

### 3. THE NUCLEAR FUEL CYCLE

#### 3.1 General

The fuel cycle of a nuclear power plant can be divided into three main stages:

- a) the so-called front-end which extends from the mining of uranium ore until the delivery of fabricated fuel elements to the reactor site;
- b) fuel use in the reactor, where fission energy is employed to produce electricity, and temporary storage at the reactor site;
- c) the so-called back-end, which starts with the shipping of spent fuel to away-from-reactor storage or to a reprocessing plant and ends with the final disposal of reprocessing VHLW or the encapsulated spent fuel itself.

Costs of the second stage, b), are not dealt with in this report, because they are conventionally covered under the capital or operating and maintenance costs of the nuclear power plant. For the analyses presented in this study, two PWR fuel cycle options are considered. The fuel cycle option in which the spent fuel from the reactor is reprocessed, to separate plutonium and remaining uranium from the wastes produced in the fission process, is identified as the reprocessing option. The second option, which is generally known as direct disposal, involves disposing of spent fuel following appropriate treatment after a period of, usually, long-term storage. Figures 3.1 and 3.2 illustrate the two fuel cycle options and also give an indication of the quantities of the material involved in the different stages, for each tonne of uranium fed into the study's reference reactor.

#### 3.2 The front-end of the fuel cycle

##### 3.2.1 Uranium mining and milling

Uranium is the fuel used in nearly all existing nuclear reactors. It is very widely distributed in the earth's crust and oceans, but can only be economically recovered where geological processes have locally increased its concentration. Almost all economically workable uranium-bearing ores have in the past typically contained less than 0.5 per cent of uranium, and in some cases ores were mined with grades as low as 400 parts per million. On the other hand, some uranium deposits exhibit uranium concentrations of several percent and the trend with new discoveries has been towards higher grades. The quantity, quality and geographical distribution of uranium resources are discussed in detail in regular OECD/NEA and IAEA publications<sup>(8)</sup>.

Uranium ore is mined either by conventional open-pit or underground mining methods and the uranium is extracted from the crushed ore in a processing plant (mill) using chemical methods appropriate to the specific mineral form. These usually extract some 85 to 95 per cent of the uranium present in the ore. The radioactivity of the separated uranium is very low. The radioactive daughter products are left with the mill tailings, stabilized and put back into the mine or otherwise disposed.

In some cases it is possible to pass chemical solutions through the ore bodies and dissolve the uranium directly. This process is known as solution mining, or in-situ leaching. Uranium can also be recovered as a by-product of the extraction of other metals from their minerals, for example copper and gold, and as a by-product of phosphoric acid production from phosphate rocks. Solution mining has been increasingly used during recent times.

The uranium concentrate ( $U_3O_8$ ) produced in the ore processing plant is known as *yellowcake* and usually contains between 60 and 85 per cent uranium by weight. Depending on its quality, the concentrate is sometimes further purified in a refinery near the mine before being shipped in metal containers to a conversion plant.

### 3.2.2 Conversion

The high purity required for nuclear fuel is achieved by dissolving the uranium concentrate in nitric acid, filtering and treating the solution with chemical solvents. The resulting uranyl nitrate is more than 99.95 per cent pure.

The uranyl nitrate is reconverted to uranium oxide and this, in turn, is converted to readily volatile uranium hexafluoride ( $UF_6$ ) which is used in the enrichment process. If enrichment is not required, for example for heavy water reactor fuel, then uranium dioxide ( $UO_2$ ) is produced from the uranyl nitrate and shipped directly to a fuel fabrication plant.

### 3.2.3 Enrichment

Uranium occurring in nature consists largely of  $^{238}U$  which acts predominantly as a neutron absorber. The fissile  $^{235}U$ , an isotope with a lighter atomic nucleus, occurs to the extent of only 0.71 per cent in natural uranium. Reactors such as the graphite moderated Magnox reactor and the heavy water cooled and moderated reactor (CANDU) are able to function with fuel containing only the naturally occurring proportion of  $^{235}U$ . Light water cooled and moderated reactors (LWR) as well as advanced gas-cooled reactors (AGR) contain a greater proportion of neutron absorbing materials and this has to be compensated for by increasing the concentration of the  $^{235}U$  isotope in the fuel from 0.7 per cent to around 3 to 4 per cent.

Although the isotopes of a given element have identical chemical properties, the nuclei of their atoms have slightly different masses and these differences provide a means whereby a given element can be separated into portions containing different relative proportions of heavy and light isotopes. The process by which the concentration of the  $^{235}U$  isotope is increased is known as enrichment.

The enrichment techniques generally involve separation in the gas phase hence the conversion to readily volatile uranium hexafluoride. This compound has the additional advantage that fluorine has only one isotope, so that molecular mass differences are entirely due to differences in the masses of the uranium atoms they contain.

Gaseous diffusion through porous membranes is the most widely used technique but a number of countries have installed gas centrifuges. An alternative process which may be used commercially in the future relies on separation in streams of gas flowing through specially-shaped nozzles.

Laser excitation techniques, in which advantage is taken of small differences in the light absorption characteristics of uranium atoms or their compounds, and enrichment through chemical processes, are being

actively pursued in many laboratories. Laser enrichment and advanced gas centrifuge techniques are quite likely to be introduced within the timescale considered by this study. Their energy consumption is only a fraction of that required by the gaseous diffusion process.

After passing through the enrichment plant, the uranium hexafluoride has been separated into two fractions. The smaller of these is enriched in the  $^{235}\text{U}$  isotope and is shipped to the fuel fabrication plant in metal cylinders with suitable precautions to guard against inadvertent criticality. The larger fraction (enrichment tails) is depleted in  $^{235}\text{U}$  and is stored. It may be used in MOX fuel or in commercial breeder reactors in the future. Economic and technical changes may make the recovery of some of the residual 0.2 to 0.3 per cent  $^{235}\text{U}$  contained in the tails worthwhile.

### **3.2.4 Fabrication**

The enriched uranium hexafluoride is chemically converted to pure uranium dioxide powder which is then pressed into pellets and sintered in a furnace at high temperature to produce a dense ceramic fuel. The PWR fuel pellets are stacked together and then they are sealed in tubes of corrosion resistant zirconium alloy with a low neutron absorption. These loaded tubes, called fuel pins, are put together in a lattice of fixed geometry called a fuel assembly (289 pins per assembly for the study's reference reactor). A similar procedure is adopted for unenriched uranium oxide fuel for CANDU reactors and for the fuel for advanced gas-cooled reactors, although in the latter case stainless steel, which resists corrosion by the carbon dioxide reactor coolant, is used in place of zirconium alloy to contain the fuel pellets.

### **3.2.5 Wastes arising in the front-end of the fuel cycle**

Uranium mining produces waste rock with a lower uranium content than that of the ore. Milling wastes include radium and other naturally occurring radioactive substances. These wastes are generally disposed of in engineered geological facilities which are covered on top and sealed underneath and on the sides in order to reduce radon emissions and the movement of ground water.

Wastes from the conversion process may contain uranium, acids and some organic chemicals. Some conversion facilities recycle such wastes to uranium mines in order to recover the uranium content while others directly dispose their waste.

Wastes arising from the uranium enrichment and fuel fabrication processes contain essentially small amounts of uranium and the associated naturally occurring radioactive elements.

Currently, the tails that result from the enrichment process (of fresh uranium or reprocessed uranium) are stored in the form of uranium hexafluoride, a high vapour pressure solid at ambient temperature. Later, these tails may be recycled in MOX fuel for thermal fast reactors. Control and management of the fluorine gas and the  $\text{UF}_6$  tails poses a more difficult task than dealing with the radioactive waste products. To ensure even greater safety it is likely that  $\text{UF}_6$  tails will be converted to  $\text{U}_3\text{O}_8$  powder form in future.

During fuel fabrication, it is important to distinguish between scraps and wastes. Scraps are recycled through dry or wet routes (the latter allowing chemical purification). The volume of scraps usually represents a few per cent of the initial material. Wastes comprise contaminated materials; they arise in much greater volume but contain far fewer quantities of initial materials than scraps and therefore are not recycled. In the case of  $UO_2$ , the low radioactivity of the product allows a very simple management of both the scraps and the wastes.

Although uranium has a low radio-toxicity, the same is not true for plutonium. Thus, in the case of MOX fuel fabrication greater care has to be taken in the management of the wastes. The treatment of wastes in order to separate the plutonium and uranium, and the subsequent waste conditioning are fully mastered. A typical value for the quantity of plutonium finally present in wastes is 0.01 per cent of the initial plutonium.

### **3.3 Fuel at reactor**

New fuel arriving at the reactor site is placed in a store designed to contain sufficient stock to cover the reactor operator's needs and to guard against any short term supply problems.

From the store, the fuel assemblies are transferred to the reactor and placed in the core where they remain for about three to five years, depending on the selected refuelling schedule. During this time a proportion of the uranium atoms undergo fission to produce energy and fission products. In addition plutonium is also produced from uranium atoms and is, in turn, partly fissioned<sup>(f)</sup> in the reactor. As a consequence, the discharged fuel is highly radioactive and has to be heavily shielded. A typical PWR fuel assembly also generates, immediately after discharge, many hundreds of kW of heat from the radioactive decay of the fission products within the fuel. For these reasons it is normal practice to store the newly discharged PWR fuel assemblies in the reactor pool for at least a few years, to allow the radioactivity to decline naturally. Two meters of water above the fuel assemblies provides adequate protection against radiation; the water in the pool also acts as a good heat transfer medium.

In addition to the spent fuel, a reactor produces, during its normal operation, some liquid and solid wastes containing much lower levels of radioactivity. The costs of storing, treating and ultimately disposing of these wastes are relatively small and are regarded as operational costs rather than fuel cycle costs.

### **3.4 The back-end of the fuel cycle**

#### **3.4.1 Transport and interim storage of spent fuel**

##### **3.4.1.1 Transport**

After a cooling period of a few years at the reactor site, the most highly radioactive fission products will have decayed and the rate of heat production from the spent fuel will have declined appreciably. Although the fuel assemblies are still highly radioactive and produce significant quantities of heat, safe transport of the spent fuel is now more readily accomplished. For transport the spent fuel is loaded into heavily shielded transport casks in which it is shipped to the interim storage facility or to the reprocessing plant. These transport casks, which provide cooling for the fuel elements and shielding for workers and the public against the emitted radiation, are designed to withstand transport crashes and fires so that the protection they afford would be maintained even in the event of a major accident.

##### **3.4.1.2 Interim storage**

The interim storage period is the time interval after the minimum cooling period following discharge from the reactor until reprocessing or spent fuel encapsulation prior to disposal. Interim storage of spent fuel could take place at the reactor site in cooling pools or in cask storage. In this case, storage costs are often an integral part of the power plant operating costs. Alternatively, it could take place at a separate interim storage site or in storage pools at the reprocessing site. In the latter case, interim storage costs are usually included in the reprocessing price.

A number of different approaches have been developed for interim storage in which the fuel assemblies, either intact or dismantled to reduce the volume they take up, are stored in cooling pools situated either on the reactor site or at separate sites. Additionally, dry stores have been developed in which the fuel assemblies, with or without pre-treatment and special packaging, can be safely held in either an air or inert gas atmosphere.

### **3.4.2 Reprocessing option**

#### **3.4.2.1 Reprocessing**

Reprocessing involves dissolving the spent fuel to enable the re-usable plutonium and uranium content to be separated from the residual waste fission products and actinides. PWR spent fuel typically contains 1.15 per cent (by weight) plutonium, 94.3 per cent uranium and 4.55 per cent waste products. The separated uranium may then be re-enriched prior to re-use and the plutonium incorporated with MOX fuel. In this manner, about 30 per cent of the potential energy in the initial fuel can be re-utilised in thermal reactors and more if fast reactors were used.

Operations at the reprocessing plant are conducted remotely in facilities with adequate shielding to protect the workforce from the effects of radiation exposure. The fuel assemblies are chopped up and placed in nitric acid. This enables the fuel content, which dissolves in the acid, to be separated from the insoluble zirconium alloy or stainless steel cladding.

The solution of uranium, plutonium, other actinides and fission products is then chemically treated in a series of stages which are designed to produce solutions of plutonium nitrate and uranyl nitrate of high chemical purity. The waste products (other actinides, fission products and unwanted impurities) are stored as a highly radioactive solution in water cooled double-walled high integrity stainless steel tanks before further conditioning. The separate solutions of uranyl nitrate and plutonium nitrate are further processed. The uranium can be converted to uranium dioxide for storage or for the production of new fuel, by blending with fissile material or conversion to uranium hexafluoride for return to the enrichment plant. The plutonium nitrate is converted to plutonium dioxide for storage or for incorporation into mixed oxide fuels for thermal or fast reactors.

#### **3.4.2.2 Waste management**

Conditioning of the wastes produced by reprocessing is a well established operation that has been rigorously examined and approved by regulatory authorities in several countries. The removal of the plutonium and the uranium via reprocessing reduces the volume of high level waste, but leads to the production of low and intermediate level wastes (see below and Annex 3). Operating experience has been accompanied by a strong downward trend in the volume of wastes produced. In addition, there are important programmes in hand to further diminish these volumes. For instance, the volume of French wastes for deep

disposal is expected to decrease from the current volume of 1 400 l/tHM to a volume less than 465 l/tHM by around the year 2000.

i) Process wastes

The process wastes are primarily fission products and actinides which represent about 99 per cent of the total radioactivity in spent fuel. These products have been vitrified on a commercial scale since 1978. The volume of VHLW is only 115 l/tHM.

The second source of process wastes is hulls and end fittings. These wastes are embedded in cement and belong to the category of Intermediate Level Waste (ILW).

The operation of reprocessing plants results in the discharge to the environment, after appropriate treatment, of very low level airborne and liquid effluents arising from various process stages.

ii) Technological wastes

These are the wastes coming from the operation of the plant:

- used equipment and parts;
- degraded solvent;
- "trash bins" (gloves, etc.);
- metallic drums containing small contaminated parts.

They are either liquids or solids. Liquid wastes are concentrated and today embedded on liner bitumen (ILW) or are precipitated to form a solid waste. Solid wastes are either embedded in cement (ILW) or packaged in drums (LLW).

iii) Interim storage of wastes

In most countries, interim storage is also required for the wastes during the period between conditioning and final disposal. Specially constructed facilities already exist for this purpose.

### **3.4.3 Direct disposal option**

#### **3.4.3.1 Encapsulation of spent fuel**

After removal from the reactor, the spent fuel will normally be stored in pools at the reactor site and then be transferred to an interim store.

Fuel assemblies may, after a period of cooling, which may be 30 to 50 years, be encapsulated directly or be disassembled using remote handling techniques so that the fuel pins can be packed together more closely prior to encapsulation. The encapsulation process involves placing the spent fuel in a canister of metal, such as copper, steel or titanium, or of ceramic material. After that the canister is tightened, e.g. by welding a lid. Intermediate storage and encapsulation results in 0.2 m<sup>3</sup> of medium level waste per tonne of uranium.

#### **3.4.4 Final disposal of waste**

In both the reprocessing and the direct disposal options, disposal of the wastes arising at the front-end of the fuel cycle and from interim storage are included for costing purposes with the appropriate fuel cycle components.

#### *3.4.4.1 Reprocessing*

Following conditioning and, in most cases, interim storage for a number of decades to allow further reduction of radioactivity and heat generation, vitrified HLW, suitably encased, can be transported to and placed in a deep geological repository. Here, it can be held under supervision and, when considered appropriate, sealed off permanently. The glass matrix in which the highly radioactive wastes are incorporated, the method of encapsulation and the geological formation chosen to isolate the radioactivity from the biosphere, are carefully selected to ensure long term safety.

ILW fixed in a concrete or a bitumen matrix within a steel container can also be consigned to geological disposal.

Conditioned solid LLW is usually transported to shallow land burial sites or placed in geological repositories under carefully controlled and monitored conditions which seek to ensure that there is no risk of significant radiation exposure to any member of the general public. Very low level liquid wastes are discharged to the sea or to rivers. The level of liquid wastes discharged to the sea or rivers complies with stringently enforced regulations.

#### *3.4.4.2 Direct disposal*

Following encapsulation, the entire amount of spent fuel is treated as HLW and is disposed of in a range of ways paralleling those for the vitrified high level waste from reprocessing. In general this will involve placing the encapsulated fuel in deep geological repositories, possibly surrounded by a buffer material (e.g. bentonite) to prevent ground water coming into contact with the outer container forming the encapsulation.

### *3.4.5 Plutonium and uranium recycling*

#### *3.4.5.1 Plutonium recycling*

Plutonium can be used in MOX fuel in thermal reactors, such as PWR or BWR, or in fast reactors. Other reactors, such as the ATR, can also use plutonium (see section 8.2). Fast reactors hold considerable promise for the next century and the use of MOX in PWRs and BWRs is currently well developed. The first MOX assembly was loaded in a PWR in Belgium for demonstration purposes almost 30 years ago. The present global production capacity for thermal reactor MOX fuel is about 70 tonnes p.a. with almost 350 tonnes p.a. forecasted for 2000. An international market for MOX fuel already exists, with countries such as France, Germany and Switzerland having experience with thermal MOX fuels, and other countries, such as Japan and Belgium, planning to load MOX in their reactors in the future. The use of MOX fuel leads to changes in reactor core properties; shut-down margins

are reduced, compared to conventional fuel. In current LWRs, the largest licensed fraction of MOX fuel which may be loaded is approximately 50 per cent. In the future, however, it should be possible to design LWRs utilising up to 100 per cent MOX fuel.

The quantities of high neutron absorbing isotopes of plutonium increase with fuel burn-up.  $^{238}\text{Pu}$  produces significant quantities of heat and neutrons and is one of the factors to be considered in the transport and storage of plutonium and mixed oxides. Countries that have chosen reprocessing manage the stocks and flows of plutonium while taking into account the above constraints.

Plutonium production ceases when fuel is removed from the reactor. Thereafter radioactive decay becomes the critical factor in plutonium recycle as it produces a decrease in the fissile isotope content and a build-up of gamma-emitting decay products which, progressively, make handling of Pu during MOX fuel fabrication increasingly difficult and more expensive.

The 1989 NEA Plutonium Study<sup>(5)</sup> has recommended limits, based on practical experience, regarding suitable storage periods for materials containing plutonium recovered from LWR spent fuel. Maximum indicative storage periods for  $\text{PuO}_2$  powders, MOX fuel rods and fresh MOX fuel assemblies are 2 years, 10 to 13 years and 13 to 20 years, respectively. In general, a short time interval should occur between reprocessing and MOX fuel fabrication.

New plants, such as the German SIEMENS MOX plant in Hanau and the French MELOX plant, will be capable of dealing with much older plutonium powders (about 5 to 6 years after reprocessing) because of increased automation and better worker protection. If needed, it is possible to gain more flexibility by subjecting "old" plutonium to further chemical purification.

Second generation MOX plants (e.g. the Sellafield MOX plant) which will start operation later this decade, have been designed to handle even older plutonium powders from high burn-up fuels (10 years old plutonium from 60 GWd/t fuel).

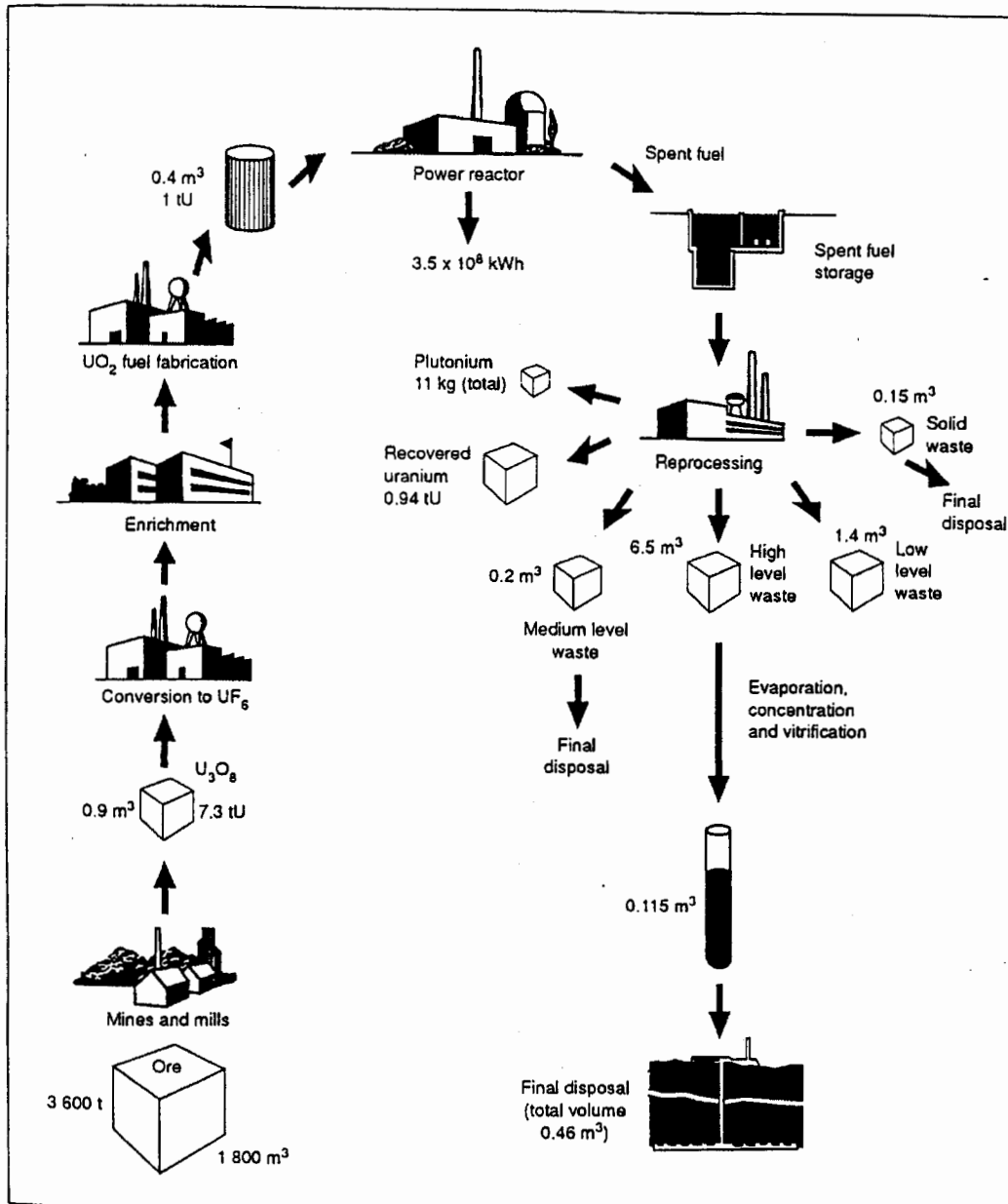
#### 3.4.5.2 Uranium recycling

The present economic situation of the uranium market limits the interest in uranium recycling. Nevertheless, some electric utilities (e.g. in France, Japan, Germany and Switzerland) show some interest in developing recycling programmes.

The technology for making reprocessed uranium fuel is well established so there should be no technical limits on these programmes. In addition, the coming into operation of AVLIS enrichment will provide a very efficient means for re-enrichment of reprocessed uranium.

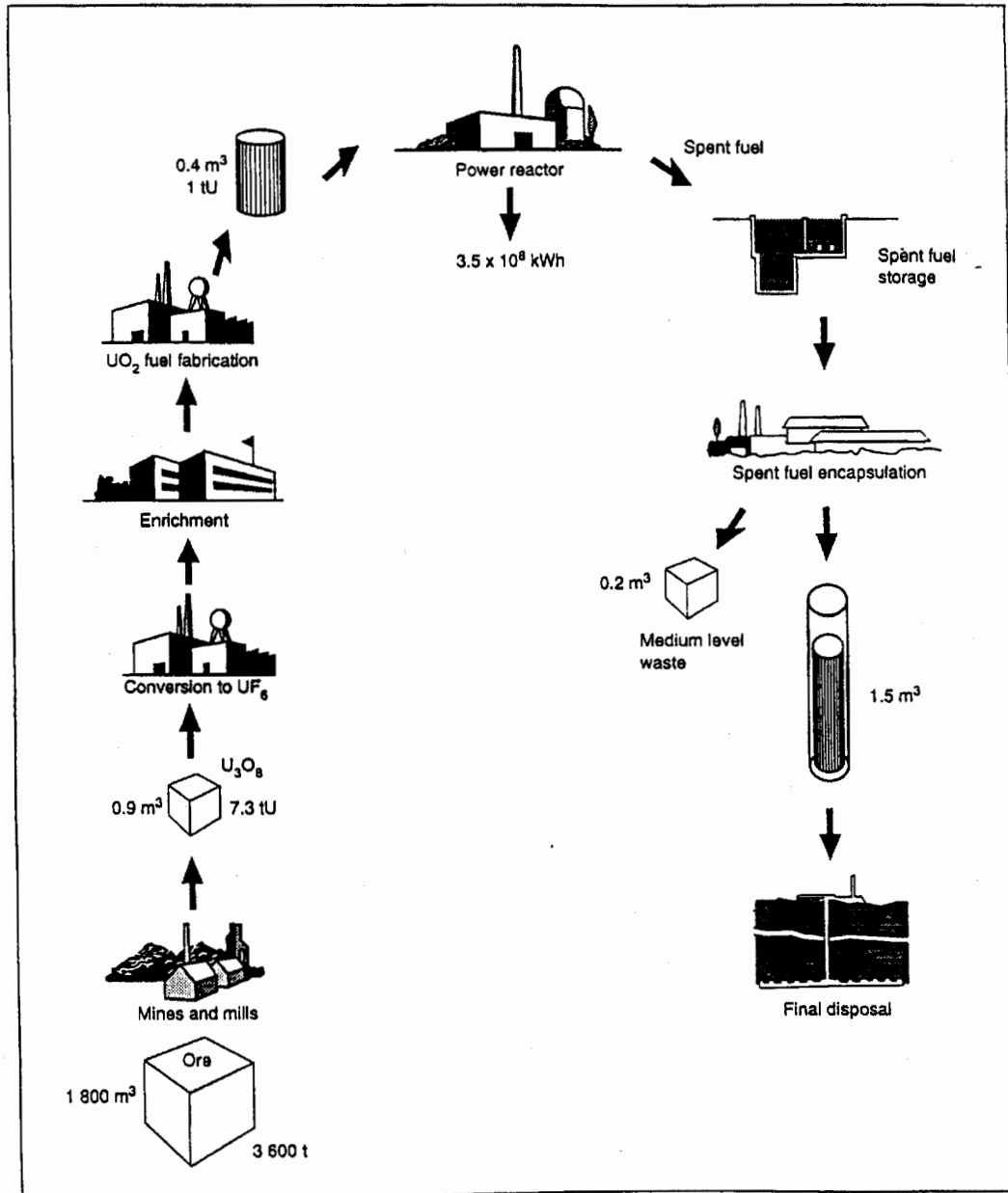


Figure 3.1 Material flow of the PWR reprocessing option  
 (the figure is an example and the numbers are approximate only)



Recovered uranium and plutonium can be recycled.  
 Source: COGEMA, HORIZON 2000.

Figure 3.2 Material flow of the PWR direct disposal option  
 (the figure is an example and the numbers are approximate only)



Source: COGEMA, HORIZON 2000 and information provided by SKB.