



**AUSTRALIAN ATOMIC ENERGY COMMISSION  
RESEARCH ESTABLISHMENT  
LUCAS HEIGHTS**

**WASTES AND WASTE MANAGEMENT IN THE URANIUM  
FUEL CYCLE FOR LIGHT WATER REACTORS**

by

**J.M. COSTELLO**

August 1975

ISBN 642 99700 4



AUSTRALIAN ATOMIC ENERGY COMMISSION  
RESEARCH ESTABLISHMENT  
LUCAS HEIGHTS

WASTES AND WASTE MANAGEMENT IN THE URANIUM FUEL CYCLE  
FOR LIGHT WATER REACTORS

by

J.M. COSTELLO

ABSTRACT

The manufacturing processes in the uranium fuel cycle for light water reactors have been described with particular reference to the chemical and radiological wastes produced and the waste management procedures employed. The problems and possible solutions of ultimate disposal of high activity fission products and transuranium elements from reprocessing of irradiated fuel have been reviewed. Quantities of wastes arising in each stage of the fuel cycle have been summarised. Wastes arising from reactor operation have been described briefly.

National Library of Australia card number and ISBN 642 99700 4

The following descriptors have been selected from the INIS Thesaurus to describe the subject content of this report for information retrieval purposes. For further details please refer to IAEA-INIS-12 (INIS: Manual for Indexing) and IAEA-INIS-13 (INIS: Thesaurus) published in Vienna by the International Atomic Energy Agency.

FUEL CYCLE; ISOTOPE SEPARATION; ORE PROCESSING; RADIOACTIVE WASTE DISPOSAL; RADIOACTIVE WASTE PROCESSING; RADIOACTIVE WASTE STORAGE; REPROCESSING; URANIUM; URANIUM HEXAFLUORIDE; URANIUM ORES; WATER COOLED REACTORS; WATER MODERATED REACTORS

## FOREWORD

The information presented in this report has been based mainly on environmental surveys and analyses of the fuel cycle published by the Environmental Protection Agency and Atomic Energy Commission of the United States. The report was originally the subject of a lecture delivered to the Australian Ionizing Radiation Advisory Council at Lucas Heights on 4th April 1975.



## CONTENTS

	Page
1. INTRODUCTION	1
2. THE URANIUM FUEL CYCLE	1
3. URANIUM MINING	3
3.1 Mining Operations	3
3.2 Mining Wastes and Effluents	4
4. MILLING OF URANIUM ORES	4
4.1 Description of Process	5
4.1.1 Acid leach process	5
4.1.2 Carbonate leach process	5
4.2 Milling Effluents	6
5. MANUFACTURE OF URANIUM HEXAFLUORIDE	7
5.1 Description of Processes	7
5.2 Hexafluoride Process Effluents	8
5.2.1 Wet process	8
5.2.2 Dry process	8
6. URANIUM ENRICHMENT	8
6.1 Enrichment by Gaseous Diffusion	9
6.2 Enrichment by Gas Centrifuge	9
6.3 Effluents from Enrichment Processes	10
7. FUEL MANUFACTURE AND FABRICATION	10
7.1 Process Operations	10
7.2 Effluents from Fuel Fabrication	11
8. LIGHT WATER REACTORS	12
8.1 Effluents from Operation of LWRs	12
8.2 Treatment of Reactor Effluents	12
8.2.1 Gaseous effluent	12
8.2.2 Liquid effluent	13
8.2.3 Solid wastes	13
8.3 Activity in Reactor Effluents	13
9. REPROCESSING OF IRRADIATED FUEL	14
9.1 Process Description	14
9.2 Effluents from Fuel Reprocessing	15

## CONTENTS (continued)

	Page
10. TREATMENT AND STORAGE OF HIGH LEVEL WASTE	16
11. ULTIMATE DISPOSAL OF HIGH LEVEL WASTE	18
11.1 Irretrievable Burial of Waste	18
11.2 Transmutation of Waste	20
11.3 Extraterrestrial Disposal of Waste	20
11.4 Summary - Ultimate Waste Disposal	21
12. QUANTITIES OF WASTES FROM THE URANIUM FUEL CYCLE	21
13. FINAL COMMENTS	22
14. ACKNOWLEDGEMENTS	22
15. ABBREVIATIONS	22
16. REFERENCES	23

Table 1 Uranium Radioactive Decay Series

Table 2 Radioactivity of Major Fission Product Isotopes in Wastes From Reprocessing One Tonne of Irradiated Uranium From a Light Water Reactor

Table 3 Radioactivity of Major Transuranic Isotopes in Wastes From Reprocessing One Tonne of Irradiated Uranium From a Light Water Reactor

Table 4 Maximum Annual Fuel Cycle Requirement For a Modern 1000 MW(e) Water Reactor

Table 5 Chemical and Thermal Effluent From the Annual Fuel Cycle Requirements of a 1000 MW(e) LWR

Table 6 Radiological Effluent From the Annual Fuel Cycle Requirements of a 1000 MW(e) LWR

Figure 1 The uranium fuel cycle

Figure 2 Artist's impression of an open cut uranium mine and associated treatment plant [Ranger Uranium Mines 1974]

Figure 3 Uranium mill - acid leach process simplified block flow diagram [USAEC 1974a]

Figure 4 Uranium mill - carbonate leach process simplified block flow diagram [after Merritt 1971]

## CONTENTS (continued)

- Figure 5  $UF_6$  production - wet solvent extraction simplified block flow diagram [USAEC 1974a]
- Figure 6  $UF_6$  production - dry hydrofluor process simplified block flow diagram [USAEC 1974a]
- Figure 7 Enrichment processes
- Figure 8 Fuel fabrication - ADU process simplified block flow diagram [USAEC 1974a]
- Figure 9 Boiling light water reactor (BWR)
- Figure 10 Pressurised light water reactor (PWR)
- Figure 11 Transport flask for irradiated fuel elements [USAEC 1974d]
- Figure 12 Fuel reprocessing simplified block flow diagram [USAEC 1974a]
- Figure 13 Sketch of storage vessel for high level liquid waste [Nakajima 1972]
- Figure 14 Typical canister for solidified high level waste [USAEC 1974d]
- Figure 15 RSSF sealed cask concept for storage of high level waste canisters [USAEC 1974b]
- Figure 16 RSSF air-cooled vault concept for storage of high level waste canisters [USAEC 1974b]
- Figure 17 Artist's impression of a repository for disposal of high level waste in bedded salt deposits [Rogers 1971]



## 1. INTRODUCTION

A nuclear power reactor, in effect, replaces the boiler of conventional coal or oil burning electricity generating stations. The light water moderated and cooled nuclear power reactor (LWR) appears likely to be the predominant nuclear power plant over the next 20-30 years. A tonne of the element uranium contained in ore deposits is capable of producing heat energy in a thermal reactor equivalent to that from over 20 000 tonnes of black coal.

Unlike coal in a conventional station, uranium ore cannot be fed directly into a reactor; it has to be refined, concentrated, and fabricated into a suitable form. Spent fuel discharged from reactors requires further complex treatment to recover and reconstitute plutonium and residual uranium into fresh fuel, and to remove and immobilise the radioactive fission products for interim storage and ultimate disposal.

The manufacturing processes for the production of fresh uranium fuel from ores and for the treatment, storage and disposal of spent fuel are termed the uranium fuel cycle (UFC). These operations generate process wastes contaminated with radioactivity to a lesser or greater extent, depending on the stage of UFC involved. This paper indicates the contribution of each part of UFC to wastes in the industry, and outlines waste management procedures employed. Reference is also made to the problems and possible solutions of long-term waste disposal.

## 2. THE URANIUM FUEL CYCLE

A schematic outline of UFC operations is shown in Figure 1.

Uranium is mined typically as a low-grade ore, usually containing greater than 0.1% wt. uranium. It is processed at a mine site to concentrate the uranium into an economically transportable form. Commercial concentrates, called yellowcake, may contain over 90% of oxides of uranium. Natural uranium contains only 0.7% of the uranium-235 isotope, which is capable of generating power in an LWR through thermal nuclear fission. The remaining 99.3% is the isotope uranium-238, which is effectively non-fissile in a LWR but contributes to power generation through production of small quantities of fissile plutonium.

The use of natural uranium as a fuel places some restrictions on the choice of constructional materials, moderator and coolant for reactor designs; use of uranium enriched in the 235 isotope allows a wider range of design options and a more compact reactor core, resulting in lower capital costs. LWRs require fuel enriched in the uranium-235 isotope.

Commercial enrichment processes require the uranium to be in the form of a gas. The crude yellowcake is converted into pure uranium hexafluoride ( $UF_6$ ), a compound of uranium readily vaporised by heating. The uranium isotopes in the  $UF_6$  vapour are separated in an enrichment plant, typically into a product stream containing about 2 to 4% uranium-235, and a reject 'tails' stream depleted to about 0.2-0.3% uranium-235.

Fuel fabrication, the next step in UFC, involves processing of the enriched  $UF_6$  into a ceramic oxide ( $UO_2$ ), which is pressed and sintered into fuel pellets. The pellets are loaded and sealed into metallic tubes, which are assembled into a precise mechanical array to form the fuel element assembly ready for loading into the reactor.

Burnup of fissile material and the buildup of neutron-absorbing fission products require the periodic replacement of fuel assemblies. The discharged assemblies are reprocessed to separate and recover uranium containing residual uranium-235, plutonium bred during the nuclear reaction, and the fission product waste material, including isotopes of transuranic elements other than plutonium. A portion of the uranium-235 may be recovered by the enrichment process and re-used in fuel manufacture. Plutonium may be recycled with uranium as fuel for LWRs, or stockpiled for use in fast breeder reactors. Fission products and waste transuranic elements are concentrated for storage as liquid wastes in tanks, and in the future will be immobilised for storage in a stable form.

All UFC operations differ from conventional metallurgical processes in that they generate chemical effluents contaminated with radioactivity; the radioactive content may range from thousands of curies\* per litre ( $Ci\ l^{-1}$ ) to fractions of microcuries per litre ( $\mu Ci\ l^{-1}$ ) - a range of  $10^9$  to  $10^{10}$ . From uranium mining operations to fabrication of uranium fuel, the radioactivity is derived from uranium and its decay products, and is at a relatively low level; effluents from reprocessing of irradiated fuel discharged from reactors contain intensely radioactive fission products and transuranic elements, and are arbitrarily termed high, intermediate, or low level wastes according to the concentration of radioactive isotopes present.

The objective of waste management in UFC is the control of environmental hazards latent in the chemical and radioactive nature of the waste material.

---

\* Curie: The amount of a radioisotope which decays at the rate of  $3.7 \times 10^{10}$  disintegrations per second

Three basic approaches to radioactive waste management are in current application [Keher & Miles 1971]:

- . Concentration and Containment, typically applied to high level wastes from fuel reprocessing which are relatively small in volume but remain intensely radioactive for over 1000 years.
- . Delay and Decay, applied to some intermediate and low level wastes of short half-life\*, containing radioactive isotopes which are converted into stable (non-radioactive) forms during short-term storage.
- . Dilution and Dispersion, employed for some chemical wastes containing trace levels of radioactivity.

The method or combination of methods applicable to a specific waste management problem is dependent on the UFC stage from which the waste is derived, and upon the capacity of the environment to accept wastes.

### 3. URANIUM MINING

#### 3.1 Mining Operations

Uranium may be mined by three methods:

(a) Opencut mining is employed when the orebody lies close to the surface under an overburden which can be removed easily and economically. The operation involves the mining, removal and stockpiling of all the material associated with the ore deposit. Mining by opencut methods is proposed for some deposits in the Northern Territory (NT) discovered by Ranger Uranium Mines Pty. Limited.

(b) Underground mining is generally employed for orebodies at depths greater than 100 m, or when the ore lies under rock strata which would otherwise require substantial blasting for excavation. The operation is more selective for ore and produces less waste rock than in opencut mining. Some uranium ores in NT are suited to underground mining operations.

(c) Solution mining, in which the ore is leached in situ by injection of acid into the underground deposit and the product solution recovered by pumping to a treatment plant, may be employed where an ore bed is horizontal and lies on relatively impervious strata. In addition the ore must lie below the static water table, and extreme care must be taken to avoid contamination of ground water flows [USEPA 1974].

---

\* Half-life: The time needed for a number of atoms of a nuclide to be reduced by half through radioactive decay

### 3.2 Mining Wastes and Effluents

Solid waste produced in opencut mining operations is the earth overburden above the orebody and the barren rock in which the ore is dispersed. To give an example, production of 2500 tonnes of uranium per year from some NT deposits can involve the annual mining of over one million tonnes of ore and about four million tonnes of barren rock [Ranger Uranium Mines 1974].

Initially, waste rock from excavation may be used for construction of earthworks, foundations and roads; excess rock is transported to a stockpile in the vicinity of the mine. Other solid waste material, separately stockpiled, consists of rock bearing traces of uranium below the economic cutoff grade. An impression of a typical opencut uranium mining development is shown in Figure 2.

Liquid effluent from uranium mining consists of surface water runoff from the waste rock and ore stockpiles, water seepage through the waste rock and stockpiled ore, and water seepage into the opencut. This effluent may contain suspended solids and dissolved minerals, including uranium and its decay products; treatment practices involve impoundment in retention ponds for settling of solids and surface evaporation of some water, the use of mine drainage water as a process feed to the uranium mill, and the controlled dilution and discharge of some impounded water during periods of heavy rainfall.

Gaseous effluents from uranium mining consist of some uranium and its decay products released to atmosphere during exposure and breakup of the orebody, and chemical effluents from the combustion of petroleum fuels used by the mining equipment. These effluents are rapidly diluted by air circulation in the opencut, or by forced draught circulation in the case of underground mining. The radioactive decay series for uranium is shown in Table 1.

### 4. MILLING OF URANIUM ORES

In the milling operation the ore is processed mechanically and chemically to extract the bulk of the uranium content and to produce an impure concentrate of uranium oxides, called 'yellowcake'. The uranium content of yellowcake is generally expressed in terms of the oxide  $U_3O_8$ .

There are essentially two alternative methods of uranium milling. Uranium is leached from the finely ground ore either by sulphuric acid or by sodium carbonate, the choice of process being determined mainly by the acidic or alkaline nature of the host rock containing the uranium minerals

[Hartley 1972]. The acid leach process is suitable for some ores discovered in NT, while carbonate leaching is applicable to some uranium deposits in Western Australia.

#### 4.1 Description of Process

##### 4.1.1 Acid leach process (Figure 3)

- . Ores from the mine are stockpiled into grades based on uranium content and leaching characteristics.
- . Ores are blended and reduced in size by coarse and fine crushing, followed by wet grinding to produce a slurry of finely divided ore.
- . The ore is usually heated with sulphuric acid and an oxidant to leach over 90% of the uranium into solution. The leach liquor is washed from the insoluble residue, called tailings, by counter-current decantation (CCD) and transferred to a purification stage. The tailings residue is transferred after neutralization to a retention system.
- . In the purification step, the uranium is extracted from the bulk of other soluble minerals by solvent extraction or ion exchange, and concentrated into aqueous solution. Part of the acid raffinate may be recycled for use in washing the tailings, and the remainder is neutralized and transferred with the tailings to a retention system.
- . The uranium is precipitated with an alkali (for example, ammonia) and the slurry of ammonium diuranate thickened and centrifuged to separate the solid concentrate.
- . The diuranate is decomposed into  $U_3O_8$  by calcination, followed by pulverising and packaging into drums for transport.

##### 4.1.2 Carbonate leach process (Figure 4)

- . Ores are stockpiled into grades and blended, crushed, and wet ground to a fine slurry.
- . The ore is heated with sodium carbonate solution containing some sodium bicarbonate to leach over 90% of the uranium and retain it in soluble form. Air or oxygen may be fed to the solution as an oxidant. The leached liquor is decanted or filtered from the undissolved solids, which are washed and transferred to a retention system.
- . Generally, the carbonate solution of uranium does not require purification; uranium may be precipitated selectively by addition

of sodium hydroxide with close control of pH to retain contaminant ions in solution.

- . The precipitated slurry of sodium diuranate is thickened and centrifuged. The barren solution is treated with carbon dioxide to regenerate sodium carbonate for re-use in the leaching step.
- . The sodium diuranate may either be dried and packaged as crude yellowcake or treated to reduce its sodium content by redissolution in sulphuric acid and reprecipitation with ammonia, followed by separation and calcination of the ammonium diuranate product.

#### 4.2 Milling Effluents

The tailings slurry is the major chemical and radiological effluent from the milling process. This stream is a slurry of leached solid ore and waste solutions containing spent chemicals from the grinding, leaching, uranium purification, precipitation and washing circuits of the mill. The solution from an acid leach process predominantly contains sulphate ions with quantities of soluble metal ions and traces of organic solvents. The sodium removal step in a carbonate leach process will also give rise to a sulphate waste solution. The neutralized solid tailings consist of unleached rock and precipitated mineral hydroxides, and contain about 85% of the radioactivity originally in the ore [Tsivoglou & O'Connell 1962].

Both acid and carbonate leach processes result in nearly all the radium present in the ore being retained in the tailings and discharged into the tailings retention system. The yellowcake product from acid and carbonate leaching mills contains respectively up to 0.1 and 2% of the original radium present in leached ore; this radium will appear in the waste stream from the subsequent UFC stage, hexafluoride manufacture [USEPA 1974].

The tailings slurry is pumped into a retention system designed to contain all solids, to prevent discharge of waste liquid into the surface water system and to minimise percolation into the ground. Monitor wells are located around retention systems to detect and monitor surface seepage for ionic content; excessive seepage would be returned to the retention system, and the ground faults responsible grouted by injection.

Airborne effluents from milling include radon gas from ore stockpiles, leaching tanks and the tailings retention system, and dusts containing uranium and its decay products blown from ore stockpiles. Dusts produced in processing operations such as crushing and grinding of ore and calcining and packaging of yellowcake are, in the main, removed by the ventilation extract scrubber.

Airborne chemical contaminants released to the environment include combustion products ( $\text{CO}_2$ , nitrogen oxides) from the process steam boilers, sulphuric acid fumes in small concentrations from the leach tanks, oxides of sulphur and sulphuric acid fumes from the sulphuric acid plant, and traces of vaporised organic reagents from the solvent extraction ventilation system [USAEC 1974a].

## 5. MANUFACTURE OF URANIUM HEXAFLUORIDE

The crude uranium ore concentrates require to be converted into pure uranium hexafluoride ( $\text{UF}_6$ ), the feed stock for uranium enrichment plants.

### 5.1 Description of Processes

Two major process types are in current use for  $\text{UF}_6$  manufacture: so-called 'wet' processes in which the uranium is purified before conversion to  $\text{UF}_6$ , and a 'dry' process in which complete purification is not achieved until after the  $\text{UF}_6$  has been produced. Alternative process technologies were reviewed by Alfredson [1972]. Simplified block flow diagrams of these processes are given in Figures 5 and 6.

Both process types stipulate virtually complete recovery of uranium, total utilisation of fluorine, and high utilisation of a number of other process chemicals, such as hydrogen, hydrogen fluoride, and ammonia [USEPA 1973a].

Operations in a typical wet process (Figure 5) are:

- . sampling, analysis and storage of received yellowcake;
- . dissolution of yellowcake in nitric acid;
- . purification of the uranyl nitrate solution by solvent extraction;
- . concentration and thermal decomposition of uranyl nitrate to uranium trioxide ( $\text{UO}_3$ ), with recovery of nitric acid;
- . reduction with hydrogen to form  $\text{UO}_2$ ;
- . reaction with hydrogen fluoride,  $\text{HF}$ , to form uranium tetrafluoride  $\text{UF}_4$ ; and
- . fluorination to  $\text{UF}_6$  with elemental fluorine.

The alternative dry process (Figure 6) involves the following stages:

- . sampling, analysis and storage of yellowcake;
- . treatment of the yellowcake for processing in a fluidised bed; treatment processes may include crushing and pelleting yellowcake and possibly removal of excessive sodium impurity;
- . reduction of the  $\text{U}_3\text{O}_8$  to  $\text{UO}_2$  with hydrogen;
- . reaction with hydrogen fluoride to  $\text{UF}_4$ ;
- . fluorination to  $\text{UF}_6$  with elemental fluorine; and

- . fractional distillation to purify the crude  $UF_6$  product from other volatile fluorides.

## 5.2 Hexafluoride Process Effluents

The effluents from the wet and dry processes differ substantially. The bulk of the impurities entering with the yellowcake is rejected in the wet process raffinate solution from solvent extraction, whereas in the dry process, the bulk of the yellowcake impurities is contained in solid wastes from the fluorination and distillation stages.

### 5.2.1 Wet process

Effluents from a typical wet process (Figure 5) consist of

- . neutralized aqueous raffinate from solvent extraction,
- . caustic effluents and residual fume from recovery of nitric acid and hydrogen fluoride and treatment of general off-gas streams, and
- . a small quantity of solid calcium fluoride from the fluorination step (assuming fluorination in a fluidised bed).

The raffinate stream amounts to about  $5 \text{ m}^3$  volume  $\text{t}^{-1}$  of uranium processed [USAEC 1974a]. It may contain substantial dissolved solids, radium and thorium-230 entering with the yellowcake feed, and about 0.2% of the uranium processed. This stream is neutralized and impounded in a retention pond. Proposals in the USA call for evaporation of the supernatant water and either burial of the sludge, or its transfer to a uranium mill retention system. Some scrubber effluents are treated with lime to precipitate fluoride ion for packaging and burial as calcium fluoride.

### 5.2.2 Dry process

The major part of the chemical solid effluent from the dry process (Figure 6) occurs as non-volatile ash containing iron, calcium, magnesium, copper and other fluorides. This residue can amount to about 0.1 tonne per tonne of product  $UF_6$ ; it is reacted with fresh fluorine to recover uranium and the final residue packaged and buried as low activity solid waste [USAEC 1974a]. Scrubber effluents from treatment of hydrofluorination off-gas stream arise and are treated as in the wet process.

## 6. URANIUM ENRICHMENT

Natural uranium contains about 0.7% of the fissionable isotope uranium-235. LWRs, however, require uranium enriched in the range 2-4% in uranium-235. Commercial enrichment technologies are based on gaseous

diffusion or centrifugation of uranium isotopes in the form  $UF_6$ . A simplified block diagram of enrichment operations is shown in Figure 7.

### 6.1 Enrichment by Gaseous Diffusion

In this process, gaseous  $UF_6$  is compressed and passed over a porous membrane. Molecules of  $UF_6$  containing the lighter isotope, uranium-235, diffuse through the membrane at a faster rate than molecules containing the heavier uranium-238 isotope; consequently the  $UF_6$  passing the membrane contains a slightly greater proportion of the uranium-235 molecules, and is said to be 'enriched' in uranium-235. The degree of enrichment for one membrane is minute and over a thousand successive diffusion stages are necessary to separate the proportion of uranium-235 from the naturally occurring level of 0.71% to that of 3% required for LWR fuel and 0.25% for a reject 'tails' stream. Each stage requires recompression of the gaseous hexafluoride which passes through the membrane, involving considerable electrical power.

Uranium enriched by gaseous diffusion requires approximately 98% of the electrical energy used in the entire UFC [USAEC 1974a]. A fossil fuelled power station would consume over 90 000 tonnes of coal to generate about 230 000 MWh of electricity needed to enrich the uranium for the annual fuel requirement of a light water reactor of 1 000 MW(e) capacity. In perspective, however, the nuclear reactor would produce about 28 times this quantity of electrical power.

### 6.2 Enrichment by Gas Centrifuge

This process involves the centrifugation of uranium hexafluoride gas in rapidly rotating cylinders. Inertia causes a greater proportion of the molecules containing the heavier isotope uranium-238 to migrate to the wall of the tube, and a consequent 'enrichment' of the gas near the tube axis in the lighter uranium-235 isotope. Separation factors are greater than in the diffusion process, and the two streams removed from the tube axis and wall require only tens of stages arranged in a cascade to produce the required percentage of uranium-235 in product and reject tails. The high centrifugal stresses limit the size of the equipment, and many parallel cascades involving hundreds of thousands of centrifuges are required to achieve the separative capacity of a commercial enrichment plant.

However, the gas centrifuge enrichment process is more economical on energy input, requiring about one tenth of the electrical power used by a diffusion plant of similar capacity; in perspective, the nuclear reactor could produce about 280 times the fossil electrical power required to

manufacture its uranium fuel.

### 6.3 Effluents from Enrichment Processes

The primary source of environmental impact associated with the enrichment of uranium is related to oxides of nitrogen and sulphur and particulate emissions from combustion of fossil fuel to generate the required electrical power. About 5 000 tonnes of waste gas emissions (including particulate) may be associated with the production of the annual fuel requirement for a 1 000 MW(e) LWR by the diffusion process, based on U.S. estimates [USAEC 1974a].

The enrichment plant itself generates small quantities of airborne fluorides and oxides of nitrogen and sulphur from the process and auxiliary systems. Liquid effluents are produced from process cooling systems, process cleanup operations, on-site steam plant and auxiliary production facilities. These may contain low concentrations of sulphate, chloride, fluoride, and nitrate radicals and metallic ions such as sodium, calcium, chromium and iron. Uranium losses in this effluent are very low - typically about 0.005% of throughput, equivalent to a loss of less than 10 kg out of about 190 tonnes of uranium feed required to manufacture the annual fuel requirement of a LWR [USAEC 1974a]. These effluents are disposed of by discharge and dilution. Some sludges arising from clean-up operations are retained on site.

## 7. FUEL MANUFACTURE AND FABRICATION

The feed material for the manufacture and fabrication of fuel for a LWR is uranium hexafluoride ( $UF_6$ ) enriched to about 2 to 4% in the uranium-235 isotope. The  $UF_6$  is converted to uranium dioxide powder ( $UO_2$ ) and the  $UO_2$  is formed into pellets which are sintered to achieve the desired density and ground to the required dimensions. Fuel pellets are loaded into tubes of Zircaloy or stainless steel, which are sealed at both ends. Fuel rods are spaced in fixed parallel arrays to form reactor fuel elements.

### 7.1 Process Operations

The process steps involved in making  $UO_2$  powder, shown in Figure 8, illustrate the conventional route by which  $UF_6$  is converted first to ammonium diuranate (ADU) and then to  $UO_2$ :

- . the  $UF_6$ , a solid at normal ambient temperature, is received in sealed cylinders from the enrichment plant;
- . the  $UF_6$  is vaporised by heating electrically or with steam;
- . the gaseous  $UF_6$  is hydrolysed with water to form a solution

- of uranyl fluoride ( $\text{UO}_2\text{F}_2$ );
- . ammonium hydroxide is added to precipitate ammonium diuranate (ADU);
- . the slurry of ADU is centrifuged or filtered; and
- . the ADU is decomposed by heating, pyrohydrolysed with steam to remove traces of fluoride, and reduced to  $\text{UO}_2$  powder with hydrogen.

The fabrication operations involve the following stages:

- . preparation of  $\text{UO}_2$  powder of desired size distribution by comminution, compaction and granulation;
- . manufacture of  $\text{UO}_2$  pellets;
- . sintering of the pellets in hydrogen gas;
- . grinding of the sintered pellets to the required size;
- . washing, drying and loading the fuel pellets into tubes of Zircaloy or steel and sealing the ends with welded caps; and
- . locating fuel rods in fixed parallel arrays forming the reactor fuel elements.

Off-specification material produced during fabrication is fed into a scrap recovery cycle in which uranium oxide is dissolved in nitric acid, the resultant uranyl nitrate is purified by solvent extraction, and the uranium reconverted into  $\text{UO}_2$ .

## 7.2 Effluents from Fuel Fabrication

The effluents with greatest potential environmental impact are chemical in nature [USAEC 1974a]. Fluorine discharged as solid fluorides is the only significant airborne chemical effluent from fuel fabrication. The fluorine used for manufacture of  $\text{UF}_6$  becomes a waste product during production of enriched  $\text{UO}_2$  powder. The gaseous fluoride wastes generated are effectively removed from the air stream by scrubbing and filtration systems before discharge to atmosphere.

Liquid effluents from fuel manufacture contain nitrogen compounds formed by ammonium hydroxide in production of  $\text{UO}_2$  powder and by nitric acid in the scrap recovery operations. Water from the air scrubber systems is combined with liquid wastes and the fluoride content precipitated with lime as calcium fluoride ( $\text{CaF}_2$ ) which is filtered and stored on site. The annual fuel requirement for a LWR results in production of about 22 tonnes of solid  $\text{CaF}_2$ , occupying a volume of about  $7 \text{ m}^3$ ; this contains a total of about 0.2 curie of uranium [USAEC 1974a].

The small percentage of fluorine not removed by lime treatment is diluted and discharged at a low concentration. Very low quantities of uranium are released in the effluent gases and liquids.

## 8. LIGHT WATER REACTORS

Reactor operation does not strictly form part of UFC cycle but is discussed briefly.

There are two basic types of light water reactor - the boiling water reactor (BWR) and the pressurised water reactor (PWR). These systems are fundamentally different. In the BWR, steam generated directly from the primary water coolant in the reactor vessel is used to drive the turbine generators producing electricity (Figure 9); in the PWR, the primary coolant is maintained as water at a high pressure, and steam to drive the turbines is generated in a secondary circuit isolated from the primary reactor coolant (Figure 10).

### 8.1 Effluents from Operation of LWRs

Nuclear reactors generate radioactive materials

- . in the fuel as a consequence of fission of uranium and plutonium.
- . in the coolant and in structural materials by neutron absorption leading to induced activity.

Activity directly induced in the coolant by activation of water and air generally consists of gases (argon-41, fluorine-18, nitrogen-13 and -16, and oxygen-19) which have short half-lives. Activity of considerably longer half-life may enter the coolant from corrosion or erosion of structural materials comprising active isotopes of zinc, manganese, nickel, iron, cobalt, chromium and copper. The coolant may also receive fission product activity from traces of uranium on the outer surface of the fuel cladding, or through release of fission products through defects in cladding integrity; this activity may contain isotopes of krypton, xenon, caesium, rubidium, barium, strontium, iodine and bromine.

### 8.2 Treatment of Reactor Effluents

#### 8.2.1 Gaseous effluent

The main gases encountered are krypton and xenon isotopes, and halogens including radioiodine [Carlson 1974].

Treatment methods involve

- . hold-up for decay of activity in tanks or on charcoal absorbers or in cryogenic distillation units;
- . filtration of particulate matter from gas streams; and
- . recombination of dissociated hydrogen and oxygen; this

is most significant in BWR plants where an off-gas flow of  $5 \text{ m}^3\text{m}^{-1}$  can be reduced to the condenser air in-leakage flow of about  $0.5 \text{ m}^3\text{m}^{-1}$ .

### 8.2.2 Liquid effluent

In both PWRs and BWRs, treatment processes for liquid wastes handle fission products and activated corrosion products which are bled, or leak, from the system. The trend in current reactor design is to recycle up to 90% of the liquid and to discharge the remainder within authorisation limits [Carlson 1974].

Treatment methods in both reactor types include

- . hold up for decay of short-lived isotopes;
- . filtration to remove suspended solids;
- . demineralisation with ion-exchange resin to remove a large proportion of the activity, except for tritium or dissolved krypton or xenon;
- . evaporation to separate some dissolved solids; and
- . centrifugation to dewater sludge from evaporators in preparation for solidification and storage.

### 8.2.3 Solid wastes

Solid wastes such as ion-exchange resins, evaporator concentrates and contaminated non-combustible waste are disposed of in a fixing agent (cement, bitumen) in drums and transported off site for storage.

## 8.3 Activity in Reactor Effluents

Comparison of measured releases of activity from some old and newer BWR and PWR reactors with generating capacities exceeding 400 MWe which were fully operational during 1973 showed substantial variation [USNRC 1974]. This variation of releases reflects the improvements made in waste retention systems in modern reactors.

	<u>Gaseous effluent</u>	<u>PWR</u>	<u>BWR</u>
(a)	Kr and Xe Ci $\text{y}^{-1}$	32 - 576 000	79 000 - 900 000
(b)	Halogens and particulates Ci $\text{y}^{-1}$	$10^{-4}$ - 1.8	0.1 - 31
(c)	Tritium Ci $\text{y}^{-1}$	0.1 - 269	0 - 27

	<u>Liquid effluent</u>	<u>PWR</u>	<u>BWR</u>	
(a)	Mixed fission and corrosion products	Ci y <sup>-1</sup>	<10 <sup>-3</sup> - 28	10 <sup>-4</sup> - 41
(b)	Tritium	Ci y <sup>-1</sup>	154 - 4 070	0.1 - 47

#### Solid wastes

Typical annual waste volumes for an LWR in 1973 were generally several hundreds of cubic metres, containing up to several thousand curies of radioactivity.

### 9. REPROCESSING OF IRRADIATED FUEL

Irradiated fuel discharged from LWRs is reprocessed to recover plutonium and residual uranium for return into the fuel cycle. Reprocessing additionally enables the highly radioactive waste products from the fission reaction to be converted into a compact, stable, solid form for decay storage or ultimate disposal [Buckham 1975].

#### 9.1 Process Description

Irradiated fuel elements are stored at the reactor site for about 90 days to allow fission products of short half-life to decay. The elements are then transported to the reprocessing plant in heavily shielded casks (Figure 11), reinforced to retain activity in the event of a transport accident, and designed for dissipation of the heat from fission product decay, which may exceed 25 kilowatts per tonne of irradiated uranium.

The fuel is stored for a total period of at least 150 days from reactor discharge, to permit decay of iodine-131 before reprocessing. The present lack of commercial reprocessing capacity for oxide fuel in the Western World has resulted in a buildup of fuel which will be stored for periods much greater than 150 days before it is reprocessed. This situation has arisen because existing reprocessing plants are shut down for modifications to increase capacity and/or improve safety features while new plants are still being constructed. A simplified block diagram of reprocessing operations is shown in Figure 12. The fuel elements are chopped into short pieces to expose the irradiated fuel. The fuel is leached into boiling nitric acid, and the solution is removed for chemical treatment. The residual chopped 'hulls' of Zircaloy or steel tubing, are washed with nitric acid and water to remove adherent fuel in solution, and the washings combined with the main product of leaching.

The solution of dissolved fuel is sampled and the quantity of contained uranium and plutonium determined. The solution is treated by counter-current

solvent extraction and ion-exchange processes to separate the fission products, uranium and plutonium into three streams. The purified product of uranium nitrate is converted into  $UF_6$  and is transported to a plant for enrichment in the  $^{235}U$  isotope and recycle into fuel manufacture. The purified plutonium nitrate product is stored for conversion into plutonium dioxide ( $PuO_2$ ) for use in fast breeder reactor fuel elements, or for manufacture of mixed oxide fuel ( $UO_2$ - $PuO_2$ ) for LWRs.

## 9.2 Effluents from Fuel Reprocessing

Reprocessing gives rise to several different waste streams, of which the most significant in terms of biological hazard is the raffinate from the first cycle of solvent extraction. This raffinate contains about 99.8% of the fission products present in the dissolved fuel and is termed 'high level' waste. After concentration, the high level waste stream may contain up to several megacuries of activity per cubic metre.

Reprocessing also gives rise to solid, liquid and gaseous wastes containing less than 0.2% of the fission products in a large volume relative to the high level waste; these wastes have been arbitrarily termed 'intermediate', and 'low' level, depending on the concentration of contained activity. Procedures for management of wastes from fuel reprocessing have been reviewed by Hardy & Hespe [1972].

Solid wastes from reprocessing include the metal cladding of the fuel elements which was activated by the neutron flux of the reactor and which also contains traces of undissolved irradiated fuel. Other solid wastes are used ventilation filters and discarded ion-exchange materials, contaminated equipment, tools, and protective clothing. Intermediate level solid waste typically contains activity at concentrations in the range  $350$ - $35,000$  curies  $m^{-3}$ ; wastes with lower concentration of activity are termed low level [Keher & Miles 1971]. Intermediate level solid wastes are either stored in shielded silos or are reduced in volume by incineration or compaction and encapsulated in concrete or bitumen before burial; low level solid waste may be treated similarly or buried without treatment. Intermediate and low level solid wastes produced in reprocessing the annual fuel requirements for a 1 000 MWe LWR may occupy a volume of  $20$ - $60m^3$  [USAEC 1974a].

Low and intermediate level liquid wastes are produced by chemical treatments other than the first cycle of solvent extraction; these wastes are usually concentrated by evaporation and blended with the high level waste stream, while the radioactivity in the distillate may be reduced by ion exchange to a level acceptable for discharge into a storage

lagoon or to the environment, for example, into coastal waters.

Gaseous wastes arise from vessel and cell ventilation extract systems; the contained activity consists of the fission product noble gases (notably krypton-85), tritium and radioiodine, together with an aerosol containing mixed fission products. Gaseous wastes are discharged to atmosphere through off-gas treatment and filtration systems to remove substantially all radioiodine and entrained aerosol activity. Reprocessing plants up to the present have discharged all the krypton and the bulk of tritium contained in the fuel into the biosphere, where dilution and dispersion occurs. With the growth of nuclear power, the increased quantities released could be undesirable, and methods for removal of the activity are being developed [USEPA 1974b].

#### 10. TREATMENT AND STORAGE OF HIGH LEVEL WASTE

The high level waste from modern fuel reprocessing plants consists of a nitric acid solution containing the fission products and transuranium elements other than plutonium, together with up to 1% of uranium and plutonium not recovered by the process.

The solution is initially concentrated for short term storage in liquid form at the reprocessing plant. About 29 tonnes of irradiated uranium fuel discharged annually from a 1000 MW(e) LWR produces approximately 33 m<sup>3</sup> of high level waste concentrate. This could contain about 130 million curies of fission product activity if the fuel is reprocessed at 150 days after discharge from the reactor [ORNL 1970]. The solution also contains about 300 kg of uranium and 3 kg of plutonium. The concentrate from some older reprocessing plants for the US military program was neutralized and stored in carbon steel tanks inside concrete vaults, buried in the ground. Twenty of these tanks have leaked and over 120 000 curies of activity has been absorbed in the soil surrounding the tanks [Lennemann 1972]. The first commercial plant for reprocessing power reactor fuels in the USA stored neutralized concentrate in mild steel tanks of improved design, for which no leakage has been reported. Reprocessing plants in the United Kingdom and France have stored acidic concentrate in stainless steel tanks without leakage. The trend in modern reprocessing plants is to store the acidic waste in stainless steel tanks which are located in concrete cells with walls up to 2 m in thickness for radiation shielding. The cell floors and part of the walls are lined with stainless steel, forming a sump for retention of spillage in the event of tank failure. The tanks are provided with cooling systems to

remove fission product heat which may amount to  $16 \text{ kW m}^{-3}$  in fresh concentrate, falling to about one third of this value after a year's storage in the tank. A typical storage tank and containment cell are shown in Figure 13.

The practice of storing high level waste in liquid form demands high standards of construction and inspection for the storage tanks. Although structural failure of tanks built to modern specifications is considered unlikely, the possibility that leakage may occur from some tanks during their operational lifetime cannot be excluded. The system relies on multiple containment, provision of spare tankage, and continual surveillance to prevent spread of activity in the event of tank failure.

Processes have been developed to convert the high level wastes into a calcined solid or a glass for encapsulation in steel canisters, to reduce the risk of activity leakage inherent in storage of liquid waste. One year's operation of a 1000 MW(e) LWR would produce up to six tonnes of solidified waste [USAEC 1974b] occupying a volume of about  $2 \text{ m}^3$ , equivalent to 10 storage canisters each 3 m high by 30 cm in diameter (Figure 14).

Under existing USA Federal Regulations all high level liquid waste must be solidified within five years of its production, and the canisters of waste transferred into a national repository for storage in a retrievable form within ten years of production of the waste in liquid form. No such repository exists at present.

The period of storage of liquid waste allows fission product activity to decay and can simplify operations and reduce the cost of solidification. However, Clelland [1974] noted that about 45% of the cumulative activity from a large reprocessing program would be present in the liquid wastes stored for one year before solidification, increasing to 75% for a three-year pre-solidification period. Solidification must therefore be effected with minimum delay to make a significant improvement in the safety of the overall system.

The USA intends to store solidified high level waste in retrievable form until a permanent method for removal of the wastes from the environment is established. An intended lifetime of up to 100 years for retrievable storage has been proposed [USAEC 1974a]. Proposals being considered for a Retrievable Surface Storage Facility (RSSF) include storage of waste canisters in individual shielded flasks (Figure 15) or in a shielded structure (Figure 16). In the latter case fission product heat could be

removed from the canisters by air convection or by their immersion in a water pond.

The storage period required for radioisotopes to decay into stable elements depends on the half-lives of the isotopes and of the successive radioisotopes produced by decay until stability is reached.

Tables 2 and 3 show respectively the radioactivity of major fission products and transuranium elements in high level waste resulting from the reprocessing of one tonne of irradiated uranium from a routine LWR discharge [ORNL 1970]. An initial activity of over 4 megacuries in the fresh waste from reprocessing will decay to about 30 kilocuries in 100 years, when the majority of the activity results from strontium-90 and caesium-137, and their decay products, yttrium-90 and barium-137. After 1000 years' storage, the activity will consist of about 20 curies of long-lived fission products with half-lives exceeding 200 000 years (technetium-99, zirconium-93, caesium-135 and iodine-129), and about 80 curies of transuranium isotopes of half-lives in the range 400-25 000 years (notably isotopes of neptunium, plutonium and americium). This activity could require over one million years for substantially complete decay to stable elements or isotopes of extremely long half-life (e.g. uranium).

It should be noted that this approximate 100 curies of activity remaining after a storage period of two years was derived from a quantity of uranium ore containing about 30 curies of activity.

#### 11. ULTIMATE DISPOSAL OF HIGH LEVEL WASTE

Alternatives for ultimate disposal of the high level waste are being studied intensively overseas and have been reviewed by Harries & Wilson [1975]. The options seen for permanent removal of the radioactivity in the waste from the terrestrial environment fall into three categories:

- (i) Burial of the waste in non-retrievable form in deep geological formations, in trenches on the sea-bed, or below the antarctic ice cap, for long-term decay of radioactivity.
- (ii) Transmutation of radioactive isotopes into stable nuclei by neutron bombardment.
- (iii) Extraterrestrial disposal to a solar orbit, solar impact, or escape to deep space.

##### 11.1 Irretrievable Burial of Waste

Disposal by irretrievable burial of solidified waste containing the

very long-lived radioisotopes appears one of the lower cost alternatives for waste disposal. A prerequisite for this system is the identification of a site which can be guaranteed to retain the waste until substantially all the active isotopes have decayed into stable or extremely long-lived elements, and this could require a period of storage measurable on a geological timescale. Burial in salt beds or shale deposits, or in deep holes drilled in some igneous or metamorphic rock formations, is being investigated. An artist's impression of an underground disposal system is shown in Figure 17. The repository would require to be sited in an area free from seismic or volcanic disturbances.

A major problem in this method lies in guaranteeing the stability of the site for extremely long periods on the basis of past events. Storage sites should not be of interest for other types of exploitation. Sites may be adversely affected by human endeavours; thus a salt mine in Kansas was rejected as a result of water ingress and salt erosion from mining oil, gas and salt [Harries & Wilson 1975]. A recent proposal has been made for the investigation of salt deposits in New Mexico for initial underground storage of wastes in retrievable form, with the longer term aim of conversion into a permanent repository [Pittman 1975].

Other burial proposals under consideration include disposal of the waste canisters into the Antarctic ice sheet, or into deep trenches in the sea-bed, where folding of the ocean floor could engulf the waste permanently. These systems require further research; the long-term existence of the ice sheet has been queried and information on the rates of ice movement within the sheet is sparse. Proposals involving disposal on the sea-bed require development of canister materials to resist corrosion by sea water and extensive environmental and other surveys to identify a suitable site.

Alternatively, if the long-lived fission products and transuranium isotopes were removed from the high level waste for special treatment, the residual activity would decay in about 1000 years (Table 2). This could substantially reduce the long-term risks and uncertainties for the bulk of the waste activity, and permit decay to stable isotopes to be effected in a shielded building; the Egyptian pyramids have existed for more than five times the period necessary to effect decay of the bulk of the activity, including the moderately long-lived strontium-90 and caesium-137. Removal of between 99.9 and 99.99% of the transuranium elements from the waste is thought to be required [USAEC 1974c]; it is not

certain whether this level of separation could be achieved with existing extraction technology, and further development of solvent extraction or ion-exchange systems would be essential for this management scheme.

The special treatment referred to for the separated long-lived fission products and transuranic elements could involve their transmutation ultimately into stable isotopes by recycling in a reactor, or their discharge from the earth into a space trajectory.

### 11.2 Transmutation of Waste

Radioactive isotopes of long half-life may be converted into short-lived isotopes by absorption of neutrons into their nuclei, thereby reducing the period of storage necessary for their decay. Transmutation of all the fission products to be produced by generation of power from nuclear fission is not practicable. Modern reactors, however, generate sufficient excess neutrons at a flux capable of effective transmutation of the transuranium elements and the very long-lived fission products, provided these are separated from the bulk of the fission product waste [Harries & Wilson 1975]. The activity in the residual waste would decay to a low level in 1000 years, and selective transmutation of wastes is a possible alternative to the selection of storage sites requiring guaranteed integrity for over a million years. The transuranium elements to be transmuted could contain up to 1% of the total fission products.

### 11.3 Extraterrestrial Disposal of Waste

Proposals for discharge of waste into space are being studied in the USA and include placement of some waste into orbit around the Earth, moon, and sun, and trajectories leading to solar impact or discharge of the waste from the solar system. Existing space technology could dispose of the very long-lived fission products and transuranium elements separated from the bulk of the waste, but it would not be practicable economically to dispose of all the high level waste in this way, and the residual activity would require storage for about 1000 years to decay [USAEC 1974c].

Failure of a launching mission could result in return of the waste to the environment through volatilisation in re-entry to Earth's atmosphere or by ground impact. A waste package featuring multiple encapsulation of the waste in an inert matrix has been designed to remain integral in the event of re-entry, but substantial testing is necessary to validate the concept. The waste for disposal could contain up to 1% of the bulk fission products.

#### 11.4 Summary - Ultimate Waste Disposal

Most of the proposals for ultimate disposal of the high level waste appear currently to be technically feasible and preliminary estimates of cost suggest that none of the disposal concepts being studied seems likely to increase the cost of nuclear electrical power by a major amount [USAEC 1974c]. However, stringent guarantees on safety and performance are required for the proposals, and the cost estimates remain to be validated.

All proposals which are irreversible require careful examination to resolve environmental uncertainties and conflicts of policy, and to produce a system which is safe and acceptable to international authorities.

The decision to operate a particular mode of ultimate waste disposal should not be taken under pressure to find an immediate solution through the need for power generation. However, nuclear power stations are in operation, and substantial quantities of high level waste have already been produced. The major advantage of the engineered storage project (Section 10) for receipt of solidified waste is that the activity is retrievable, accountable, and available for whichever ultimate disposal route is finally taken [Alder 1975].

#### 12. QUANTITIES OF WASTES FROM THE URANIUM FUEL CYCLE

Table 4 shows estimates of the maximum annual UFC requirements of uranium and separative work for support of a modern light water reactor of 1000 MW(e) capacity [Plotnikoff 1975]. The annual requirements of the 'model' LWR were determined by averaging the requirements of PWRs and BWRs of similar generating capacity in a ratio of two to one respectively, which corresponds to approximately the ratio of the reactors being installed. The 'lifetime average' annual fuel requirement inclusive of initial core and annual reloads was assessed on the basis of a 40 y reactor lifetime.

These requirements were used to revise data [USAEC 1974a] on the waste arisings for each stage of the fuel cycle. The quantities of chemical effluents are listed in Table 5, and the corresponding activities in Table 6.

The bulk of the chemical effluents discharged to the environment are oxides of sulphur and nitrogen, and particulate emission from combustion of fossil fuels providing energy of the fuel cycle processes. The power-intensive enrichment stage accounts for over 97 per cent of these fossil combustion wastes.

Solid tailings and raffinate solution from processing of uranium ores represent the largest sources of solid and liquid effluent respectively; however, these effluents are neutralized and stored in a system designed to retain all solids and minimise seepage of liquid to the environment.

The greatest release of radioactivity from the fuel cycle into the environment consists of the krypton and tritium discharged in gaseous and liquid effluents from reprocessing of irradiated fuel. More than 99.8 per cent of radioactivity present in all effluents from the cycle is contained in the high level solid wastes, and the irradiated fuel cladding material, which are isolated from the environment.

### 13. FINAL COMMENTS

The preceding sections have attempted to place in perspective the procedures of waste management for each sector of the uranium fuel cycle and of the light water reactors served by the cycle. These procedures are continually being reviewed and improved to reduce the impact of the wastes on the environment.

Compliance with waste management criteria is effected by a regulatory and licensing process. Proposed plant designs and operating procedures are examined before plant construction through the production of an environmental impact statement. Plant performance during operating life is periodically reviewed and necessary changes to plant equipment and operating procedures recommended and implemented.

### 14. ACKNOWLEDGEMENTS

The author wishes to acknowledge a contribution by Mr. W.W. Plotnikoff in estimation of the fuel cycle requirements for LWRs, and also helpful discussions with Mr. R.W. Carlson on the management of wastes arising during reactor operation.

### 15. ABBREVIATIONS

LWR	Light Water Reactor
UFC	Uranium Fuel Cycle
NT	Northern Territory of Australia
CCD	Counter-current decantation
RSSF	Retrievable surface storage facility
U <sub>3</sub> O <sub>8</sub>	Tri uranium octoxide, 'yellowcake'
UO <sub>2</sub>	Uranium dioxide
UF <sub>4</sub>	Uranium tetrafluoride
UF <sub>6</sub>	Uranium hexafluoride
UO <sub>2</sub> F <sub>2</sub>	Uranyl fluoride

ADU	Ammonium diuranate
HF	Hydrogen fluoride
NH <sub>3</sub>	Ammonia
CaF <sub>2</sub>	Calcium fluoride
PuO <sub>2</sub>	Plutonium dioxide

## 16. REFERENCES

- Alder, K.F. [1975] - Nuclear Energy Resources for Electrical Power Generation. Atomic Energy in Australia, 18, (1)2.
- Alfredson, P.G. [1972] - Review of methods and technology for the production of uranium hexafluoride. AAEC Symposium on Uranium Processing, Lucas Heights, 20-21 July, AAEC/E238.
- Buckham, J.A. [1975] - 'Why Reprocess Nuclear Fuels?' Chemical Engineering Progress 71, (3) 21.
- Carlson, R.W.S. [1974] - A comparison of PWR and BWR radwaste systems. AAEC internal document.
- Clelland, D.W., [1974] - Present methods of storing highly radioactive waste in the United Kingdom and proposals for the future, Ninth World Energy Conference, Detroit, 22-27 September.
- Hardy, C.J. & Hespe, E.D. [1972] - Management of wastes from irradiated nuclear fuels. Paper presented to the Symposium on the Role of Nuclear Energy in Australia's Development, Canberra, 1-2 June, AAEC/TM628.
- Harries, J.R. & Wilson, D.J. [1975] - The storage and disposal of high level radioactive waste: A critical review. AAEC internal report No. NSTB/PE6.
- Hartley, F.R. [1972] - Conventional processes to produce yellowcake. AAEC Symposium on Uranium Processing, Lucas Heights, 20-21 July, AAEC/E238.
- Keher, L.H. & Miles, G.L. [1971] - The nature and disposal of radioactive wastes. Proc. Australian Waste Disposal Conference, University of New South Wales, Sydney, August 1967.
- Lennemann, W.L. [1972] - Management of radioactive aqueous wastes from US Atomic Energy Commission's fuel reprocessing operations: experience and planning. Proceedings of a Symposium on the Management of Radioactive Wastes from Fuel Reprocessing. OECD-OCDE, Paris, 27 November-1 December.

- Merritt, R.C. [1971] - 'The extractive metallurgy of uranium'. Colorado School of Mines Research Institute.
- Nakajima, K. [1972] - Management of fuel reprocessing wastes in Japan, Proceedings of a Symposium on the Management of Radioactive Wastes from Fuel Reprocessing, OECD-OCDE, Paris, 27 November-1 December.
- ORNL [1970] - Siting of fuel reprocessing plants and waste management facilities. Compiled and edited by ORNL, July, ORNL-4451.
- Pittman, F.K. [1975] - Testimony to the Joint Congressional Committee on Atomic Energy for Fiscal Year 1975, 4-5 March 1974, US Government Printing Office, Washington, Part 4 p 1921.
- Plotnikoff, W.W. [1975] - Internal Communication, AEC.
- Ranger Uranium Mines Pty. Limited [1974] - Environmental Impact Statement, February.
- Rogers, W.M. [1971] - Nuclear fuel reprocessing requirements in the Western United States, Western Interstate Nuclear Board, December.
- Tsivoglou, E.C. & O'Connell, R.L. [1962] - Waste guide for the uranium milling industry, U.S. Department of Health, Education and Welfare, Public Health Service, and Division of Water Supply and Pollution Control.
- USAEC [1974a] - Environmental survey of the uranium fuel cycle.  
USAEC Fuels and Materials Directorate of Licensing, WASH-1248.
- USAEC [1974b] - The nuclear industry, 1974. WASH-1174-74.
- USAEC [1974c] - High-level radioactive waste management alternatives, USAEC, May, WASH-1297.
- USAEC [1974d] - Everything you always wanted to know about shipping high-level nuclear wastes. USAEC August, WASH-1264.
- USEPA [1973a] - Environmental analysis of the uranium fuel cycle. Part I - fuel supply, US Environmental Protection Agency, October, EPA-520/9-73-003-B.
- USEPA [1973b] - Environmental analysis of the uranium fuel cycle. Part III - nuclear fuel reprocessing, USEPA, October, EPA-520/9-73-003-D.
- USEPA [1974] - State of the art. Uranium mining, milling and refining industry, National Environmental Research Center, Office of Research and Development, US Environmental Protection Agency, June.

USNRC [1974] - Summary of radioactivity released in effluents from  
nuclear power plants in 1973. U.S. Nuclear Regulatory  
Commission, NUREC 75-001.



TABLE 1  
URANIUM RADIOACTIVE DECAY SERIES  
(Minor branches not shown)

Isotope	Radiation	Half-life	Remarks
<sup>238</sup> Uranium 92	α, γ	4.5 x 10 <sup>9</sup> y	
↓			
<sup>234</sup> Thorium 90	β, γ	24.1 d	β-rays in uranium metal and ore come from these
↓			
<sup>234</sup> Protoactinium 91	β, γ	1.14 min	
↓			
<sup>234</sup> Uranium 92	α, γ	2.5 x 10 <sup>5</sup> y	
↓			
<sup>230</sup> Thorium 90	α, γ	80 000 y	
↓			
<sup>226</sup> Radium 88	α, γ	1620 y	
↓			
<sup>222</sup> Radon * 86	α	3.83 d	gaseous
↓			
<sup>218</sup> Polonium * 84	α	3.05 min	collects on dust in mine
↓			
<sup>214</sup> Lead 82	β, γ	26.8 min	β-and γ-rays in mine come from these
↓			
<sup>214</sup> Bismuth 83	β, γ	19.7 min	
↓			
<sup>214</sup> Polonium * 84	α	1.64 x 10 <sup>-4</sup> s	collects on dust in mine
↓			
<sup>210</sup> Lead 82	β	22 y	
↓			
<sup>210</sup> Bismuth 83	β	5 d	
↓			
<sup>210</sup> Polonium * 84	α	140 d	
↓			
<sup>206</sup> Lead 82 (stable)	-	-	

\* Constitutes the α hazard in mining, etc.

**TABLE 2**  
**RADIOACTIVITY OF MAJOR FISSION PRODUCT ISOTOPES IN WASTES FROM**  
**REPROCESSING ONE TONNE OF IRRADIATED URANIUM FROM A**  
**LIGHT WATER REACTOR\***  
**[ORNL 1970]**

Isotope	Half-life	Radioactivity of fission products (Ci t <sup>-1</sup> U)**			
		Time after discharge from reactor			
		150 days	10 years	100 years	1,000 years
tritium	12.3 y	690	400	2.5	0
krypton-85	10.76 y	1.1 x 10 <sup>4</sup>	6.1 x 10 <sup>3</sup>	18.0	0
strontium-89	51 d	9.6 x 10 <sup>4</sup>	0	0	0
strontium-90	28 y	7.6 x 10 <sup>4</sup>	6.0 x 10 <sup>4</sup>	6.5 x 10 <sup>3</sup>	1.4 x 10 <sup>-6</sup>
yttrium-90	2.60 d	7.6 x 10 <sup>4</sup>	6.0 x 10 <sup>4</sup>	6.5 x 10 <sup>3</sup>	1.4 x 10 <sup>-6</sup>
yttrium-91	59 d	1.6 x 10 <sup>5</sup>	0	0	0
zirconium-95	65 d	2.7 x 10 <sup>5</sup>	0	0	0
niobium-95	35 d	5.2 x 10 <sup>5</sup>	0	0	0
technetium-99m	2 x 10 <sup>5</sup> y	14.0	14.0	14.0	14.0
ruthenium-103	40 d	8.9 x 10 <sup>4</sup>	0	0	0
rhodium-103m	56 min	8.9 x 10 <sup>4</sup>	0	0	0
ruthenium-106	1 y	4.1 x 10 <sup>5</sup>	550.0	0	0
rhodium-106	2 h	4.1 x 10 <sup>5</sup>	550.0	0	0
antimony-125	2 y	8.1 x 10 <sup>3</sup>	690.0	0	0
iodine-131	8 d	2.2	0	0	0
caesium-134	2 y	2.1 x 10 <sup>5</sup>	8.3 x 10 <sup>3</sup>	0	0
caesium-136	13 d	20	0	0	0
barium-137m	2 min	1.0 x 10 <sup>5</sup>	8.0 x 10 <sup>4</sup>	1.0 x 10 <sup>4</sup>	0
caesium-137	30 y	1.0 x 10 <sup>5</sup>	8.0 x 10 <sup>4</sup>	1.0 x 10 <sup>4</sup>	9.3 x 10 <sup>-6</sup>
barium-140	12.8 d	430.0	1.0 x 10 <sup>-3</sup>	0	0
lanthanum-140	1.6 d	500.0	0	0	0
cerium-141	32.5 d	5.7 x 10 <sup>4</sup>	0	0	0
cerium-144	285 d	7.7 x 10 <sup>5</sup>	150.0	0	0
praseodymium-144	17 min	7.7 x 10 <sup>5</sup>	150.0	0	0
promethium-147	2.6 y	9.9 x 10 <sup>4</sup>	7.8 x 10 <sup>3</sup>	0	0
others		1.3 x 10 <sup>5</sup>	2.1 x 10 <sup>4</sup>	1.0 x 10 <sup>3</sup>	7.0
<b>TOTAL</b>		<b>4.4 x 10<sup>6</sup></b>	<b>3.2 x 10<sup>5</sup></b>	<b>3.4 x 10<sup>4</sup></b>	<b>21.0</b>

\* Irradiation parameters: Burnup 33 000 MWD t<sup>-1</sup>U; Power 30 MW t<sup>-1</sup>U

\*\* A zero value is < 10<sup>-6</sup>

TABLE 3

RADIOACTIVITY OF MAJOR TRANSURANIC ISOTOPES IN WASTES FROM REPROCESSING  
ONE TONNE OF IRRADIATED URANIUM FROM A LIGHT WATER REACTOR\*  
 [ORNL 1970]

Isotope	Half-life	Radioactivity of transuranic isotopes (Ci t <sup>-1</sup> U)			
		Time after discharge from reactor			
		150 days	10 years	100 years	1000 years
neptunium-237	2.1 x 10 <sup>6</sup> y	0.5	0.5	0.5	0.6
neptunium-239	2 d	17.4	17.4	17.2	15.9
plutonium-238	88 y	35.7	102.0	51.9	0.1
plutonium-239	24 400 y	1.7	1.7	1.7	2.1
plutonium-240	6 500 y	2.4	4.5	8.9	8.2
plutonium-241	15 y	574.0	344.0	2.9	0.0
americium-241	433 y	172.0	176.0	162.0	41.5
americium-242	16 h	4.0	3.9	2.5	0.04
americium-243	7 400 y	17.4	17.4	17.2	15.9
curium-242	163 d	1.5 x 10 <sup>4</sup>	3.2	2.1	0.03
curium-244	17.6 y	2.5 x 10 <sup>3</sup>	1.7 x 10 <sup>3</sup>	55.0	0.0
others		320.6	33.8	11.2	0.2
TOTAL		1.8 x 10 <sup>4</sup>	2.4 x 10 <sup>3</sup>	330.0	84.5

\* Irradiation parameters: Burnup 33 000 MWD t<sup>-1</sup>U: Power 30 MW t<sup>-1</sup>U

TABLE 4  
MAXIMUM ANNUAL FUEL CYCLE REQUIREMENT FOR  
A MODERN 1000 MW(e) WATER REACTOR

	Initial core			Annual reload			Lifetime average annual fuel requirement LWR
	PWR	BWR	LWR	PWR	BWR	LWR	
Irradiation level $MWd_t/t U \times 10^{-3}$	22.6	17.0	19.4	32.6	27.5	29.8	
Fresh fuel assay wt.% uranium-235	2.26	2.03	2.15	3.21	2.73	2.97	
Spent fuel assay wt.% uranium-235	0.74	0.86	0.80	0.90	0.84	0.87	
Ore supply t ore $\times 10^{-3}$	174	208	185	66.7	64.7	66.0	69.0
Yellowcake t U	398	477	424	152.8	148.1	151.2	158.0
Natural + recycle $UF_6$ , t U	396	474	422	176.5	176.1	176.6	182.7
Separative work kg SWU $\times 10^{-3}$	177	197	184	96.9	76.1	90.0	92.4
Enriched $UF_6$ t U	83.0	112.7	92.9	26.6	31.4	28.2	29.8
Fuel loading t U	82.2	111.6	92.0	26.3	31.1	27.9	29.5
Reprocessed fuel t U	79.5	107.9	89.1	24.6	29.5	26.2	27.7

Basis of estimate

1. Reactor plant load factor 75%
2. Reactor thermal efficiency 32%
3. LWR requirements derived from averaging those of PWR and BWR in ratio 2:1
4. Ore grade - 0.3%  $U_3O_8$ ; recovery efficiency 90%.
5. Enrichment tails assay 0.3% Uranium-235
6. No plutonium recycle
7. Reloads include recovered uranium
8. Processing losses:  $UF_6$  production - 0.5%  
Fuel manufacture and fabrication 1%  
Fuel reprocessing - 1% U and Pu
9. Reactor life assumed - 40 y

TABLE 5

CHEMICAL AND THERMAL EFFLUENT FROM THE ANNUAL FUEL CYCLE REQUIREMENTS OF A 1000 MW(e) LWR

Operation	Mining	Milling	UF <sub>6</sub> Prod.	Enrichment	Fuel Fab.	Reprocessing	Reactor Operation	Total
<u>Effluents (t)</u>								
<u>Chemical</u>								
<u>Gases</u> (1)								
SO <sub>x</sub>	6.4	28.1	29.0 (3)	3420	19.3	5.2 (4)	-	3510
NO <sub>x</sub>	3.8	12.1	10.0 (2)	900	5.1	6.0	-	937
Hydrocarbons	0.2	1.0	0.8	9	0.05	0.02	-	11
CO	0.02	0.2	0.2	23	0.13	0.03	-	24
Particulates	-	7.4	7.6	900	5.1	1.4	-	922
<u>Other Gases</u>								
F	-	-	0.1	0.4	-	0.1	-	0.6
<u>Liquids</u>								
SO <sub>4</sub>	-	-	4.5	5.4	-	0.3	-	10.2
NO <sub>3</sub>	-	-	0.1	2.7	19.4	0.8	-	23.0
Fluoride	-	-	-	-	3.5	-	-	3.5
Ca	-	-	-	5.4	-	-	-	5.4
Cl	-	-	0.2 (5)	8.2	-	0.2	-	8.6
Na	-	-	3.9	8.2	-	4.5	-	20.1
NH <sub>3</sub>	-	-	-	-	8.4	-	-	8.4
Tailings Solutions	-	182 000	24 000 (6)	-	-	-	-	206 000
Fe	-	-	-	0.4	-	-	-	0.4
<u>Solids</u> (other than high level high level waste)	-	69 000	40	-	22	(20 - 60 m <sup>3</sup> ) 5.7	(< 300 m <sup>3</sup> )	< 73 000
<u>Thermal</u> GWh	-	15.0	6.0	746	2.0	15.0	13 960	14 740

(1) Estimated effluents based upon combustion of equivalent coal for power generation  
 (2) Combined effluents from combustion of coal and natural gas and process tankage; contains 0.2 MT of hexane  
 (3) 25% from natural gas use  
 (4) 77% from process  
 (5) Contains about 80% potassium  
 (6) 'Wet' process only

TABLE 6  
RADIOLOGICAL EFFLUENT FROM THE ANNUAL FUEL CYCLE REQUIREMENTS OF A 1000 MW(e) LWR

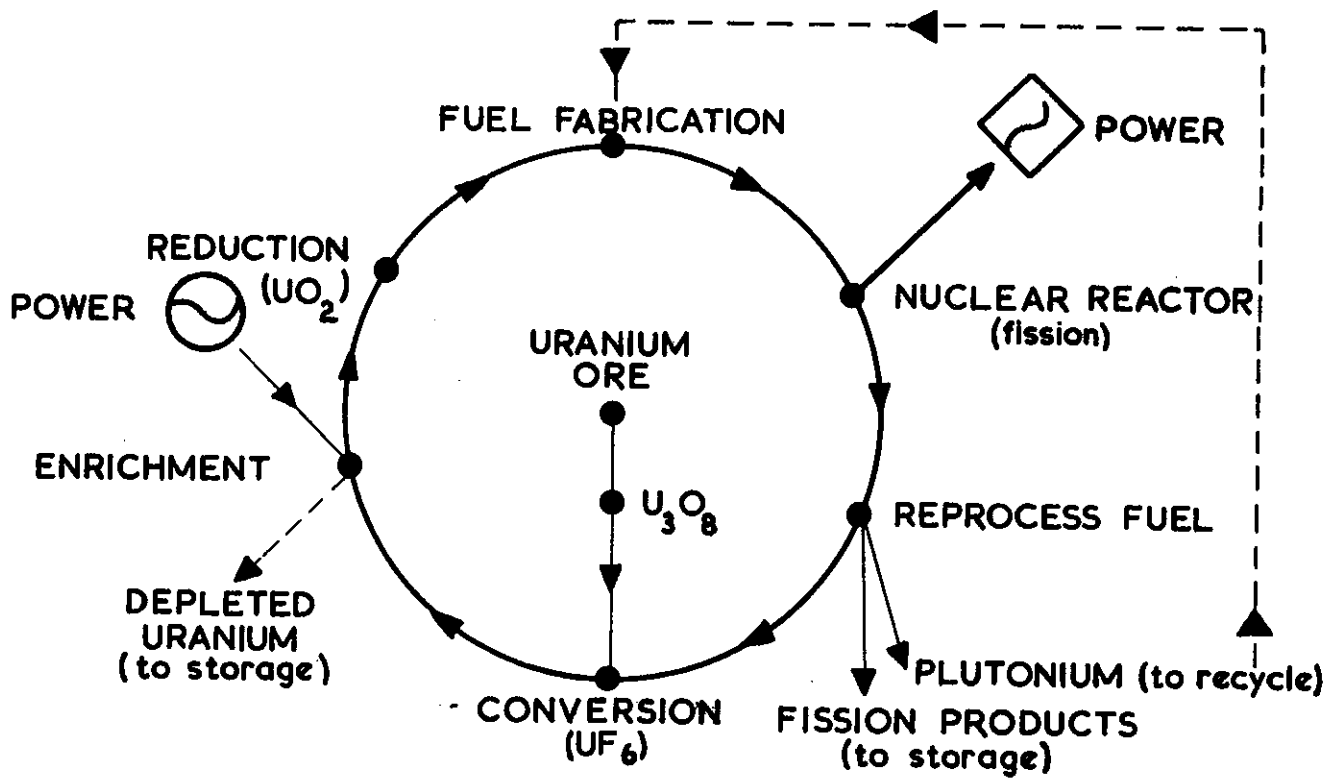
Operation	Mining	Milling	UF <sub>6</sub> Prod.	Enrichment	Fuel Fab.	Reprocessing	Reactor Operation	Total
<u>Effluents</u>								
<u>Radiological (curies)</u>								
Gases (including entrainment)								
Radon-222	-	76.3	-	-	-	-	-	76
Radium-226	-	0.02	-	-	-	-	-	0.02
Thorium-230	-	0.02	-	-	-	-	-	0.02
Uranium	-	0.03	0.0015	0.002	0.0002	-	-	0.03
Tritium	-	-	-	-	-	14 100	< 270	< 14 400
Noble gases (isotopes of Kr and Xe)	-	-	-	-	-	295 000	< 900 000	< 1.2 x 10 <sup>6</sup>
Iodine-129	-	-	-	-	-	0.002	-	< 31
Iodine-131	-	-	-	-	-	0.02	-	< 31
Fission Products	-	-	-	-	-	0.84	-	0.003
Transuranics	-	-	-	-	-	0.003	-	0.003
<u>Liquids</u>								
Uranium & Daughters	-	2.1	0.044	0.02	0.017	-	-	2.2
Radium-226	-	-	0.0034	-	-	-	-	0.003
Thorium-230	-	-	0.0015	-	-	-	-	0.002
Thorium-234	-	-	-	-	0.01	-	-	0.01
Tritium	-	-	-	-	-	2100	< 4100	< 6200
Ruthenium-106	-	-	-	-	-	0.11 <sup>(1)</sup>	< 41 <sup>(2)</sup>	< 41
<u>Solids (buried)</u>								
Other than high level	-	614 <sup>(3)</sup>	0.86	-	0.20	8 x 10 <sup>5(3,4)</sup>	< 3000	8 x 10 <sup>5(3,4)</sup>
High level waste	-	-	-	-	-	130 x 10 <sup>6(3)</sup>	-	130 x 10 <sup>6(3)</sup>

(1) Cs-137 (0.075 Ci/AFR) and Sr-90 (0.004 Ci/AFR) are also emitted

(2) Mixed fission and corrosion products

(3) Isolated from the environment

(4) Activity in fuel cladding

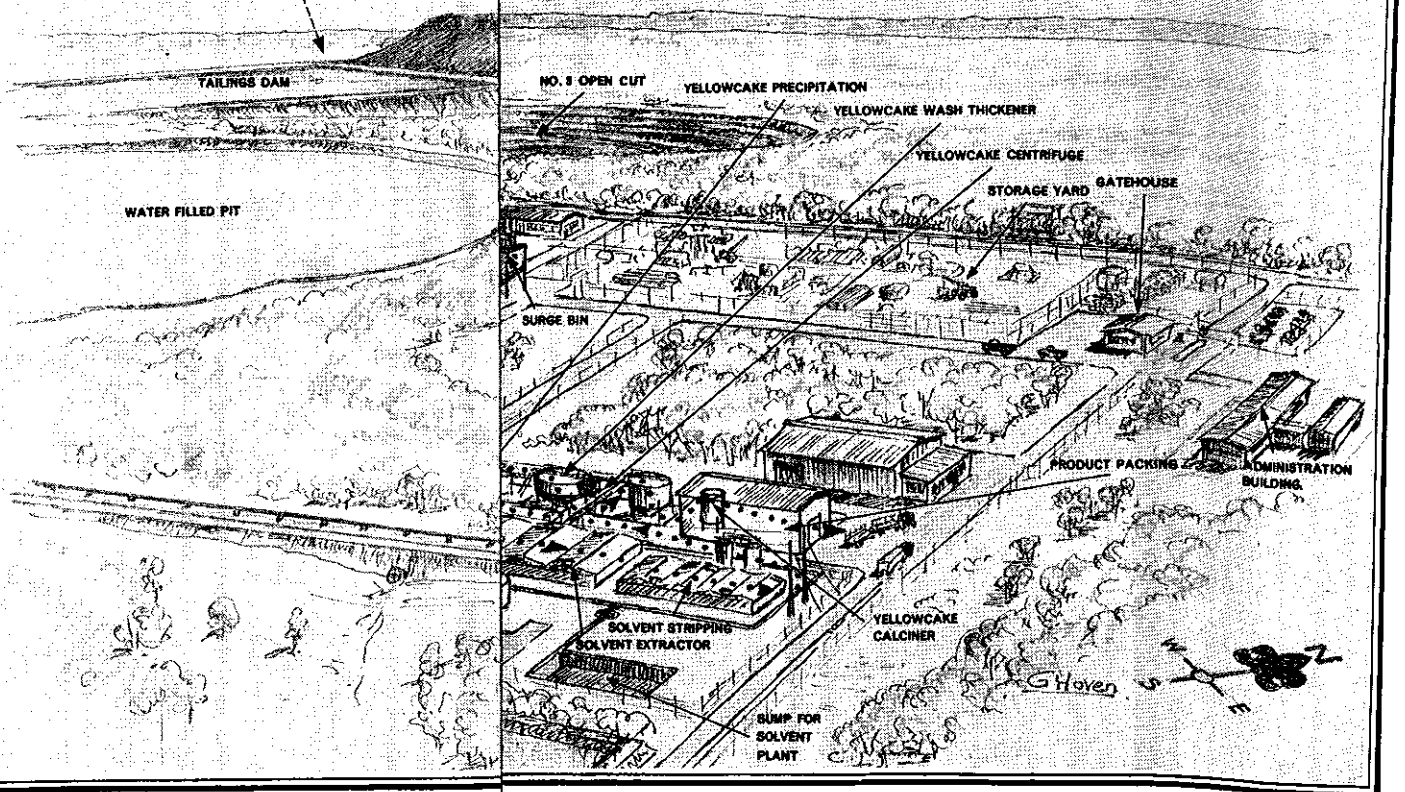


**FIGURE 1. THE URANIUM FUEL CYCLE**



**TAILINGS DAM**  
*Radon and dissolved radionuclides and heavy metals released to acceptable levels*

**OF NO. 3 OREBODY**  
*Radon emission of acceptable levels*



**IATED**



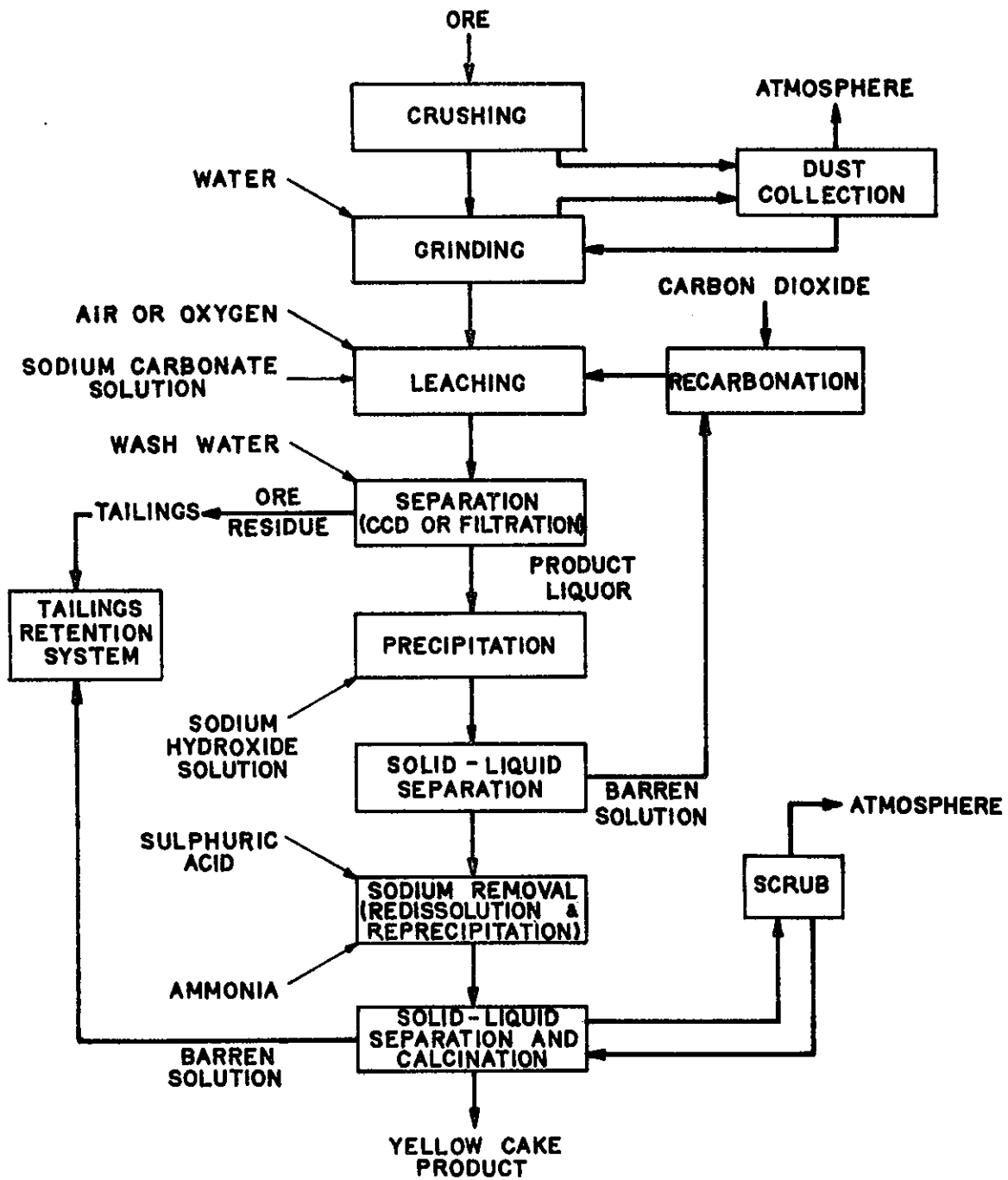
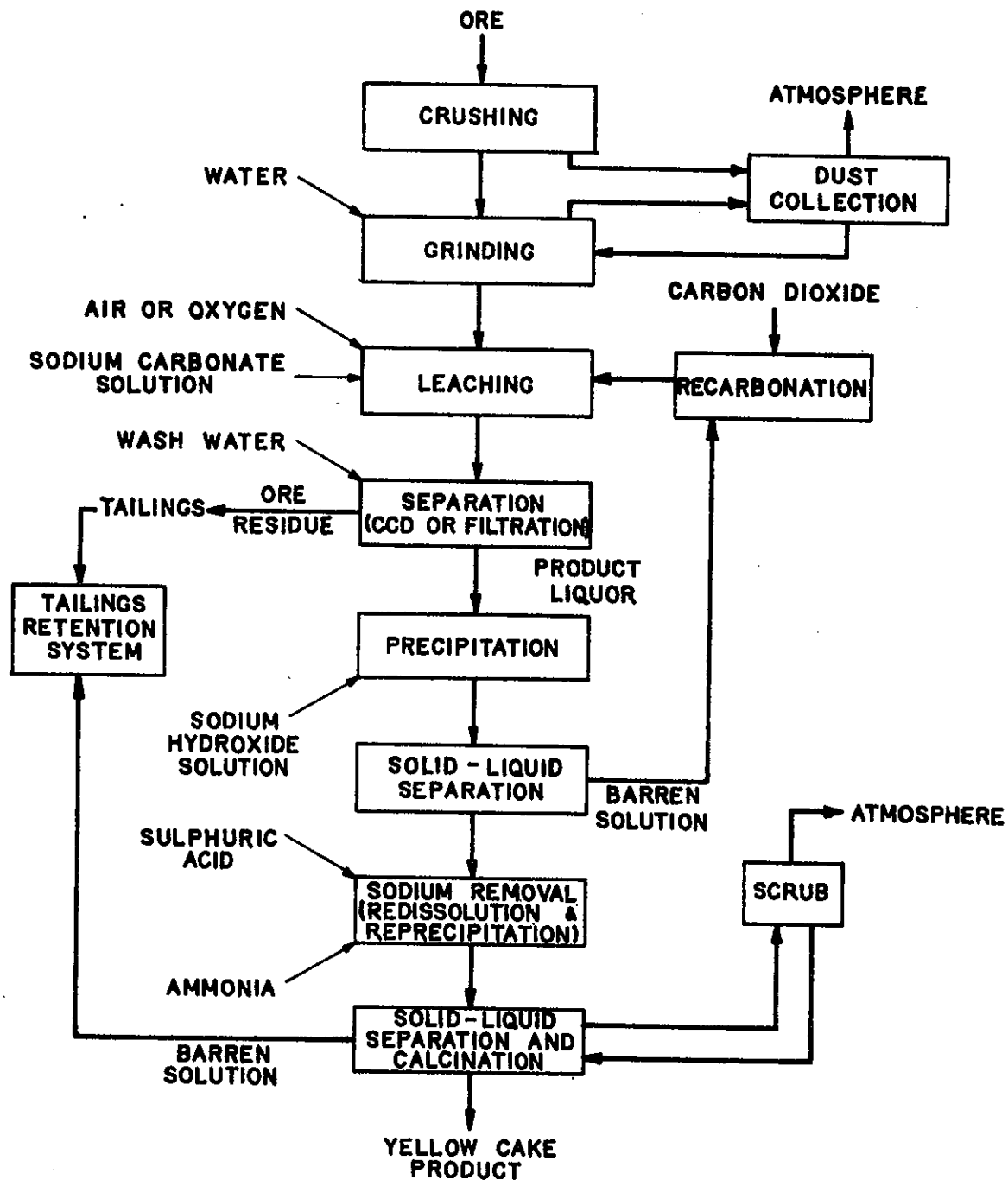


FIGURE 3. URANIUM MILL - ACID LEACH PROCESS SIMPLIFIED BLOCK FLOW DIAGRAM (USAEC 1974a)



**FIGURE 4. URANIUM MILL - CARBONATE LEACH PROCESS  
SIMPLIFIED BLOCK FLOW DIAGRAM  
(after Merritt 1971)**

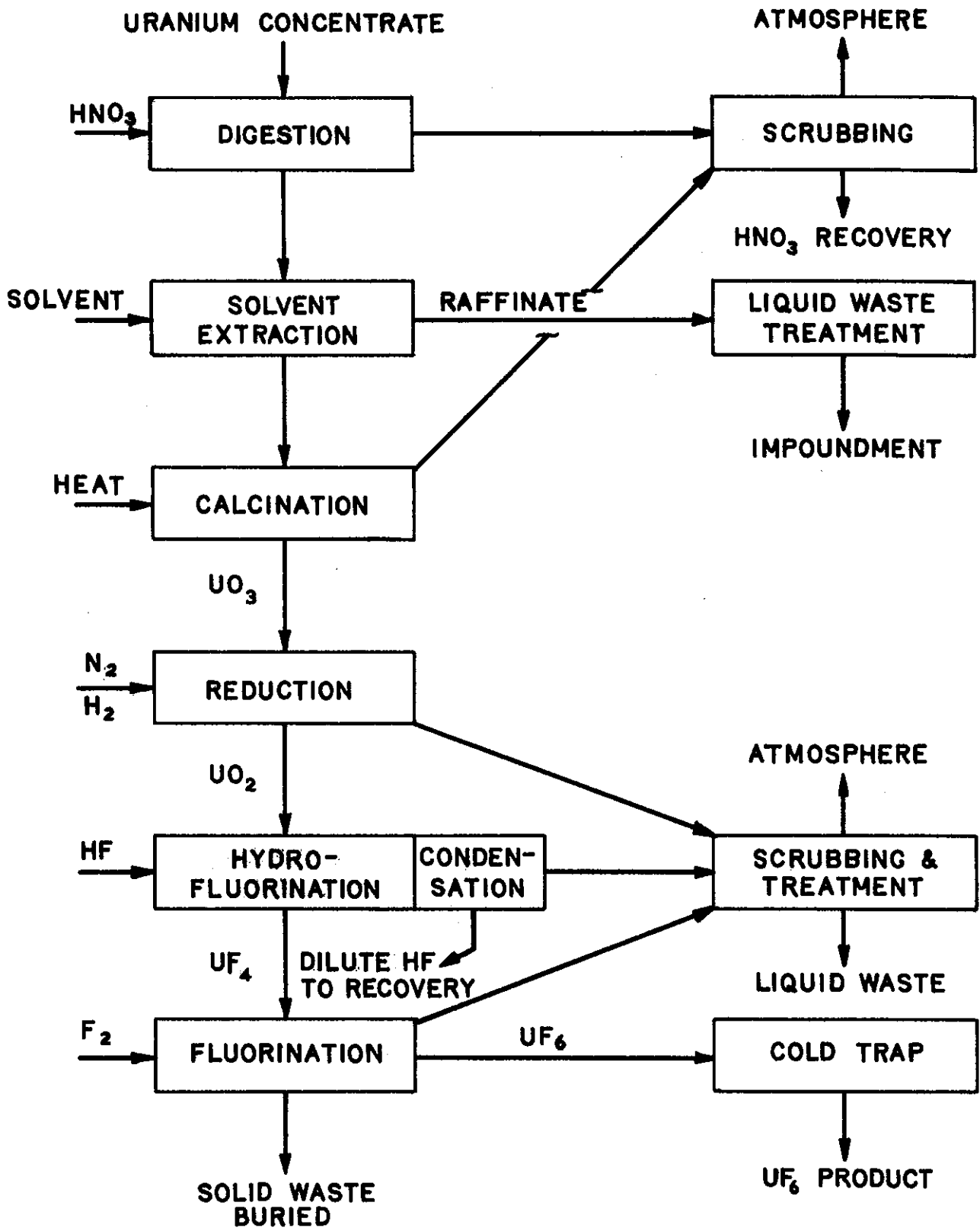


FIGURE 5.  $UF_6$  PRODUCTION - WET SOLVENT EXTRACTION  
SIMPLIFIED BLOCK FLOW DIAGRAM (USAEC 1974a)

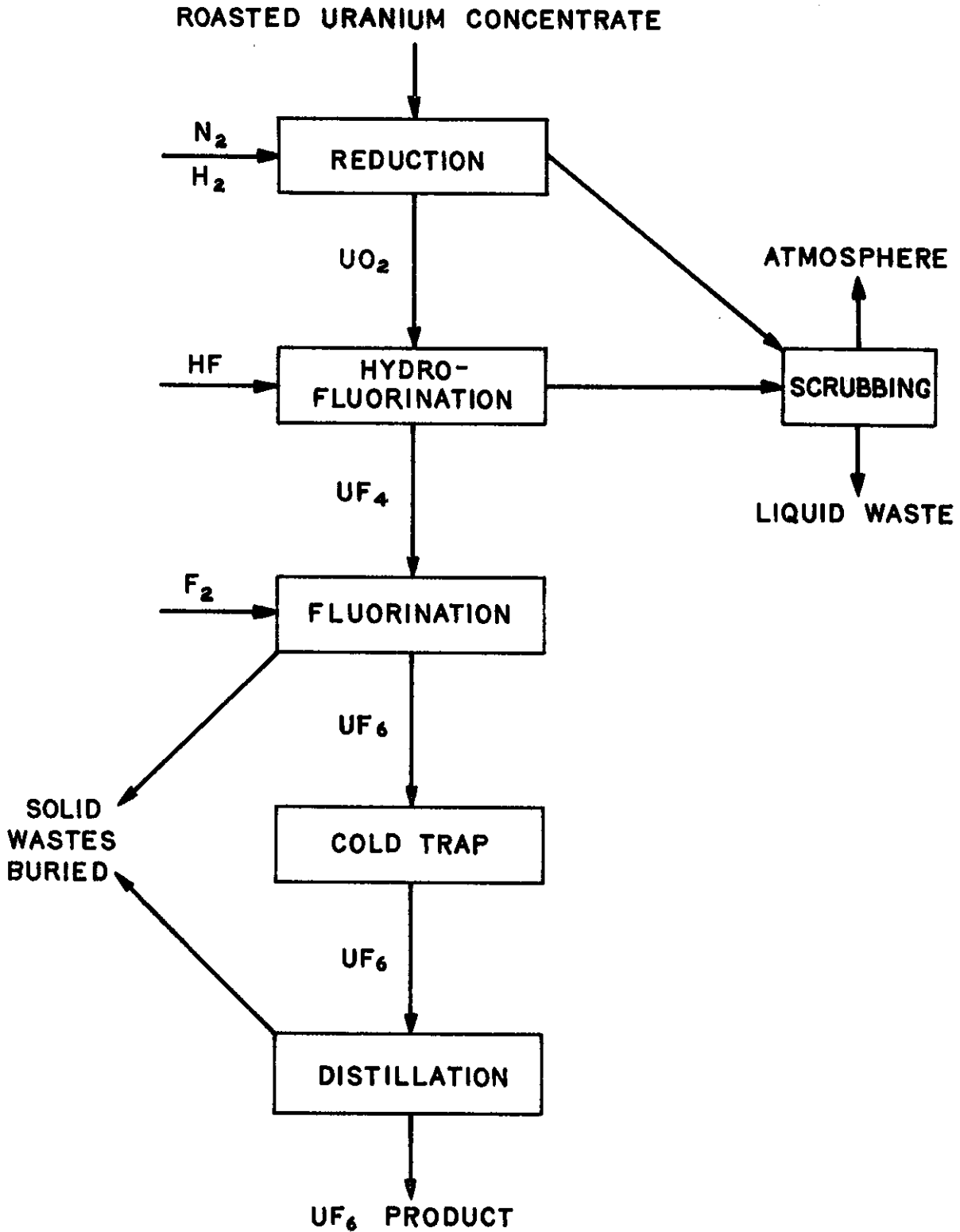
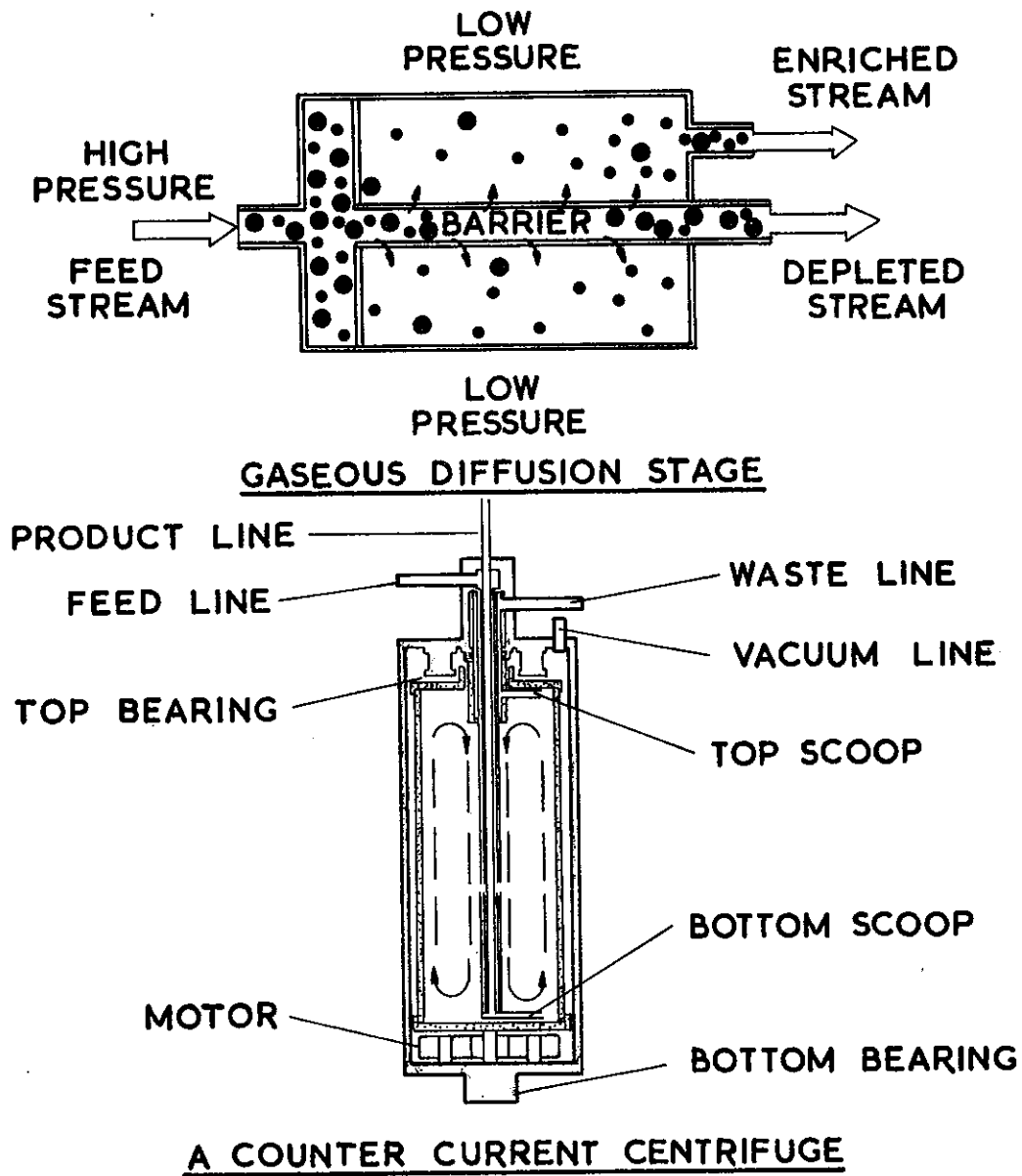


FIGURE 6.  $UF_6$  PRODUCTION - DRY HYDROFLUOR PROCESS  
SIMPLIFIED BLOCK FLOW DIAGRAM (USAEC 1974a)



**FIGURE 7. ENRICHMENT PROCESSES**

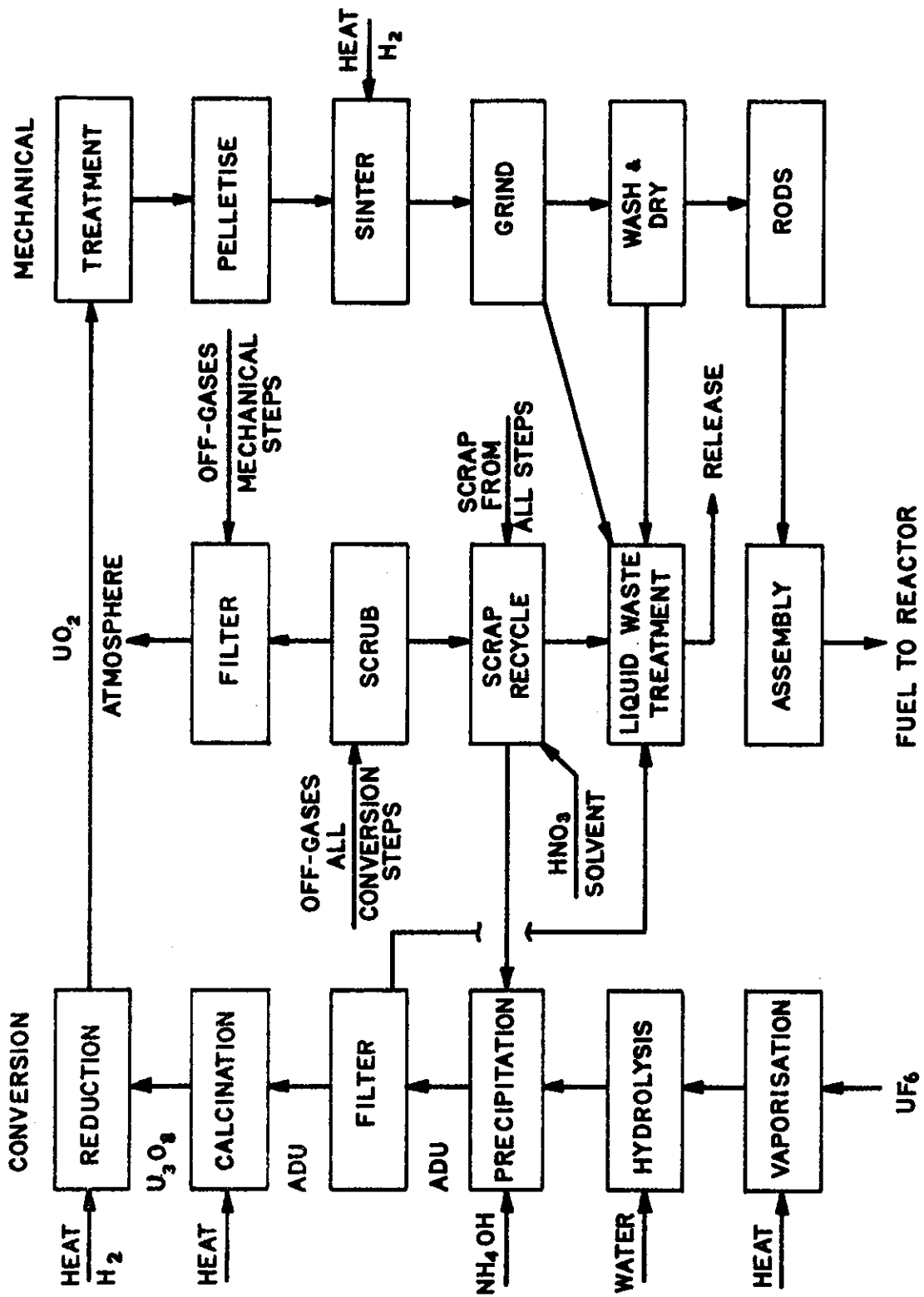
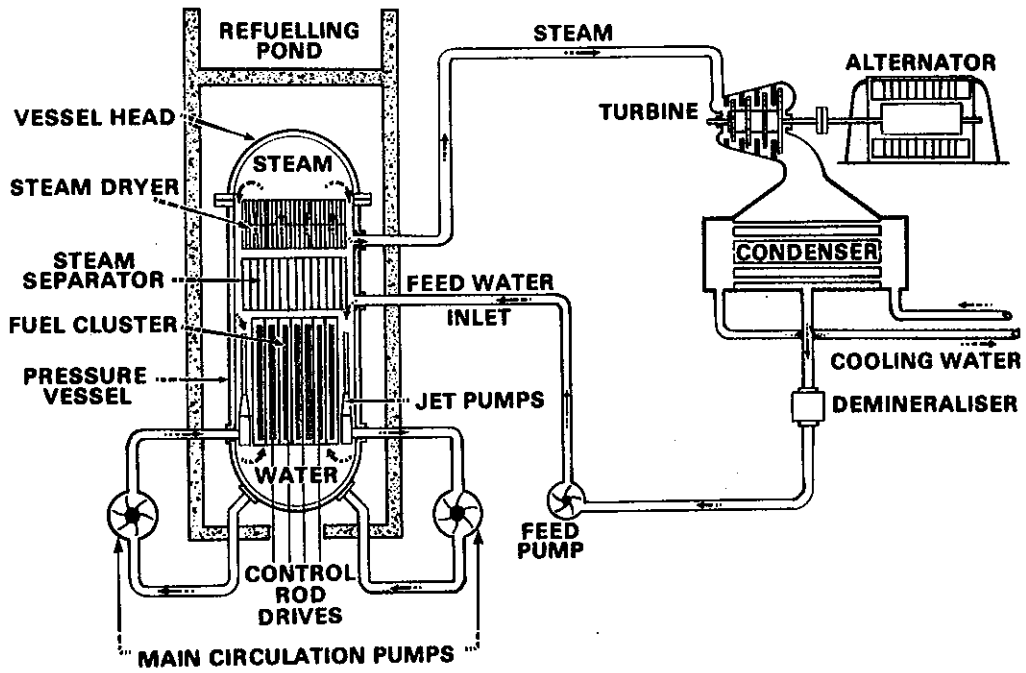
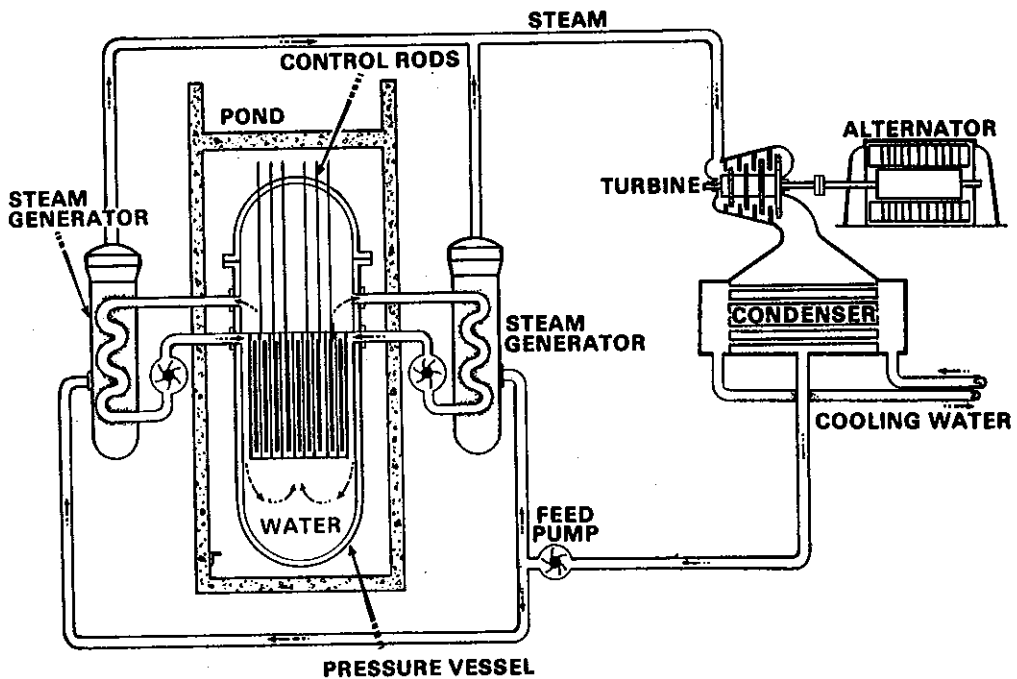


FIGURE 8. FUEL FABRICATION - ADU PROCESS SIMPLIFIED BLOCK FLOW DIAGRAM (USAEC 1974a)



**FIGURE 9. BOILING LIGHT WATER REACTOR (BWR)**



**FIGURE 10. PRESSURISED LIGHT WATER REACTOR (PWR)**

# SHIPPING CASK

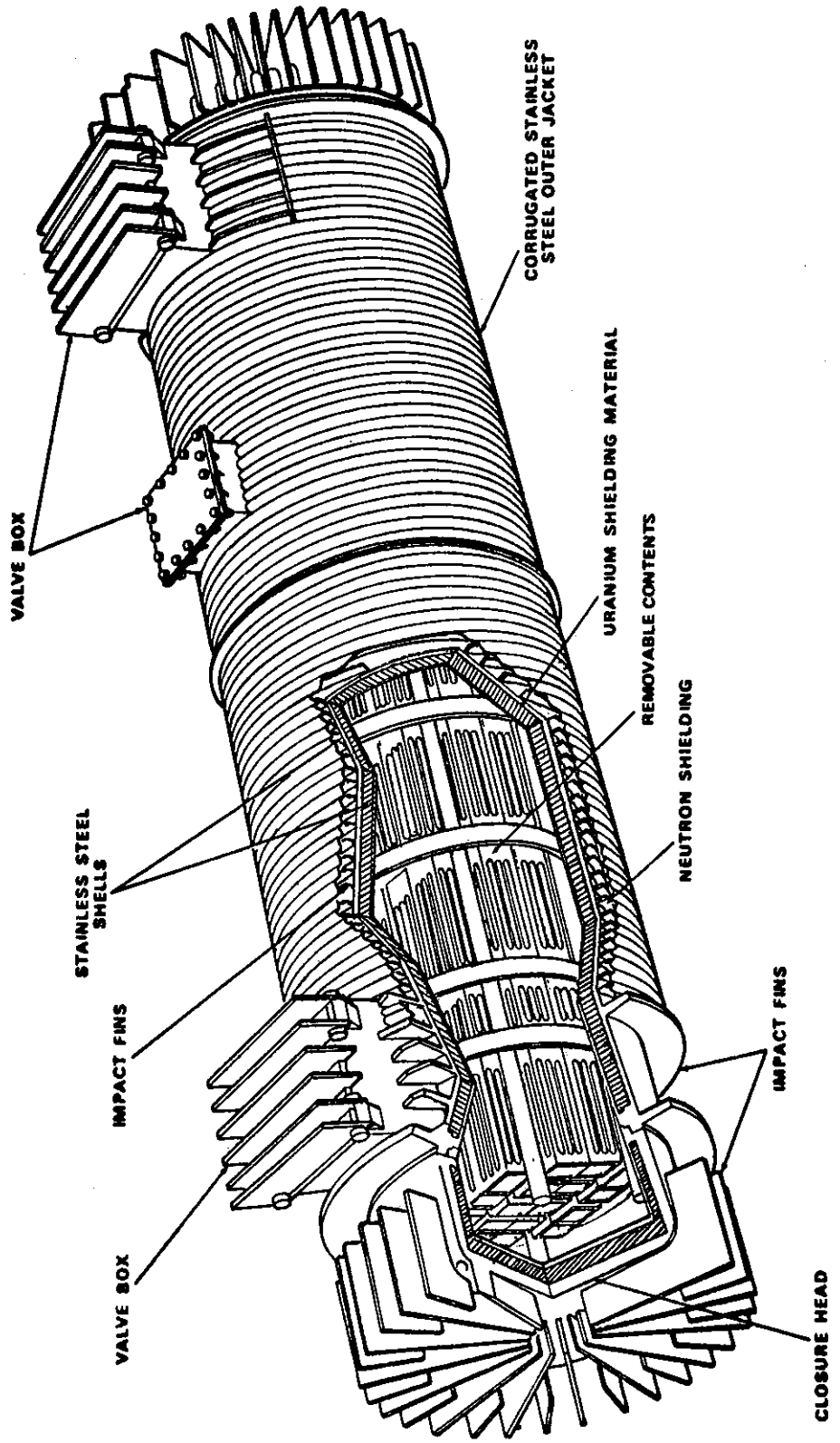


FIGURE 11. TRANSPORT FLASK FOR IRRADIATED FUEL ELEMENTS (USAEC 1974a)

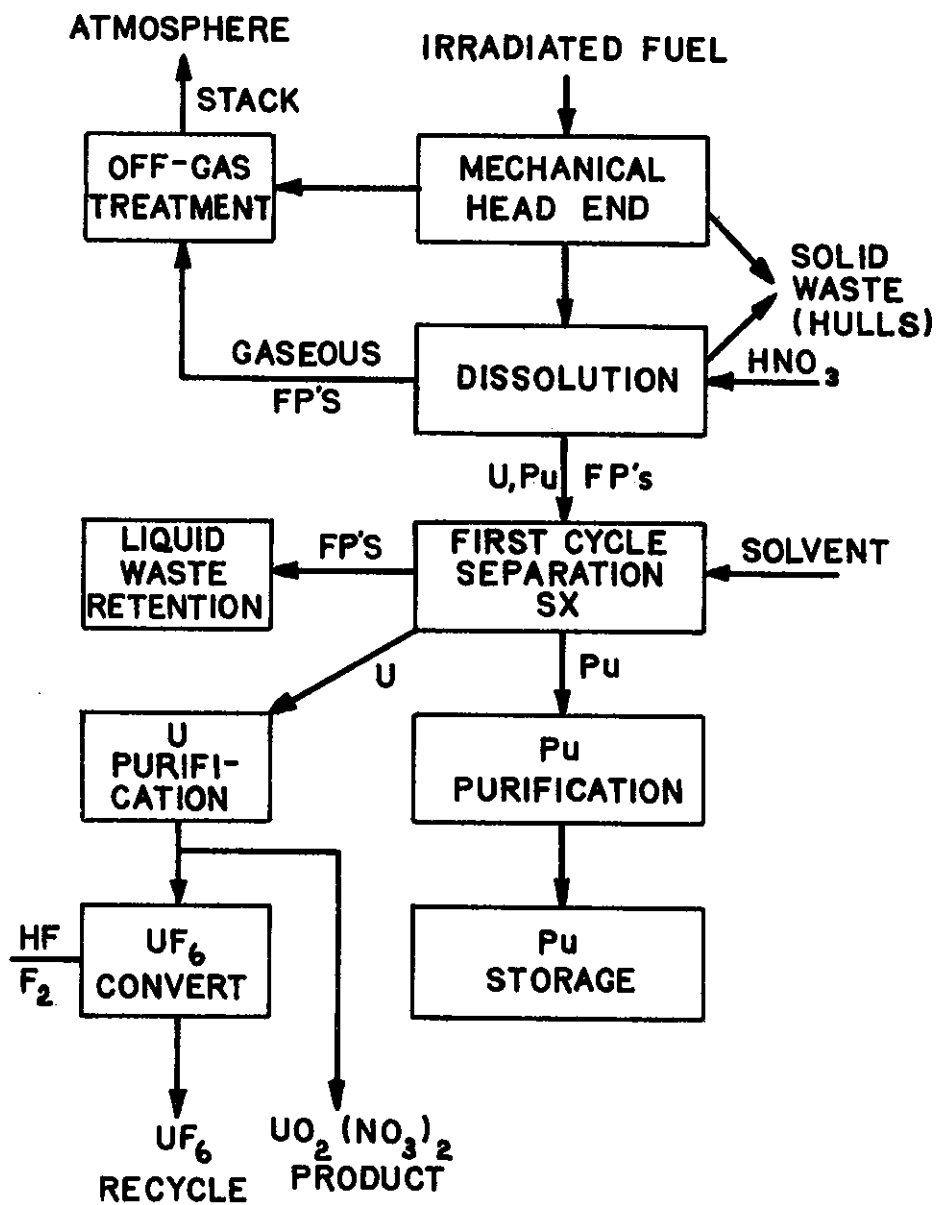
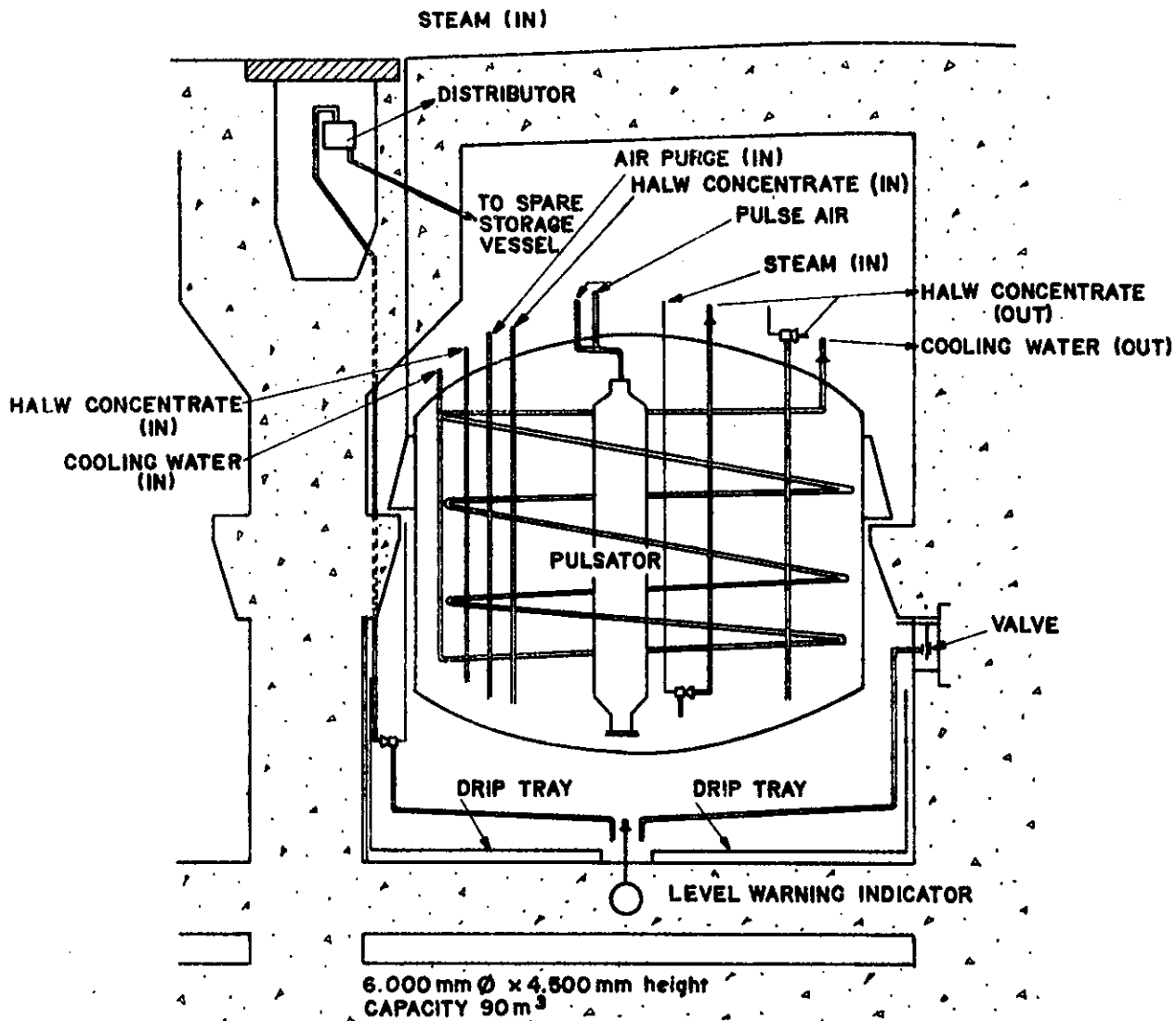
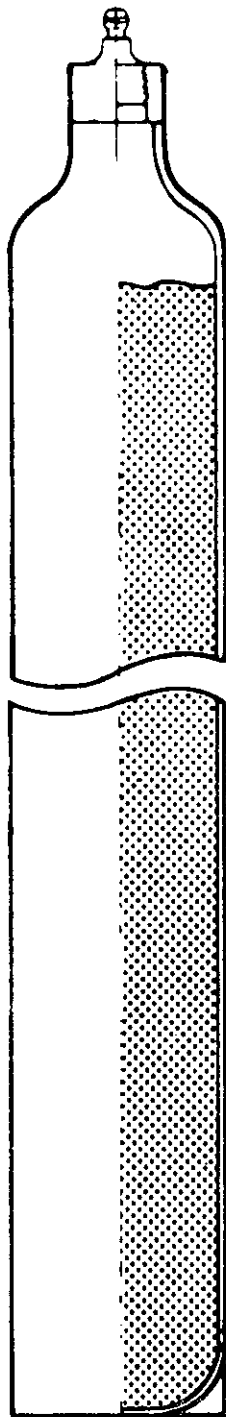


FIGURE 12. FUEL REPROCESSING SIMPLIFIED BLOCK FLOW DIAGRAM (USAEC 1974a)



**FIGURE 13. SKETCH OF STORAGE VESSEL FOR HIGH LEVEL LIQUID WASTE (Nakajima, 1972)**



	DIAMETER	LENGTH
TYPICAL	30 cm	3 m

Contents of Typical Canister:

Volume:	~	0.2 m <sup>3</sup>
Weight:	~	1.8 t
Heat Output:	~	5 kilowatts
Curies:	~	5 x 10 <sup>6</sup>

**FIGURE 14. TYPICAL CANISTER FOR SOLIDIFIED HIGH LEVEL WASTE (USAEC 1974d)**



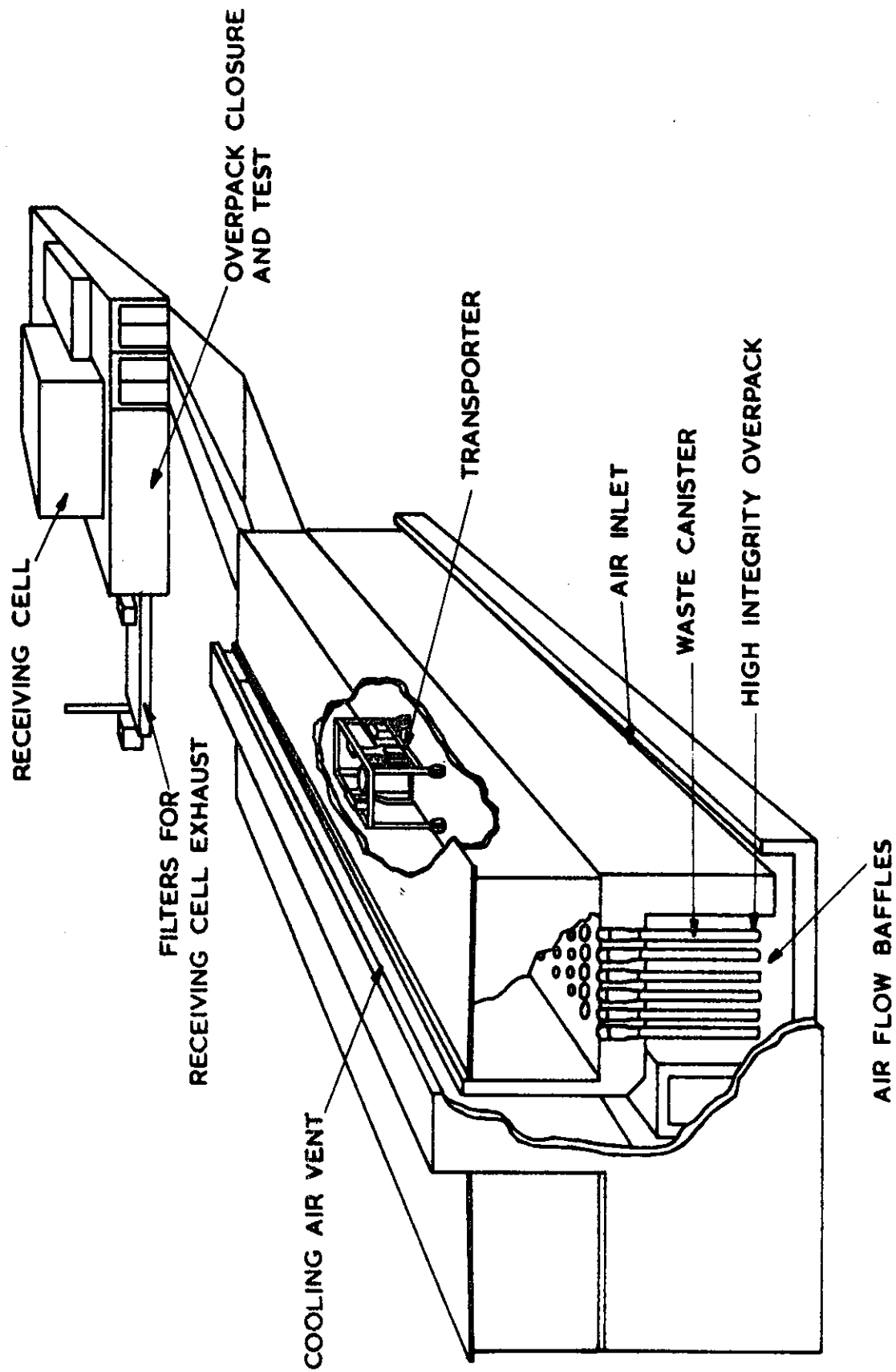


FIGURE 16. RSSF AIR-COOLED VAULT CONCEPT FOR STORAGE OF HIGH LEVEL WASTE CANISTERS (USAEC 1974b)

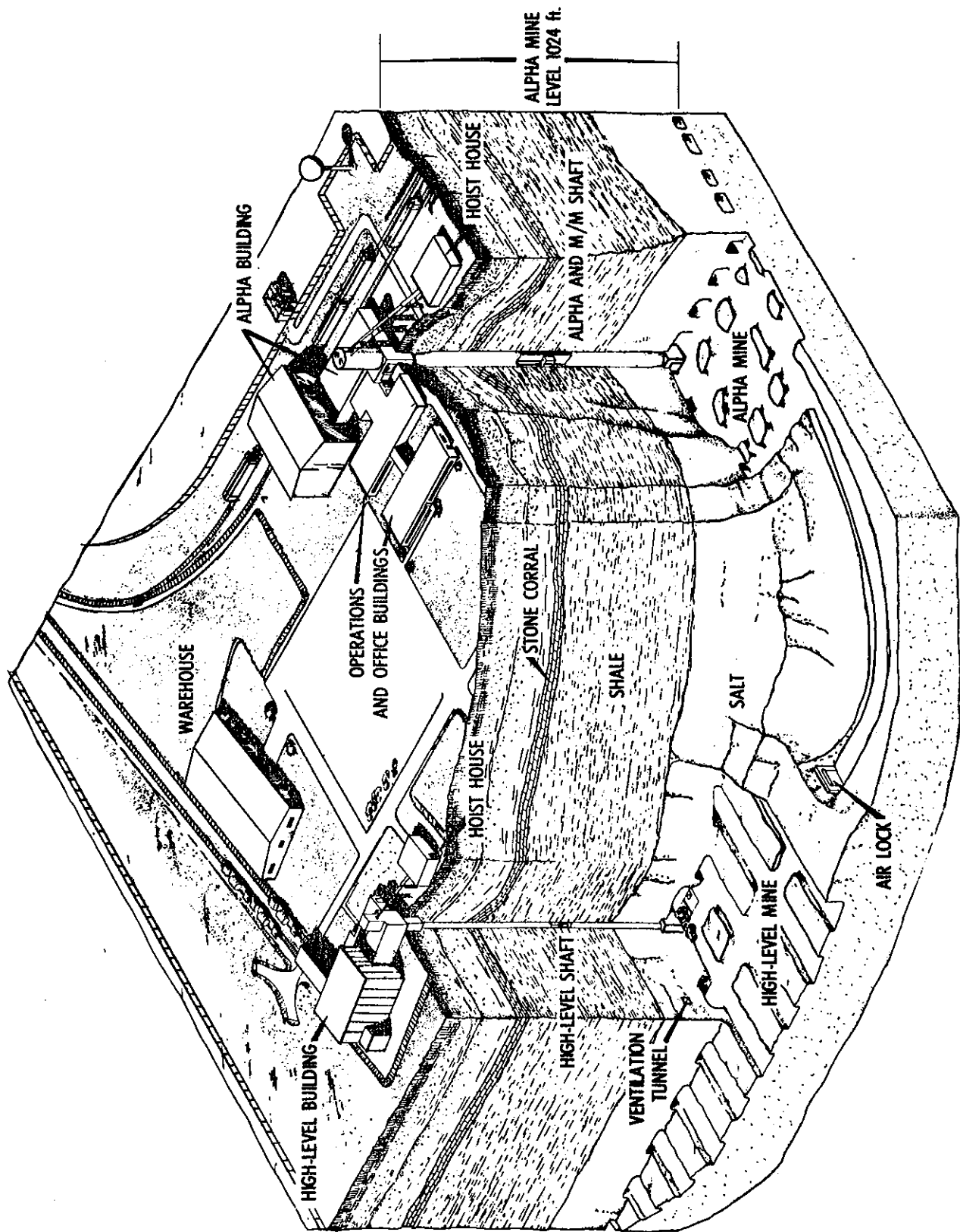


FIGURE 17. ARTIST'S IMPRESSION OF A REPOSITORY FOR DISPOSAL OF HIGH LEVEL WASTE IN BEDDED SALT DEPOSITS (Rogers, 1971)