much more systematic and predictable ways.

With the solutions to these technical problems it has been possible to produce centrifuges with very high separation factors, at least 1.5, and with separative powers of anywhere from 5 to 100 kg SWU/yr [23a]. The large variation in this latter rating reflects substantial differences in the sizes and rotational speeds of the centrifuges. European and Japanese machines tend to be relatively small, while the United States has developed substantially larger machines with separative powers about 10 times those of the European and Japanese designs [2u].

The major factor limiting the separative power of a centrifuge is the very low throughput. This is limited by the slow diffusion rate in the gas and by the requirement that the pressure of the UF₆ at the centrifuge wall
be below its sublimation vapour pressure at the operating temperature, usually normal room temperature, 20°C. If this latter condition is not satisfied, then solid UF₆ will deposit on the walls of the centrifuge, an obviously undesirable situation.

The vapour pressure of UF₆ at room temperature is only about 0.1 atmosphere [2v], and if this is taken to be the upper limit of the pressure at the wall, the pressure on the axis must be about 2.5 × 10⁻⁹ atmosphere, a very good vacuum. Using these limiting pressures and the exponential form for the density similar to equation 6.2, it can be shown that the total amount of uranium in a centrifuge 1.5 m long and 20 cm in diameter spinning at 800 rev/s is only about 0.25 g.

The separative power of a centrifuge is optimized by determining the proper ratio of product withdrawal to countercurrent flow, that is, the reflux ratio. With this criterion and certain assumptions about efficiency, it is possible to derive all the properties of a model centrifuge [24a]. Using these results the hypothetical centrifuge described in this section can be shown to have a separation factor of 1.51 and an optimum separative power of 15.2 kg SWU/yr. This puts it somewhat higher than the values typically quoted for European and Japanese centrifuges, but considerably lower than US values. The properties of this model centrifuge are summarized in table 6.2. It must be emphasized that these values are both hypothetical and approximate. They are hypothetical because the detailed operating characteristics of actual centrifuges are classified, and they are approximate because the formulae needed to derive them are accurate only for values of g ≤ 1 [24b]. However, they should be close enough to realistic values to give a reasonably reliable description of the capabilities of a modern gas centrifuge.

Table 6.2. Properties of a hypothetical centrifuge

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radius</td>
<td>10 cm</td>
</tr>
<tr>
<td>Length</td>
<td>150 cm</td>
</tr>
<tr>
<td>Rotational frequency</td>
<td>800 rev/s</td>
</tr>
<tr>
<td>Peripheral speed</td>
<td>500 m/s</td>
</tr>
<tr>
<td>Separation factor (q)</td>
<td>1.51</td>
</tr>
<tr>
<td>Separative power</td>
<td>15.2 kg SWU/yr</td>
</tr>
<tr>
<td>Inventory</td>
<td>0.26 g U</td>
</tr>
<tr>
<td>Throughput</td>
<td>600 kg U/yr = 0.019 g U/s</td>
</tr>
<tr>
<td>Hold-up time</td>
<td>13.7 s</td>
</tr>
</tbody>
</table>
A centrifuge cascade

The values of table 6.2 can now be applied to the design of one of the cascades of table 5.1. For example, a commercial plant producing 3 per cent product with 0.2 per cent tails would require roughly 11 stages. Note that with \( g = 0.51 \) the approximation \( g \approx 1 \) is not really appropriate, and a more complicated calculation would have to be performed to get a better value. If the more precise \( \ln q \) is used instead of \( g \) in table 5.1 to compute the number of stages the number is increased from 11 to 13.

Notice also that it is appropriate to use ideal cascade rather than square cascade values in table 5.1 because the use of many identical centrifuges allows the stages to be adjusted in width to approximate an ideal cascade (see figures 6.6(a) and (b)).

For a plant rated at 1 000 t SWU/yr about 66 000 of the centrifuges of table 6.2 would be required. The flow in the widest stage would be \( 5.9 \times 10^6 \) kg U/yr, requiring that just under 10 000 centrifuges be placed in this stage. In practice the plant would probably be divided into modules, as is the Urenco facility at Almelo [25]. For example, a 1000 t SWU facility could be built up of ten 100 t modules, with each module being brought into production as it is completed. This kind of flexibility is one of the factors that makes the centrifuge so attractive compared to gaseous diffusion.

Another important advantage of centrifuges is their relatively low energy consumption. Quoted values range from 100 to 300 kWh/kg SWU, roughly a factor of 10 better than gaseous diffusion [23b]. If it is assumed that one of the centrifuges in table 6.2 uses 200 kWh/kg SWU, then the operating power of the centrifuge is only 350 W, and 66 000 such machines would require only a relatively small 23 MW power plant.

Finally, the equilibrium time of a commercial cascade is essentially negligible, only of the order of 2.5 minutes. It takes much longer than this to bring the centrifuges up to operating speed. The total plant inventory is also very small. The 0.26 g per centrifuge becomes a total of only 17 kg of uranium in the entire facility. This not only substantially reduces capital costs but also allows for much greater accuracy in accounting for material input and output. This has important implications for the application of safeguards to centrifuge plants (see chapter 3). The very short equilibrium time and small inventory suggest that a small centrifuge cascade might be easily used in a batch recycle mode to produce highly enriched uranium. However, this would require stopping and cleaning out all the centrifuges before restocking them with enriched feed material. This stopping and restarting of the centrifuges is both time-consuming and potentially damaging to the centrifuges, which must be accelerated through critical frequencies to bring them to operating speed. It would seem that commercial centrifuges are designed to suffer this experience only once and then to run at their nominal speed for many years without stopping [25]. Only systematic testing would determine how resistant they are to
Figure 6.6. Two centrifuge cascades
(a) Cascade hall at Almelo, the Netherlands

(b) Section of a Japanese cascade
repeated accelerations through the critical frequencies. Presumably such tests have been carried out, but the results are not available.

This discussion of the centrifuge can be summarized by stating that the technique seems to have reached maturity and is ready to compete on very favourable terms with gaseous diffusion. It is also clear that the centrifuge presents a qualitatively different and substantially more serious problem with respect to nuclear weapon proliferation than does gaseous diffusion. This aspect of the centrifuge is examined further in chapter 2.

IV. Aerodynamic separation methods

Basic principles

There are a large number of techniques for separating isotopes which can be classified as 'aerodynamic'. A good definition which includes most of them is: "... aerodynamic separators are characterized as those involving preferential diffusion of disparate masses either driven principally by a pressure gradient generated by streamline curvature (Type I) or through molecular processes that involve large perturbations from an equilibrium distribution (Type II)" [26a].

The concept of diffusion across a streamline is quite analogous to the diffusion against a gravitational or centrifugal force which was discussed in the previous section. A streamline in a flowing fluid is a line across which no net material transport takes place. So all separation processes in the fluid occur in directions perpendicular to streamlines. For example, in a centrifuge the gas is moving in circular paths, so the streamlines are concentric circles. The isotope separation takes place in the radial direction, perpendicular to these lines. If a streamline is curved, this implies that the gas is being accelerated, and that a pressure gradient (i.e., force) must exist perpendicular to the streamline. This situation is common to all Type I aerodynamic processes, and both of the aerodynamic processes which have achieved or are approaching commercial viability (the German nozzle process and the South African advanced vortex tube process) are of Type I.

The large perturbations which characterize Type II processes can either be strong density or pressure gradients or the interaction between two rapidly moving jets of gas. These methods have been studied for some time under laboratory conditions, but so far none has been developed to even a prototype level for the separation of uranium isotopes. Economic studies based on early experimental data have generally concluded that the Type II aerodynamic methods are not likely to compete successfully with currently workable methods [27a, 28a]. Although experience warns that such assessments should be viewed with caution, there is at present no
compelling reason to consider these methods in detail in this book. However, a few will be described briefly in section VIII of this chapter.

The jet nozzle process

The aerodynamic nozzle process invented and developed by E. W. Becker and his associates has been the most successful to date of all the aerodynamic processes. In the Becker process a jet of gas consisting of roughly 96 per cent hydrogen and 4 per cent UF₆ is allowed to expand through a narrow slit [2w]. The gas moves at high speeds (comparable to those at the periphery of a modern centrifuge) parallel to a semicircular wall of very small radius of curvature (see figure 6.7). If the speed of the gas is 400 m/s, and the radius of curvature is 0.1 mm, then the centrifugal acceleration achieved is $1.6 \times 10^9$ m/s² or 160 million times gravity. The accelerations exceed even the high values achieved in centrifuges by a factor of a thousand or more, and they are achieved in an apparatus with no moving parts. The centrifugal forces on the molecules cause the streamlines of the heavier components of the gas to move closer to the curved wall than those of the lighter components as the gas flows around the semicircle. At the other side, where the gas has changed direction by $180^\circ$, a sharp 'skimmer' separates the flow into an inner light fraction and an outer heavy fraction.

Figure 6.7. A separation nozzle

A cross section of the separation nozzle system currently in use. The knife edge skimmer at the right is placed so that 25 per cent of the UF₆ in the feed goes into the light fraction and 75 per cent into the heavy fraction.
Figure 6.8. A separation nozzle element
(a) Stack of photoetched metal foils forming a separation nozzle structure. The diameter of the curved deflection wall is 0.2 mm

(b) Assembly of separation nozzle elements by stacking metal foils into chips which are then set into a tube. A jet nozzle stage comprises a large number of such tubes (see figure 6.10)
The position of the skimmer is arranged so that one-quarter of the total UF$_6$ content is extracted in the light fraction. This is the 'cut'. The other three-quarters in the heavy fraction is depleted in $^{235}$UF$_6$ and forms the tails from the separating element.

The hydrogen gas plays two crucial roles in amplifying the separative effect of the nozzle. First, its low molecular weight greatly decreases the average molecular weight of the process gas. This increases the sonic velocity of the gas and allows the flow through the nozzle to be subsonic. This eliminates shock effects which would otherwise absorb energy and create turbulence and some remixing of the separated isotopes. The UF$_6$ molecules are dragged along by the fast-moving hydrogen and achieve much greater peripheral speeds than could be achieved in pure UF$_6$ gas.

The second beneficial effect of the hydrogen is also the result of drag forces it exerts on UF$_6$ molecules. As the latter move around the curve they tend to 'fall' to the outside, and if it were not for the hydrogen they would fall at the same rate. However, the drag forces exerted by the hydrogen act unequally on the light and heavy isotopes and cause the $^{235}$UF$_6$ molecules to fall more slowly than the $^{238}$UF$_6$. This is a non-equilibrium effect, which substantially enhances the separation as long as the skimmer takes its cut before equilibrium is achieved [29].

The extremely small size of the separative nozzle allows the process to operate at relatively high pressures and velocities and to also have the gas flow 'laminar', or non-turbulent. In fluid mechanics an index called the Reynolds number is used to indicate the transition from laminar to turbulent flow, and the transition is at a value of around 2 000. The Reynolds number is proportional to the linear dimensions of the nozzle, and in order to keep the value at about 100, well under the onset of turbulence, the slit can be only about 0.05 mm wide or about one-half of the curvature radius (see figures 6.8(a) and (b)). This implies that the rate of material flow must be very small in a single nozzle, and that many thousands of nozzles are necessary to make up an enrichment stage of reasonable size.

The tiny nozzle elements are produced on thin metal foils by a photoetching process [2x]. The foils are then stacked to make elements or chips with slit lengths of a few millimetres and these are enclosed in two cover plates. One cover plate has rows of holes which open into the feed chambers of all the elements and the other cover plate has holes which connect to the tails chambers. The light, enriched fraction emerges from the sides of the strip. Then 80 chips are mounted in a tube which is two metres long. The feed is admitted to the chamber on one side of the tube and the tails extracted from the other. The enriched product emerges from the space between the two tube halves. It is claimed that such a tube can produce a separation gain of 0.0148 with a cut of 0.25, and a separative work capacity of 50 kg SWU/yr [30].

Before these data can be used to construct a cascade the cascade theory developed in chapter 5 must be modified to take account of the
Figure 6.9. An asymmetric cascade
A cascade assembled from elements with a cut of one-quarter. Product from stage \( n \) is sent forward to stage \( n + 3 \) while tails are sent back one stage. Note the three product streams proceeding upward in parallel.

3 Product streams

1 Tails stream

asymmetrical cut of the nozzle process. The essential criterion that must be satisfied is that streams of differing concentrations are not mixed. Applying this criterion leads to a set of equations for an ideal asymmetric cascade whose solutions are similar in form to those of the ideal symmetric cascade but which require a different arrangement of process streams [2y]. In particular, if the cut is \( 1/n \) then the product stream from a stage must be sent forwards \( n-1 \) stages and the tails stream backwards one stage. When \( n = 4 \), which corresponds to the nozzle process, this results in a cascade like that of figure 6.9. The figure shows that in an asymmetric cascade there are several process streams moving up the cascade in parallel. As a general rule for a cut of \( \theta = 1/n \) there will be \( n-1 \) such streams.

If the separation factor of the stages is close to 1, then a full analysis of the asymmetric cascade shows that all of the formulae of chapter 5 and table 5.1 can be used directly, with the following two adjustments [2z].

1. Product flow \( P \) must be taken as the sum of the product flows from all parallel streams.

2. The separation gain \( g \) is to be replaced by \( 2\theta g \).

As an example consider a jet nozzle facility for production of 3 per cent product and 0.2 per cent tails. With \( g = 0.0148 \) and \( \theta = 0.25 \) the number of stages in an ideal cascade comes out to be 743. For a square cascade the result is 649. This can be compared with the data given for the
Figure 6.10. A prototype jet nozzle stage
Feed enters from the right centre and passes through the separation elements arranged around the top chamber. The light, enriched fraction is drawn down through a heat exchanger and compressor and sent on to another stage. This test prototype has the tails being recycled directly and mixed with the feed.

conceptual design of a large enrichment plant based on the nozzle process and designed to produce 3 per cent product with 0.34 per cent tails [15b]. This is a squared-off cascade having two different sizes of cascade element and containing a total of 570 stages. Considering the higher tails assay, this compares quite well with the estimate in table 5.1.

The higher than usual tails assay quoted for this demonstration plant is almost certainly connected to the high energy costs of SWUs in a nozzle plant. The pumping and pressurizing of large quantities of gas demands a great deal of energy, even more than for gaseous diffusion. This results mainly from the fact that only 4 per cent of the process gas is UF₆. The energy costs for the nozzle process are generally quoted as between 3 000 and 3 500 kWh/SWU [23b]. As was shown in chapter 5 such high energy costs could dictate higher tails assays, since the cost of the extra ²³⁵U thrown away could be less than that of the energy which would be needed to retrieve it. Whether or not this will be true in full-sized commercial jet nozzle plants remains to be seen.

Figure 6.10 shows a prototype stage element. It has many features in common with a gaseous diffusion stage including a compressor, heat exchanger and 'diffuser' section. It is in the latter that the actual separation takes place. Given these similarities it seems likely that the hold-up time for a jet nozzle stage is comparable to or possibly somewhat less than that of a gaseous diffusion stage. In the absence of any published information on hold-up time it will be estimated that the value is between 1 and 5 seconds.² Since the effective separation gain \(2\theta_g = 0.0074\) is larger than that for gaseous diffusion, and the hold-up time possibly somewhat shorter, the inventory requirements and equilibrium time for a nozzle cascade should be lower than for gaseous diffusion.

One special problem which arises in the aerodynamic processes is the handling of hydrogen. As was mentioned in chapter 5, UF₆ reacts strongly with hydrogen at elevated temperatures, so care must be taken to keep this reaction from occurring. In addition, the separation nozzles tend to separate the hydrogen from the UF₆. It turns out that this problem is not serious when the elements are connected in a cascade, presumably because the product stream which has been mildly enriched in \(^{235}\)UF₆ but strongly enriched in \(\text{H}_2\), is mixed at the entrance to the next stage with a tails stream which has been depleted in \(\text{H}_2\). But there is still a problem at the product outlet of the cascade where the hydrogen has to be purified and recycled to the bottom of the cascade [2aa].

An interesting recent development which will be applied in the new Brazilian facility is the use of a more elaborate nozzle system to divide the gas into three fractions instead of two (see figure 6.11) [32]. The intermediate fraction is then recycled within the stage in a form of internal reflux. Just as in other uses of reflux, this has the effect of increasing the

² A hold-up time of two seconds has been given by one source [31].
Figure 6.11. Advanced separation nozzle
This nozzle system employs a double deflection of the jet to produce a light, intermediate and heavy fraction. The intermediate fraction is recycled within the stage as an internal reflux.

separation gain while reducing the throughput, but optimization of the reflux ratio can produce significantly greater separative power in a given stage element. It also changes the optimum cut from one-quarter to one-third, reducing the number of stages needed.

The Helikon process

The most succinct description of the South African enrichment process has been the following: “The UCOR process may best be described as a combination of a stationary-wall centrifuge with a highly asymmetric cut and a cascade system (the helikon system), which is eminently suited to asymmetric separation” [13c]. The ‘stationary-wall centrifuge’ is thought to be related to a device called the vortex tube [26b]. Just how close the relation is is difficult to judge, especially since a group of the project’s workers has said that while the separating element was originally based on the vortex tube its present form is “far removed” from it [13d].

With this caveat in mind it may still be useful briefly to examine the working of a vortex tube. This is a tube into which a mixture of 1–2 per cent UF₆ and 98–99 per cent H₂ gases [33] is injected at high velocity tangential to an inner wall, but with an axial component of velocity as well (see figure 6.12). As the gas spirals around the tube the lighter component tends to concentrate near the axis just as it does in the rotating centrifuge or jet nozzle. When the gas reaches the other end of the tube the outer and inner portions are drawn off separately.

Very little work on isotope separation with the vortex tube has been done outside South Africa, and one early assessment of the process was quite negative [27b]. However, this study apparently underestimated the improved performance which can be obtained by diluting the UF₆ with a light carrier gas such as H₂. The same enhancing effects which occur in the jet nozzle process also seem to operate in the vortex tube to give greatly improved separation factors over what can be obtained with pure UF₆. The
A mixture of UF₆ and H₂ is injected tangent to the tube’s inner wall and spirals down the tube. At the end the heavy and light fractions are separated by a skimmer. In this tube the cut is only one-twentieth.

The separation factor most often quoted for the South African process is $q = 1.03$ [34a].

However, this large separation factor can be obtained only at the price of taking a very small cut, $\theta = 0.05$ [34b]. This means that 19 separate streams of product, each with a slightly different enrichment, must be moved forward without mixing. Such a process is economically prohibitive if separate transmission pipes and compressors must be provided for every stream.

A very imaginative solution to this problem has been developed by South African researchers, who discovered that many separate streams can be sent simultaneously through the same axial-flow compressor without significant mixing among them. An axial-flow compressor consists of a rapidly rotating shaft driving a series of blades similar to turbine blades (see figure 6.4). These are mounted in a casing to which are attached arrays of stationary blades which alternate with the rotating ones. As gas is drawn into the front of the compressor it is alternatively speeded up by the rotating blades and deflected by the stationary ones in such a way that it emerges from the back of the compressor at a higher pressure. The most familiar application of such compressors is in jet engines, and it was on a jet engine that the South Africans did their original experiments on flow mixing [34b].

The use of this technique allows the combining of all 19 elements of a given cascade level into a single module utilizing only two compressors. This compact design, coupled with the relatively high separation gain associated with the small cut, allows the design of a commercial-type cascade with only 100 modules [34a]. It should be emphasized that these 100 modules actually constitute 1,900 enrichment stages. This is consistent with a separation gain of $2\theta g = 0.0026$, about two-thirds of the value.
obtainable in gaseous diffusion. One important advantage claimed for the modules is that they are flexible in the way the various streams are organized. This allows for a squared-off cascade to be built up entirely of identical modules with only the connections being altered to allow for changes in flow rate and enrichment factor [34c].

A single module would contain two axial-flow compressors with associated heat exchangers, the separation elements and the piping necessary to move the 19 feed streams into and out of the module while the single tails stream is deflected within the module through each stage in succession (see figure 6.13). A module with a capacity of 10 t SWU/yr would be about 3.6 m in diameter, and if figure 6.13 is roughly to scale, about 10 m long [13d]. Larger modules with diameters of 6.5 m have been proposed and have been rated at 50–65 t SWU/yr [28b]. Recent improvements are said to have raised this to between 80 and 90 t SWU/yr for a module of the same size [13e]. These modules would be comparable in size to the largest gaseous diffusion stages, but only about one-tenth as many would be needed to make a plant of the same capacity.

The energy consumption of the South African process is comparable to that of the jet nozzle, somewhere in the range 3 000–3 500 kWh/SWU [13f]. The equilibrium time is stated to be low, only 16 hours for a plant producing 3 per cent product [28b]. Using table 5.1 and the value 20g = 0.003 gives a stage hold-up time of between 0.05 and 0.2 seconds. This is the hold-up time per stage, and since there are 19 stages in a module, the modular hold-up time is probably an order of magnitude

Figure 6.13. A prototype Helikon module
This module is 3.6 m in diameter and is rated at 6–10 t SWU/yr. It contains two axial flow compressors with associated heat exchangers. The central region contains the separating elements and the piping to route the tails stream through the stages in a helical pattern. The drawing does not illustrate these details clearly.
larger. The small hold-up time also leads to a small inventory, making the Helikon process a relatively convenient one for batch recycling to produce higher enrichments. This may be the means by which South Africa was able to produce uranium enriched to 45 per cent for its research reactor at Pelindaba, an achievement which was announced on 29 April 1981 [35].

V. Chemical-exchange methods

Basic principles

The chemical-exchange process is an excellent object lesson in the difficulty of making reliable predictions about the development of enrichment processes. Many attempts were made in the early years after World War II to find chemical enrichment techniques, but in all cases the methods were found to be far too slow and expensive to compete with gaseous diffusion [36]. Although chemical-exchange techniques had proved very successful in separating lighter isotopes, it was known that the separating effect decreases with isotopic mass. This implied that chemical separation of uranium was likely always to be difficult, and that finding a successful process could require substantial further research in uranium chemistry. This was the general situation described in a review of these methods in 1972 [27c].

However, in 1968 French researchers had discovered a new technique [37], and after nine years of development the process was announced at the 1977 IAEA Conference at Salzburg [13g]. More recently the Japanese have announced a successful process developed by the Asahi Chemical Company, which is currently planning construction of a pilot plant [38a]. So, in a very short period of time chemical enrichment of uranium has become a prominent candidate for future development, one with some important implications for the proliferation issue.

The chemical-exchange method of isotope separation depends on the very small tendency of different isotopes of an element to concentrate in different molecules when there is an opportunity for exchange between the molecules. As an illustration consider two uranium compounds AU and BU which are mixed together and allowed to come to equilibrium. In general each compound will have two isotopic forms: A\(^{235}\)U and A\(^{238}\)U; and B\(^{235}\)U and B\(^{238}\)U. Therefore, in any mixture of the two uranium compounds there will be four different species present. If these species are chosen properly, they can exchange uranium atoms. Chemists depict such an exchange equilibrium in the form of an equation

\[ A^{238}\text{U} + B^{235}\text{U} \rightleftharpoons A^{235}\text{U} + B^{238}\text{U} \]
where the double arrows imply that the reaction can proceed in both directions.

When the system is in equilibrium each species is present in a certain concentration, usually measured in moles per litre and denoted by the symbol \([AU]\). The equilibrium is characterized by an equilibrium constant \(K\) which gives the relation among the four concentrations

\[
K = \frac{[A^{235}U][B^{238}U]}{[A^{238}U][B^{235}U]} \quad (6.6)
\]

The chemical equation is customarily written to make \(K\) greater than 1, so this implies that in equilibrium the product of concentrations with \(^{235}\text{U}\) in A and \(^{238}\text{U}\) in B is greater than the product with \(^{238}\text{U}\) and \(^{235}\text{U}\) interchanged. Therefore if the two compounds \(AU\) and \(BU\) initially contain identical isotopic ratios, after the equilibrium is established compound \(A\) will be slightly enriched and \(B\) slightly depleted in \(^{235}\text{U}\).

Unfortunately it is not easy to explain why the value of \(K\) is different from 1. Indeed, the equipartition theorem of statistical mechanics, which was used to explain the separation effect in gaseous diffusion, predicts that \(K\) should always equal 1, and that no isotope separation should occur. The fact that it does occur is explained by the theory of quantum mechanics and the connection between the energy of a molecule and its vibrational frequencies. No such connection exists in the older classical physics.

The quantum theory states that the unit of vibrational energy of a molecule is proportional to its frequency, and that the molecule can only absorb energy in amounts which are a multiple of this basic unit. This means that the classical equipartition theorem cannot be valid for such a system, since the theorem requires that any amount of energy, no matter how small, which is added to the system must be shared equally among all the molecules. This situation exists to a good approximation if the temperature is high, but as the temperature is reduced the equipartition principle breaks down. In practice this results in the lighter isotope tending to concentrate in the more loosely bound molecule, while the heavy isotope is more likely to be found in the tightly bound molecule [11b].

This explanation immediately suggests two requirements on the compounds for a good chemical-exchange separation:

1. The two compounds should be very different, with the uranium tightly bound in one and loosely bound in the other. Free uranium ions fit the second requirement best of all.
2. The equilibrium should be established at the lowest possible temperature to maximize the separation effect.

Unfortunately, both of these requirements are incompatible with the requirement of easy exchange of the uranium. Two very different compounds do not exchange components effectively, and reducing the temperature reduces the rate at which equilibrium can be established.
This problem can be dealt with by using suitable catalysts. These are compounds or materials which do not participate directly in the chemical reaction but whose presence in some way facilitates or speeds up the reaction. The ultimate success of the French and Japanese chemical enrichment processes seems to have been largely the result of the discovery of suitable catalysts. These will be discussed in more detail below.

The equilibrium constant $K$ of equation 6.6 is identical to the single-stage separation factor $q$. This can be demonstrated as follows. Consider a stage to be a single operation of mixing the two compounds AU and BU, with both having equal proportions of light and heavy uranium isotopes. After equilibrium is achieved the compound AU will be slightly enriched in the light isotope and BU will be slightly depleted. Compound AU can therefore be called the product, and BU the tails coming out of this stage.

The relative isotopic abundances $R_p$ and $R_w$ in the two compounds are simply the ratios of the concentrations of the two isotopic species in each compound:

$$R_p = \frac{[A^{235}U]}{[A^{238}U]}$$
$$R_w = \frac{[B^{235}U]}{[B^{238}U]}$$

(6.7)

and the separation factor is given as in chapter 5 (p. 95) by

$$q = \frac{R_p}{R_w}$$

(6.8)

Inserting equations 6.7 into 6.8 and rearranging the factors reproduces equation 6.6 and shows that

$$q = K$$

(6.9)

This relationship holds as long as each compound contains only one atom of uranium. In more complicated compounds the formula must be modified, but such refinements are not needed for a basic understanding [18b].

For isotopes of light elements the value of $q$ can be quite large, and in fact isotopes of hydrogen, boron and nitrogen are most effectively separated by chemical means [18c]. For uranium, however, $q$ is limited to values at or below 1.003, and a great many stages are required to achieve useful enrichments. For example, if $q = 1.002$, over 2,700 stages are required in an ideal cascade to produce 3 per cent enriched product with 0.2 per cent tails. But chemical separation ‘stages’ are quite a bit simpler than the other types discussed so far, and this number is not as prohibitive as it may seem.

A chemical separation stage consists simply of the thorough mixing of two substances for a sufficient time to allow chemical equilibrium to be established. This is most efficiently done in a device called a ‘counter-current column’ in which one of the compounds is carried upward and the
other downward. Any level in the column is associated with a given isotopic composition of both compounds, so that only compounds of the same composition are brought into contact. The very slight isotopic transfer causes one compound to be slightly enriched and the other depleted as each moves on to the next contact.

For an efficient exchange the two compounds AU and BU must be both easily brought together and easily separated. This is accomplished most efficiently if the two compounds are in different phases. One might be a liquid and the other a gas; or both could be liquids which are immiscible; or one could be a liquid or gas and the other a solid. The liquid–gas system operates very much like a standard fractional distillation column, the liquid–liquid system like a solvent extraction process, and the liquid–solid system works like an ion-exchange column or ‘chromatography’. All of these techniques are widely used and very well understood by the petroleum and chemical industries, and this great wealth of experience adds to the attractiveness of chemical separation. Another attractive feature is the potentially very low energy consumption of the process. Since chemical exchange is an equilibrium process there is no need for powerful compressors or pumps for the preparation of phases for contacting.

These advantages are counterbalanced by the problem of processing the two compounds at the ends of the column, the problem of ‘reflux’. To illustrate this problem, suppose that compound A is moving upward in the column and B downward (see figure 6.14). This means that the highest enrichment in $^{235}$U is at the top, and the enriched uranium emerges from the top in the compound AU. Some of this can be removed as product (or sent on to another column), but most of it must be refluxed to maintain the large countercurrent flow. However, before it can be sent back down the column the AU must be converted to BU. (The opposite must be done at the lower end, the tails reflux.) This is in general a non-reversible chemical process, which can require large amounts of energy and which must be done very carefully to avoid losses of enriched uranium. Because the reflux ratio is generally very large (i.e., the amount recycled is many times the amount extracted), even a small percentage loss of material in the reflux reaction can significantly reduce the efficiency of a plant (see chapter 5, p. 115).

With this general introduction to the theory of chemical enrichment it is now possible to consider the two most promising processes in more detail. The first is the Japanese process which depends on an exchange of uranium between a liquid solution and a finely divided ion-exchange resin. The second is the French solvent extraction process which uses an exchange between two immiscible liquid phases, one aqueous and the other organic. A third process, studied extensively in the USA, but apparently considerably less developed than the first two, is also a liquid–liquid process based on exchange between solutions of UF$_6$ and NOUF$_6$. 
The Japanese process

In this process a column or series of columns is packed with an ion-exchange resin analogous to those used in many industrial purification processes or in domestic water softeners. This resin has the property of attracting and holding to its surface (adsorbing) some chemical species in preference to others, so it serves as the medium or 'phase' for holding one of the two uranium compounds to be contacted. The resin is in the form of a fine powder in which the individual particles are spheres of only 20–200 μm in diameter [39]. This provides a large area for contacting the two phases, and it is quite analogous to the function performed by a gaseous diffusion barrier.

The resin is first prepared for receiving the uranium compound by charging it with a strong oxidizing agent, called, for convenience, OxI[II] [40a]. When the resin is saturated with the oxidizing agent, a new solution is introduced containing a uranium compound in which the uranium is in the IV oxidation state (see chapter 5, p. 119). When this comes into contact with the oxidizing agent on the resin, it is oxidized to U(VI) and replaces the now reduced oxidizing agent on the resin. This reaction is quite rapid and leads to a well-defined boundary between resin saturated in OxI[II] and in U(VI) (see figure 6.15). This boundary moves slowly down the column.
as more U(IV) solution is added at the top. When an appropriate length of resin has been saturated with U(VI), a new chemical solution is introduced at the top of the column. This is a reducing agent, referred to only as Re\(^{1}\) [40b], which acts on the U(VI) adsorbed on the resin to reduce it to U(IV), and also to replace it on the resin. The released U(IV) compound goes into solution and migrates down the column, coming into contact with the

**Figure 6.15. An ion-exchange module**

The four columns on the left are filled with ion-exchange resin particles. In the regions marked “RED” and “OX”, the resin is in its reduced and oxidized states respectively, and no uranium compounds are adsorbed on it. In the regions marked “U”, a U(VI) compound is adsorbed on the resin and is in continuous contact with a U(IV) compound in solution. In the fourth column the reduced resin is being oxidized in preparation for the arrival of the leading edge of the “U” column. Rotating valves at the tops and bottoms of the columns control the flow of oxidizing and reducing agents and uranium compounds. They are also programmed to admit feed and remove product and tails at appropriate times. These arrangements allow continuous operation of the module.

The two columns on the right are used to recharge and recycle the reducing and oxidizing agents. The input of “O\(_{2}\)” symbolizes reoxidation of the oxidizing agent while “H\(_{2}\)” symbolizes reduction of the reducing agent. These symbols do not necessarily imply that oxygen and hydrogen are the actual substances used in the recharging process.

![Diagram of ion-exchange module](image_url)


U(VI) still adsorbed further down. In this region the isotopic exchange reaction

\[ ^{235}\text{U(IV)} + ^{238}\text{U(VI)} \rightleftharpoons ^{238}\text{U(IV)} + ^{235}\text{U(VI)} \]

can take place, resulting in a slight concentration of \(^{235}\text{U}\) in the U(VI) compound adsorbed on the resin. The equilibrium constant usually quoted
for this reaction is 1.0013 [40c]. This is therefore the single-stage separation factor for the process (see equation 6.9).

The reducing agent at the top of the column also creates a sharp boundary, so that the portion of the column in which the isotope exchange takes place is a well-defined, slowly descending segment. The length of this segment is determined by the desired degree of enrichment and the flow rate of the solution through the resin. Enrichment in $^{235}$U occurs at the top of the segment, since the $^{235}$U tends to concentrate in the U(VI) compound which remains adsorbed on the resin.

The segment of column between the two boundaries moves slowly downward, but it is easier to analyse if viewed from a frame of reference moving at the same speed as the boundaries. In this frame of reference the resin and its adsorbed U(VI) are moving upward; the solvent and its dissolved U(IV) are moving downward; and reflux is occurring at both ends. This makes clear the countercurrent nature of the process and permits analysis of the system by the standard methods applied to any countercurrent square cascade [40d]. Therefore the formulae derived in chapter 5 (see table 5.1) can be used directly to estimate the important parameters of the system. The value of $q$ can be taken to be 1.0013 and the cut assumed to be equal to a half. The total number of stages required for 3 per cent product, 0.2 per cent tails would therefore be 3 690 (an ion-exchange column must be treated as a square cascade) [38b].

The definition of a stage in this process is not as obvious as in those previously described. The flow of materials is continuous, so a stage is a more abstract concept similar to the notion of 'theoretical plate' in fractional distillation [11c]. The length of a stage in chemical exchange is determined by the rate of flow of materials and the equilibrium time of the reaction. Recent statements give the equilibrium time as "less than a second" [40e], and the thickness of a stage can be estimated from the statement that "several hundred theoretical separation stages per meter" are achievable [38b]. So a stage will be only a few millimetres long and the velocity of flow will be of the order of millimetres per second. A typical U(IV) molecule would therefore take roughly one hour to migrate from the top of the exchange band to the bottom. Note that this velocity has no simple connection to the rate at which the exchange region boundaries move.

The key to making this process commercially attractive has been the reduction of the equilibrium time by the use of catalysts, in particular resins which lowered previous reaction times by a factor of 1 000 [38c]. It also required many years of research to find suitable oxidizing and reducing agents as well as economical methods of recharging and recycling them. Considering the fact that quite negative assessments were being made of this method as recently as 1975 [15c], this example can serve as a useful reminder that even the most frustrating technological problems have a way of being solved if sufficient incentive is present.
As figure 6.15 shows, this process does not use a single column, but is carried out in a series of three columns while the resin is being recharged in the fourth. By programming the valves at the top and bottom of the column the moving liquid column can be recycled indefinitely through the four-column module. As the leading edge of the uranium band passes a valve, a quantity of tails can be removed and, similarly, product can be removed and feed added as the appropriate portions of the band pass through the valves. All of this has apparently been achieved by the design of appropriate valves, detectors for determining separation band boundaries, and computerized control systems [40f].

The amounts of material and sizes of equipment involved can also be estimated with the help of table 5.1. If the exchange band is 'several' metres long, figure 6.15 suggests that each column should be a 'few' metres in height. From table 5.1 it can be shown that the total inventory of uranium in the enrichment band is

$$I = 44 \frac{t_h}{g^2}$$

(6.10)

Note that for this process the values for a square cascade are the most appropriate, since the column has a constant diameter. Taking $t_h = 1$ s and $g = 0.0013$ in equation 6.10 gives an inventory of $2.6 \times 10^7$ kg U for every kg/s of product, and since the latter represents 4.3 kg SWU/s the specific inventory can be shown to be 0.19 kg U/kg SWU/yr. These values are roughly twice the corresponding number for gaseous diffusion (see p. 133). This means that an ion-exchange plant will necessarily have a large inventory relative to its production rate, but it should also be pointed out that ion-exchange facilities need not be as large as gaseous diffusion plants to be economically viable [38d].

A rough idea of the size of a column can be obtained from the inventory values. Consider a four-column module with a capacity of 1 000 kg SWU/yr designed for 3 per cent product and 0.2 per cent tails. Such a module would contain 190 kg of uranium, and if this is present in solution with a concentration of the order of 0.1 mole/litre [40b], a total of about 8 000 litres of liquid must be present. If this is in the form of a column 4 m long then the diameter would be 1.6 m. Allowing for the volume taken up by the resin particles one might guess that a typical 1 t SWU/yr module would consist of four columns each about 3 m high and something under 2 m in diameter. A 1 000 t SWU/yr plant would have 1 000 such modules, or fewer larger ones.

Since the chemical-exchange process is reversible, very little energy is required for the ion-exchange columns (unless they must be kept at an elevated temperature, but this is not indicated in the most recent literature). The main energy expenditure comes at the regeneration of the oxidizing and reducing agents and in the pumping of recycled solutions. It is not possible to estimate the specific energy consumption without more data [40g], but it seems reasonable to assume that it is considerably less than gaseous diffusion, but probably somewhat larger than the centrifuge.
The French process

This process utilizes chemical exchange between two uranium compounds dissolved in two immiscible liquids. One of the phases is aqueous and the other an organic solution, and they are made to flow through each other in a countercurrent column. The contacting of the two phases is achieved by agitating them so that the organic liquid breaks up into small droplets suspended in the aqueous solution. The effect is quite similar to that achieved by shaking a bottle of oil and vinegar salad dressing. In an industrial separation column this agitation can be achieved in several possible ways, but the French process employs a "pulsed-column" [37]. This is a column in which the contents are agitated either by an external mechanical device or by a series of reciprocating discs installed in the column [41a].

The isotope exchange rate is limited by two important factors: the inherent rate of the chemical reaction and the diffusion rate of the two chemical species into and out of the volume of the oil droplets. The former can be speeded up by an appropriate choice of compounds and by catalysts and/or temperature increases. The key to the success of this process seems to be the use of an unusual kind of compound called a 'crown ether' [42a]. An example of a crown ether, dibenzo-18-crown-6, is shown in figure 6.16 [43]. The name 'crown' was given to this compound because of the shape of the molecule, which in three dimensions has the oxygen atoms arranged in a circle above the ring much like the points of a crown.

The crown ether is part of a larger class of compounds called chelating agents which are very useful in many branches of chemistry [41b].

Figure 6.16. A molecule of dibenzo-18-crown-6

This crown ether has 18 atoms in its basic ring structure, six of which are oxygens. The structures on each end are benzene rings, each containing six carbon atoms and associated hydrogens.
agents work by forming co-ordination compounds with metal ions. For example, the molecule of figure 6.16 will very readily react with potassium ions so that each of the oxygen atoms in the ring contributes a pair of electrons to a co-ordination bond with the potassium ion. The metal ion is then said to be ‘sequestered’, and in this form it can either be inhibited from undergoing reactions it might otherwise participate in, or encouraged to do things it would not otherwise do, for example, to dissolve in an organic solvent [41c].

Crown ethers can be made in a variety of sizes, and the key to making a crown ether effective for a particular ion is to match the size of the opening in the crown to the size of the ion in solution. Presumably this is one of the discoveries which underlies the successful French process. If a crown ether has been made which forms a co-ordination compound with uranium ions, then this compound can serve the dual function of extracting the uranium from one liquid phase to another and enhancing the separation effect. This latter property follows from the fact that the separation effect is directly proportional to the number of co-ordination bonds connected to the uranium [44].

The second factor which limits the exchange rate is the rate of diffusion of uranium compounds into and out of the oil droplets. This can be increased by making the droplets as small as possible, just as the ion-exchange reaction is enhanced by using very small resin particles. However, it does not seem to be feasible to agitate the two liquids violently enough to produce droplets smaller than a few millimetres in diameter [45a]. This factor seems to be the main one in making the stage hold-up times in the French process at least 20 times as long as those in the Japanese process, that is, of the order of 20–30 seconds [45b].

The separation gain has been given as either “greater than \(2 \times 10^{-3}\)” [46a], or “about twice higher than the best ones [previously] known” [45c]. Given that values of 0.0013 to 0.0016 are well known for other processes [38b, 47], it is reasonable to speculate that a value between 0.0025 and 0.0030 has been achieved in the French process. This suggests that a square cascade designed to produce 3 per cent product and 0.2 per cent tails should have between 1 600 and 1 900 stages. If the total height of the reaction column is taken to be 30 m (see below) then a single stage has a length of between 1.6 and 1.9 cm. Note that both the stage height and column height are about an order of magnitude larger than those of the Japanese process. The lower separation factor of the latter seems to be more than compensated for by its much faster equilibrium time.

The proposed French enrichment facility would be constructed of units each consisting of two identical columns connected as shown in figure 6.17 [48a]. Note that this mode of connection has the effect of turning two square columns into a squared-off cascade. This increases the cascade efficiency (i.e., the approximation to an ideal cascade) to about 85 per cent, thereby reducing somewhat the very long equilibrium time [48b].
Each column will be more than 20 m high and more than 1 m in diameter (a volume of 15.7 m³) and will be capable of producing 3 000–5 000 kg SWU/yr [48c]. This implies a product flow rate of 1 400–2 350 kg/yr from a two-column unit. Using a hold-up time of 25 s (see below) and \( g = 0.0028 \) in table 5.1 the total uranium inventory in the two columns can be estimated as between 6 and 10 t. Using the total volume of 31.4 m³ the average concentration of the uranium solutions is found to be roughly 1 mol/l, considerably more concentrated than those employed in the Japanese process. This makes for a very large uranium inventory, acknowledged to be an order of magnitude larger than that for a gaseous diffusion plant of similar capacity [48b]. This same ratio holds approximately for the equilibrium times as well, with a Chemex plant requiring 15 months to begin producing 3 per cent enriched product [46b]. This implies a hold-up time of 20–30 s.

It is not only the inventory of uranium which is going to be large in a Chemex plant. The volumes of all other necessary chemicals must also be correspondingly large, and some of them, such as the crown ethers, may be relatively expensive. There is also the problem of refluxing these enormous
quantities of chemicals with very high efficiency. This problem is claimed to have been solved, and ‘established technology’ has been adopted wherever possible. The energy consumption is claimed to be less than 600 kWh/kg SWU [37].

The only secrets in the process seem to be the uranium compounds themselves, the catalysts, and the specific reflux reactions. It is noted that corrosion problems require all components to be made of “appropriate plastic materials” [48d]. It is also emphasized that the expensive process of converting uranium oxide to uranium hexafluoride is unnecessary in the Chemex process [48d].

The American process

The characterization of this process as ‘American’ is a bit arbitrary [23c], since it is clear that it has been studied by researchers in many countries. It is also clear that this process is not nearly as close to commercial realization as the Japanese and French processes mentioned previously. Therefore only a brief summary of its current status will be given here.

The process involves the exchange of UF₆ molecules in the following reaction:

\[ \text{NO}^{238}\text{UF}_6 + ^{235}\text{UF}_6 \rightleftharpoons \text{NO}^{235}\text{UF}_6 + ^{238}\text{UF}_6 \]

The most extensive experiments have been done with two liquid phases: NOUF₆ dissolved in anhydrous hydrogen fluoride (HF) and the UF₆ dissolved in freon or some other saturated fluorocarbon. The measured equilibrium constant for this reaction is between 1.0010 and 1.0016 [27d] and the equilibrium time seems to be comparable to that of the French process, that is, of the order of one minute [15d]. These data suggest that the process might be potentially attractive, but all efforts to develop it past the laboratory stage seem to have encountered severe difficulties with the reflux reactions.

The major problem seems to be the reaction which recycles NOUF₆ [27d, 15e]. No way has yet been found to accomplish this reflux in a reasonable time and without excessive energy costs and losses of material. It is not clear from the open literature whether or not these problems are still being studied, but in any event this process appears to be far behind the other two discussed above. The process does not seem to have a high priority in US research and development plans [49a].

Summary

This survey of chemical enrichment methods has shown that at least two processes have evolved to the point where they have begun to look commercially attractive. Supporters of both the Japanese and the French
processes point out the advantages of the relative simplicity and low energy costs of the technology, and also emphasize that the process lends itself to the economical construction and operation of small or moderately sized units [38d, 46c]. But at the same time they argue that the large inventories, long equilibrium times and criticality dangers (the danger of an accidental chain reaction) all operate to prevent the misuse of such small facilities for the production of highly enriched uranium [38d, 50a]. These claims are examined in chapters 2 and 3.

VI. Laser isotope separation

Basic principles

Just as in chemical exchange, laser isotope separation depends on the quantum mechanical connection between energy and frequency in a molecule or atom. In chemical exchange this dependence is quite subtle and indirect, but, in contrast, the use of lasers to separate isotopes exploits this connection in an elegantly simple and direct way. This is made possible by the unique properties of the laser.

According to the quantum theory an atom or molecule cannot have an arbitrary energy but can exist only in a set of discrete ‘states’, each having a well-defined energy. The number of such states can be very large, but they are all separated from each other by finite energy differences. The atom or molecule can be induced to make transitions from one such state to another by the absorption or emission of electromagnetic radiation. This radiation is in the form of a ‘photon’, an entity which exhibits either particle-like or wave-like properties, depending on the way it is observed. Among its wave-like properties is a frequency of vibration, \( v \), related to its energy by the simple equation

\[
E = hv
\]

where \( h \) is Planck’s constant equal to \( 6.63 \times 10^{-34} \) joule seconds (J s).

A laser is a device which can produce large numbers of such photons, all having almost precisely the same frequency. If this radiation can be focused on a gas of atoms or molecules which have a pair of states differing in energy by just this amount, then the atoms or molecules can absorb the laser light and be ‘excited’ to the higher energy state. The most important characteristic of this excitation process is that it can only occur with large probability if the photon energy is ‘tuned’ precisely to the energy difference of the atomic or molecular transition. In this way the process resembles the operation of an ordinary radio or television set, which can be tuned to receive and amplify only one particular frequency out of the many
which simultaneously impinge on the antenna.

In an atom the energy differences between states depend on the detailed structure of the electron cloud as well as on the properties of the nucleus. As was pointed out in chapter 1 the structure of the electron cloud is determined almost entirely by the number of protons in the nucleus, but changing the number of neutrons does produce small effects. For uranium, typical shifts in the absorption frequencies are only about 1 part in 100,000. For example, the energy of an important transition for isotope enrichment of uranium is about 2.1 eV,\(^3\) and the difference between the energies for this transition for \(^{235}\text{U}\) and \(^{238}\text{U}\) is 4.2 \(\times\) 10\(^{-5}\) eV [51a]. This means that a laser must be tunable to an accuracy of 1 part of 10\(^5\) in order to excite one of the isotopes without affecting the other. Such fine tuning is possible with modern lasers. In particular a class of lasers called dye lasers operate in the visible part of the electromagnetic spectrum using organic dyes. These dyes can be designed to produce radiation of any desired colour, and within a given colour they can be tuned to very narrow frequency bands [2bb]. For example, the light which induces the above-mentioned transition is red-orange in colour and produced from a very common dye called rhodamine 6G [52].

If molecules are used instead of atoms, the transitions involve changes in vibrational energy of the molecule rather than electron energy states. Generally vibrational transitions require considerably less energy than electronic transitions, so the laser light required is in the infra-red rather than the visible portion of the spectrum. An important example is a vibrational transition in UF\(_6\) which has an energy of 0.065 eV, quite far out in the infra-red [2cc].

Powerful infra-red lasers which use carbon monoxide or carbon dioxide are available at a number of frequencies, but it has not been as easy to produce this particular frequency as it was in the atomic case. There is some disagreement in the literature about how close this problem is to solution [49b, 42b]. Meanwhile some researchers have taken another track and instead of attempting to bring the laser to the molecule they are attempting to design a molecule to fit the laser. The most promising laser is the reliable and powerful carbon dioxide laser, which operates at a wavelength of about 10 µm (photon energy approximately 0.1 eV). A number of attempts have been made to design molecules containing uranium which will have strong vibrational transitions in this energy range [53, 54]. This is not unlike the problem in chemical exchange of trying to design uranium molecules or complexes with large chemical isotope effects.

Currently the development of laser enrichment is proceeding along several parallel paths. For the more detailed descriptions which follow it is

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\(^3\) The symbol eV stands for electron volt, an energy unit convenient for atomic and molecular processes. One electron volt equals 1.6 \(\times\) 10\(^{-19}\) J.
convenient to divide the field into three categories: those which use atomic uranium, those which use molecular species, in particular UF₆, and a newer and much less developed class which might be called laser-assisted aerodynamic or diffusion processes.

**Atomic vapour laser isotope separation (AVLIS)**

Research and development on the AVLIS process is under way in many countries, but the system on which the most information is publicly available is the one which was until recently undergoing development by Jersey Nuclear–AVCO Isotopes (JNAI) in the USA. This project was terminated in March 1981 because of a substantial cut in financial support by the US Department of Energy which decided to direct most of its support to a similar project at the Lawrence Livermore Laboratories (LLL) [55]. The reasons offered for the decision were technical, but given the history of competition between the two laboratories and the fact that the decision directly overruled the recommendations of a technical advisory panel [49c], one cannot escape the suspicion that the reasons were at least as political as they were technical.⁴

Despite the abandonment of the JNAI programme our description will be based largely on the JNAI technique. The rather small amount of information available on the LLL process strongly suggests that it is quite similar to the JNAI process, although the Livermore group seems to be keeping open a number of alternative options for laser wavelengths and pumping schemes [56]. However, a description based on the JNAI process will certainly be adequate for the purposes of this survey. It should also be noted that research on laser enrichment is being actively pursued in a number of other countries, including, among others, the USSR and Israel. There have also been some reports of an Indian laser research programme (see chapter 8). However, very little information is available on these programmes.

**An AVLIS module**

The basic features of an AVLIS module are illustrated in figure 6.18 [51b]. Such a module might measure about one metre in height and one to three metres in length, and it would be entirely enclosed in a system capable of

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⁴ As this book was going to print, the US Department of Energy announced that it had chosen the Lawrence Livermore AVLIS process over its two competitors, the Los Alamos MLIS process and the TRW Corporation plasma process, for further development [105]. The Livermore AVLIS process will now be the only advanced enrichment method brought to the pilot plant stage in the USA. It is also being explored for possible use in isotopically purifying plutonium for the US nuclear weapon programme (see below, p. 175 and ref. [82]).
Figure 6.18. An AVLIS module

Uranium vapour is produced by electron beam evaporation from the crucible at the bottom. The vapour expands outward until it reaches the irradiation region where the $^{235}$U atoms are excited and ionized by laser light. The ions are deflected and collected on the vertical plates by a combination of electric and magnetic fields. Neutral atoms continue outward and are collected on the horizontal plate at the top. The laser light is shown being reflected several times through each collection volume by a system of mirrors.

![Diagram of AVLIS module](image)

maintaining a high vacuum. All laser light sources and mirrors would be outside the volume containing uranium vapour. Since the path length of the laser light in the vapour may be more than 100 m [57a], the light must be reflected back through the chamber many times, possibly as many as 300 times [42c].

The uranium to be enriched is first converted to the pure metallic form and formed into a long ingot. It is then melted in a crucible by heating it with a beam of electrons directed to the surface of the ingot by a magnetic field. Since hot liquid uranium is extremely corrosive, a rather elaborate mechanism must be used to keep the molten uranium from contacting the support structure and to prevent rapid dissipation of the heat supplied to the uranium ingot [2dd]. The thin strip of molten uranium provides a line source of uranium atoms which then diverge radially outwards towards the top of the chamber. The atoms are allowed to move undisturbed through the lower portion of the chamber during which time they lose much of the excess energy given to them in the evaporation process. The purpose of this is to get as many atoms as possible into their lowest energy states and to allow atoms ionized by the electron beam to recombine into neutral...
form. Creation of this radially diverging vapour involves considerable losses, and only 50 per cent of the evaporated atoms reach the irradiation zone [42d]. The rest are deposited on various surfaces inside the chamber, and this material must be periodically collected and recycled. It represents a kind of internal reflux analogous in principle to the recycled intermediate fraction in the advanced Becker nozzle process (see p. 149). However, in contrast to the efficiency improvements this reflux leads to in the nozzle process, in the AVLIS process it represents a significant waste of both energy and time.

The vapour that does reach the irradiation zone is then illuminated by laser light carefully tuned to excite transitions only in \(^{235}\text{U}\) atoms. Four lasers are used, all with slightly different colours, but all in the same red-orange portion of the spectrum. The need for four lasers can be understood by referring to figure 6.19 [51c].

The total energy required to remove an electron from a uranium atom is 6.2 eV. This is very difficult to supply with a single laser in an isotopically

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**Figure 6.19. Three-step laser ionization**

The first step uses two laser beams of slightly different frequency in order to excite the large number of \(^{235}\text{U}\) atoms in the ground state and low-lying excited state. The \(^{238}\text{U}\) level scheme is similar to this one for \(^{235}\text{U}\) except that each level is shifted by an amount too small to be shown on the figure, but large enough to allow discrimination by finely tuned lasers.
selective way, although the argon fluoride laser may turn out to be useful for this purpose [56a]. In practice, at least two transitions are required, and three have been selected in order to take advantage of the readily accessible and well-established technology of rhodamine dye lasers. Notice that the ionization energy of uranium is just under three times the energy of a red-orange photon, so three steps is just the right amount. Each of these steps has a slightly different energy, so the three lasers must be tuned to slightly different frequencies. These three lasers are sufficient to ionize about 40 per cent of the $^{235}\text{U}$ atoms in the vapour. This is the fraction which is in the lowest energy state at the high temperature at which the irradiation takes place. Another 30 per cent are in the next higher energy level, which is only 0.077 eV above the ground state. It turns out to be economically advantageous to employ a fourth laser to get at this additional 30 per cent. Even so, the remaining 30 per cent of the $^{235}\text{U}$ is unaffected by the lasers and becomes part of the tails.

The $^{235}\text{U}$ atoms which are ionized by the laser light are then deflected by a pulse of strong electric and magnetic fields towards collecting plates oriented parallel to the radial flow of neutral vapour. Ideally only the charged ions would be given transverse velocities and migrate to the collector plates, and the neutral atoms would continue to move outward and be deposited on a surface beyond the irradiation-collection region. In practice, however, it is not possible to shield the collectors from all of the neutral particles, and anywhere from 3 to 15 per cent of the feed material is expected to be collected along with the enriched material. This and several other inefficiencies limit the achievable single-stage enrichment factor to 15 at the very most [42c].

If such a value can be obtained in practical enrichment facilities, it will constitute a remarkable advance over current processes, for which enrichment factors are only slightly larger than one. For example, such a facility would be capable of producing 3 per cent reactor fuel from 0.2 per cent feed in one stage, thereby creating an enormous source of fuel from the existing stockpile of gaseous diffusion plant tailings. This possibility seems to be one of the major driving forces behind the development of laser enrichment. It must be emphasized, however, that an enrichment factor of 15 is a highly optimistic goal. It is probably more reasonable to expect a value between 5 and 10.

These numbers can now be used to construct a model of an AVLIS enrichment stage (figure 6.20). The numbers on the model were derived assuming natural uranium feed, an enrichment factor of 7, and a reflux to feed ratio of 0.5. It was also assumed that 60 per cent of the $^{235}\text{U}$ which makes it to the irradiation region is ionized and collected. Amounts of feed, reflux and tails are computed on the basis of 1 kg of product. The amount of separative work done by this stage can be computed from equation 5.4 to be 7.6 kg SWU/kg product. Note that for such large enrichment factors the use of quantities such as cut and separation gain is not meaningful. For this reason it is the value of the enrichment factor, that
Figure 6.20. An AVLIS enrichment stage
All flow numbers are based on 1 kg of product and a hypothetical enrichment factor of 7. The reflux represents the 50 per cent of feed material which deposits on various surfaces of the module and is unprocessed. This material must be recovered and recycled.

is, the ratio of isotopic compositions of product to feed (see equation 5.7, p. 104) which is entered in table 6.3.

Another standard parameter which loses much of its significance is the stage hold-up time. If this is taken to be the time required for a uranium atom to move from the surface of the liquid to the collector, it is only a small fraction of a second. Obviously this is not a controlling factor in determining production rates or equilibrium time. The latter is essentially instantaneous. Far more important are the materials-handling processes which produce feed and recover product and tails. These require highly elaborate and sophisticated procedures [51d], which present important and apparently still unresolved problems [49d].

One of the major early attractions of the laser enrichment processes was their potential for great savings in energy over gaseous diffusion and even the centrifuge [58]. The specific energy consumption of the element of figure 6.20 can be estimated by assuming that 30 per cent of the $^{235}$U atoms in the feed absorb 6.2 eV of photon energy to produce 7.6 kg SWU. This represents $1.0 \times 10^5$ J or $2.8 \times 10^{-2}$ kWh. Even if the lasers are only 0.2 per cent efficient, the energy required is only 14 kWh or about 2 kWh/kg SWU. This is 1000 times less than gaseous diffusion and 100 times less than the centrifuge. However, this estimate ignores a number of other factors, such as evaporation energy, optical system inefficiencies, reflux procedures, and so on. More recent estimates of energy consumption make AVLIS comparable to or somewhat better than the most efficient centrifuges, that is, about 100 kWh/kg SWU [2ee, 56b].

**Laser requirements**

Energy costs, however, are only a part of the problem, and when it comes to capital costs and technical complexity in design and operation the laser
process looks much more problematical. As only one example of the extreme complexity of the system it is instructive to examine the requirements which the lasers must satisfy.

First, the lasers must be able to provide a certain minimum flow of photons which is determined entirely by the properties of the atomic transition itself and is independent of the density of vapour or the desired production rate. This requirement follows from the need to make sure that sufficient numbers of atoms are available at each of the energy steps to take advantage of the photons ready to boost them to the next step. Because excited atomic states decay, it is essential that they be re-excited as fast as they decay, and this puts a lower limit on the photon flux which is acceptable. In laser jargon this requirement is that the energy level be 'saturated' [59a] and this saturation requirement means that the lasers must be quite powerful, even to produce relatively small quantities of product. The minimum power density required can be shown to be of the order of tens of kilowatts per square centimetre. This can be produced by a laser-pulsed beam of 1 cm² containing tens of millijoules of energy and lasting one millionth of a second.

Another stringent requirement placed on the lasers is the pulse repetition rate. As the uranium vapour passes through the irradiation zone it is crucial that all the ²³⁵U atoms be sufficiently exposed to the laser radiation. This means that two pulses of radiation cannot be separated by a time longer than it takes a uranium atom to cross this zone. If the zone is assumed to be 5 cm thick, and the average speed of a uranium atom is 500 m/s, then it takes only \(10^{-4}\) s for an atom to cross the zone. So the lasers must be fired at least 10 000 times per second to ensure full exposure of the uranium. Since it is highly unlikely that a single laser with sufficient beam intensity could be made to fire at such a rapid rate, the AVLIS system might require 20 or more sets of four lasers each, all precisely controlled in frequency by a master laser oscillator and timed to fire in sequence, each one at a rate of 500 pulses per second [51e]. It is probably unnecessary to emphasize that such a system presents major technical problems, both in its original design and in ensuring that it can work for hours at a time in reliable, continuous operation.

One issue which has generated some controversy is the ease with which an AVLIS enrichment stage could be used for batch recycling to obtain highly enriched product. With an enrichment factor of 10, only three such recyclings would be needed in principle to produce 97 per cent enriched product. However, one evaluation of the proliferation dangers of this technique has argued that such a procedure would be extremely difficult, if not impossible [42f]. Briefly stated, the argument is as follows:

If normal uranium vapour densities are used, then as a larger fraction of the uranium becomes ²³⁵U, the laser power must be increased in order to ionize the same fraction of ²³⁵U atoms, or, if the ²³⁵U content is over 50 per cent, the lasers can be tuned to remove ²³⁸U instead. If laser powers are increased, then the ²³⁵U plasma created by the ionization becomes so
dense that the efficiency of the ion collectors drops. This is a plasma shielding effect similar to those discussed in the next section (see pp. 180–81). Tuning the lasers to excite $^{238}\text{U}$ will work only if the plasma shielding effects remain small, up to 50 per cent ionization. Finally, if the density of the atomic vapour is reduced in an attempt to solve the problem, there will no longer be enough collisions in the vapour to bring a sufficient number of atoms into their lowest states. Again the efficiency of the process will be degraded.

These arguments are all physically valid, but a question remains as to their quantitative significance. Some knowledgeable analysts seem unconvinced by these arguments and suggest that the batch-recycling procedure is feasible as long as some sacrifice in throughput is tolerable [60, 61]. It is not possible to make an independent judgement on this question using only open sources of information.

Whatever the merits of this particular argument, it can be safely said that the creation of a successful AVLIS facility will be an extremely complex and sophisticated technical achievement. The laser system will involve hundreds of powerful lasers all pulsed according to a precisely designed time schedule. The light from the lasers will have to be reflected hundreds of times through a system involving hundreds of extremely efficient mirrors and other optical devices, all of which must be shielded from any contact with the uranium vapour. The optical system is further complicated by the need to prevent the propagation through the system of frequencies corresponding to other uranium transitions. If these are allowed to pass through the system, then the uranium vapour can itself become a laser and lose most of the energy pumped into it without even being ionized. Methods exist for dealing with this problem, but they add considerably to the complexity of the laser optical system [62].

There are even more subtleties and difficulties which could be mentioned, but the above should suffice to demonstrate that the AVLIS process is enormously complex and nowhere near commercial viability. This has been dramatized most vividly by the recent withdrawal of the JNAI effort from the competition.

**Molecular laser isotope separation (MLIS)**

Although some efforts have been made to use molecules better suited to existing lasers (see p. 159), the major efforts to develop MLIS have concentrated on $\text{UF}_6$. The reasons for this are obvious: $\text{UF}_6$ is a well-understood material with a high volatility and convenient chemistry. It is not an easy substance to handle in practice, but so much experience has been gained in its management from the gaseous diffusion and centrifuge programmes that it presents no serious difficulties.

The basic principle of the MLIS process is similar to that of AVLIS. $^{235}\text{UF}_6$ molecules must be made to absorb energy from laser beams while
\(^{238}\text{UF}_6\) molecules remain unaffected. However, the different nature of the absorption process makes the use of dye lasers inappropriate, and MLIS relies instead on either all infra-red light or a combination of infra-red and ultraviolet. The first process is often called ‘infra-red multiphoton dissociation’ and the latter ‘two-step dissociation’. In each case the object is to inject enough energy into a \(^{235}\text{UF}_6\) molecule so that it loses one of its fluorine atoms:

\[
\text{^{235}UF}_6 \xrightarrow{\text{energy}} \text{^{235}UF}_5 + \text{F}
\]

The \(^{235}\text{UF}_5\) which is produced condenses rapidly into a fine powder which can then be filtered from the \(\text{UF}_6\) gas [49e].

In the AVLIS process the use of hot uranium vapour reduces the efficiency of the process since only a minority of the uranium atoms can be excited out of the lowest energy state. This problem requires an extra laser and still results in a loss of 30 per cent of the potential \(^{235}\text{U}\) atoms. This same problem occurs in the molecular process, and in fact is far worse. At ordinary temperatures the collisions between \(\text{UF}_6\) molecules are so violent that virtually all of the molecules are excited out of their lowest vibrational states. The molecules have a wide range of vibrational energies which makes it very difficult to get any significant selectivity by tuning to a particular vibrational transition. Only by cooling the \(\text{UF}_6\) to very low temperatures can this problem be solved. But at very low temperatures \(\text{UF}_6\) is normally a solid.

The solution to this problem is to supercool the \(\text{UF}_6\), that is to cool it to temperatures where under normal conditions it would condense, but to trick it into believing it is still a gas. This is done by diluting the \(\text{UF}_6\) with an inert carrier gas such as nitrogen or argon and expanding the mixture suddenly through a nozzle (see figure 6.21). The expanding gas cools rapidly, and as the molecules collide downstream of the nozzle the vibrational energy in the \(\text{UF}_6\) is converted into the translational motion of the gas [57b]. This is quite similar to the process which occurs in the atomic vapour between the evaporation point and the irradiation region. However, the molecular process is more successful, and roughly 95 per cent of the \(\text{UF}_6\) molecules can be put into the lowest vibrational state. Therefore, only a single isotopically selective laser is needed to excite most of the \(^{235}\text{UF}_6\).

The wavelength of the light required from this first laser is 16 \(\mu\text{m}\), and a number of possible candidates already exist. One is a combination carbon dioxide (\(\text{CO}_2\)) and carbon tetrafluoride (\(\text{CF}_4\)) system developed by a group at the University of Southern California [63]. Another uses Raman scattering in hydrogen to step up the wavelength of \(\text{CO}_2\) laser light from 10 \(\mu\text{m}\) to 16 \(\mu\text{m}\) [64, 65, 66].

A third potential candidate is the recently developed free-electron laser, a remarkable new kind of light source which shows promise of both high efficiency and precise tunability [67]. There are many incentives aside from uranium enrichment for the development of these lasers, so it is reasonable to assume that one or more of them will be perfected.
Figure 6.21. An MLIS stage

This is a schematic illustration of the components of an MLIS stage. UF₆ and the inert carrier gas are mixed and expanded at supersonic speeds through a nozzle. Just downstream of the nozzle the gas is irradiated by an isotopically selective infra-red laser which vibrationally excites the $^{235}\text{UF}_6$, and a powerful ultraviolet laser which dissociates the excited UF₆ molecules. The $^{235}\text{UF}_5$ 'laser snow' is filtered from the gas stream and sent on to be refluorinated back to UF₆. The remaining gas is cleaned up and sent on for further tails stripping.

Source: Dr N. Haberman, US Department of Energy, Washington, D.C., USA.

When the $^{235}\text{UF}_6$ molecules have absorbed a 16 $\mu$m photon they can then be further excited either by the absorption of many more infra-red photons, or by a single ultraviolet photon which carries enough energy to dissociate the molecule directly (see figure 6.22). The tendency of complex molecules to absorb a number of infra-red photons of the same frequency was first noticed in 1974 [68] and has since been a topic of intense experimental interest [59b]. One of the strong advantages of this method seems to be that once the molecule has absorbed a small number of 16 $\mu$m photons the rest of the infra-red absorption can be supplied by the powerful and efficient CO₂ laser at 10 $\mu$m [69]. On the other hand the multiphoton absorption process is still not very well understood, and
Figure 6.22. Two molecular dissociation processes

The horizontal lines represent vibrational excitation levels, and the vertical arrows transitions induced by laser radiation. In the scheme on the left a large number of infra-red photons are used to excite the molecule up through many vibrational levels to dissociation. In the process on the right only one infra-red photon is used to excite $^{235}\text{UF}_6$, and the dissociation is accomplished in one step with an ultraviolet photon.

DISSOCIATION

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VIBRATIONAL ENERGY LEVELS

INFRA-RED MULTIPHOTON ABSORPTION

TWO-STEP INFRA-RED ULTRAVIOLET DISSOCIATION

certain features of it suggest that it may be difficult to apply the process to the large quantities of UF$_6$ required in a commercial-sized enrichment plant [70]. One particularly troublesome phenomenon is the tendency of irregularities in laser beam intensity to become magnified by a non-linear process called self-focusing. This can cause the beam to shrink down to very small irradiation volumes or to break up into many 'filaments' [71, 72]. The recent discovery of this phenomenon calls into question the validity of previous interpretations of infra-red absorption experiments and raises questions about the applicability of the process to large-scale processing of UF$_6$ [72a].

The alternative of using an ultraviolet laser for the second step is the method currently being pursued most actively by the Los Alamos Laboratory in the USA. There seems to be some doubt that this process may be successfully developed for large-scale enrichment. The basic problem is the very low absorption probability for ultraviolet photons in vibrationally excited UF$_6$ molecules. This implies that even under favourable conditions the UV excitation process will be very inefficient, and that the UV lasers will have to be large and powerful. The open
literature suggests a genuine dispute among researchers concerning the prospects for UV photo-dissociation. Even a relatively optimistic assessment points out that “the ... UV laser system ... will require substantial development to achieve low-cost, reliable devices” [49e]. Others are quite negative, even to the point of excluding ultraviolet photo-dissociation from a list of promising processes for collecting vibrationally excited UF₆ molecules [73]. There is no way for an outsider to decide who is right in this argument, but it is safe to say that arguments of such a fundamental nature generally do not take place over systems which are close to commercial application.

As difficult as the laser problem may be, the advantages of the MLIS process, as well as the many other potential uses for such lasers, guarantee that the search for adequate lasers will go on. The MLIS process promises much smaller irradiation volumes than AVLIS and therefore less extensive and complex optical systems for the laser beams. This follows from the higher densities possible in the irradiated gas [57c]. There is also the advantage of handling UF₆ instead of hot, corrosive uranium vapour. Finally, there is the possibility that the molecular process can be conveniently operated in stages. This follows from the simple and clean method by which the enriched product in the form of UF₅ ‘laser snow’ can be filtered from the gaseous tails stream [74]. The latter can immediately be sent on for further depletion while the former can be quickly refluorinated (see figure 6.21) and sent on for further enrichment.

The actual feasibility of this will depend on how sensitive the process is to ²³⁵U assay. However, there is no obvious analogy with the problems encountered for higher assays in the AVLIS process (see pp. 171–72). In that process there was no buffer gas to dilute the ²³⁵U concentration to match the laser power capabilities while still promoting vibrational cooling. So there seem to be no obvious obstacles to cascading the MLIS process to high enrichments. This means that one solution to the ultraviolet laser problem may be the use of many lower power lasers in a cascaded system rather than a single very powerful UV laser in a one-stage system. It is perhaps significant that the most recent theoretical work on non-ideal cascades with high single-stage enrichment factors has been done at Los Alamos [75, 76].

One other important difference between the MLIS and AVLIS processes is in the product collection mechanism. Not only does the formation of UF₅ snow facilitate the segregation of product from tails, but it also removes the need for very high laser pulse rates. These are needed in the AVLIS process to ensure irradiation of all of the vapour, since non-irradiated vapour is collected at a fixed rate along with the ²³⁵U ions. But in the MLIS process any UF₆ gas which is not irradiated simply continues on into the tails stream and has no effect on the product assay of the irradiated vapour. In addition it would seem that for higher feed assays less degradation of product would occur as a result of fluorine-exchange reactions. This exchange process
can take place when the photo-dissociated $^{235}\text{UF}_5$ collides with $^{238}\text{UF}_6$ molecules before condensation. Such collisions are of course less frequent the higher the percentage of $^{235}\text{UF}_6$ in the gas. So one expects that enrichment efficiency would improve at higher assays, again adding to the attraction of the cascading. The use of a buffer gas for vibrational cooling means that the $^{235}\text{UF}_6$ density can easily be made independent of feed assay and thereby always matched to the capabilities of the lasers.

No data are available, so the best that can be done is to estimate that the separation factors achievable in the MLIS process will be comparable to those of the AVLIS process. If not, the greater ease of cascading should redress the balance. Energy consumption in the laser systems of both processes is roughly equivalent, and the evaporation energy of AVLIS is replaced by pumping energy in MLIS, so it seems reasonable to assume that the MLIS process will share the energy-saving virtues of AVLIS.

All of these factors suggest that, if the laser problems can be solved, the MLIS process may actually prove to be effective for producing highly enriched uranium at significant rates in relatively small facilities. This has clear implications for proliferation control, which are examined in chapter 2.

**Laser-assisted processes**

The foundation of most laser-assisted processes is the ability of the laser to selectively deposit energy in one kind of molecule while leaving all others unaffected. In the previously discussed methods this energy is used to ionize an atom or dissociate a molecule, but even if smaller amounts of energy can be deposited selectively, some very interesting results can occur. In particular, this energy can be used to rapidly raise the temperature of one component of a gas mixture while leaving the temperature of other components relatively unaffected.

The concept of thermal equilibrium was discussed in section II in connection with the theory of gaseous diffusion (see p. 121). Any dense mixture of gases will rapidly achieve thermal equilibrium and an equipartition of energy by virtue of the many collisions between molecules. This equipartition of energy fixes the ratios of average velocities of the molecular species, and in the case of $^{235}\text{UF}_6$ and $^{238}\text{UF}_6$, this ratio is only very slightly different from one. This very slight difference then leads to the requirement of many enrichment stages in a gaseous diffusion or aerodynamic separation facility.

Suppose, however, that it were possible to raise the temperature of the $^{235}\text{UF}_6$ relative to the $^{238}\text{UF}_6$. This average velocity ratio could then be substantially increased and far fewer stages might be needed. This selective increase in temperature can be achieved with an infra-red laser similar to
the one used in the MLIS process. The laser excites $^{235}\text{UF}_6$ molecules into a high vibrational state, and, then, as the molecules collide with others, usually a buffer gas, much of this vibrational energy can be converted into translational energy, that is, higher speed. The $^{235}\text{UF}_6$ will then diffuse much more rapidly than $^{238}\text{UF}_6$.

One proposal to this effect has been made by JNAI [77]. The process involves injecting a subsonic flow of a $\text{UF}_6$–argon mixture into a tube in which it can be irradiated with an infra-red laser. At the downstream end of the tube are a set of cryogenically cooled plates through which all the gas must pass. Since the $^{235}\text{UF}_6$ molecules have been given higher velocities, they are more likely to collide with the plates where they condense to form solid $\text{UF}_6$. After some period of operation the tube is evacuated and the plates are heated to release the accumulated $\text{UF}_6$ product. This can then be sent directly on to another stage without any reflux chemistry having to be performed.

Another suggestion has been to combine a laser with the Becker nozzle process or other aerodynamic processes [78]. Since the selectivity of these processes depends on the rate of diffusion of $^{235}\text{UF}_6$ across curved streamlines, the separation effect can be enhanced by selectively heating the $^{235}\text{UF}_6$ as it passes through the curved region, thus enhancing its rate of diffusion.

A third proposal uses atoms of uranium instead of $\text{UF}_6$ and proposes that the laser photons be used directly to impart a transverse momentum to the $^{235}\text{U}$ atoms in a collimated atomic beam [79]. When an atom absorbs a photon it acquires that photon's momentum; so if the beam is not too dense and an adequate collision-free drift region can be maintained, it may be possible to separate the deflected beam from the undeflected one and obtain an enriched product.

No evidence exists that these methods are much more than interesting ideas at this time, and their ultimate fate will take a long time to be determined. Meanwhile new variations on this basic theme and others are appearing with undimining regularity. The number of possible variations seems to be very large.

**Summary**

This survey has shown that a large number of isotope separation techniques have been made possible by the advent of the laser. The essence of the laser's attractiveness for this purpose lies in its highly precise ability to process only $^{235}\text{U}$, leaving the vast vast majority of the uranium unaffected. This increases selectivity and greatly reduces energy costs. However, it achieves these improvements at the cost of high technical sophistication and component costs. Substantial questions still remain as to whether or not stable, reliable, and economical laser and materials handling systems can be developed.
If they are developed the implications for weapon proliferation of laser isotope separation are quite serious. Attention was called to these implications in 1977 [57, 80, 81], but since that time there has still been relatively little public discussion of the costs and benefits of continued development of these systems. As usual, concern about proliferation seems to be subordinated to commercial and bureaucratic interests.

One more important aspect of laser processes deserves mention. This is the ability of the laser to also separate plutonium isotopes, something which is not generally considered feasible in any of the other processes discussed previously [82]. The most important plutonium isotope for weapons is $^{239}$Pu, but this tends to be heavily contaminated by $^{240}$Pu when the plutonium has been produced in normal nuclear reactor operations. The presence of the $^{240}$Pu degrades but does not destroy the material’s usefulness as a nuclear explosive [50b].

Since the two isotopes differ in mass by only one unit instead of three (as in uranium), and since other isotopes ($^{238}$Pu, $^{241}$Pu, $^{242}$Pu) are also often present, the ability of all the previously described methods to separate them is greatly reduced. In addition, plutonium presents much more severe problems of radioactivity, toxicity and criticality than uranium.

Only the laser, and possibly the electromagnetic methods discussed in the next section, show some promise of separating plutonium isotopes. Presumably plutonium metal can be vaporized like uranium metal, but criticality dangers would require that the amounts used be much smaller. Plutonium also forms $\text{PuF}_6$, a substance which seems to have properties very similar to $\text{UF}_6$ [1c]. Research on laser plutonium separation is going on in the USA, and presumably elsewhere as well [42f, 49f, 83]. Given these developments there can be no doubt that continued progress in laser isotope separation will greatly complicate efforts to control nuclear weapon proliferation.

VII. Electromagnetic and plasma processes

Basic principles

All of the methods in this category depend on the ionization of all or part of the feed material and the use of electric and magnetic fields to accelerate the ions and separate the isotopes. All are characterized by relatively high separation factors but relatively low throughput, caused mainly by the inherently low densities of the ion beams or plasmas used.

A feature common to all electromagnetic processes is the use of magnetic fields to accelerate uranium ions. When a charged particle enters such a field it can experience a force, and this force has two special
properties:

1. It only occurs if the particle has some motion perpendicular to the field lines.
2. The force itself is perpendicular to both the field lines and the velocity of the particle.

These two characteristics of the magnetic force lead to the result that the paths of all charged particles (electrons or ions) in a uniform magnetic field are circles or helices, with the plane of circular motion perpendicular to the magnetic field lines. The frequency of the circular motion, the so-called 'cyclotron frequency', depends only on the charge and mass of the particle and the strength of the magnetic field.

\[ \nu_c = \frac{eB}{2\pi m} \]  

Equation 6.11 shows that the frequency of rotation is independent of the particle velocity. This means that all particles complete one revolution in the same amount of time. For this to be true there must be a direct proportionality between the speed of a particle and the radius of its orbit, called the 'radius of gyration'. So, for a given type of particle the faster it is moving the larger the circle it will describe. This is the physical basis for the ion cyclotron resonance technique for isotope separation which will be described more fully below.

Another way of utilizing the magnetic force is to give all the ions a very well-defined velocity perpendicular to the field. Then the magnetic force will act with equal strength on all of them, but according to Newton's laws the lighter particles will experience greater accelerations than the heavier ones, implying that the lighter particles will move in circles of smaller radius. After one-half of a circular orbit, the beam will have separated into a number of distinct beams, each containing only particles of a given mass (assuming that all have the same charge). This is the principle underlying the technique of mass spectroscopy, employed for isotope separation in a device called the calutron.

An even more interesting phenomenon can be observed if an electric field is added to the magnetic field. For example, consider a situation such as shown in figure 6.23 in which a radially directed electric field has been superimposed on the axial magnetic field of a solenoid. The electric field will accelerate positive ions outward, but as the ions move outward their interaction with the magnetic field will cause them to deflect into circular orbits. After a very short time the ions will acquire a uniform speed called a drift velocity in the azimuthal direction, that is, perpendicular to both the electric and magnetic fields. This motion differs from cyclotron motion because the direction of the drift velocity is independent of the charge of the particle.

The magnitude of the drift velocity is also independent of both the mass and the charge of the particle, but the drift motion is superimposed on the cyclotron motion. This results in a net azimuthal velocity which does depend on the mass of the particle. If one now injects into the solenoid a
gas of ions and electrons, that is, a plasma, the entire gas will be set into rotation. Very high rotational speeds can be achieved in this way. This is
the principle underlying several suggested designs for 'plasma centrifuges' which separate isotopes in a manner very similar to the mechanical centrifuge of section III but with no moving parts.

Before proceeding to more detailed discussions of particular processes, it is useful to describe the general features of a plasma. A plasma is
a gas of charged particles — generally positive ions and negative electrons. It must have a very high temperature, at least of the order of thousands of
degrees Celsius, so that the high relative velocities of ions and electrons will prevent their rapid recombination to neutral atoms. It is possible to
apply many of the same laws of statistical mechanics which apply to neutral gases to plasmas, in particular the equipartition law. This immediately
leads to the prediction that the electrons in a plasma must have much
greater average velocities than the ions. In a uranium plasma this
difference is particularly dramatic since the mass of a uranium ion is over
400,000 times that of an electron. This implies that electron velocities are
about 650 times greater than ion velocities on the average.

This enormous disparity in velocities suggests that electrons might quickly escape from the plasma, but electrons which do emerge from the
plasma are attracted back by the residual positive charge of the ions left behind. These electrical forces are very strong and tend to preserve the overall charge neutrality of the plasma. The result is that the plasma is surrounded by a thin sheath of electrons held in place by the attraction of the positive charge in the interior. The thickness of this sheath can be estimated from simple electrostatic principles, and it is found to be comparable to the Debye shielding distance \[84\]. Strictly speaking, a plasma is defined to be a gas of charged particles whose dimensions are large compared to this distance. In the uranium plasmas used for isotope separation the Debye shielding distance is typically 1–10 μm.

Plasmas can be produced in a number of ways. One method is to inject a beam of high energy ions or electrons into a neutral background gas. Collisions cause the ionization of the background gas, and a plasma can be formed. Another method is to create an electrical discharge in a gas between two electrodes by applying a voltage difference. This is a phenomenon observed in nature: in electrical storms lightning discharges create temporary plasmas along the path of the discharge, and the strong electric fields present in the humid air can cause other strange effects which depend on the formation of plasmas, one of which is called ‘ball lightning’ \[85\].

A third method of obtaining a plasma is to allow a neutral beam of atoms to strike a hot, glowing metallic surface, which strips the electrons off the incoming atoms and forms a plasma. Such a device is called a ‘Q-machine’ \[2ff\].

With these general introductory comments as background, it is now possible to consider the three classes of electromagnetic methods in more detail.

The calutron

The name ‘calutron’ says a great deal about the historical origins of this process. The calutron was invented in the laboratory of Ernest O. Lawrence of the University of California (hence ‘cal’) in the early 1940s. In the late 1930s Lawrence had invented the cyclotron, a device for accelerating charged particles to high energies for nuclear physics research, and the calutron concept was based on the same technology (hence ‘tron’). In fact, with the start of World War II and the intense interest in the United States in rapidly producing atomic bombs, Lawrence turned much of his laboratory and its equipment over to the attempt to make highly enriched uranium \[86a\].

The major piece of equipment which made it possible for Lawrence to begin his new project rapidly was a large magnet, 4.67 m in diameter. This magnet consisted of two circular pole faces separated by a gap in which a vacuum chamber was inserted. Inside this chamber could be placed a source of uranium ions and, 180° around the circle, a collector. The source
Figure 6.24. An early two-beam calutron

One of the two-beam calutrons designated Alpha-1 at the Oak Ridge facility in 1944. The unit rests on its door on a storage dolly. The covers have been removed to show the double source at the right and the two receivers at the left. Some four-beam units were also put into service before the end of the war.


would accelerate the ions to a velocity such that the curvature of their trajectory in the magnetic field would cause them to arrive at the detector (see figure 6.24).

The radius $r$ of the path of an ion in a uniform magnetic field is given by

$$r = \frac{mv}{eB}$$

where $m$ and $e$ are the ion's mass and charge, respectively, $v$ is the ion velocity, and $B$ is the magnetic field strength. If the ions are accelerated by a voltage $V$ then their kinetic energy will be $eV$ and their velocities will be

$$v = \sqrt{2eV/m}$$

When these two equations are combined, the radii of curvature of the ions in the magnetic field turn out to be

$$r = \frac{1}{B} \sqrt{2Vm/e} \quad (6.12)$$
Since all quantities but \( m \) are the same for different ion species, it follows that the radius for a particular isotopic ion is proportional to the square root of its mass. This implies that two ions whose masses differ by 1.27 per cent will have radii that differ by only 0.64 per cent. Given typical values [86b]: \( B = 0.34 \text{ tesla}, V = 35 000 \text{ volts}, e = 1.6 \times 10^{-19} \text{ coulomb}, m = 3.9 \times 10^{-25} \text{ kg} \), the beam radius comes out at \( r = 1.22 \text{ m} \), and the separation between the \(^{235}\text{U}^+\) and \(^{238}\text{U}^+\) beams would be a little over 15 mm. This is just large enough to allow a collector to be made which will admit only the \(^{235}\text{U}^+\) beam and reject the \(^{238}\text{U}^+\) beam with a high degree of selectivity [87a]. It even proved to be possible to put two or even four source and collector pairs into the same vacuum tank to make more efficient use of the magnetic fields (see figure 6.24).

What appears in theory to be a very simple and precise method for isotope separation turns out in practice to be extremely difficult if any more than laboratory-sized amounts of product are desired. The major problems encountered in the Y-12 calutron project were the severe limitation on ion beam strength created by space charge effects, and the inability to convert more than a small fraction of the feed material into product in any single run. Space charge problems result from the tendency of particles with similar charges to repel each other and destroy the beam. This can be overcome to some extent by allowing the positive ions to collide with residual background gas in the calutron and thereby release electrons which neutralize the repulsion. In effect, a neutral plasma is created along the beam path [11d]. However, this technique is limited to low densities of background gas, since too high a density would destroy the beam by causing too many collisions with the beam particles. So the currents in a typical calutron beam were limited to a few hundred milliamperes, leading to a collection rate of only about 100 mg/day of \(^{235}\text{U}^+\). At this rate it would take over 400 years to collect a critical mass. This explains why the Y-12 project employed more than 1 100 calutrons [87b].

The actual enrichment factors achieved in the Y-12 calutrons were at least 20 [86c] and probably somewhat over 30. Various improvements since 1945 have resulted in the capability to get enrichment factors anywhere from 30 to as high as 80 000 in a single pass [87c]. However, the very high values can only be obtained using special geometrical arrangements and very low throughput. It seems reasonable to infer that at the throughput rates needed for the production of significant amounts of highly enriched uranium a range of 20 to 40 should adequately describe the capabilities of the calutron.

The high enrichment gain is offset by a very low efficiency in feed utilization. This is due to the tendency of ionized uranium vapour to deposit on all available surfaces inside the vacuum chamber. Only 10 to 15 per cent of the feed material is actually processed [11e], meaning that the cut is below 0.01 for low-enriched feed.

This problem was particularly serious in the early calutrons which operated on feed material that had already been partially enriched and was
therefore extremely valuable. The insides of these machines had to be periodically cleaned and the material recovered with high efficiency.

The low collection efficiency also leads to a very high specific energy consumption because of the high energy to which the full beam must be accelerated (≈ 35 000 eV) to get separation. Using this energy value and an enrichment factor of 30, and assuming that 15 per cent of the $^{235}\text{U}$ in the beam is captured, the specific energy consumption can be estimated at 3 800 kWh/\text{SWU}, greater than for gaseous diffusion. This number applies to the early calutrons, so it could possibly be improved upon with modern technology.

In the end it proved possible for over 1 000 calutrons to produce enough uranium for one bomb, but only after three years of intense effort and roughly a billion dollars of expense. It is clear that if it had not been for the overwhelming desire to achieve a usable weapon before the end of the war, the calutron project would never have been pursued as far as it was. Indeed, once the gaseous diffusion plant at Oak Ridge began to produce highly enriched product, the large calutrons were closed down. Now this process is considered suitable only for the production of small quantities of very pure isotopes. It is perhaps worth noting that the calutron method, like the laser methods, is one of the few suitable for separating plutonium isotopes [88]. But the limitations of the method make it extremely unlikely that it would ever be feasible for the production of kilogram quantities of pure $^{239}\text{Pu}$ from irradiated reactor fuel.

**Ion cyclotron resonance**

A schematic diagram showing the essential features of this process is given in figure 6.25. The method uses selective excitation of $^{235}\text{U}$ cyclotron motion in a plasma. The plasma is created either by an electrical discharge or by a Q-machine and caused to flow axially through a region containing a strong magnetic field created by a superconducting solenoid [89a]. Typical experimental values of the field strength have been about 2 tesla [49g].

As they pass through the magnetic field the ions are subjected to an alternating electric field whose frequency is tuned precisely to the cyclotron frequency of the $^{235}\text{U}$ ions (see equation 6.11, p. 174). Assuming that the uranium ions are singly charged ($e = 1.6 \times 10^{-19}$ coulomb), and using an ion mass of 235 atomic mass units (1 amu = $1.66 \times 10^{-27}$ kg), the cyclotron frequency of a $^{235}\text{U}$ ion turns out to be 130 kHz in a field of 2.0 tesla. The difference between the $^{235}\text{U}$ and $^{238}\text{U}$ frequencies is about 1.25 per cent of this, or about 1.65 kHz.

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5 Superconductivity is a phenomenon which occurs in many metals at very low temperatures, that is, a few degrees above absolute zero. It is characterized by zero electrical resistance and therefore a zero rate of resistive energy loss. Only liquid helium can be used to cool the magnet coils, since only helium remains a liquid at such low temperatures.
Figure 6.25. Schematic illustration of the plasma separation process based on ion cyclotron resonance

Since the rotation of the ions is taking place in the plane perpendicular to the axis of the solenoid, the alternating electric field must also be in this plane so that it can add energy to the ions. Figure 6.26 illustrates this acceleration process. Consider an ion which is moving in the $x$ direction when the electric field is also in this direction. In this situation the field will accelerate the ion and add to its energy, causing it to move in a circle of larger radius but with the same frequency. One half-cycle later, the ion is moving in the opposite direction, but if the alternating field has also changed direction, the acceleration can occur again, and the radius of motion of the ion will be further increased. It is this precise timing of the oscillating frequency of the field to the cyclotron frequency of the ion which is called cyclotron ‘resonance’.

The frequency must be stable to an accuracy of 1 per cent or better to distinguish $^{235}\text{U}$ from $^{238}\text{U}$. This is not difficult to achieve, but it is more difficult to produce a magnetic field which is uniform to better than 1 per cent over a large area. Modern magnet technology can achieve this goal only with relatively sophisticated and expensive solenoid designs (see figure 6.27).

The exciting electric field can be applied in a number of ways, but all must take into account the strong tendency of plasmas to prevent electric fields from penetrating very far into their interior. As was mentioned above, a uranium plasma has a characteristic shielding length of only a few
micrometres, so externally applied electric fields cannot penetrate deeper into the plasma than this distance. For this reason the electric field must be applied indirectly, either inductively by modulating the axial magnetic field, or by inducing an ion cyclotron resonance wave in the plasma with properly placed electrodes [89b]. Both of these methods create some problems for application, and it is possible that a third, more recently proposed method may prove superior to both [90].

However the electric field is applied, its purpose is to oscillate in phase with the rotating $^{235}\text{U}$ ions. Ions whose cyclotron frequencies are slightly different will fall out of synchronization with the field and will therefore experience no net gain in energy. The effect of increasing the kinetic energy of the $^{235}\text{U}$ ions is to greatly increase their radius of gyration in the field. A typical unexcited ion will have a radius of gyration of the order of a millimetre, but excited $^{235}\text{U}$ ions can be made to move in circles several centimetres in diameter (see figure 6.25). It is not difficult to think of collector designs which would allow most of the $^{238}\text{U}$ ions rotating in their tiny circles to pass through, while capturing most of the $^{235}\text{U}$ ions moving in large circles.

The limitations on density and temperature of the plasma can be inferred from the requirement that the $^{235}\text{U}$ ions be allowed to go through
many cyclotron orbits as they pass from one end of the tube to the other. Because the cyclotron frequency of $^{238}\text{U}$ is so close to that of $^{235}\text{U}$, the two must make many revolutions in order to ensure that the $^{238}\text{U}$ will be out of phase with the electric field for a substantial number of revolutions. The number of cyclotron periods needed to ensure this is of the order of $M/\Delta M$ or about 80. If the separating tube is about 1 m long and the period of a cyclotron oscillation is 7.7 $\mu$s (1/130 kHz) then the axial velocity of the ions cannot be greater than 1 600 m/s. This corresponds to a plasma temperature of about 75 000 K.

The plasma density will be limited by the requirement that a $^{235}\text{U}$ ion must be able to make all of its 80 or so revolutions without suffering a collision with another ion. Such a collision would obviously throw it out of phase with the electric field and cause it to lose energy. This requirement will be satisfied if the average ion collision frequency is about 1/80 of the cyclotron frequency, or under 2 kHz. Using a typical collision cross-section of $10^{-14}$ cm$^2$ for uranium ions [57c], it can be shown that the ion density in the plasma cannot be greater than $10^{12}$/cm$^3$.

Knowing the ion density in the plasma and the average speed down the tube it is a simple matter to compute the particle flux. This is of the order of $10^{17}$ ions/cm$^2$s. If the plasma beam has a diameter of about 100 cm, if the feed material is assumed to be natural uranium, and if all the $^{235}\text{U}$ ions are assumed to be collected, then the rate of collection of $^{235}\text{U}$ would be $5.7 \times 10^{18}$ atoms/s or about 70 kg $^{235}\text{U}$/yr. In order to separate 1 t of $^{235}\text{U}$ per year (enough to make 30 t of reactor fuel) the diameter of the beam would have to be increased by almost a factor of 4, to 3.8 m. This might explain the research and development now being carried out on a
superconducting solenoid with a diameter of 4 m [49g].

It must be emphasized that this result is only coincidental, and that all of the numbers in the above calculations are highly uncertain. The eventual values of magnetic field, plasma density and temperature, and collection efficiency will depend on many factors which are either unknown at the present stage of development, or, if they are known, not available in published sources. So the estimates made here should be seen as only rough, order of magnitude approximations. They do, however, fall more or less within the ranges mentioned in the literature [89, 2gg].

There are no reliable data on the enrichment factors achievable with the plasma resonance process. One source suggests “about 10” [33]. A possible lower limit to the acceptable enrichment factor can be inferred from the stated mission of the US Advanced Isotope Separation (AIS) programme, of which the plasma separation process (PSP) is a part. The purpose of the programme is “treatment of uranium tails from conventional [enrichment] plants to produce feed-grade material at costs below natural uranium prices” [91]. This suggests that unless enrichment factors of at least 3.5 per cent in one pass (converting 0.2 per cent feed to 0.71 per cent product) are attainable, the process is unlikely to be considered worth pursuing further.6

Very little more can be said about how this process will look if and when it becomes commercially or militarily viable. The size of an enrichment unit is comparable to that for the AVLIS process, since the atomic vapour and plasma densities and flow rates are comparable. Both processes will also have to have similar facilities for materials processing and handling. The plasma process will not require the elaborate laser infrastructure of AVLIS, but in its place will be the need to supply large amounts of liquid helium to maintain the magnets in their superconducting state. The energy consumption of the plasma process will probably be somewhat larger than that of AVLIS, but no firm estimates are available. One source does mention a possible specific energy consumption of “a few hundred kWh/SWU” for anticipated plasma facilities [92].

Plasma centrifuges

There are a number of variations of the plasma centrifuge concept, but all involve the creation of a rapidly rotating plasma. This is achieved by creating an electrical discharge in a direction perpendicular to a strong magnetic field [2hh] (see figure 6.28). The geometry in the figure is simplified for illustration purposes. Actual plasma centrifuges can be much more elaborate [93] (see figure 6.29), and they differ in the arrangements

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6 This decision has now apparently been made, at least in the USA (see footnote 4, p. 160).
A flowing discharge is created by applying a voltage between the anode and cathode. The discharge flows across magnetic field lines created by a solenoid which surrounds the discharge tube. Rotation of the plasma is induced by the mechanism shown in figure 6.23.

of the discharge electrodes, field configurations and methods of feeding in and collecting uranium [15f]. But all share the very great difficulties associated with the complicated behaviour of plasma discharges in magnetic fields. These have been summarized as follows: “... so far no gas discharge operated in the presence of a magnetic field is understood well enough that all the properties of the plasma may be calculated. It is even more difficult to obtain reliable information about the role of the neutral particles. Evidently the state of the art in isotope separation with plasmas is directly related to the extent that the used plasma is understood” [2ii].

The isotope separation effect in a rotating plasma depends on the collisions which occur between the two ionic species in the plasma. As was shown in the introduction to this section, ions of different masses have different rotational speeds, so at a given radius they will have a relative velocity with respect to one another. When they collide the heavier species tends to speed up and the lighter to slow down. The result is a tendency for the heavy ions to drift to the periphery of the rotating plasma while the light ions concentrate on the inside [93]. In this way the results are quite similar to the behaviour of UF₆ gas in a mechanical centrifuge, but the physics of the process is far more complicated and difficult to manipulate.

One intuitive prediction which is borne out is that it will require considerably more energy to set the entire plasma into rotation than to selectively excite only ²³⁵U ions. Early experiments on isotope separation in krypton gas gave specific energy consumptions 10 times greater than for gaseous diffusion [2jj]. It is suggested that this can be reduced considerably by optimization of design, but the numbers suggested are still substantially larger than those for mechanical centrifuges.

Figure 6.29. Rotating plasma device with curved magnetic field lines
The plasma occupies the shaded area in which an electric field $E$ produces a rotation around the axis of symmetry and a centrifugal force by which ions with different masses are separated.


One suggestion to reduce energy consumption is to use a partially ionized plasma with a low degree of ionization. In this variation the charged particles can be accelerated into rotation, and this rotation can then be transferred to the neutral gas by viscous forces (i.e., collisions) [94]. However, there is no evidence that this idea has progressed much beyond the original suggestion.

Another factor which suggests that the plasma centrifuge may have a difficult time competing with existing methods is the comparison of theoretically obtainable single-stage enrichment factors with those of the mechanical centrifuge. One early estimate for uranium was 1.134 [95a], and one experimental determination gave values in the range of 1.05–1.10 [2kk]. Experiments on krypton gave values in the range 1.05–1.15 depending on the cut [2jj]; and up to 1.10 was achieved in experiments on neon [15g], although this was improved to 1.20 in a later experiment [96]. A Soviet group has achieved an enrichment factor of 1.06 in a $^{129}$Xe–$^{136}$Xe mixture [97]. Given that the mass of uranium ions is considerably greater than that of krypton or neon, and that the xenon isotopes had a mass difference of seven units compared to the three of uranium, these latter two results must be upper limits for uranium.

Of course these numbers are not directly comparable with modern centrifuges, because the latter use internal countercurrent flow to enhance the separative effect. This possibility also exists in some forms of plasma centrifuge [95b]. Separation factors can in principle also be increased quite dramatically in plasma centrifuges by increasing the rotational speed of the plasma. There are no bearings to wear out or walls to explode if the rotational speeds are increased to 100 or more times those achievable with mechanical centrifuges. However, such speeds in a plasma require very
strong magnetic fields and intense discharge currents, and as the peripheral
speed increases, energy consumption and problems with ion-neutral
collisions also increase.

Comparisons are also difficult between the plasma centrifuge and
cyclotron resonance techniques. While the attainable separation factors for
the latter seem to be substantially larger, the former are not limited by
collision effects (indeed they require collisions for their operation) so
densities and therefore throughputs can in principle be much larger.
Densities in plasma centrifuge experiments seem to have been of the order
of $10^{15}$ particles/cm$^3$ [2 mm], 100–1 000 times those of cyclotron reso-
nance.

Summary

There is a kind of historical symmetry about the electromagnetic processes.
The first successful enrichment method, the calutron, belonged to this
class, and now (35 years later) as the enrichment industry moves towards
maturity, these processes are again under serious consideration. None of
the modern methods are yet at a stage where either their commercial
potential or proliferation implications can be reliably assessed. The
numbers quoted and computed in this section (except for the calutron)
are certainly the most uncertain of any in this study and must be
used with great caution in any comparisons among different types of
technology.

VIII. Miscellaneous methods

The previous six sections have described the enrichment processes which
have either established themselves as capable of large-scale enrichments or
are considered likely enough to do so in the near future that they have been
given substantial research and development support. However, this list of a
dozen or so methods represents only the tip of the iceberg, the submerged
portion of which is made up of dozens, possibly hundreds, more ideas,
gadgets, patents, and so on, which have been proposed over the past 30
years.

Wherever a strong commercial or military incentive exists this kind of
creativity is certain to follow, and for many years it was believed that the
world would beat a path to the door of the nation or corporation which
built a better enrichment plant. These expectations have faded consider-
ably in the past few years, but the creativity goes on, and new enrichment
ideas appear with surprising regularity.
There is neither need nor space to survey this broad field here, but a few examples might give some idea of the wide variety of proposals which have been made. Since this is intended as a qualitative survey, no numbers will be given or evaluations made. Such numbers or evaluations which do exist can be found in the references.

One group of methods has already been mentioned briefly in section IV. These are the aerodynamic techniques, and there are many variations with names like ‘separation probe’, ‘jet membrane’, and ‘crossed jet’ [26, 28a, 98]. The first directs a supersonic flow of UF₆ in a background gas against the end of an open tube. A shock wave is created at the mouth of the tube, and the separation depends on the differential rate of diffusion of $^{235}$UF₆ and $^{238}$UF₆ across the shock front. The heavier molecules are more likely to penetrate the front, so the tube draws off the depleted fraction, leaving behind an enriched fraction.

The jet membrane is analogous to gaseous diffusion except that the gas is forced to diffuse through another gas rather than through a solid, porous membrane.

The crossed beam technique directs a jet of UF₆ gas at right angles to a jet of some other gas. Collisions occur in the region where the jets intersect, and the lighter $^{235}$UF₆ molecules are deflected slightly more on average than the $^{238}$UF₆. This process is similar in concept to the laser deflection technique described in section V (p. 172), with molecules substituted for photons.

Another type of separation process relies directly on velocity differences between light and heavy UF₆ molecules. One variant called the ‘velocity slip’ technique accelerates UF₆ in a light background gas through an expanding nozzle. This is done in pulses, and a rotating velocity selector is used to skim off the lighter, faster molecules as they emerge from the nozzle. Another variation on this theme is the so-called ‘garden hose’ technique, which again relies on average velocity differences between light and heavy UF₆ and uses a complex spherical sweeping device to skim the enriched fraction [99, 100].

There have been many variations on the centrifuge concept, and a relatively recent one is the ‘HAGA radial-separating-nozzle centrifuge’, designed and built in Austria [101, 102]. This device is a very complex, multi-chambered centrifuge which relies on vortex and eddy effects at the periphery of a rotating disc to get the separation effect. Very high separative capacities are claimed for rather small devices, but the theoretical and experimental results are very preliminary.

Finally, one of the more recent contributions to the field has been a device that uses a vibrating bar, something like a tuning fork, placed in an atmosphere of UF₆ [103]. When the bar vibrates in one of its resonant modes, the lighter UF₆ tends to concentrate at the locations where the amplitude of vibration is maximum (the antinodes) while $^{238}$UF₆ migrates to the static nodal regions.
### Table 6.3. Summary of enrichment process characteristics

<table>
<thead>
<tr>
<th>Process</th>
<th>Working material</th>
<th>Single-stage separation factor (q=1+g)</th>
<th>Stage cut (\theta = P/F)</th>
<th>Stage hold-up time (s)</th>
<th>Specific energy consumption (kWh/SWU)</th>
<th>Stage reflux mechanism</th>
<th>Status of technique</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gaseous diffusion</td>
<td>UF₆</td>
<td>1.0040–1.0045</td>
<td>1/2</td>
<td>5–10</td>
<td>2 300–3 000</td>
<td>None</td>
<td>Mature</td>
</tr>
<tr>
<td>Centrifuge</td>
<td>UF₆</td>
<td>1.3–1.6</td>
<td>~1/2</td>
<td>10–15</td>
<td>100–300</td>
<td>Internal countercurrent flow</td>
<td>Mature</td>
</tr>
<tr>
<td>Aerodynamic Nozzle</td>
<td>0.04UF₆ + 0.96H₂</td>
<td>1.015²</td>
<td>1/4</td>
<td>~2</td>
<td>3 000–3 500</td>
<td>Recycle intermediate fraction</td>
<td>Demonstration</td>
</tr>
<tr>
<td>Helikon</td>
<td>0.01–0.02UF₆ + 0.98–0.99H₂</td>
<td>1.025–1.030²</td>
<td>1/20</td>
<td>0.05–0.2</td>
<td>3 000–3 500</td>
<td>None</td>
<td>Demonstration–Production</td>
</tr>
<tr>
<td>Chemical Solvent extraction</td>
<td>Aqueous and organic solutions of U compounds</td>
<td>1.0025–1.0030</td>
<td>1/2</td>
<td>20–30</td>
<td>≤600</td>
<td>Chemical conversions U(IV)⇔U(VI)</td>
<td>Pilot plant</td>
</tr>
<tr>
<td>Ion exchange</td>
<td>Aqueous U solution and ion-exchange resin</td>
<td>1.0013</td>
<td>1/2</td>
<td>~1</td>
<td>(400–700)²</td>
<td>Chemical conversions U(IV)⇔U(VI)</td>
<td>Pilot plant</td>
</tr>
<tr>
<td>Laser</td>
<td>Molecular UF₆–N₂</td>
<td>(5–15)³,⁴</td>
<td>NA</td>
<td>NA</td>
<td>10–50</td>
<td>None</td>
<td>R&amp;D</td>
</tr>
<tr>
<td></td>
<td>Atomic U vapour</td>
<td>5–15³</td>
<td>NA</td>
<td>NA</td>
<td>10–50</td>
<td>Recover and recycle U metal</td>
<td>R&amp;D</td>
</tr>
<tr>
<td>Method</td>
<td>Source Material</td>
<td>Separation Factor</td>
<td>Recovery</td>
<td>Cost (k$)</td>
<td>Remarks</td>
<td></td>
<td></td>
</tr>
<tr>
<td>------------------------</td>
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<td>----------------------------------------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Electromagnetic Calutron</td>
<td>UCl₄&lt;sup&gt;e&lt;/sup&gt;</td>
<td>20–40&lt;sup&gt;d&lt;/sup&gt;</td>
<td>NA</td>
<td>NA</td>
<td>Recover and recycle UCl₄</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ion cyclotron resonance U plasma</td>
<td>3.5–10&lt;sup&gt;d&lt;/sup&gt;</td>
<td>NA</td>
<td>NA</td>
<td>200–600</td>
<td>Recover and recycle U metal</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup> In order to use these values in table 5.1, the value of $g$ given here should be multiplied by twice the value of the stage cut.
<sup>b</sup> Estimate based on rough comparison with solvent extraction method.
<sup>c</sup> Estimate based on rough analogy with AVLIS method.
<sup>d</sup> The value entered for the last four techniques is the enrichment factor $\alpha$ (see equations 5.7) rather than the separation factor $g$.
<sup>e</sup> UCl₄ is the usual charge material used in the ion source (see reference 11f and reference 87d). Uranium ions are produced by passing a strong electric discharge through the UCl₄ vapour in a heated oven.
This list could go on much longer, but this sample should suffice to support the assertion that the field of isotope separation is far from being exhausted. This suggests that the possibility remains that one day a simple, inexpensive and easily concealable method for uranium enrichment will be discovered. Once the physical principles of a process are understood, efforts to maintain technological secrecy can never be more than temporary stopgaps [104]. And, it can safely be said that where there is a commercial or military will, there is usually a technological way. Any efforts to prevent nuclear proliferation by primarily technological means are therefore constantly threatened with rapid obsolescence.

IX. Summary

The important quantitative data on the techniques described in this chapter are collected in table 6.3. This table is intended for use in conjunction with table 5.1 (p. 113), and the data on separation factor, stage cut and hold-up time necessary for table 5.1 can be taken directly from table 6.3. For an example of how this is done see chapter 5, following table 5.1.

The exceptions to this rule are the techniques with high enrichment factors which are separated from the others in table 6.3 by a double line. Table 5.1 is not applicable to analysis of these, but most of the formulae needed to describe their operation are presented in chapter 5.

References to the sources of the numbers in table 6.3 are not displayed in the table but can be found in the relevant section of this chapter. The few cases where no public data could be found, and where the number represents an educated guess by the authors, are denoted in the table by parentheses. These values should, of course, be used with caution, but this warning should also be made for other numbers as well, especially near the bottom of the table. For those techniques below the double line which are still in their research and development phase, all quantitative estimates must be viewed as highly tentative.

The purpose of the descriptions in this chapter and the organization of the data in table 6.3 has been to provide the reader with sufficient information to make his or her own evaluations of the proliferation implications of each technique. For example, in Part One a possible diversion scheme is mentioned in which a quantity of 3 per cent enriched material is sent to a small clandestine centrifuge facility where it is converted into a small amount of weapon-grade material (90 per cent $^{235}$U) with the tails being returned quickly to the large facility from which the diversion took place.

Starting with an assumed requirement of 20 kg of 90 per cent $^{235}$U for a nuclear weapon one can first use table 5.1 to compute the quantity of
feed material required. In the last column of table 5.1 it is found that 44.5 kg of feed are required for every kilogram of product. So in the present example at least 890 kg of 3 per cent enriched feed would have to be diverted. It can also be seen from the same table that 52.2 \times 20 \text{ or } 1044 \text{ kg SWU must be performed to get 20 kg of product.}

Next assume that the clandestine facility uses centrifuges like those described in table 6.2. If we assume that 4000 centrifuges are used and that each has a capacity of 15.2 kg SWU/yr, then the facility has a total capacity of 60.8 t SWU/yr and can produce the required 1.04 t SWU in about 1/60 of a year or roughly six days.

Each centrifuge has an inventory of 0.26 g of uranium, so the total inventory of the plant is just over 1 kg, a negligible fraction of the feed required. If each centrifuge has a hold-up time of 13.7 s and a separation factor of 1.51, then table 5.1 can also be used to compute the equilibrium time. Again, referring to the last column and recognizing that a cascade made of centrifuges can be very close to an ideal cascade, we can choose a multiplying factor close to the lower limit of 30, for example, 50. The equilibrium time is then

\[
13.7 \text{ s}/(0.51)^2 \times 50 = 2630 \text{ s} \text{ or } 44 \text{ minutes}
\]

Finally note that a conservative estimate of the size of the facility can be made by allowing about one square metre of floor space for each centrifuge. This gives an area of 4000 m\(^2\), or a square of 63-m sides. A facility of this size would be far easier to conceal than a gaseous-diffusion or chemical-exchange plant.