

## *Chapter 5*

### **SAFETY OF THE FRONT END OF THE FUEL CYCLE**

#### **1. Uranium mining and milling**

The mining and milling of uranium do not have the same level of potential safety problems that are associated with operation of nuclear reactors and other fuel cycle facilities. The uranium mines do have the same safety and environmental issues as other mines in addition to the precautions required to deal with the radioactivity of the ore. Uranium mines are more publicly scrutinised because of these radiation issues. The results are that the safety, radiation, and environment programmes are highly successful.

There have been over 40 years of experience in successfully applying international radiation safety regulations at uranium mines. The ICRP has established recommended standards of protection. Risk factors were determined from low levels of radiation [1]. The scientific evidence does not indicate any cancer risk or immediate effects at doses below 20 mSv/y (millisievert per year). The ICRP recommended levels of effective dose are 1 mSv/y for the general public and 20 mSv/y for the radiation workers averaged over 5 years, with no individual year above 50 mSv. This is in addition to the background and medical exposures. Most countries have adopted these standards, including the two largest producers Australia and Canada. These standards are based on the following principles:

- justification: no practice involving exposure to radiation should be adopted unless it produces a net benefit to those exposed or to society in general;
- optimisation: radiation doses and risks should be kept as low as is reasonably achievable (ALARA), economic and social factors being taken into account;
- limitation: the exposure of individuals should be subject to dose or risk limits above which the radiation risk would be deemed unacceptable.

Mining has been successful at meeting these principles and standards. Radiation dose records show that the maximum doses are about half of the annual exposure limit of 20 mSv/y compared to natural doses in some areas of the world of up to 50 mSv/y. The mine and milling facilities are designed with the ICRP principles in mind, particularly ALARA. The proposals are closely reviewed by the government regulatory agencies to ensure the principles are adhered to. Regular reviews and inspections occur during construction, operation, and decommissioning to ensure the accepted plans are implemented and that the high standards are maintained in radiation protection. Public input is part of the process.

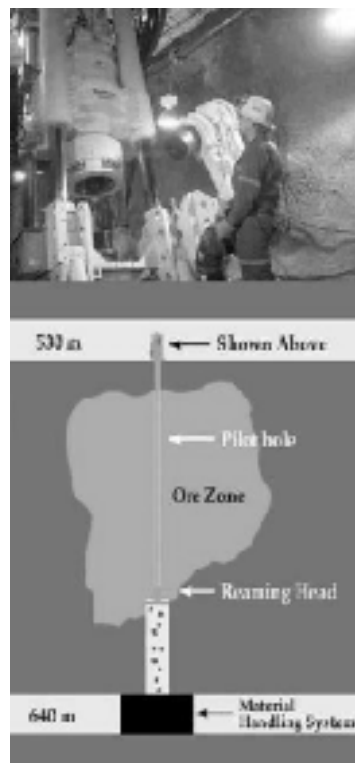
The three main sources of radiation exposure are radioactive dusts that are breathed in, gamma radiation from the ore that irradiates the body, and radon gas that is breathed in. Strict hygiene is practiced in the mines and process areas to prevent ingestion of uranium or uranium oxide. This includes restricted eating areas, washing prior to eating, regular washing of work clothes, and a urine sampling programme to monitor any ingestion.

The dusts are generated by drilling, crushing, and dry conditions where fine particles exist. The drilling is now done wet with the drill cuttings contained by wet scrubbers or some other form of containment to minimise contamination. Crushing is either done wet or with hooding and exhaust ventilation to contain the dust. Fines in areas such as process or roadways are kept to a minimum with prompt cleanup and keeping areas wet. Equipment such as scoop trams, trucks, and surface loaders has enclosed cabs with filtered air systems. Dust masks with the appropriate dust filter are used where necessary. Mines have area and personnel dust samplers to monitor the success of the programmes and where improvements are necessary.

Gamma radiation protection follows the three principles of shielding, time, and distance. The main goal is to keep the worker away from the source of the gamma radiation. This is achieved by doing the access and mining in the waste rock, where possible, particularly in higher grade underground mines. This usually involves using a non-access type of mining method that has the extraction chamber and drilling chambers in the waste rock. Blast hole stoping is a method conducive to this. The ore drilling is done from a drift in waste rock above or to the side of the ore zone to be extracted. The extraction is done at the bottom of the stope through draw points in waste rock such as at Rabbit Lake. The high grade McArthur River property uses this principle as well.

The raise bore mining method is schematically shown on Figure 5.1. The top photo shows the raise bore. The raise bore first drills a pilot hole downwards from a waste rock drift through the ore zone to the extraction chamber in waste rock. The drilling is done wet with water that carries the drill cuttings by pipes to the process. The 3 m diameter reamer head is then attached and pulled back up through the ore zone. The raise cuttings are collected through a chute into a remote controlled scoop tram for transport to the process.

**Figure 5.1. Ore extraction – Raise bore method**



Shielding can also be done in mining by concreting or shotcreting over the gamma source.

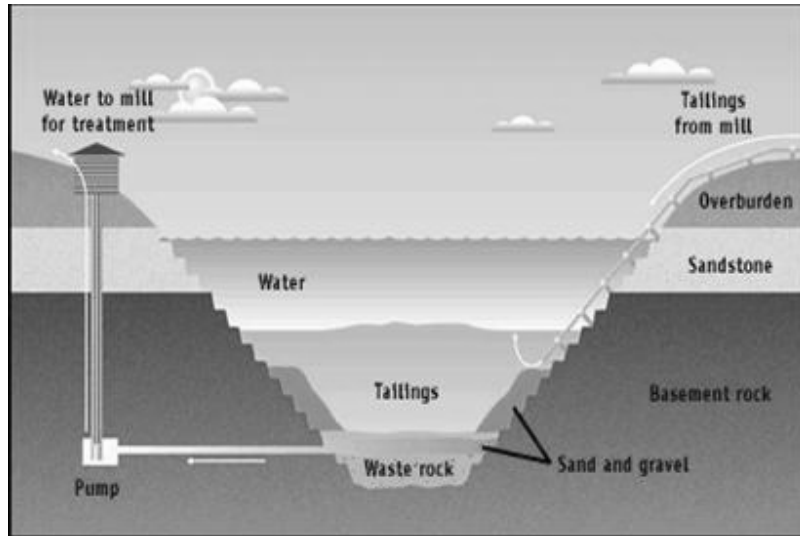
Cluff Lake Mine uses shotcrete in their under hand cut and fill method. The ore is less than 1% and allows an entry mining method. The shotcrete and concreted backfill provide sufficient shielding to mine the ore safely. The tanks and pipes are designed with sufficient shielding thickness of steel and concrete to reduce the gamma radiation to acceptable levels. Access to process and mining areas, particularly during operation, are also controlled to minimise potential exposure times. Direct reading dosimeters are used to help minimise worker exposures, to increase the awareness of the workers to potential radiation sources, and to aid in the expedition of corrective measures.

Radon gas inhalation is a potential hazard as it decays to the solid radon daughter which is significantly alpha radioactive. The radon comes from mine water that is rich with radon directly from the ore or from the uranium ore body. This is usually not a problem in open pits. However, this can be a serious problem in underground mines if the appropriate controls, ventilation, and instrumentation are not effectively in place. Radon gas is controlled by preventing radon-bearing water from entering the underground workplace by designing the openings in dry areas. If this is not feasible, then grouting the cracks in the surrounding rock or applying an impervious layer to contain the water can reduce the radon. Radon-rich water can be dealt with effectively by containment in air tight enclosures or enclosures that are exhaust ventilated to remove the contaminated air to an exhaust pipe or airway. Where there are potential sources of radon in the workplace, single pass fresh air ventilation exhausting directly to a restricted exhaust way keeps the worker in fresh air. It is important to keep mechanical air control devices in fresh air so that repairs can be done in fresh air. In areas that have potential for radon, instruments that can continuously monitor the radon level can alert the workers to any changes so that corrections can be implemented to maintain the safe working conditions. The instrument displays should be located so that all workers in the area, as well as those entering the area, can be aware of the conditions. These methods effectively prevent worker exposures from radon.

Environmental safety issues are basically similar to other mines with an additional factor, radioactive contamination. The waste products are the waste rock, the tailings, and the contaminated water from mining or the process. The waste rock usually has three categories: uranium bearing but uneconomical, acid generating, or barren. The first two require containment to prevent water runoff to the environment. Approved designs for construction, operation and decommissioning are required. Barren waste rock can be stockpiled, or used for fill and roadways. The tailings must be stored in a manner to minimise the potential contamination of the water table as well as the generation of radioactive dusts. Historically, mines have used natural depressions or constructed tailings impoundments. There have been problems with low-level radioactive dust if the impoundments are not covered quickly in decommissioning.

The current thinking is to dispose of the tailings in water-tight surface containments designed to eliminate the pore water pressure problem or submerge them in highly impervious mined out pits constructed to allow the groundwater to flow around the tailings as shown in Figure 5.2.

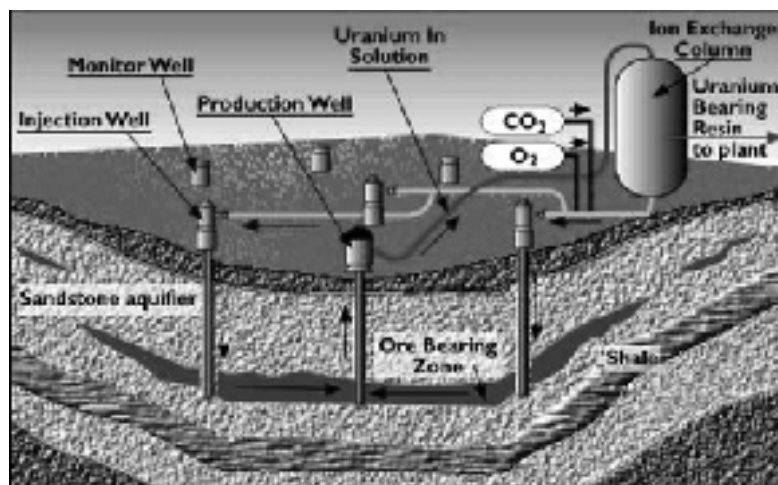
**Figure 5.2. Tailings management facility at Key Lake**  
 (Source: Cameco – Canada)



Contaminated water is water that has been in contact with the ore or has been contaminated in the process. The excess water that cannot be used in the mine or process is treated to meet the local government discharge standards. The contaminated water from a uranium mine also has radium that is removed usually with barium.

*In situ* leach process facilities have monitoring wells around the periphery of the mining area to ensure there is no contamination to the surrounding groundwater (Figure 5.3). After the mining is complete, the mining area is restored to the baseline water quality before decommissioning, which includes the sealing of the wells and removal of all facilities. Revegetation of the area is done as a final step.

**Figure 5.3. Idealised *in situ* leach process in section at Crow Butte site**  
 (Source: Cameco – Canada)



## 2. Uranium refining and conversion

The mining and milling processes produce uranium ore concentrate (UOC), sometimes known as yellowcake, which, chemically, is impure  $U_3O_8$  or uranium diuranate. This has to undergo refinement, in processes described in Section 2-2, to convert it to nuclear-purity uranium hexafluoride, tetrafluoride or uranium metal, which are then processed to uranium oxide, or uranium metal, fuel respectively.

The potential hazards to workers and the public from the refining stages are similar in many ways, but there are two important differences. These are:

- The solvent extraction stage in the mining/milling process removes most of the radioactive daughter products of uranium, present in secular equilibrium with natural uranium. This means that operator and public doses due to short-lived beta radiation are much lower, and it effectively eliminates the impact of the daughter product radon;
- The most significant potential hazard is chemical, not radiological, arising from the handling of bulk quantities of hydrogen fluoride (HF) and the fact that a release of  $UF_6$  (hex) would also generate HF on contact with moisture in the atmosphere.

The early process for refining in the United Kingdom, involving conversion of pitchblende ore, placed emphasis on complete changes of protective clothing, radiation monitoring badges, and frequent medical checks to protect the operators [2]. The substitution of UOC for pitchblende, although significantly reducing the hazard, did not in any way detract from the rigor of radiological protection that has continued, with incorporation of many improvements, to the present day. Some of these are described subsequently.

Emergency plans, for workers, on-site facilities, and for the general public, focus on dealing with the potential chemical hazard. These plans are publicly available documents, available from the refining utilities, and lodged in United Kingdom in places such as public libraries [3].

### 2.1 Radiological protection

The main potential radiological hazards in refining arise from the possibility of inhalation or ingestion of compounds of uranium, leading to internal dose. The potential for external dose is generally low except where the process leads to concentration of daughter products of uranium that have grown back in after the mining/milling stage. Regardless of the magnitude of the dose, however, the United Kingdom operators implement a continuing system of dose reduction, on the ALARP principle, wherever improvements can be identified and made. One of the stages at which such accumulation of daughter products can occur is the casting of uranium billet into crude rods for uranium fuel fabrication. Although not directly a part of the refining process, casting in the United Kingdom is carried out in the same building, and by the same workforce, as the preceding production of uranium from uranium tetrafluoride. For this reason, and to illustrate the general principles of dose reduction and management that are used, it is useful to consider the measures taken at the casting stage. A combination of capital investment in engineered dose reduction improvements and many other solutions stemming from joint management/workforce suggestions and initiatives, had resulted in a lowering of average dose (sum of internal and external) from about 18 mSv/y (1986) to about 7 mSv/y (1990) [4,5]. Since that time, many further workforce and management initiatives have been put in place with the effect that the average dose of the (now) combined uranium production and casting stages has been reduced to about 3 mSv per year [6,7]. Although operator doses at the UOC refining stages have always been generally at the lower end of these ranges (about 1-2 mSv/y in

modern times), the same principles of dose reduction have been, and continue to be, employed wherever practicable. In France, casting is carried out in separate plants but comparable general principles of dose reduction and management are employed in refining plants, where the average individual dose (including internal dose) is about 2 mSv/y.

The main thrust of dose management and reduction at UOC refining stages has, for many years, been in the area of minimisation of internal dose from the inhalation or ingestion of uranic powders such as UOC,  $\text{UO}_3$ ,  $\text{UO}_2$ , and  $\text{UF}_4$ . This is achieved firstly by aiming to reduce contact by personnel with these compounds by engineered methods; then to contain all operations with potential for generating airborne contamination inside enclosures equipped with air extraction and to ensure that operators engaged in such operations wear respiratory protection, which is also mandatory for some other non-enclosed operations. The change from fluidised beds to rotary kilns, for production of  $\text{UF}_4$ , in the late 1970s significantly reduced the degree of operator contact with process materials at this stage and was an important landmark in this respect. This principle of containment, used for many years, and now augmented by incorporation of operator entry lock chambers in which powder can be washed or vacuumed off protective clothing, has proved very successful in reducing operator internal dose uptake. This success is demonstrated by reference to actual operator doses which now average around 1.0 mSv/y (or less, sum of internal and external) against the statutory limit of 50 mSv/y [8] and are summarised in Section 12-1.

All of this is underpinned by a sophisticated system of dose monitoring, recording, and follow up consisting of the following main features [9,10]:

- radiation film badges for operators (these can be augmented by other types of dosimeter as appropriate) to assess external dose;
- static air samplers positioned according to the outcome of comprehensive studies of operations and likely contamination levels (these can be augmented by operator-personal samplers as appropriate) which, combined with operator occupancy, are used to assess internal dose;
- urinalysis;
- whole body monitoring.

Though rare, all cases of unusual dose are rigorously investigated. Finlayson [11] describes how this is done.

All other refining companies, in other countries, use similar procedures in principle.

## 2.2 *Conventional safety*

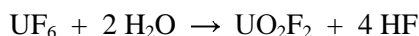
The production of hex has been carried out since the early 1940s in the United States, since 1952 in the United Kingdom and France, and since 1969 in Canada. The design and operation of the facilities in this part of the cycle require that not only are radiological concerns satisfactorily addressed, but also that the main processing chemicals, HF, fluorine, and others, are safely handled. In fact, the primary concern from a possible major emergency viewpoint arises from the handling of these materials, so plans for dealing with such situations are based thereon. These are discussed below.

The CSNI Specialist Meeting [12] on the safety problems associated with the handling and storage of hex, held in Boekelo, concluded that the existing processes for refining UOC and conversion to hex gave rise to no significant radiological hazards, and that the safety problems were essentially those of a conventional chemical industry dealing with toxic chemicals. That conclusion

has stood the test of time and remains true to this day. However, adequate precautions are necessary and are implemented to protect workers from radiation exposure and the inhalation of uranic dust, as described above. This is further exemplified by the need to protect against radiation from flame reactor operations in some countries outside the United Kingdom.

Uranium hexafluoride at ambient temperature (at which it is stored) is a colourless, crystalline solid with a low, but significant, vapour pressure. When heated, for example to transfer it from vessel to vessel, the crystals sublime without melting, giving a vapour pressure of 760 mm Hg at about 56°C. At higher temperatures, melting, accompanied by a substantial increase in specific volume, occurs. It is the handling of hex, in the vapour and liquid states, that represents the greatest potential for high release rates in the event of a failure of containment.

Hex is a very reactive substance. It reacts with water, most organic compounds, and with many metals. However, it does not react with oxygen, nitrogen, or dry air. Its reactivity with moisture in air is what leads to the principal hazard if it is released to atmosphere. Two toxic substances, hydrogen fluoride and uranyl fluoride (UO<sub>2</sub>F<sub>2</sub>), are produced according to the equation:



With gaseous hex, this reaction proceeds rapidly, liberating heat and accompanied by a substantial volume increase at atmospheric pressure. Emergency plans are structured to deal with this situation also.

Hex is stored in cylinders containing a maximum of 12 t, which is the limiting upper quantity that could be released from one cylinder. However, temporary in-process storage of hex in plant condensers could result in accidental release of a larger amount. Also, accidents involving more than one cylinder could occur, for example during transport or storage. At these stages, though, it must be recognised that the hex is in solid form and therefore much less likely to be released than when it is being produced, or transferred, in the liquid or vapour phases. The possibility of fire, raising the temperature sufficiently to vaporise hex, has to be taken into account, as do more remote scenarios such as accidental plane crashes onto a hex facility. These are all addressed in emergency plans that conclude that the main hazard is from chemical toxicity rather than immediate radiological hazard.

Some countries have chosen to reduce the risk associated with the storage of hex by converting depleted UF<sub>6</sub> to UF<sub>4</sub>, a powder which can be stored more safely and at less cost. Decisions of this nature will be made after taking due consideration of potential risks from both situations on the ALARA principle. Depleted means either “pile depleted” that is having been used in a reactor, or “tails depleted”, being the depleted fraction from an enrichment process. In France, both pile and tails depleted UF<sub>6</sub> are converted into U<sub>3</sub>O<sub>8</sub> powder, which is also a very stable form suitable for safe storage.

As far as actual experience is concerned, relatively few accidental releases of substantial quantities of hex have occurred. A review of those that have occurred is presented in Section 12-3 and discussed further in [12,13,14].

The main chemical hazard in the production of hex arises from the use of HF at the UF<sub>4</sub> and UF<sub>6</sub> production stages. Fluorine, though certainly highly toxic and damaging to human tissue, presents less of a hazard since its production can be stopped fairly quickly, for example, in the event of a release, and it is not stored. The use of the highly reactive, and explosive, chlorine trifluoride for hex production ceased many years ago. In the United Kingdom, the risk from HF has been reduced dramatically since the mid 1990s by significantly lowering the onsite inventory in favour of direct

feeding to user plants, and by the introduction of larger supply tankers which allow lower numbers of deliveries and fewer connecting/disconnecting operations. Despite this, the most stringent controls of, and procedures for, handling of this very toxic substance are still employed (and always have been). These are based on selection of correct materials for plant and equipment, containment, workforce protection and education, and regularly rehearsed emergency plans for all stages from local plant to offsite areas.

Other potential large-scale hazards can include, depending on the processes used, magnesium, hydrogen, and nitric acid. Magnesium is stored in a bonded store, with fire and explosion prevention measures, and is subject to similar protection measures during use in the process. Hydrogen inventory is minimised by a “just in time” supply system and its usage takes advantage of all up-to-date safety systems and procedures. Nitric acid is still stored and used in bulk quantities, but the inherent risk associated with it is very much less than that of HF. Nevertheless, its use is still subject to the same standards of care and containment.

Very few fatal, or other truly serious, accidents have occurred in the history of UOC to hex refinement. However, there is an incidence of the kind of accident that can occur on any type of chemical plant – falls, chemical burns, hot and cold burns, accidents due to electricity, release of stored energy, or failure to wear protective clothing, as well as many other types. Refining operators have always paid particular attention to this type of accident but, toward the end of the 1980s, the United Kingdom operator felt that its standards in this area did not match those for radiological protection. Accordingly, it embarked on a programme aimed at reducing the incidence of this type of accident. There were many reasons why this was necessary, and these, together with the results up to the mid-1990s, have been described in [15]. The rate of accidents that resulted in a person having to lose more than three consecutive days work had, by then, been more than halved. In the United Kingdom, this is a legally reportable measure of accident prevention performance. Since that time, the programme has moved forward based on systems for improvement of safety culture and personal safe behaviour to the extent that this operator now ranks with the best chemical facility operator in the world ([8] and Chapter 12).

### **2.3 *Emergency plans***

As stated above, these centre on the need to respond to toxic chemical releases though, on multifunctional sites such as the United Kingdom one, there is also an element related to action in the event of an uncontrolled nuclear reaction (a criticality incident) [16,17]. In the United Kingdom, as in other countries, the emergency plan is a formal requirement of the licence to operate [18]. It covers local plant on-site and off-site emergency scenarios and a key feature is the requirement to demonstrate to the regulator the effectiveness of the plan by means of scheduled exercises at all levels.

More recent legislation, although aimed primarily at conventional chemical plants, places additional commitments on nuclear chemical plants [19]. In addition, application of the new Radiation Emergency Preparedness and Public Information Regulations 2000, has resulted in a slight extension of the emergency planning distance for sheltering from radiation as part of the overall response [20].

### **2.4 *Safety management and assessment***

The United Kingdom refining operator implements a structured and comprehensive safety management and assessment system which covers requirements for existing and future plants, and for plants that have been shut down for decommissioning. It is based on a process of categorisation of a



project's safety significance followed by a rigorous assessment of safety implications and needs, the depth of which increases according to the safety significance. All projects, from very minor modifications to existing processes, up to the most major new projects, are covered by the system. All internationally recognised techniques for this kind of work are used and assessments are subject to audit and independent peer review. Full details of the system in use up to the mid-1990s were presented in [21]. Since that time, this operator has introduced a number of further improvements to the way in which it prepares its safety cases. The most significant of these is the Continued Operation Safety Report (COSR) for existing plants. The purpose of the COSR is to summarise the safety arguments and to highlight the main systems (engineering and procedural) that ensure that a plant (or other facility) remains safe. It does this in a simpler format, with a greater emphasis on continued safety of plant engineering functions, than did its predecessors [22].

## **2.5 Waste management and environmental monitoring**

The wet process for refining UOC includes a solvent extraction stage in which remaining impurities in the UOC are separated from the uranium. The radioactive component of these impurities consists of alpha active  $^{230}\text{Th}$  and  $^{232}\text{Th}$  isotopes, with beta active  $^{234}\text{Th}$  and  $^{234}\text{Pa}$  arising in an aqueous waste stream known as raffinate. It is accepted practice to dispose of this raffinate to the tidal waters of a river estuary because of its very low environmental impact. In the United Kingdom, this has been carried out for many years under the terms of an authorisation granted by the relevant regulatory authority, currently the Environment Agency [23,24]. The operator carries out a wide-ranging programme of environmental monitoring (which is a combination of a Statutory Environmental Monitoring Programme and additional voluntary work) in order to quantitatively determine the impact of this and other disposals and publishes the results of this programme annually [24]. This shows that the radiation dose received, predominantly from refining operations on this site, to the very small numbers of persons most exposed, is of the order of 10 mSv/y. To other people, it is very much less than this. These results are summarised in Sections 12-1 and 12-2. Their significance can be gauged from the fact that average exposure to the sum of natural radiation, and radiation for medical purposes, in the United Kingdom is about 2 600  $\mu\text{Sv}$  per person, with much higher figures than this in some parts of the country [25].

Discharges of airborne radioactivity, disposals of solid waste, and direct radiation are all also measured and their effects estimated. They result in a lower impact than that of liquid disposals.

A number of independent organisations also monitor the impact of these disposals and predominant amongst these is the regulatory authority, the Environment Agency [26]. Although not all of the authority's determinations of radioactivity levels agree with those of the operator, the majority does, and the differences can, in most cases, be explained by decay of short-lived beta activity or different methods of analysis. They are not significant in terms of impact and there is general agreement that arrangements in place to monitor radioactivity levels are effective.

The UOC used for refining also contains a range of non-radioactive impurities which are separated and discharged in the raffinate along with the radioactive component. Details of these discharges are published together with those of the radioactive species [24].

During 1993, the United Kingdom operator, at the request of the regulatory authority, carried out a Best Practicable Environmental Option (BPEO) study on its liquid radioactive disposals [27]. A BPEO study considers all parts of the environment so that an assessment of impact on the environment as a whole, rather than just one part of it, can be made. A large number of possible options for treatment or discharge of the liquid waste stream were considered and assessed and the outcome was

that the only change that should be made was to set out to select for refinement those UOCs with relatively low thorium content. Since then, this has been put into practice with the effect of reducing  $^{230}\text{Th}$  and  $^{232}\text{Th}$  discharges to the nearby tidal river. A review of this study is expected to conclude that this practice should continue.

### 3. Uranium enrichment

Most nuclear power plants use enriched uranium to improve reactor power density, increase operating time between refuelling, reduce spent fuel quantities, and allow the use of light (normal) water for the moderator and coolant. Enrichment uses physicochemical methods to increase the concentration of the fissile uranium isotope ( $^{235}\text{U}$ ) from its naturally occurring level of about 0.71% to the 3-5% range for power reactor use. A DU stream containing less than natural assay uranium (typically 0.25-0.4%  $^{235}\text{U}$ , depending mainly on economic conditions) is also generated as a by-product and is sometimes referred to as uranium tails. The enrichment plants are large facilities because of the small differences in physicochemical properties between  $^{235}\text{U}$  and the other uranium isotopes (primarily  $^{238}\text{U}$ ) that require many unitary enrichment stages. A listing of the large-scale, operating enrichment facilities is given in Appendix 1. These are based on GD and GC technologies. Advanced GC and laser-based processes are also being investigated in active R&D programmes.

#### 3.1 Separative work unit (SWU) – A metric for enrichment plants

The concept of separative work is used to understand and compare the various enrichment processes. Separative work is a quantitative measure of the amount of isotope separation obtained by a separator. A separator is a process or device which accomplishes the enrichment of the  $^{235}\text{U}$  uranium isotope. For uranium enrichment, separative work is usually expressed in units of kg SWU or simply SWU. A given amount of separative work can significantly enrich a small amount of uranium or slightly enrich a greater amount of uranium. Separative work is proportional to the amount of uranium processed. Separative work increases with the degree of enrichment in  $^{235}\text{U}$  in the enriched product (i.e. more work is required to recover more  $^{235}\text{U}$  from the same amount of natural uranium) or the depletion of the isotope in the DU tails.

Separative work is defined by the following equation:

$$\text{SWU} = [P \cdot V(x_p) + W \cdot V(x_w) - F \cdot V(x_f)] \cdot T$$

where:

- T = time (usually 1 year);
- P, W, F are product, tails, and feed flow, in kg;
- V(xi) = separation potentials.

The separation potentials are sometimes called value functions, and are defined as:

$$V(x_i) = (2 \cdot x_i - 1) \cdot \ln[x_i / (1 - x_i)]$$

where  $x_i = x_f, x_p$ , or  $x_w$  represent assay levels (enrichment) in the feed, product, and tails streams respectively.

The flow rates can be calculated from:

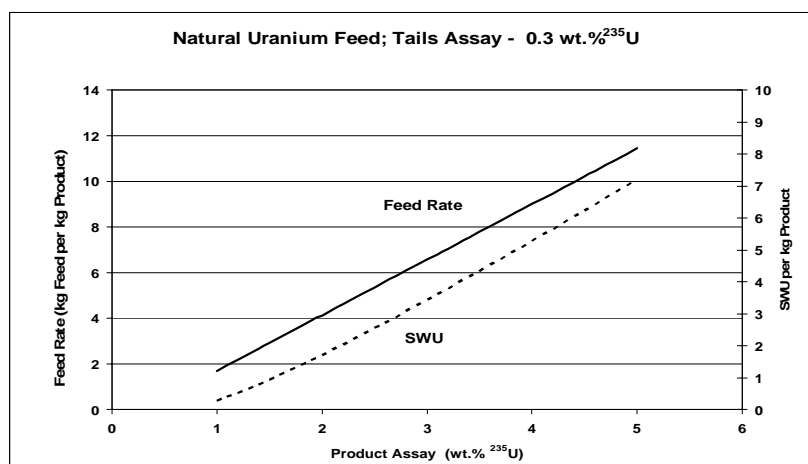
$$[F/P] = (x_p - x_w)/(x_f - x_w)$$

$$F = P + W$$

For uranium enrichment, a feed material is introduced to the separator and the resulting product is produced with a higher enrichment while the generated depleted stream is produced with a lower enrichment. The uranium enrichment process usually utilises multiple separators arranged in stages to achieve useful amounts of product enrichment. Stages are arranged in series to increase enrichment and in parallel to increase throughput. The assembly is termed a cascade. The assay of the product is the percentage of uranium atoms that are  $^{235}\text{U}$ . The process of enrichment increases the assay while the process of depletion decreases the assay. Through a succession of stages, the enrichment of  $^{235}\text{U}$  is increased until the assay of  $^{235}\text{U}$  reaches the desired level for the particular application. For most commercial applications, the  $^{235}\text{U}$  is enriched to something less than 10%  $^{235}\text{U}$  (3-5% is typical for nuclear power reactors) and is termed low-enriched uranium (LEU).

Figure 5.4 provides a curve for the feed rate (kilograms of feed for each kilogram of enriched product) and a curve for the SWU factor (SWU per kilogram enriched) as functions of the product enrichment level, for a 0.3% tails assay. Typically, for LEU with about a 3% assay, around 5.5 kg of feed and 4.3 SWU are required per kilogram of product, with a tails assay of 0.2%. With a tails assay of 0.3%, 6.6 kg of feed are needed, with only 3.4 SWU. In contrast, increasing the assay to around 4.5% can require about 10.2 kg of feed and 6.2 SWU per kilogram of product, at a tails assay of 0.3%.

**Figure 5.4. Feed rate and SWU relationships**



A large commercial nuclear power plant requires 30-35 t of uranium per refuelling outage. Plants on annual cycles use LEU around 3% assay, and plants on longer cycles (18 months) use 4-4.5% assay LEU. These parameters translate into approximately 100 000 to 120 000 SWU per reactor year.

In contrast, some research reactors use HEU fuel. Around 200 SWU are needed to produce 1 kg of 90% HEU (i.e. containing 90% of the  $^{235}\text{U}$  isotope) from 200 kg of natural uranium. However, HEU is a small market (most research reactors have been modified to work with 20% assay uranium, for proliferation reasons) and most enrichment facilities are designed for producing LEU for nuclear power production and cannot produce HEU.

Economical and reliable operation of enrichment facilities depends upon balancing facility capital costs (number of stages), feed quantities (tails assays), and operating costs (power and equipment maintenance).

Separative work depends only on the end states and is independent of the type of enrichment process. The separation factor is the ratio of the amount of enrichment achieved to the amount of depletion achieved, and is usually based upon one stage or separator. The separation factor is sometimes referred to as alpha and is intrinsic to a specific process. For processes utilised commercially, the separation factor is approximately independent of the feed assay. Ideally, a separator for an enrichment process would have a high separation factor, a high throughput, and a relatively high ratio of product rate to feed rate.

### 3.2 Enrichment technologies

A number of different enrichment technologies have been proposed to achieve <sup>235</sup>U enrichment. The principal technologies include GD, GC, laser isotope separation, and electromagnetic isotope separation. Table 5.1 provides a list of uranium enrichment methods and their parameters that have undergone significant testing and/or use. Of these principal technologies, uranium enrichment by the GD and GC technologies have proven to be the most commercially viable alternatives and have been implemented at the plant scale. Enrichment by GC methods is becoming more prevalent.

#### 3.2.1 The gaseous diffusion enrichment process

The GD processes for uranium were first developed in the 1940s. The first successful, large-scale facility was constructed and operated in Oak Ridge, Tennessee, in the United States. Since that time, several plants have been built worldwide. Gaseous diffusion plants (GDPs) have a large floor area under roof; the GDPs in the United States have over 160 ha (400 acres) within the process buildings. In the past decade, several GDPs have been placed into standby operation because of an overabundance of enrichment supply, due in part to the success of the GC technology (discussed later) and the downblending of weapons HEU to reactor-grade LEU.

**Table 5.1. Overview of uranium enrichment methods**

Method	Working medium or fluid	Approximate number of stages to 5% LEU	Operating mode	Potential hazards with respect to GDPs
Thermal other physical	UF <sub>6</sub>	>1 000	Continuous	Higher
Electromagnetic calutrons	UCl <sub>4</sub> , U	2-10	Batch	Higher
Gaseous diffusion (GDP)	UF <sub>6</sub>	700	Continuous	[Baseline for comparison]
Gas centrifuge	UF <sub>6</sub>	50-100	Continuous	Slightly lower
AVLIS	U-Fe Metal Alloy	1-5	Batch <sup>1</sup>	Higher
SILEX	UF <sub>6</sub>	1-5 – under development <sup>2</sup>	Batch <sup>1</sup>	Slightly lower

1. The AVLIS and SILEX process were developed using a batch operation, however, they have been demonstrated to operate in the continuous mode on a pilot level. Additional work will be required before they can be routinely operated in the continuous mode.

2. Currently, USEC is doing tests at SILEX to determine the separation factor for the process which will allow the calculation of the number of stages required; it is too early to comment on the number of stages required.

The GD process depends on the separation effect from the molecular effusion of gas molecules through a membrane. The gas utilised is uranium hexafluoride. On average, lighter gas molecules travel and diffuse faster than heavier gas molecules. Therefore, the flow through a membrane of the uranium hexafluoride with a  $^{235}\text{U}$  atom travels slightly faster than the uranium hexafluoride gas with a  $^{238}\text{U}$  atom. As such, the  $^{235}\text{U}$  uranium hexafluoride molecules strike the separator barrier more often, and if the barrier is properly engineered, thereby diffuse through the barrier to produce a uranium hexafluoride gas downstream of the barrier which is slightly enriched in  $^{235}\text{U}$  uranium hexafluoride. The separation factor is about 1.004 per stage. This is a small value and many successive stages are required to achieve useful enrichment of  $^{235}\text{U}$ . For power reactor grade LEU, between 500 and 700 stages in series are needed.

The main components of a GD stage include a large cylindrical vessel that is mounted either horizontally or vertically, which contains the barrier. A compressor compresses the  $\text{UF}_6$  gas to a pressure sufficient to create flow through the barrier. The compressors are typically driven by electric motors and the  $\text{UF}_6$  gas, once it passes through the barrier, is passed through a heat exchanger to remove the heat of compression prior to being introduced and compressed in the subsequent stage. The compressors require seals to prevent outleakage of  $\text{UF}_6$  and inleakage of air. Outleakage would result in contamination of the facility and would pose a hazard both chemically and radiologically to the operating personnel. Inleakage of air must be avoided to preclude the introduction of moisture into the process since the moisture interacting with the  $\text{UF}_6$  would produce toxic and corrosive acids. In addition, the introduction of moisture could compromise the safety envelope used to preclude criticality since most diffusion systems rely on moisture exclusion as one of the multiple barriers applied to preclude criticality.

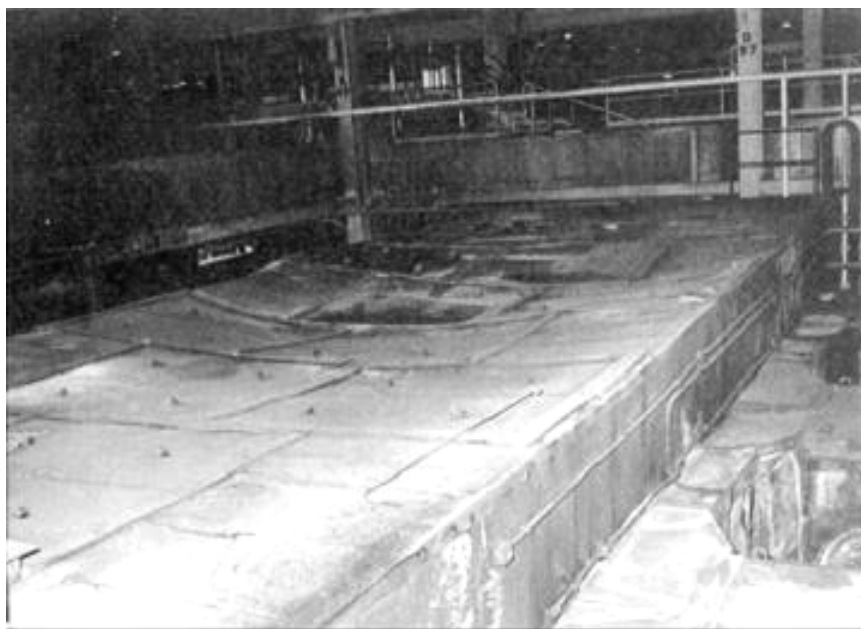
In addition to the major operating systems, GDPs require a mechanism to feed the  $\text{UF}_6$  and withdraw the depleted and enriched  $\text{UF}_6$ , as well as a means to maintain the desired operating temperatures of the facility. The temperature is maintained by large reservoirs of cooling water which are used to dissipate the waste process heat. The electrical load requirement for GDP facilities is significant.

Diffusion plants are designed for economic reasons to be run continuously for periods of several years at one time. Reliability of process equipment and components is high. From a safety standpoint, too, maintenance should be minimised in all areas of the plant where  $\text{UF}_6$  is present and particularly in high-enrichment areas where radiation doses could be significant. Bearing in mind that a diffusion plant comprises both pressure systems and vacuum systems, driven by massive pumps, it is not surprising that the most common problems associated with operation of diffusion plants involve vibrations leading to pump and valve leakages and possibly even failure. Vibration could lead to blade failure, and missiles could sever  $\text{UF}_6$  lines and lead to a release into the atmosphere. Figure 5.5 shows the effect of a fire initiated by a metal- $\text{UF}_6$  incident at the Portsmouth plant in the United States (9 December 1998); safety systems activated and plant personnel responded appropriately and contained the incident (summary report is given in Section 12-3). Such a release is by far the greatest threat to operating personnel from enrichment plants. The feed and withdrawal systems contain liquefied  $\text{UF}_6$  and represent the greatest potential for offsite impact. However, with many years of operational experience to draw on, potential releases are now much less frequent. Modularisation of diffusion plants can ease on-line maintenance on one isolated module at a time.

Operational experience with diffusion plants has been good. The facility operations have generally been reliable and have performed well with low levels of  $\text{UF}_6$  release during normal operations. Since the overall hazard associated with enrichment plants is the toxicological effects of  $\text{UF}_6$ , it is of paramount importance that any diffusion plant to have in place a comprehensive leak detection and alarm system. The EURODIF plant in Tricastin, France, and the USEC plant in

Paducah, Kentucky, United States, all have systems for detecting and containing accidental  $\text{UF}_6$  releases.

**Figure 5.5. Portsmouth cell housing after fire**

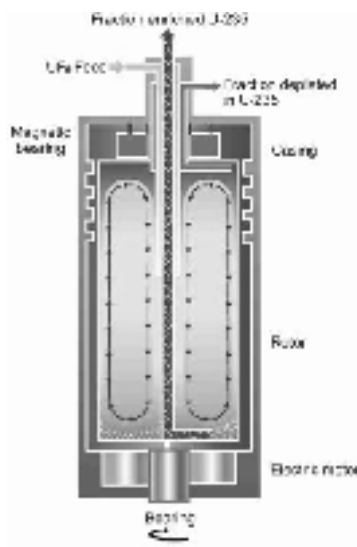


### 3.2.2 Gas centrifuge enrichment process

The use of centrifugal methods for isotope separation was first discussed in the early 1900s. Uranium enrichment by centrifuges was first discussed in the early 1940s. Working centrifuges of about 1 SWU/machine were developed and tested in the mid-1940s but were overshadowed by the large-scale success of GD, easier to implement in a short time. Experimental work continued on centrifugal methods leading to successful implementation on a large scale in the 1970s.

The gaseous centrifuge (GC) process utilises gaseous  $\text{UF}_6$  which is fed into a cylindrical rotor that is spinning at a high rate. The spinning cylinder can be operated at peripheral speeds of several hundred meters per second. The cylinder is evacuated and maintained at low pressures. Figure 5.6 provides a diagram of a gaseous centrifuge machine. The gaseous  $\text{UF}_6$  is introduced at the centre of the cylinder. Centrifugal force causes the heavier  $^{238}\text{UF}_6$  molecule to move closer to the spinning cylinder wall than the lighter  $^{235}\text{UF}_6$  molecule, thus tending to separate the two uranium isotopes. Enriched  $\text{UF}_6$  and depleted  $\text{UF}_6$  are removed near the ends by special scoops and baffles. Connections are usually made coaxially. Separative efficiency is greatly enhanced by increasing the diameter and length of the cylinder, the peripheral speed of the centrifuge rotor, and by counter current flow induced by internal baffles and thermal gradients. However, these changes also increase the potential hazards and consequences of failures. The tensile strength of the material of construction and resonant vibration frequencies physically limit the size and SWU per machine. Typical machines are 25-30 cm (10-12 ") diameter and 4-6 m (15-20 ft) high. Thus, typical rotational speeds of 20 000 to 25 000 rpm translate into edge velocities of 300-400 m/s (1 000-1 300 ft/s). In the United States, larger machines were used in pilot programmes during the 1970s and early 1980s, and these designs may form the basis for a revised testing and pilot programme.

**Figure 5.6. Diagram of a gaseous centrifuge**



Several large facilities with individual capacities in the MSWU range now exist in Europe and in Japan. Operational experience with centrifuge plants has been good. Plants have generally been very reliable and performed well with low levels of  $UF_6$  release during normal operations. There have been very few and only minor mechanical failures. Furthermore, the electrical consumption of a GC facility is much less (typically more than 20 times) than that of a GDP which generally translates into lower operating costs.

A failure of a GC unit operating at the high operating speed of a GC would generate shrapnel as a result of the destruction of the rotor and other spinning components. Prevention of such a failure is minimised by good design, materials of construction, and assembly of the unit. Research into new constructional materials continues in order to develop safer and higher performance centrifuges. Management of these material stresses is at the heart of the safety of centrifuge plants. Figure 5.7 shows a portion of the many machines in a cascade. A 1-1.5 MSWU/y GC facility might have up to 42 000 machines. At an annual failure rate of 1%, there would be about one machine failure per day. Thus, reliability and failure mitigation are very important, and the facility must accommodate GC machine failure while maintaining safety.

**Figure 5.7. Cascade of centrifuges**



Gas centrifuge enrichment plants pose hazards associated with handling UF<sub>6</sub> and enriched uranium, as well as hazards that are also found in other industries – particularly those posed by the use of high-speed rotating equipment. The plants are designed to be run continuously for periods in excess of ten years, and reliability and ease of maintenance are of paramount importance both for safety and economic reasons.

### 3.2.3 Chemical exchange processes

Different isotopes affect chemical reaction kinetics. In general, the compound containing the heavier isotope has a slower reaction rate than the lighter isotope. Isotopes of heavier elements also experience lesser relative differences in the reaction rates, resulting in lower separation factors and more cascade-like steps as compared to isotopes of lighter elements. However, this kinetic isotope effect can be used to enrich a heavy element such as uranium. Separation factors are roughly comparable to those of GD and many stages would be required, but this could be balanced by the potential for simpler operating conditions and equipment that exists.

Enrichment by ion exchange is a chemical process which has been studied, and a pilot plant has been successfully tested in Japan, but has not been followed by industrial development. This process does not have any of the associated hazards of high-speed rotating equipment and pressure/vacuum systems. It does not utilise UF<sub>6</sub> and so none of its hazards are present. The criticality problems of handling enriched uranium do apply, however, and these are covered by concentration limits.

The choice of ion exchange medium must also be made carefully, bearing in mind the reagents used in the columns. In particular, the use of nitrates and nitric acid as regeneration agents for the ion exchange system must be carefully monitored to avoid oxidation reactions with organic ion exchange materials. Rapid oxidation can produce explosions and several have occurred due to these ion-exchange resins/nitrate/nitric acid reactions in other installations.

### 3.2.4 Laser enrichment

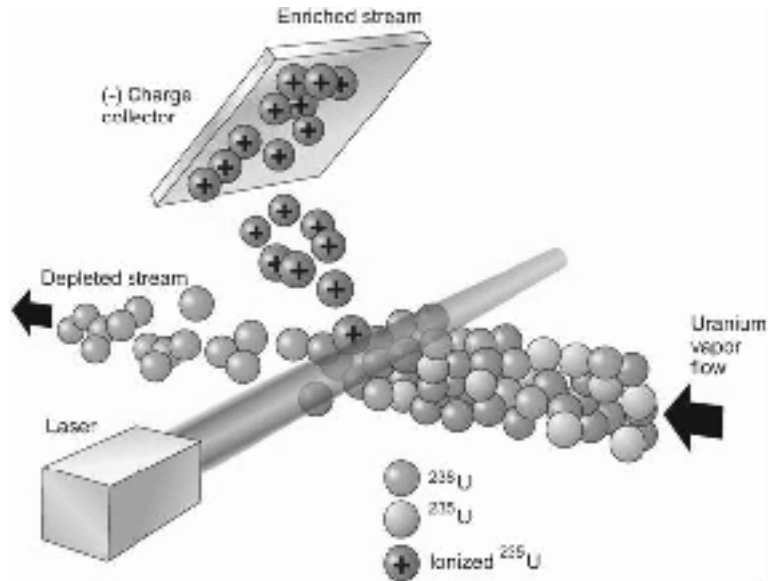
There are two principal methods of enriching uranium by means of lasers – the atomic route and the molecular route. The atomic route is generally designated as AVLIS. In this route, the working material is uranium metal, usually as an alloy. Uranium metal vapour, at around 3 000°C, is produced by electron-beam heating of a platen in a vacuum chamber. The hot vapour migrates away from the platen. A multi-step photo-ionisation process is performed on the uranium vapour with tunable lasers, resulting in the selective excitation and ionisation of <sup>235</sup>U. The resultant ions are separated and extracted electromagnetically (Figures 5.8 and 5.9). Experimental and pilot testing of AVLIS has demonstrated that reactor level enrichments are achievable in one or two stages, and licensing of a large facility has been considered in the United States. However, significant engineering is needed to improve the reliability of key components and demonstrate the economics of the overall process, including the conversion plants required to integrate an AVLIS plant with existing fuel cycle facilities. Thus, active licensing and larger demonstrations or plants are not currently planned. The research and development of AVLIS has been carried out in the 1980s to the early 2000s in Japan.

In the molecular route, the laser is used to selectively excite one isotopic form of a molecular compound of uranium. Most methods have focused on gaseous UF<sub>6</sub>. The compound is used in its vapour state and requires a very powerful laser with an energy flux of some 100 MJ/cm<sup>2</sup>. The gaseous UF<sub>6</sub> has to be cooled to less than 80 K, usually by expansion through a nozzle when mixed with a carrier gas, such as nitrogen. Several processes were under consideration in different countries. In

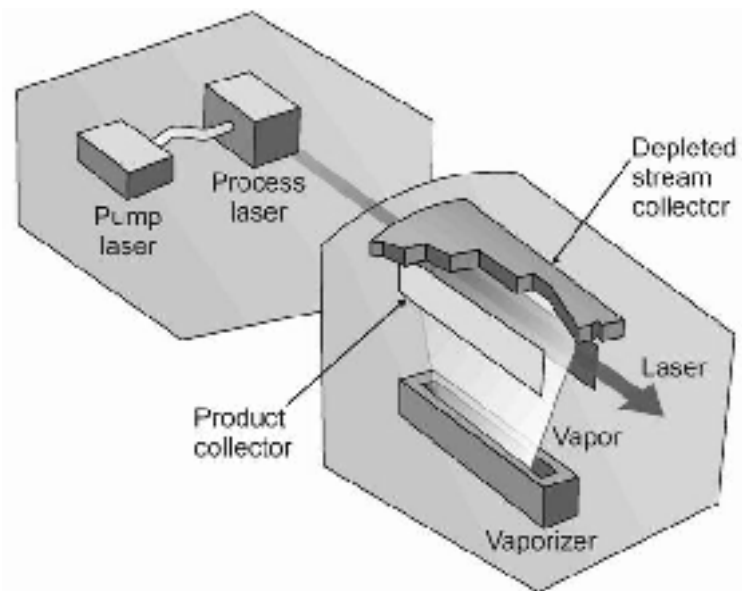


Japan, Molecular Laser Separation Test programme started in 1988 and continued until 1998. Elemental technologies which include power laser have been developed during this period and there still exist research and development items. At the present time, the most active programme involves the SILEX process in Australia. This uses  $UF_6$  and demonstration testing is planned.

**Figure 5.8. The principle of the AVLIS process**



**Figure 5.9. The AVLIS system**



Conventional hazards associated with the use of high-power lasers (i.e. high voltages and high-power laser beams) are common to both methods. In addition, the possible toxicity of laser dyes and potential fire/explosion hazards associated with high-pressure dye/solvent circuits are taken into account during safety assessments. Containment of high-temperature corrosive uranium vapour poses some metallurgical problems for the vacuum chambers. Uranium accumulation within the vacuum system may require periodic cleanout and decontamination. The vessel will have to be designed to withstand or prevent any reactions and over-pressurisation resulting from contact of hot, liquid uranium with cooling water. An additional problem with the atomic route is the removal of the  $^{235}\text{U}$  metal from the collector plates. Finally, AVLIS routes present the potential for dense uranium phases and will require more attention to criticality controls than the GDP and GC processes. Hazards associated with molecular routes include the combination of the corrosive nature of  $\text{UF}_6$  and thermo-mechanical constraints which make it difficult to find suitable materials for this route. In addition, heat exchangers and separation/recycle of carrier gases will be necessary.

### 3.3 *Uranium hexafluoride hazards*

The only uranium compound that is suitable for diffusion, centrifuge, or laser enrichment (by the molecular route), and which is gaseous at reasonably low temperatures (40 to 60°C), is uranium hexafluoride,  $\text{UF}_6$ . This substance has the additional advantage that fluorine is mono-isotopic. However, uranium hexafluoride  $\text{UF}_6$  or “hex” is a powerful fluorinating agent. It reacts with almost all metals to form metal fluorides. It reacts, often explosively, with organic material to form fluorinated compounds and hydrogen fluoride. With water or almost any level of water vapour, it forms highly corrosive and toxic hydrogen fluoride gas, together with a number of solid and toxic compounds of uranium, oxygen, fluorine, and water of hydration.

Containment of  $\text{UF}_6$  is of prime concern, from the point of view of safety, at all stages of the enrichment process. Typical containment materials, such as stainless steel and copper/nickel alloys, retain their long-term integrity by virtue of the immediate formation of a passive, impervious, protective fluoride layer when first contacted by hexafluoride. This layer prevents further corrosion and thinning of the containment.

Processing facilities are designed to a high degree of leak-tightness, particularly where hexafluoride is under reduced pressure. This prevents the ingress of moisture and the formation of solid  $\text{UO}_2\text{F}_2$  and related compounds which could accumulate and cause blockages. Deposits can also form and require additional monitoring and controls to address criticality safety.

It is those two requirements, i.e. to contain  $\text{UF}_6$  within a plant and to keep air and moisture out, which tend to dominate most of the tasks performed in enrichment plants.

$\text{UF}_6$  is supplied to the plant in large containers, typically 12 tHM. At room temperatures  $\text{UF}_6$  is solid, and the containers are warmed to generate  $\text{UF}_6$  as a gas. Overfilling  $\text{UF}_6$  storage containers can lead to rupture due to expansion of  $\text{UF}_6$  solid on melting. This has resulted in at least one incident at enrichment plants. At the end of the process the depleted hexafluoride, or tails, is recondensed into containers which may be stored in the open. Having little commercial value as long as FBR are not widely used, stocks of depleted  $\text{UF}_6$  have substantially increased over the years, and they can present a hazardous inventory to local populations in the event of a failure of containment accompanied by a source of heat, e.g. an accidental aircraft crash. Storage of the tails as a DU oxide would appear to be more acceptable, longer-term alternative, although there are hazards associated with the conversion. Some countries have chosen this route.

The primary hazard associated with enrichment plants is the toxicological effects of  $UF_6$  should it get into the environment. Therefore, of paramount importance in such plants is a comprehensive leak detection and alarm system.

### 3.4 Nuclear/radiation hazards

#### 3.4.1 Criticality

Nuclear criticality is always a concern when working with enriched uranium. Criticality is not a major concern in centrifuges themselves because the nuclear inventory in the machines and process piping is insufficient to form a critical mass under all credible scenarios. Criticality is controlled in other areas of the centrifuge plant, and in other enrichment processes, through the use of geometrically safe equipment, control of moderation (or a combination of the two), as well as adherence to administrative control of the process. In view of the size of equipment used in diffusion plants, criticality remains a potential problem. In other enrichment processes, the criticality problems can generally be resolved by engineered means. Criticality assessments, based on an assumed maximum level of enrichment, could be invalidated by an accidental over-enrichment in the process. Structured fault identification techniques such as HAZOP could highlight areas where this is a potential problem.

#### 3.4.2 Recycled uranium

Until recently most enrichment was carried out only on uranium supplies that have not been irradiated in a reactor. However, in some nations, including the United States and France both to a limited extent, progress has been made within existing nuclear fuel cycles to make use of recycled uranium recovered from the reprocessing of spent nuclear fuel.

Recycled uranium differs significantly from natural uranium in five respects:

- $^{232}U$ , which does not occur naturally, is present in recycled uranium. It has a greater specific activity than  $^{235}U$ , and its decay chain includes a 1.9 y half-life  $^{228}Th$  alpha-emitter and a number of hard gamma emitters, notably  $^{208}Tl$ . The gamma activity increases with time, reaching a secular equilibrium much greater than natural uranium in about ten years.
- $^{234}U$ , a naturally occurring alpha emitter, is also enriched in the fuel before irradiation. It is only partly burnt-out during irradiation. As a result, recycled uranium has a higher alpha activity than natural uranium.
- $^{236}U$ , a neutron poison, accumulates. Therefore, a slight increase in the enrichment level is needed in order to compensate for the poison effect.
- Traces of plutonium and neptunium will also contribute to the alpha activity of recycled uranium.
- Traces of fission products (primarily  $^{99}Tc$  but others are possible, such as  $^{106}Ru$ ) may contribute to a higher gamma background.

Thus, recycled uranium adds these radiation dose and contamination concerns. The traces of plutonium, neptunium, and fission products can be substantially eliminated by efficient separations at the reprocessing plants. The presence of  $^{232}U$  should encourage utilities to recycle material quickly, but economic factors up to now have resulted in little recycling of uranium. Most of the present stockpiles from reprocessing are already at or near equilibrium gamma dose levels.

GC process is better suited than GDP process to enrich recycled uranium, because of the much smaller dimensions of the equipment and consequent smaller hold-up: changes of feed are easier and there is much less contaminated equipment to decommission later on.

#### **4. Fuel fabrication**

For nearly five now decades the fabrication of uranium fuel elements for GCR (gas-cooled reactors) and Magnox-reactors, AGR (advanced gas-cooled reactors), LWR and HWR has been an industrial enterprise [28,29]. More than 8 800 tHM uranium oxide fuel elements are manufactured each year in OECD countries (more than 7 500 tHM for LWR and more than 1 100 tHM for CANDU). Also the fabrication of plutonium-containing MOX fuel elements for FBR and thermal reactors has reached industrial maturity on a modest scale of throughput. Very few accidents involving significant radiological hazards and substantial release of radioactive material to the environment have been reported from the nuclear fuel fabrication industry. In addition, no criticality accident has occurred in facilities linked to the commercial generation of electricity. The reasons for this very good safety record are the well-defined processing and handling operations and associated safety procedures, and the chemically stable solid state of most of the radioactive materials involved. The main exceptions to the latter are  $UF_6$  conversion and wet scrap recovery phases which require liquid processing. Potential  $UF_6$  release is also of concern in fuel fabrication plants.

MOX fuels are used for both fast reactors and thermal reactors. Due to the fact that the implementation of FBR in many countries has been postponed or even abandoned, a growing interest in recycling plutonium in LWRs can be observed in some countries. Since most of the plutonium available for thermal recycle in the future will be recovered from LWR spent fuel with increasing burn-up, the design of new planned MOX fuel fabrication facilities has to be based on typical LWR-plutonium isotopic compositions and  $PuO_2$  contents of 3 to 8% in the MOX fuel for LWR.

For the conversion of  $UF_6$  to uranium metal or  $UO_2$ , wet and dry conversion processes are used on a large industrial scale. The subsequent processes of powder production, pellet pressing and sintering, rod fabrication and assembling of fuel elements are fully developed. Completed fuel assemblies are stored at the fabrication plant before being transported to the reactor. This storage is regarded as part of the fabrication plant.

The conversion of plutonium nitrate solution to  $PuO_2$  is part of the recovery of plutonium at the reprocessing plants in most cases. Some MOX fuel fabrication plants, however, may also be equipped with facilities for plutonium nitrate conversion and for dissolution and purification of plutonium-containing scrap material. Several chemical conversion processes for plutonium nitrate solution or co-conversion processes for uranium-plutonium solutions are industrial standard. The following steps of MOX-powder and pellet fabrication and the manufacturing of MOX fuel assemblies for FBR and LWR are technically very similar to the fabrication of  $UO_2$  fuel assemblies.

##### **4.1 *Enriched uranium hexafluoride conversion to $UO_2$***

Due to the low radio toxicity of slightly enriched uranium, only limited off-site environmental consequences are to be expected following accidents. However, as in the case of the enrichment process, the safety significance of accidental releases may require a more careful assessment when recycled uranium arising from spent fuel reprocessing comes to be used on a larger scale. In general the special and inherent design safety features and carefully planned operational procedures reduce, to

very low levels, the probability of significant accidents due to plant operation. Both these and external events are considered here.

In the design phase of fuel fabrication plants internal safety hazards such as fire, explosion, release of UF<sub>6</sub> and criticality have to be considered. Adequate precautions are incorporated in the design to reduce the probabilities of these events to an acceptable level. Reliable operation is of equal importance to the safe design of the facility because operational experience clearly indicates that non-routine operations and human error are main contributors to incidents.

Various kinds of external events could have significant consequences for both the plant itself and the environment, and these too have to be considered in the safety assessment. Flooding the facility could be the cause of a criticality excursion, therefore the plants are usually built in a very low flood probability zone. However, if the absence of flooding cannot be ensured design precautions should be taken to prevent the plant from becoming critical in the flooded state. The main hazard in case of an external event such as earthquake or accidental plane crash arises from the possible release of uranium hexafluoride.

Specific safety studies have been performed and safety criteria have been established, in addition to the safety analysis, within the licensing procedure for fuel fabrication plants in some countries [30,31,32,33].

The following possibilities for in-plant-accidents are considered to be relevant:

- release of UF<sub>6</sub> caused by a pipe or valve failure or rupture of a heated UF<sub>6</sub>-container;
- criticality;
- explosion in a fluidised bed or sintering furnace.

Spills or leakages of uranium containing solutions, release of uranium dust due to filter failures, and fire in the scrap purification are other possible incidents. The main hazards, albeit small, to personnel during normal plant operations are associated with:

- the potential inhalation of uranium oxide fine particles when working with powders;
- exposure to external radiation during fuel inspection and storage.

Releases of UF<sub>6</sub> caused by failures of pipe connections or valves have occasionally occurred in fuel fabrication plants. Minor or major contamination with UO<sub>2</sub>F<sub>2</sub>, within the conversion building, from the reaction of the released UF<sub>6</sub> with moisture, was the main result of these incidents. No off-site consequences occurred. To prevent the need for major clean-up work after a UF<sub>6</sub> release, a separation or enclosure of the UF<sub>6</sub> conversion plant area is recommended. Rupture of a heated UF<sub>6</sub>-cylinder in the evaporation station at the head end of the conversion process, represents the most severe possibility for a large UF<sub>6</sub> release. In the case of enriched uranium the capacity of UF<sub>6</sub>-cylinders of type 30B is 1.5 tHM. Studies indicate the maximum release of UF<sub>6</sub> in case of a rupture of a heated cylinder for enriched uranium would not be in excess of approximately half of this quantity. Rupture of the conversion furnace itself would only give rise to a small leakage because of the restricted inflow rate used. Modern evaporation stations for UF<sub>6</sub> are equipped and operated with remotely controlled or automatic valves to keep as low as possible the quantities of UF<sub>6</sub> released in case of an incident.

One of the major safety concerns in a commercial fuel fabrication plant and the associated fuel store is the potential for a criticality. Since in most facilities only LEU is handled (with enrichments up to 5% <sup>235</sup>U), fabrication steps which are made under dry conditions are inherently safe in the absence of a moderator. In the steps in which hydrogenated fluids are used, mass or geometry controls are applied. In some cases fixed neutron absorbers are additionally used to maintain subcriticality even in the event of accidental moderation and neutron interaction. Off-site exposures from any fission

products generated in criticality excursions are likely to be insignificant. The main hazard will be the local high-radiation fields which are produced by the excursion.

The increased use of recycled uranium in the future will not increase criticality hazard. Also the increase of burn-up probably requires only a small increase of the initial enrichment so that fuels for LWR with much more than 5%  $^{235}\text{U}$  are unlikely to be fabricated in large quantities in the foreseeable future.

The use of reprocessed uranium in fuel manufacturing, with higher concentrations of  $^{232}\text{U}$ ,  $^{234}\text{U}$  and  $^{236}\text{U}$  induces a higher gamma radiation field due primarily to the formation of the decay products  $^{212}\text{Bi}$  and  $^{208}\text{Tl}$ . With increasing burn-up the concentration of  $^{106}\text{Ru}$  also increase, but this effect disappears after a prolonged storage. The radiological impact of  $^{232}\text{U}$  and  $^{106}\text{Ru}$  are both time-dependent but in an opposite way. If large quantities of reprocessed uranium are to be handled, improved shielding and encapsulation of uranium are desirable. The level of alpha or gamma-emitting impurities in reprocessed uranium call for improved purification of the  $\text{UF}_6$  at the end of the conversion steps and for a substantially improved cleaning of liquid effluents. Studies of these effects and their possible influence on accidental situations should be performed taking into account not only fuel manufacturing but the complete recycling strategy for reprocessed uranium.

In fluidised bed or sintering furnaces, hydrogen is used as a reducing agent, usually diluted with inert gas to prevent the likelihood of explosion. However explosive mixtures can develop from incomplete removal of air from a cold furnace at start-up, or from in-leakage of air into the furnace. Control mechanisms have a low probability of failure, but the possibility of the development of an explosive air-hydrogen mixture cannot be completely excluded. The pressures developed would be sufficient to blow out large quantities of  $\text{UO}_2$  powder, if an explosion occurred in a fluidised bed furnace. For a sintering furnace explosion,  $\text{UO}_2$  would be blown out of the ends of the furnace in the form of pellets. The release of  $\text{UO}_2$  to the environment would, however, be small and its off-site consequences would not be of any significance.

#### **4.2 Specific safety measures for MOX fuel fabrication facilities**

The specificity of MFFF safety is connected with the following:

- first of all, the radiotoxicity of plutonium, higher than that of uranium;
- secondly, the dry process fabrication method used in the industrial-size facilities currently in service, which has a potential for criticality and for dispersion of radioactive material;
- finally, the isotopic characteristics of the plutonium used in these facilities, which will have an effect on the criticality likelihood, the risk of external exposure, and the thermal risks.

External exposure and thermal hazards come from the neutron emission from the  $^{238}\text{Pu}$  and  $^{240}\text{Pu}$  isotopes and the gamma emission from the  $^{241}\text{Am}$  which may have “grown into” the oxide by the decay of  $^{241}\text{Pu}$  during storage.

A wide variety of hazards have determined the wide variety of adequate safety measures used in MOX fuel fabrication facilities.

#### *4.2.1 Dispersion of radioactive material*

Due to the high radiotoxicity of plutonium by inhalation, the safety measures and design characteristics related to MFFF safety are very different from those characterising the fabrication of uranium-based fuel.

The design-related measures taken to prevent the dispersion of radioactive material are based on containment systems whereby a series of static barriers is interposed between the radioactive material and the environment. The first containment system generally comprises a static barrier interposed between the personnel and the radioactive material. It may consist of glove boxes in which the process equipment is installed, the process equipment itself, welded fuel rod cladding, sealed containers intended for the transfer of the radioactive material, etc.

The second static barrier consists of the walls of the premises that contain the equipment ensuring the first barrier. The third and last barrier consists of the walls of the buildings. The second barrier contributes to the protection of the permanent workstations (control rooms in particular) and the third barrier guarantees that the environment is unaffected.

The quality of these static barriers is taken into account during the design and construction phases, particularly through the selection of materials adapted to withstand the stresses that they may be subjected to (mechanical, thermal, etc.), through the seismic design of certain structures and equipment, through the quality of construction and manufacture, and through inspections to verify this quality. The quality of design and construction of the first containment system must guarantee the absence of contamination within the plant premises during normal operation.

A dynamic containment is associated with these static containment barriers and complements them. It ensures a cascade of negative pressures directing the air circulation from the potentially less contaminated areas towards the potentially more contaminated areas. This system also ensures purification of the air before its discharge to the environment by trapping the possibly contaminated elements in a series of HEPA filters associated with each static containment barrier. The organisation of the buildings in containment systems, with their associated specific ventilation system, results in a division of the facilities into different areas according to the level of contamination risk, whereby different areas are accessed via air-locks.

The ambient activity rate of the premises where the glove boxes or radioactive material in sealed containers are installed is constantly monitored by systems that continuously sample and examine the work atmosphere, through real-time alpha activity measurements (EDGAR system) or time-delayed measurements.

Operating methods and maintenance must guarantee the observance of the design and construction measures: constant negative pressure within the glove boxes, leak-tightness and radiological cleanness of the premises, and the efficiency of the incidental contamination detection systems and of the filtering systems.

Specific practical training programmes dedicated to work within the glove boxes are provided to the plant operators. They are intended to provide the means of personal protection in case of risk of internal exposure, and to see to it that the rules to reduce the risks inherent in glove box operations (and their consequences) are observed.

In the late 90s, COGEMA has carried out significant research and development work, in collaboration with other manufacturers, to improve the mechanical performance of the gloves

equipping the glove boxes, to test the materials best suited for their use, and to master the fabrication processes, making it possible to guarantee their quality of manufacture.

#### 4.2.2 *External exposure.*

The isotopic composition of the plutonium used requires that the risks from gamma radiation and neutron emission be taken into account. The gamma radiation varies according to the  $^{241}\text{Am}$  content. The neutron emission varies according to the concentration of even isotopes in the plutonium, particularly the  $^{238}\text{Pu}$  content. It is most significant in the assembly fabrication workshop. The corresponding dose rates cannot generate a significant likelihood of external exposure in routine operation.

The design-related measures intended to reduce the exposure of the operators to radiation include automation or remote control of the operation and of the product quality monitoring techniques, installation of protection screens suited for the nature and energy of the radiation, and design of process equipment to reduce the number and duration of interventions. During operation, the principal measures must proceed from application of the ALARA principle.

#### 4.2.3 *Criticality*

The presence of significant quantities of fissile material ( $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ ) in the equipment of the industrial-size MFFF, the various stages of the elaboration of the MOX blend, and the incorporation of hydrogenated material at some of these stages call for specific safety measures regarding criticality. The prevention of criticality is firstly based on the characterisation of the fissile material during the various stages of the fabrication of the MOX fuel: isotopic composition of the plutonium and uranium, physical and chemical form (powder, granules, pellets, fuel rods), density of the blend,  $\text{PuO}_2$  content of the blend, and concentration of hydrogenated products in the blend. The full knowledge and control of these essential variables must be justified, as well as limiting their value in any situation. Second of all, the prevention of criticality is based on the determination of the control methods best suited for the process for each item of equipment or part of the facility.

The main control modes used (individually or in combination) in MFFF are the geometry of the equipment (particularly interim storage), mass, and moderation. In certain cases, neutron-absorbing fixed materials are also used to maintain a subcritical condition, even in the event of accidental moderation or neutron interactions.

The fact that the MOX fabrication process employs dry (or weakly moderated) fissile material and the use of moderation as a control mode associated to the mass or geometry calls for strict measures regarding design and operation so as to guarantee the limitation and control of the moderation in all situations. The accidental situations, in which the full control of criticality must be demonstrated, are studied by taking the other hazards into account (thermal hazards, fire, flooding, earthquake, etc.).

In addition, a criticality detection and alarm system monitors all parts of the facility where a criticality excursion is plausible.



#### 4.2.4 Thermal hazards

The thermal power of the plutonium requires that the release of heat be taken into account. This release of heat must not lead to a deterioration of the safety functions, which are essentially containment of the radioactive material, protection against ionising radiation, and prevention of criticality. The main points to be examined concern the places where significant masses of material are concentrated: powder, pellet, fuel rod, and assembly interim storage, as well as large capacity homogenisers. The possible consequences of a complete loss of ventilation, the means generally used to remove the heat, must be evaluated.

#### 4.2.5 Fire

Unlike a criticality excursion or an explosion, a fire is rarely a sudden event and often breaks out from a small source. The persons responsible for the design, construction, and operation of fuel fabrication facilities carefully consider the possibility of fires and provide for the equipment and procedures required to prevent them.

Although the nuclear materials used during the processing (uranium and plutonium oxides) are not flammable themselves, certain elements of the equipment, such as the glove box panels, may be made of more or less combustible material. In addition, a fire may be connected with the interim storage of combustible waste or fluids and break out further to flammable gas leakage.

The principal general preventive measures concern the choice of materials, methods, and equipment and the limitation of the heating load within the premises. The use of fast kinetics combustible substances must be limited as much as possible.

According to the in-depth protection concept, the next step is to provide for a quick detection of the fire by installing equipment suited for the expected events.

In order to limit the propagation of the fire, the facility is divided into fire and containment sectors. Finally, the means of intervention must be provided, including trained intervention teams and devices such as fixed CO<sub>2</sub> injection systems. The ventilation must be designed and controlled in order to protect the last filtering stage to minimise discharge to the environment.

A fire must not lead to a criticality excursion through the deterioration of a neutron absorber, a modification of dimensions, or the moderation of fissile material when a hydrogenated extinguishing substance is used. Finally, a localised fire must not jeopardise the availability of the criticality accident detection equipment. A large fire not properly subdued may lead to significant consequences on the environment. These consequences must be assessed.

#### 4.2.6 Explosion

The mixture of gases supplying the high-temperature sintering furnaces has a concentration of H<sub>2</sub> (in argon) limited to 8% in order to control the flammability and explosion of this gas in air. The mixture is either prepared outside the nuclear facility and its content verified before use, or prepared on line from component gases. In the latter case, safety features are incorporated in the gas mixing equipment, such as electromechanical isolation valves interlocked with gas analysis system.

The potential for explosion also comes from the radiolysis occurring, for the most part, in solutions containing plutonium and leading to the release of H<sub>2</sub>. However, solid waste and unsintered process rejects containing hydrogenated additives may also lead, with slower kinetics, to the production of H<sub>2</sub>. Corresponding volumes are ventilated in order to keep H<sub>2</sub> concentration always below flammability limit.

#### 4.2.7 Hazards of external origin

External hazards can become initiating events for risk-significant incidents or accidents. The following hazards must be systematically taken into account, as they are in all nuclear facilities:

- Earthquakes: in addition to the design of the facility, it is necessary to examine the risk of fire (due to the use of flammable gases) and the risk of criticality excursion, that might be both caused by an earthquake.
- Flooding: caused by tides, streams, and rainfall, possibly resulting in a dispersion of material as well as in a risk of criticality excursion.
- Risks related to the industrial environment and transport routes: the main hazards are explosions and toxic clouds. Regarding transport, accidental aircraft crashes must be considered (a probabilistic approach may be used to quantify this risk).
- External fires, extreme temperatures, wind, and snowfall.

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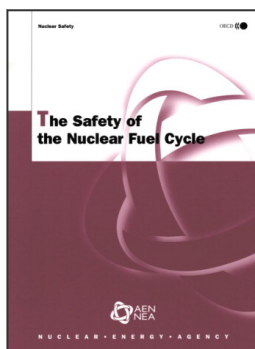
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