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NUCLEAR MATERIALS AND FUEL CYCLE SERVICES SOURCES, INVENTORIES AND STOCKPILES

Prepared for

US Arms Control and Disarmament Agency
Under Contract No. AC9NC105

Volume II

September 1979

NUCLEAR ASSURANCE CORP.
PROPRIETARY

NUCLEAR ASSURANCE CORPORATION

DATED MATERIAL

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by

Ralph Brittelli, Jr.
Dan M. Collier
Thomas A. Vizzini

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NUCLEAR ASSURANCE CORPORATION

24 Executive Park West
Atlanta, Georgia 30329
Telephone: (404) 325-4200
Telex: 549567 & 542703

Weinbergstrasse 9
8001 Zurich, Switzerland
Telephone: (01) 47 08 44
Telex: 57275

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TABLE OF CONTENTS

TABLE OF CONTENTS
(continued)

IV DESCRIPTIONS OF REACTOR SYSTEMS AND MATERIAL REQUIREMENTS (Volume II)

V GENERIC DESCRIPTIONS OF THE TECHNICAL ASPECTS OF THE NUCLEAR FUEL CYCLE (Volume II)

TABLE OF CONTENTS
(continued)

Section Number | Title of Section | Page Number

VI GENERIC DESCRIPTION OF COMMERCIAL ASPECTS OF THE NUCLEAR FUEL CYCLE (Volume III)

Section IV

DESCRIPTION OF REACTOR SYSTEMS AND MATERIAL REQUIREMENTS

1. Introduction

In this section of the report, we will present information on the dominant reactor types operating today. Section V of the report will provide a technical description of the fuel cycles of these reactor systems. After this, Section VI will provide a generic description of the commercial aspects of the nuclear fuel cycles.

The dominant reactor types operating today which will be discussed in this section are the (1) Light Water Reactors (LWRs), the (2) Magnox Reactors, the (3) Advanced Gas Cooled Reactors (AGRs), and the (5) CANada Deuterium Uranium (CANDU) Reactors. Operating characteristics and material requirements will be discussed and comparisons made between these reactors.

Light Water Reactors (LWRs)

The Light Water Reactors (LWRs) are composed of two major reactor types: the Boiling Water Reactor (BWR) and the Pressurized Water Reactor (PWR). The Pressurized Water Reactor was first developed for the US Navy Nuclear Submarine Program by Argonne National Laboratory, Westinghouse Electric Corporation, the Naval Reactors Division and the US Atomic Energy Commission in the late 1940's. The first commercial nuclear power station - Shippingport - went critical on December 19, 1957.

The operating characteristics of PWRs vary but can be summarized as follows:

- PWRs operate at intermediate temperatures up to about 650°F with pressures up to 2500 PSIA.
- PWR cores are relatively compact, and require slightly enriched uranium fuel (at equilibrium 3.2 w/o U^{235}). The majority of the modern day PWRs are fueled with uranium dioxide clad with zirconium alloys (Zircalloy-4).
- PWR refinements in fuel design, integrity fuel management and chemical shim have allowed high power densities.
- Each PWR fuel assembly contains fuel rods of only one uranium enrichment.
- PWR fuel assemblies can be of a number of physical configurations. Modern day fuel assemblies are 17 X 17, 16 X 16 and 15 X 15 arrays. These designs replaced the 15 X 15 and 14 X 14 arrays. The effect of this change was to reduce kilowatts per foot (kw/ft) and surface heat flux resulting in better licensing ability, fuel performance and reliability.
- Average fuel burnup (warranted) is 33,000 MDW/MTU.
- Plutonium generation in PWRs is between 0.6-0.7% of the initial fuel weight.
- The thermal efficiency of the PWR is approximately 32%.

The Boiling Water Reactor (BWR) was developed by Argonne National Laboratory and General Electric Company. There have been several types of BWRs in operation: direct cycle natural circulation, direct cycle forced circulation, indirect cycle natural circulation, dual cycle forced circulation, and direct cycle forced circulation with integral boiler superheater. The large commercial BWRs in operation today use the forced circulation direct cycle with steam separation inside the reactor vessel concept. Additional characteristics of the BWR are:

- BWRs operate at coolant pressures of approximately 1000 PSIA with a maximum coolant temperature of about 546°F at saturation. BWR coolant operates at a pH7 using no coolant additives, i.e., highly pure water.

- Plutonium generation in BWRs is between 0.6-0.7% of the initial fuel weight.

- BWRs use uranium dioxide fuel clad in Zircalloy-2, since earlier stainless steel cladding exhibited stress corrosion cracking and a higher neutron absorption cross section.

- G.E.'s new BWR/6 reactor fuel bundles contain sixty-four (64) rods which are spaced in a square array (8 X 8). Fifty-four (54) of these rods contain UO_2 fuel. The other ten rods are composed of eight (8) tie rods and two (2) water rods. The tie rods provide fastening capability and hold the fuel assembly together. They contain no fuel material. The two water rods contain nothing. Small holes in these rods allow water to be driven through the rod thus introducing moderating material within the fuel bundle interior.

- G.E.'s BWR fuel rods are made of Zircalloy tubing of 0.483 inches o.d., 160 1/4 inches long with a wall thickness of 32 mils. Pellets are stacked in this tubing to an active length of approximately 150 inches. (Other BWR manufacturers and fuel manufacturers use stainless steel and have different dimensional characteristics for their fuel.)

- BWR fuel bundles (initial core) contain an average enrichment ranging from approximately 1.7 w/o U^{235} to 2.1 w/o U^{235}, depending on cycle length requirements. Reload fuel bundles have an average enrichment in the range of 2.4 w/o to 3.0 w/o U^{235}.

- Different enrichments are used in fuel bundles to reduce local power peaking. Low enrichment rods (some are natural uranium) are used on the corners and higher enrichment rods are used in the central part of the fuel bundle.

- BWRs use fuel channels to direct core coolant flow through each bundle and to enclose the fuel bundle. The combination of a fuel bundle and a fuel channel is called a fuel assembly.

3. Magnox Reactors

The British nuclear program stems from the four Calder Hall reactors which were built to produce plutonium for nuclear weapons. These reactors are CO_2-gas cooled, graphite moderated reactors fueled with natural uranium metal rods clad in a magnesium alloy called Magnox. (Magnox has the following composition - magnesium alloy containing mostly magnesium with about 0.8% aluminum, 0.002 to 0.050% beryllium, 0.008% cadmium, and 0.006% iron.) The Calder Hall reactors started operating and generating electricity in May

1956. The next four Magnox reactors (Chapelcross), were also built and operated for plutonium production, with generation of electricity as a byproduct. Many variations have been made to Magnox reactors over the years due to the inherent materials limitations of the fuel and reactor system in general. Some of the characteristics of Magnox reactors are:

- The optimum average burnup for the natural uranium fuel elements has been set between 3000-4000 MWD/MTU. Higher burnups have been achieved on individual assemblies.
- Core dimensions are approximately 14m diameter x 8m high.
- Spent-fuel from the gas cooled Magnox reactors cannot be stored indefinitely in water because the Magnox alloy corrodes slowly in water.
- Fuel replacement (refueling) in Magnox reactors can be carried out while the reactors are at power using on-line continuous refueling techniques.
- Magnox fuel management procedures do not call for axial or radial shuffling of fuel. Three (approximately) fuel channels are refueled each week.
- A ton of Magnox fuel irradiated for 1000 MWDs contains about 998 kilograms of unconverted uranium and about 800 grams of plutonium.

4. Advanced Gas Cooled Reactors (AGR)

The second phase of the British nuclear program, which saw a switch in emphasis from a plutonium production program to an electrification program, saw the development and construction of an improved version of the CO_2 gas cooled reactors, the Advanced Gas Cooled Reactors (AGR). Similar to the Magnox reactors, the AGRs retained graphite as the moderator and CO_2 as the coolant. The prototype 40 MWe AGR at Windscale was on-power in 1972. As a result of simplications in the design, AGRs have less than 1/10 the number of fuel channels of earlier Magnox reactors of equivalent power. The first commercial AGR, the Hinkley B Power Station consisting of two 600 MWe reactors started operation in June 1976. Additional characteristics of AGR systems are:

- AGRs use fuel that is slightly enriched and clad in stainless steel. Equilibrium enrichment is approximately 2.3 w/o U^{235}.

- AGR fuel can operate at higher temperatures and heat output rates than Magnox reactor fuel, giving a smaller size core and a more efficient steam cycle.

- Clusters of fuel elements, more similar in construction to the CANDU than the Magnox reactor, are joined together end-to-end in a stringer, and placed in vertical holes in the graphite moderator.

- Core dimensions are approximately 9.1 meters in diameter x 8.5 meters high. Outlet (coolant) temperature is 650°C. Coolant pressure is 600 PSIA. The steam cycle efficiency is about 42%.

5. CANDU Reactors

Starting in the early 1950's, the Canadian government and industry developed a distinctive reactor type - the CANada Deuterium Uranium (CANDU) Reactor. The first commercial natural uranium, heavy water - moderated reactor was Pickering Station which came on-line in 1969.

Heavy water reactor designs, other than CANDU, have now been developed. Most have evolved from the CANDU. None have established a comparable experience base, but nonetheless they could have some influence on further HWR design. The most notable systems are:

- The British SGHWR (Steam Generating Heavy Water Reactor) which is a direct cycle hybrid having both BWR and CANDU features with possibly improved emergency cooling provisions than CANDU but a poorer neutron economy. The SGHWR is an enriched system. This system was abandoned by the British.
- The Canadian BLWHR (Boiling Light Water Heavy Water Moderator Reactor) is similar to the SGHWR but with some efforts to retain CANDU levels of neutron economy and natural uranium refueling.
- The Japanese Fugen Reactor is closer to the SGHWR than to the CANDU.
- The Canadian HWOCR (Heavy Water Moderated Organic Cooled Reactor) has a neutron economy comparable to CANDU but improved steam conditions and a low heavy water inventory, in common with direct cycle systems.

The improved fuel economy of the CANDU system stems from reduced neutron losses principally as a result of three design features:

- Use of a heavy water moderator with negligible parasitic neutron capture.
- Operation with modest fuel burnup, assisted by on-power refueling and thus a low maximum fission product inventory.

- Operation with a high neutron flux and low enrichment and thus an appreciable fast fission contribution from U^{238}.

The CANDU is at a state of commercialized development generally comparable to the Light Water Reactors. As such, it is perhaps the only other nuclear system to reach that status. Furthermore, the CANDUs are considered by some to promise unique fuel utilization features that may be attractive for those national energy programs in which there are no fast breeder installations envisioned and for developing countries that lack fuel cycle technology.

Some of CANDU's salient technological features are:

- The costs of CANDUs are estimated to be 10-20% higher than a comparable LWR. This estimate includes an initial supply of heavy water but does not include first core fuel supplies.

- The thermal efficiency of CANDU reactors is 3-5% points lower than current LWR power plants because system design limits the steam conditions.

- Satisfactory heavy water system leak tightness has been demonstrated.

- Tritium is significant but currently an acceptable contributor to on-site personnel occupational exposure.

- Steam generators are particularly critical components for maintenance of economically acceptable losses of heavy water. The contrast between CANDU steam generators and LWR steam generators experience with tube integrity has been extreme - three tube failures to 3000 LWR tube failures.

- Present CANDUs operate with a fuel burnup of just over 7000 MWD/MTU using natural uranium. This is achieved with an on-line refueling capability which is essential to avoid prohibitive refueling downtime at the relatively low burnups needed to maximize neutron economy. The resulting uranium consumption rate is approximately 25% less than that of LWRs using a once through fuel cycle.

- Uranium fueled CANDU spent fuel may not be an economic source of recycle plutonium because the fissile Pu content is low (0.27% versus 0.6-0.7% of the initial uranium weight for LWRs).

Design studies are underway to investigate ways to improve fuel efficiency using plutonium recycle, U^{235} enrichment and thorium (Th^{232} + neutron = U^{233}). At present, none of these efficiency improvement schemes have reached the stage of commercial development.

6. Material Requirements

This part of the section presents data on the material requirements of the various reactor systems characterized above. Although each individual reactor has its own definite operating requirements and characteristics, the following is presented to enable a comparison to be made between the various reactor systems. There are three basic uranium materials required for the operation of LWR nuclear stations - yellowcake, uranium hexafluoride and uranium dioxide. Yellowcake, uranium tetrafluoride and uranium metal are required for Magnox reactors. For CANDU reactors, heavy water is a required material. In all cases, plutonium is a byproduct of irradiation in a nuclear reactor. The following is a brief description of these materials.

- U_3O_8 is a granular powder, usually bright yellow. The isotopic concentration of U^{235} in yellowcake is 0.711 w/o.

- UF_6 is a white, volatile solid at room temperature. At a temperature of 147°F (64°C) and a pressure of 22 PSIA, UF_6 forms a colorless liquid of high density. UF_6 is highly reactive. Natural UF_6 is packaged and transported in steel cylinders with about 21,000 to 28,000 lbs. capacity.

- UO_2 is a dry, black, fine grained ceramic powder. It is readily oxidized by air. Sintered UO_2 is relatively stable chemically. UO_2's melting point is about 2800°C and has very low thermal conductivity. UO_2 is transported and stored in plastic containers. UO_2 contains 88% of U by weight.

- Uranium metal is a very dense, chemically reactive substance, light-gray to silvery in color. It melts at 1133°C.

- Plutonium is formed by neutron absorption in U^{238}. Pu^{239} and Pu^{241} are the fissile isotopes of plutonium. Plutonium is recovered in reprocessing operations as $Pu(NO_3)_4$. The plutonium nitrate solution has been shipped in polyethylene bottles inside steel drums. Conversion of the nitrate to ceramic PuO_2 powder is preferred during handling, shipment and long term storage.

- Heavy water is indistinguishable in physical appearance from ordinary (pure) water. Deuterium is very widely distributed in nature but only in very dilute form. The natural ratio of deuterium to hydrogen is only 1 in 7000 (143 ppm). The separation of commercial quantities involves concentrating an extremely dilute material from massive feed streams in energy - and capital - intensive processes.

Tables IV-1 and IV-2 show approximate quantities of nuclear fuel materials and heavy water (as applicable). Two tabulations are shown: first core quantities and annual reload quantities. Nominal reactor size in each case is 1000 MWe: assumed capacity factor is 70%.

TABLE IV-1

First Core Quantities

	BWR	PWR	Magnox	AGR	CANDU
U_3O_8, short tons	545	481	1533	619	175
U as UF_6, metric tons	417	368	N/A	474	N/A
Enrichment, MTSWU	250	269	N/A	244	N/A
UO_2, metric tons	142	88	N/A	196	153
MTHM (U)	125	78	1180	173	135
D_2O, metric tons	N/A	N/A	N/A	N/A	800

TABLE IV-2

Annual Reload Quantities

	BWR	PWR	Magnox	AGR	CANDU
U_3O_8, short tons	184-239	206	330	171	159
U an UF_6, metric tons	141-183	158	N/A	131	N/A
Enrichment, MTSWU	107-139	129	N/A	89	N/A
UO_2, metric tons	31-40	29	N/A	36	139
MTHM (U)	38-36	26	254	32	123
D_2O, metric tons	N/A	N/A	N/A	N/A	8 (1)
Spent fuel, MTHM	27-35	24-25	248	31	120
Fissile Pu discharges, kg.	96-124	123-174	431	102	350

Notes: (1) Annual makeup requirements are less than 1% of 1st charge (approx. 800 MT)

(2) Definitions

MTSWU = metric tons of separative work units
MTHM = metric tons of heavy metal
= 100% U for first core
= (mixed U and Pu) for annual discharges

Section V

GENERIC DESCRIPTION OF THE TECHNICAL ASPECTS OF THE NUCLEAR FUEL CYCLE

1. Introduction

This section of the report addresses the dominant fuel cycle in use today - the uranium fuel cycle - which is used in practically all nuclear power reactors in the World Outside Of Centrally Planned Economic Areas (WOCA). There are a handful of reactors that employ the uranium-thorium fuel cycle mostly through variations on the High Temperature Gas-Cooled Reactor (HTGR) design, but this cycle will not be considered in this report. Nor will the Fast Breeder Reactor (FBR) or the Steam Generating Heavy Water Reactor (SGHWR) fuel cycle be considered, since there are only three (3) FBRs and one SGHWR currently operating.

The dominant reactor type in use today is the Light Water Reactor (LWR) of which there are two variations: The Boiling Water Reactor (BWR) and the Pressurized Water Reactor (PWR). Of lesser importance but still significant are the Magnox and Advanced Gas Reactors (AGR) operating principally in the U. K. and France, and the CANada Deuterium Uranium (CANDU) Reactors operating in Canada, Pakistan, India, Argentina (under construction) and Korea (under construction).

2. Nuclear Fuel Cycles

Figure V-1 shows the basic LWR fuel cycle, indicating the physical steps involved in the cycle applicable to both BWR and PWR fuel.

The LWR fuel cycle begins with the mining and milling of uranium ore - the product being U_3O_8. While there can be various transport and stockpile steps, the U_3O_8 eventually moves to the next major fuel cycle step - conversion - where U_3O_8 is converted to UF_6. Again, there may be extended storage periods and several transport steps before the next step - enrichment - where the uranium becomes classified as a special nuclear material. From the enrichment step, the enriched UF_6 moves to a series of chemical and mechanical processes under the heading of fabrication. There can be a series of internal storage points in the fabrication process and extended storage after fabrication. The fabrication process completes the front end of the LWR fuel cycle.

Depending on the reactor type and fuel cycle plan, the fuel may reside in the core for one to four years. At discharge, the fuel is transferred to the spent fuel storage pool. This dischage is considered to begin the back-end of the fuel cycle.

The back-end has many alternatives; however, at present, the primary storage method is still at-reactor storage. Figure V-2 illustrates the many flow paths which exist or are developing. There are in fact five main classifications of temporary disposition (storage at the reactor, storage at another reactor, storage at an Away-From-Reactor (AFR) facility, and use or storage in a laboratory), and two classifications of ultimate disposal (permanent storage or reprocessing). Reprocessing ultimately leads to incentives for reuse of the uranium and plutonium in either FBRs or thermal reactors.

The Magnox Fuel Cycle deviates substantially from that of the LWR fuel cycle as shown in Figure V-3.

The Magnox fuel cycle also requires a mining and milling step to produce U_3O_8. However, U_3O_8 is fluorinated in a conversion step to UF_4 instead of UF_6. In the metal production step UF_4 is reduced to metallic uranium before fabrication of the Magnox fuel assembly. The back-end fuel cycle support facilities and alternatives are similar to that for LWRs, except that relatively low corrosion resistance of the magnesium cladding alloy in water does not permit storage in pools indefinitely. Thus, reprocessing within a relatively short period of time is essential. Reprocessing of Magnox fuel must take place no later than two years after discharge, if storage is in water.

The Advanced Gas Cooled Reactor (AGR) Fuel Cycle steps are almost identical to those for the LWR as shown in Figures V-1 and V-2. (A separate diagram has not been prepared for the AGR fuel cycle because of the similarity to the LWR fuel cycle.)

The CANDU Fuel Cycle is considerably different from the LWR fuel cycle and the Magnox fuel cycle as shown in Figure V-4. Mining and Milling is still required to produce U_3O_8, but fluorination and enrichment are not. Conversion of the U_3O_8 directly to UO_2 is necessary but this is considered part of the fabrication step.

The "back-end" alternatives are similar for CANDU except that they are less developed than that for LWRs and Magnox reactors.

It is instructive to define more precisely the steps involved in the nuclear fuel cycles of various reactor types. The following discussion provides a detailed technical description of (1) Yellowcake Production, (2) UF_6 Conversion, (3) UF_4 Conversion, (4) Enrichment, (5) LWR Fuel Fabrication,

FIGURE V-1

BASIC LIGHT WATER REACTOR FUEL CYCLE

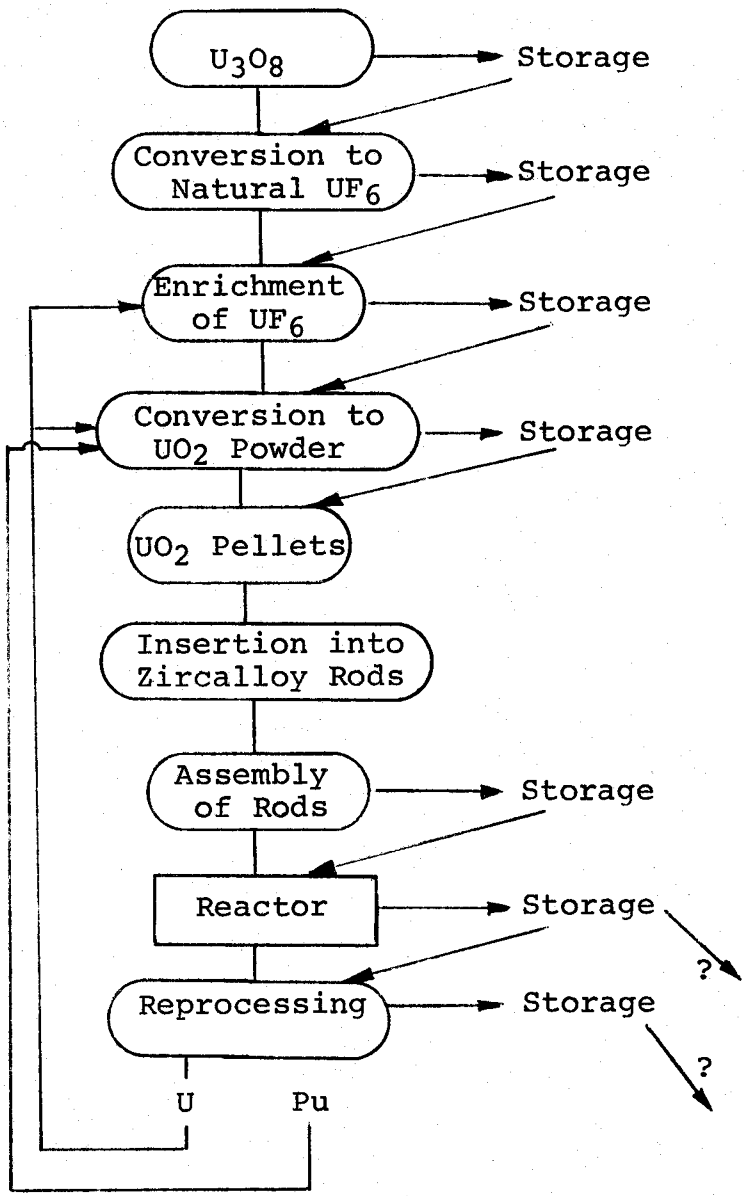


FIGURE V-2

LIGHT WATER REACTOR DETAILED BACKEND FUEL CYCLE

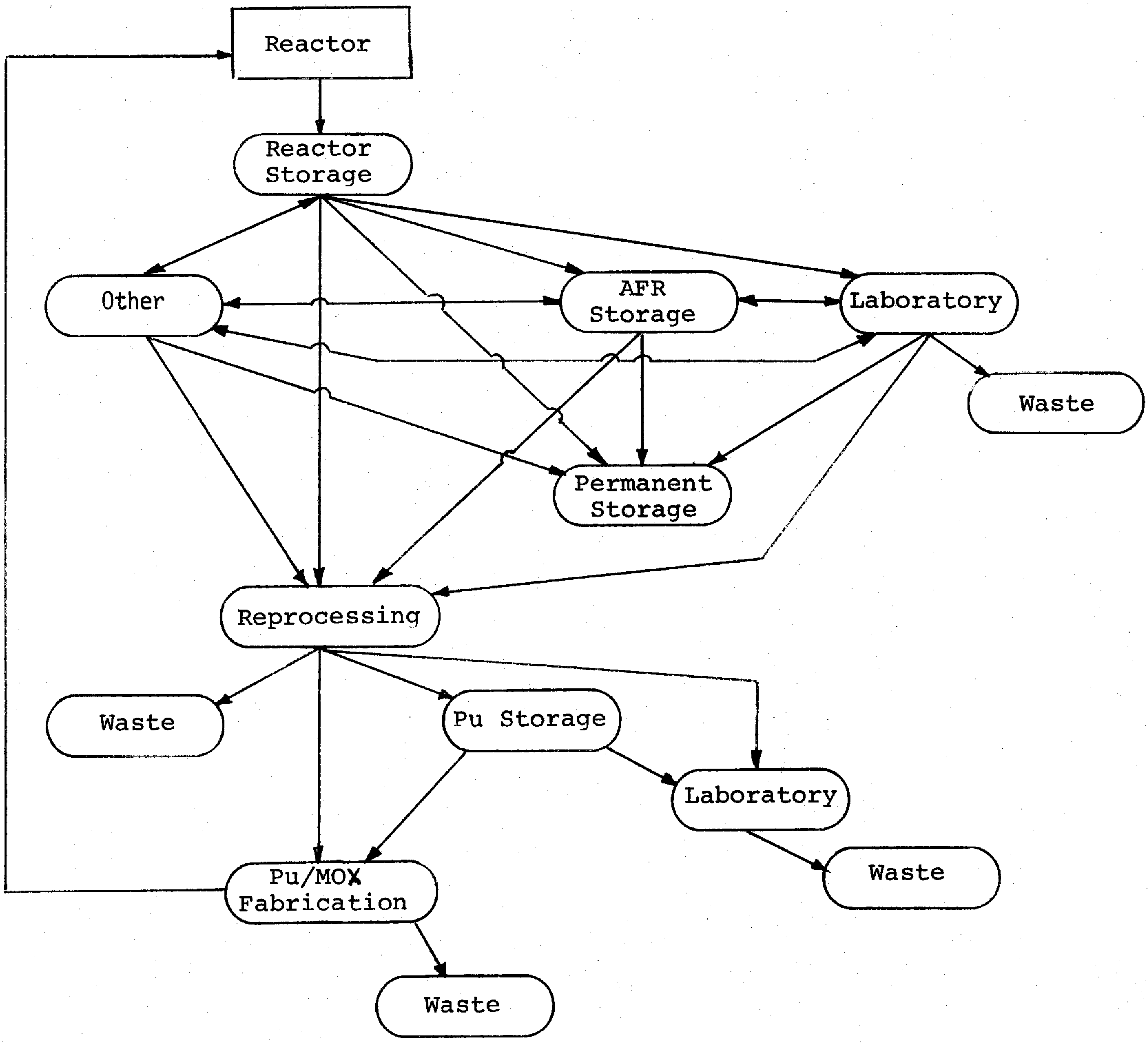


FIGURE V-3

BASIC MAGNOX REACTOR FUEL CYCLE

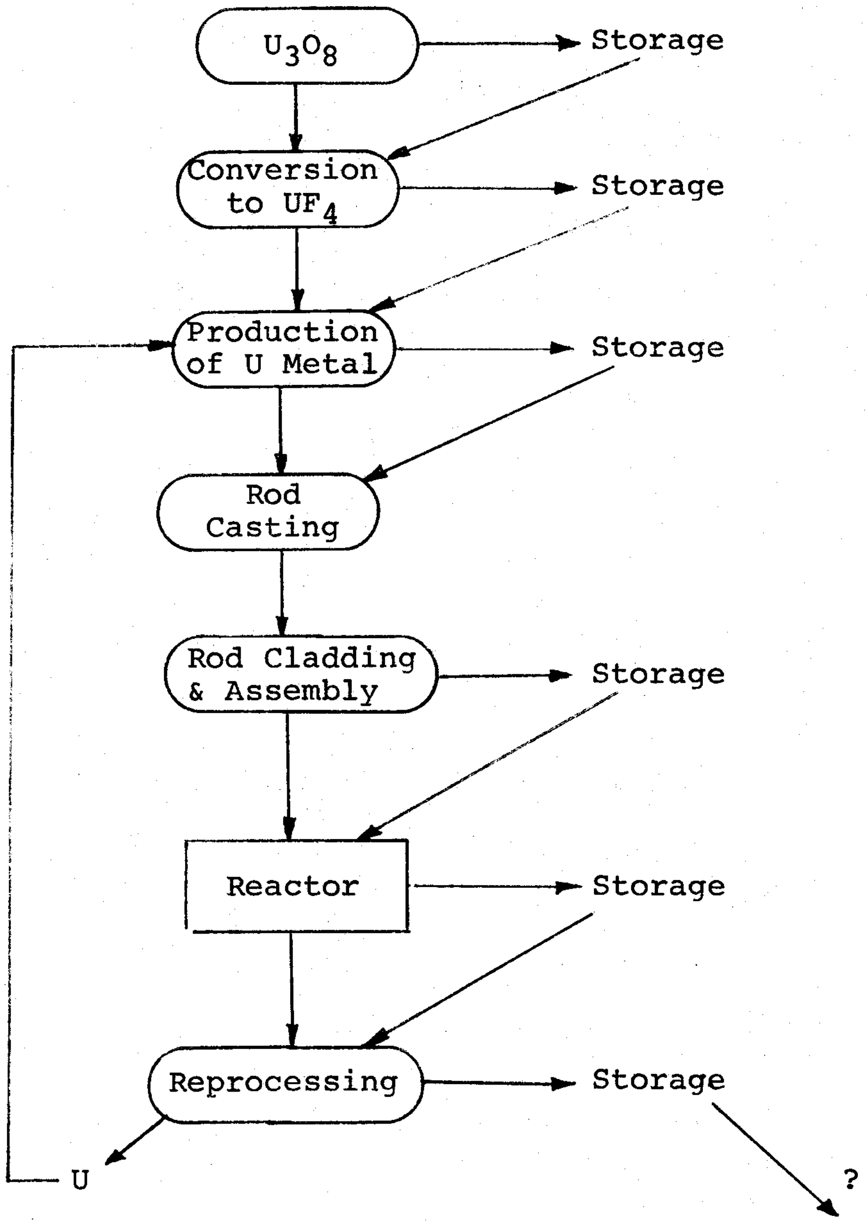
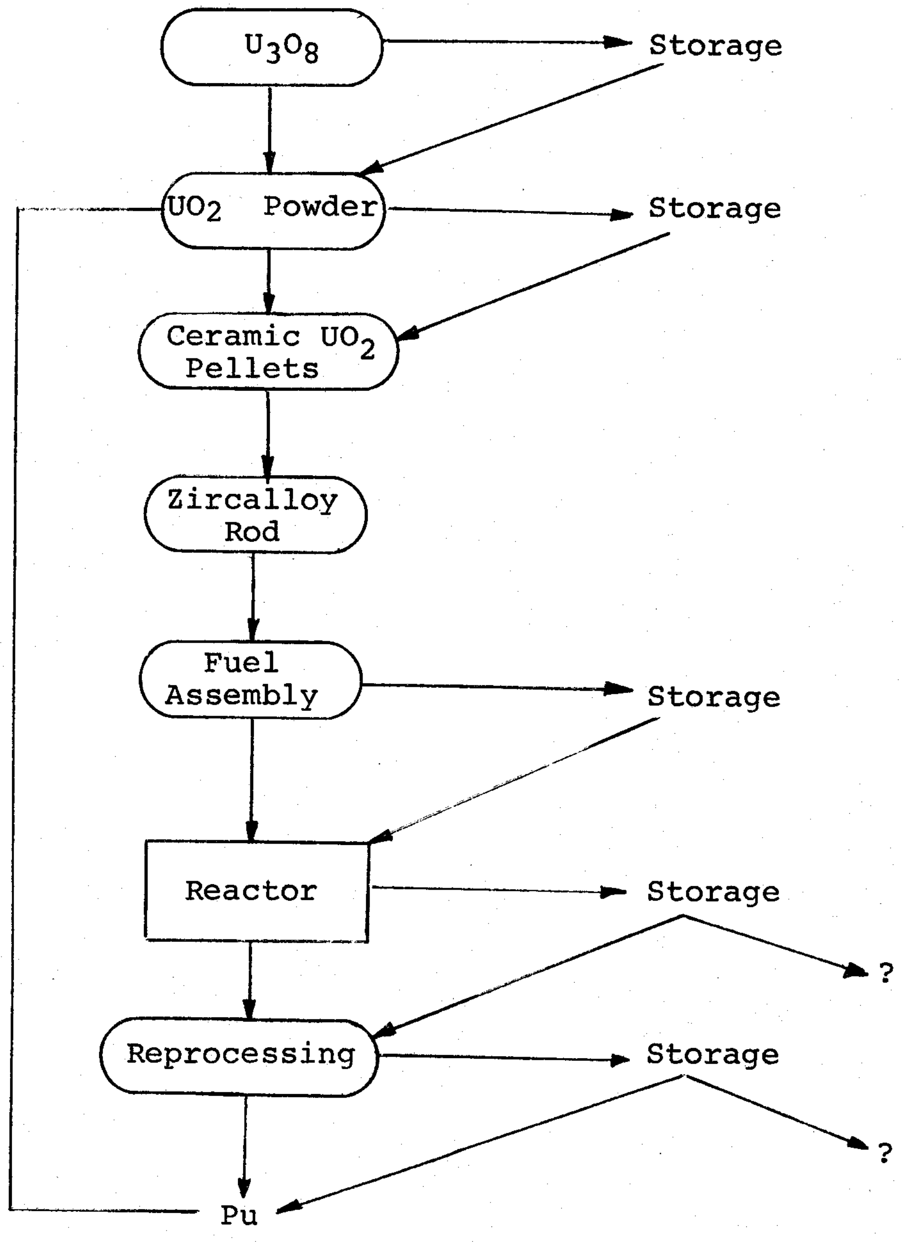


FIGURE V-4

BASIC CANDU REACTOR FUEL CYCLE



(6) Magnox Fuel Fabrication, (7) AGR Fuel Fabrication, (8) CANDU Fuel Fabrication, and (9) Reprocessing.

3. Yellowcake Production

In all cases the starting point is uranium ore. Uranium is explored for and produced much like base metals - lead, iron, zinc, copper - with a few interesting variations. In nature uranium consists of about 99.28% U^{238}, 0.711% U^{235}, and the balance other isotopes. Mined uranium ore is chemically and physically processed to produce U_3O_8 or yellowcake. Physical and chemical processing of the ore is as follows in some typical processes:

- The ore is removed from the uranium mines via a series of conveyor belts or trucks to ore crushing stations. At the ore crushing stations, the ore undergoes reduction in size to prepare it for chemical processing, blending and storage.

- Uranium ore leaving the crushing station is mixed with water and ground to a size that fits through a fine mesh screen. This process produces a mud slurry of approximately 50% solid and 50% water. The ore is now ready for chemical treatment.

- The mud slurry is pumped to leaching agitators. As the slurry enters the leaching circuit, sulphuric acid (H_2SO_4) and sodium chlorate ($NaCLO_3$) are added. Uranium and any other metals soluble in sulphuric acid are leached or dissolved. The sodium chlorate oxidizes the ore, which speeds up the leaching process and improves recovery.

- After approximately eight hours of leaching, heavier particles (sand) are separated from lighter particles (slime) by centrifugal force in classifier separations.

- The sand is removed from the bottom of the classifiers and the slime from the top. The sand, still in slurry form, is fed into a five stage rake classifier where it is thoroughly washed with an acid-water solution. As the sand is washed, the uranium, now in solution, is separated from individual grains of sand. At the end of the rake classifier circuit, the barren sand is pumped to the tailings disposal pond.

- The lighter component of the ore slurry (slime) is pumped to a thickener circuit. The slime goes through a six stage washing cycle to remove uranium attached to microparticles of slime. Barren slime is pumped to the tailings pond.

- As the acid-water-uranium solution leaves the thickener, it has advanced through five stages of slime washing. The solution now contains all the uranium that can be removed economically from the ore. This completes the second stage of ore processing.

- Next, the uranium solution enters the solvent extraction (SX) circuit to remove uranium from the acid-water solution which may contain traces of vanadium, iron, molybdenum, etc. As the uranium solution enters the one end of the SX circuit, barren solvent, usually tributylphosphate (TBP), is introduced at the other end. The two solutions advance countercurrent to each other and proceed through four stages of mixing and settling.

- The solvent extraction, or ion-exchange process, performs two basic functions. First, it selectively removes the uranium from the acid-water solution leaving the unwanted metals in solution. Second, the uranium is concentrated in the solvent threefold.

- The barren acid-water solution, or raffinate, free of uranium, leaves the last stage of the solvent extraction circuit and is pumped back into the thickener circuit as a washing solution or to the tailings disposal pond.

- From the SX circuit, the concentrated uranium in the solvent solution is pumped to the stripping circuit. Here the uranium is stripped from the solvent in a four-stage mixer-settler and again concentrated.

- Leaving the stripping circuit, the stripping solution, containing concentrated uranium, flows to a precipitation circuit. Here the uranium, which has been up to this point in solution, is caused to precipitate or "fall out" of solution. The addition of ammonia, air and heat in the precipitation circuit causes uranium to become insoluable in the acid strip solution. During precipitation, the uranium solution is constantly agitated to keep the solid particles of uranium in suspension.

- Leaving the precipitation circuit, the uranium is pumped to a thickener tank where the solid is allowed to settle to the bottom of the tank.

- From the bottom of the product thickener tank the precipitated uranium in the form of a slurry is pumped into a three stage drum filter circuit. Moving through the drum filter circuit, the uranium is thoroughly washed to remove any soluble salts that might be entrained in the product.
- The uranium leaves the last washing filter in the form of a filter cake. It is then dropped onto a perforated plate where a roller pushes the material through openings into spaghetti-like particles. The uranium in this form is fed to dryers.
- In the dryer, the product is dried by steam heat and readied for packaging. The final product - U_3O_8, or yellowcake - is packaged in 55 gallon drums for shipment.

There are a number of variations to the process for producing U_3O_8 depending upon the initial form of the ore and the location of the mill. Other processing variations are: (1) Acid-Leach/Ion Exchange (IX)/Precipitation, (2) Acid Ion Exchange (IX)/Eluex/Precipitation, (3) Acid-Leach/Resin-in-Pulp (RIP)/Precipitation, (4) Acid-Leach/Rip/Eluex/Precipitation, (5) Akaline Leach/Caustic Precipitation, (6) Akaline Leach/Rip/Precipitation, etc. Since these processing variations are similar to the one already described, no further information will beprovided other than to recognize that they do exist.

4. UF_6 Conversion

The next step in the fuel cycle is the chemical conversion to UF_6 (LWR and AGR). The conversion of uranium concentrates to UF_6 consumes tremendous quantities of industrial chemicals. Every ton of processed uranium requires more than a ton and a half of fluorspar, two thirds of a ton of hydrofluoric acid and almost two thirds of a ton of sulphur.

There are two processes used in the commercial production of UF_6 from U_3O_8. These two processes are (1) the Dry Fluoride Volatility Process and the (2) Wet Solvent Extraction Process (Refining Fluorination Process). For the purpose of this section, both of these processes will be briefly described.

As we will see, the processing of uranium concentrates (U_3O_8) into uranium hexafluoride (UF_6) has two objectives - removal of impurities, and the conversion of the concentrates into a chemical form required by enrichment. The Dry Volatility Process, developed by Allied Chemical Corporation, proceeds as follows:

- The concentrates are received at the conversion plant and undergo weighing, sampling, and moisture content determination.
- Mechanically sized concentrates are placed in a chemical reactor to react with hot "cracked" ammonia to form uranium dioxide.
- The uranium dioxide (UO_2) is placed in another reactor to react with vaporized anhydrous hydrofluoric acid to form uranium tetrafluoride (UF_4), or "green salt". (At this point, the UF_4 could be drawn off and used to produce Magnox fuel after a series of purification steps.)
- The UF_4 is combined with gaseous fluorine in a fluidized bed at temperatures approaching 1000°F. The uranium is volatized as UF_6 gas.

- The UF_6 is cooled and separated from other gaseous components, then revaporized and fractionally distilled to remove any remaining impurities.

- The UF_6 is fed as a liquid into carbon steel containers (cylinders) which hold up to 14 tons of product. On cooling, the UF_6 becomes a white crystalline solid. In this form the UF_6 is shipped to an enrichment facility to undergo the next step in the fuel cycle.

The classic process for ore concentrate conversion was developed by the US Atomic Energy Commission and is the refining-fluorination process. The refining step involves purification of a uranium solution by solvent extraction with tributylphosphate (TBP). There are two variations to the process requiring different equipment and operating conditions but overall the basic operations are similar. The Refining-Fluorination Process proceeds as follows:

- The first step in the process is digestion. The ore concentrate (U_3O_8) is digested with 40% nitric acid (HNO_3) in a heated tank to form a slurry.

- The aqueous slurry is fed to a series of pumper decanter contactors for extraction of uranium into a 30% TBP solution. The solvent and slurry are contacted in the pumper decanter contactors countercurrently. (The solvent flows in one end, the slurry from the other.) The ratio of the organic TBP to the aqueous solution is about 13:1. Raffinate leaves the last decanter with a uranium concentration less than 0.1 g U/l.

- After removal of traces of TBP, the raffinate is neutralized with a lime slurry which causes the residual trace uranium to precipitate and settle to the bottom of open storage ponds.

- The precipitate - uranyl nitrate ($UO_2(NO_3)_2$) - is pumped in solution through an evaporator where the solution is concentrated to the approximate composition of uranyl nitrate hexahydrate (UNH).

- The UNH is now fed into heated troughs with rounded bottoms to undergo denitration. In the denitration trough, rotating agitators keep the bed thoroughly mixed. The end result of denitration is uranium trioxide (UO_3).

- The UO_3 is now transferred into two vertical stainless steel reactors. Dissociated ammonia is fed into both reactors. The UO_3 is partially reduced in the first reactor and overflows into the second where reduction is completed and UO_2 is formed. The two stages are controlled at 1100°F.

- UO_2 from the fluidized bed reducers is screw fed into the first stage hydrofluorinator where it is contacted with HF exhaust gas from the second stage. The partially reacted solid is screw fed to the second stage where it reacts with anhydrous HF. The solid formed is uranium tetrafluoride (UF_4), or "green salt".

- The final step of the fluorination process is the reaction of UF_4 with elemental fluorine to produce UF_6. The UF_4 and pure fluorine are introduced at the top of a vertical tower reactor. The solid reacts with the gas almost immediately in a flame reaction having an estimated temperature of 3000°F. Essentially complete conversion of the UF_4 to UF_6 is achieved.

- The outlet gas stream, containing UF_6, fluorine, and diluent gases is cooled to 300°F and passed through cyclone filters and a sintered Monel filter to remove entrained solids. The UF_6 is then readied for packaging.

The purity of the UF_6 produced by the refining-fluorination process is exceptional. The chemical purity of the product is consistently 99.97%, and the overall yield of the process is better than 99.5%.

5. UF_4 Conversion

The Magnox reactors require that uranium concentrates be chemically converted to uranium tetrafluoride (UF_4), or "green salt". The conversion process proceeds as follows:

- Raw material, uranium ore concentrates, are sampled and checked for purity and moisture content.

- The ore is then dissolved in nitric acid (HNO_3) to form uranyl nitrate ($UO_2(NO_3)_2$). The nitric acid serves to dissolve some of the impurities but still there are a few left in suspension.

- The mixture of uranyl nitrate, nitric acid, and impurities is then filtered to remove the undissolved impurities.

- The filtered liquid passes thru a purification circuit which separates the uranyl nitrate using a solvent extraction process (similar to those described earlier).

- The pure uranyl nitrate is then passed through evaporators to remove excess water and concentrate the liquid.

- The concentrated liquid is sprayed into a chemical reactor containing a bed of about 10 tons of uranium trioxide (UO_3) powder which is electrically heated and kept "fluidized" by a stream of compressed air drawn up into the vessel from the bottom.

- The sprayed uranyl nitrate decomposes to form more uranium trioxide (UO_3) powder. As the bed builds up, the powder overflows to a second fluidized bed reactor.

- In the second fluidized bed reactor, the UO_3 is reduced by a stream of hydrogen to uranium dioxide.

- The uranium dioxide (UO_2) passes to a third fluidized bed reactor where it is converted to uranium tetrafluoride (UF_4) by introduction of hydrofluoric acid gas. The UF_4 or "green salt" is ready for shipment to a Magnox fuel production facility.

6. The Enrichment Process

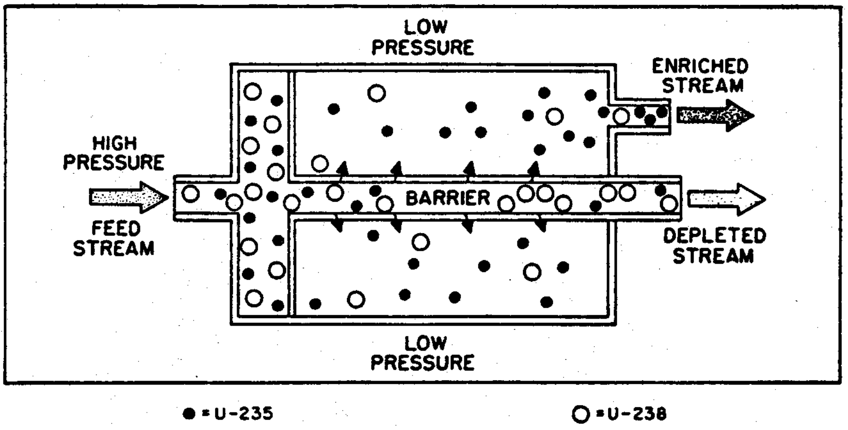
For the LWR and the AGR, enrichment in the U^{235} isotope is required. There are two major commercial methods of enrichment being used today. These processes are (1) the Gaseous Diffusion Process and (2) the Gas Centrifuge Process. There are other advanced techniques being investigated on pilot and laboratory scale that show promise commercially, but that will not be available for years. These advanced enrichment technologies involve plasma and laser applications.

The gaseous diffusion process involves the diffusion of isotopes of UF_6 through a porous membrane - the barrier. The diffusion process is based on the fact that when a gas passes through a membrane or port of a diameter smaller than the mean free path of the molecules in the gas then the flow is governed by Knudsen's Law. Knudsen's Law states that for a given pressure difference, diffusion is inversely proportional to the square root of the molecular weight of the gas. In the case of UF_6, it is 1.00429. (The point of this chemical and physical law is that $U^{235}F_6$ is slightly lower in molecular weight than $U^{238}F_6$. As a result, it is the $U^{235}F_6$ molecule that selectively passes through the barrier slightly more than the $U^{238}F_6$ molecule.) This indicates that enrichment per stage is very small and a large number of stages in series is required to produce practical enrichment. For example, production of 4.0 w/o - U^{235} from natural uranium (0.711 w/o U^{235}) requires about 1500 stages.

The gaseous diffusion process proceeds as follows:

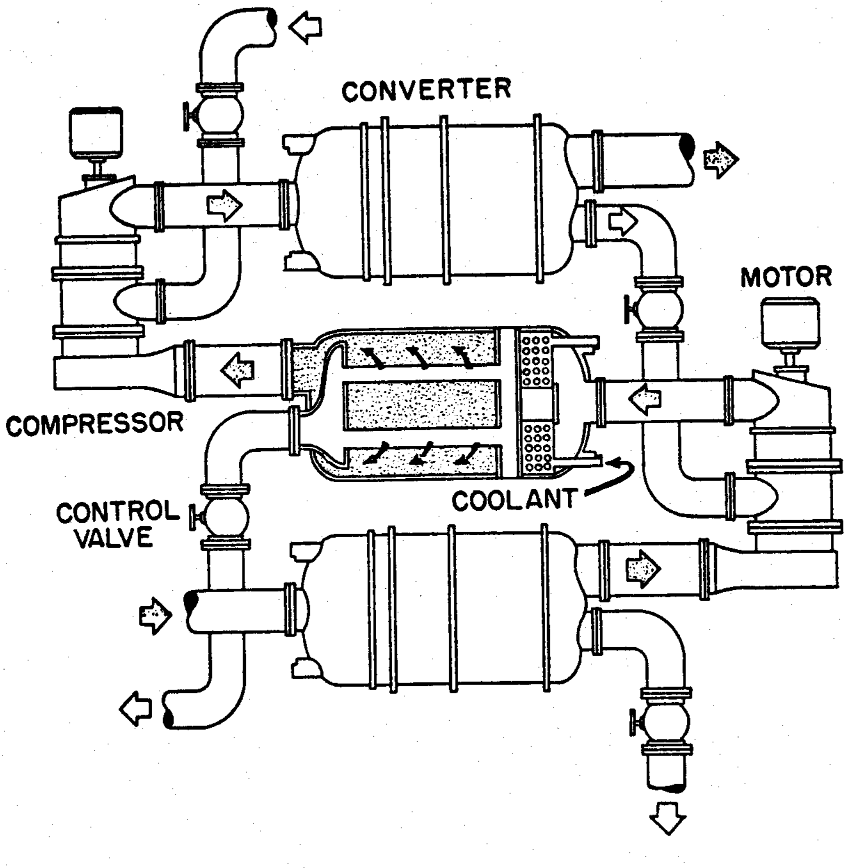
- UF_6 is introduced at high pressure inside a barrier tube. About half of the gas diffuses through the barrier and is fed to the next higher state. The reject, or undiffused stream, is recycled to the next lower stage. (Figure V-5).

A typical arrangement of gaseous diffusion equipment in a series of stages is shown in Figure V-6. The UF_6 gas is moved by axial flow compressors in the larger stages. These compressors are driven by electric motors, some as large as 2000 HP. A cooler is provided to remove the heat of compression at each stage.



Gaseous Diffusion Stage

Figure V-5



Series of Stages and Equipment

Figure V-6

The barrier tubes are contained in large cylindrical vessels called converters. To provide proper flow balance of the stream, a control valve is used in the reject stream outlet from the converter. The enriched stream from the bottom stage enters the central compressor and after being partially compressed is mixed with the depleted stream from the top stage. After further compression, the gas is then fed to the converter in the center. Groups of such stages are combined to make up operating units which, in turn, make up the diffusion cascade.

To discuss enrichment services, it is necessary to understand the concept of separative work. This concept provides a universal method for defining quantitatively in terms of a single measurement unit the enrichment services being performed.

Separative work is best understood in terms of the "separative potential" (ϕ) - a quantity associated with a unit mass of material at a given enrichment or concentration (C).

$$\phi = (2C-1) \log_e \left(\frac{C}{1-C} \right) + AC + B$$

(A & B are constants)

Passage through a separation barrier produces two fractions, an enriched product of concentration Cp and mass Mp and a depleted (waste) fraction characterized by Cw and Mw. Since the conversion of total mass and U^{235} mass is required, the following equations apply.

$$Mp + Mw = M$$

$$MpCp + MwCw = MC$$

The total separation potential times mass, however, is increased by an amount S which we term SEPARATIVE WORK.

$$S = Mp\phi p + Mw\phi w - M\phi$$

Thus, as evidenced by the above equation, a specific enrichment task can be described in terms of the quantity of material fed and the quantity withdrawn from the cascade as product and tails and the U^{235} isotope content of each of these three streams.

In essence, separative work is the work or energy required to carry out this separation of feed into product and tails. The units of separartive work are Metric Tons Separative Work Units (MTSWU). This is not a mass unit but simply the units that result from the derivation of the separative work equation.

The degree of depletion of U^{235} in the tails stream is significant in determining the amount of separative work required to accomplish the production of a stated amount of enriched product. It also has an important bearing on the amount of UF_6 required (and consequently the U_3O_8 required). With a higher tails assay, less separative work is required but more feed is required to produce specific (enriched) product. Thus, there is a trade-off between the use of feed and the use of separative work in producing enriched material.

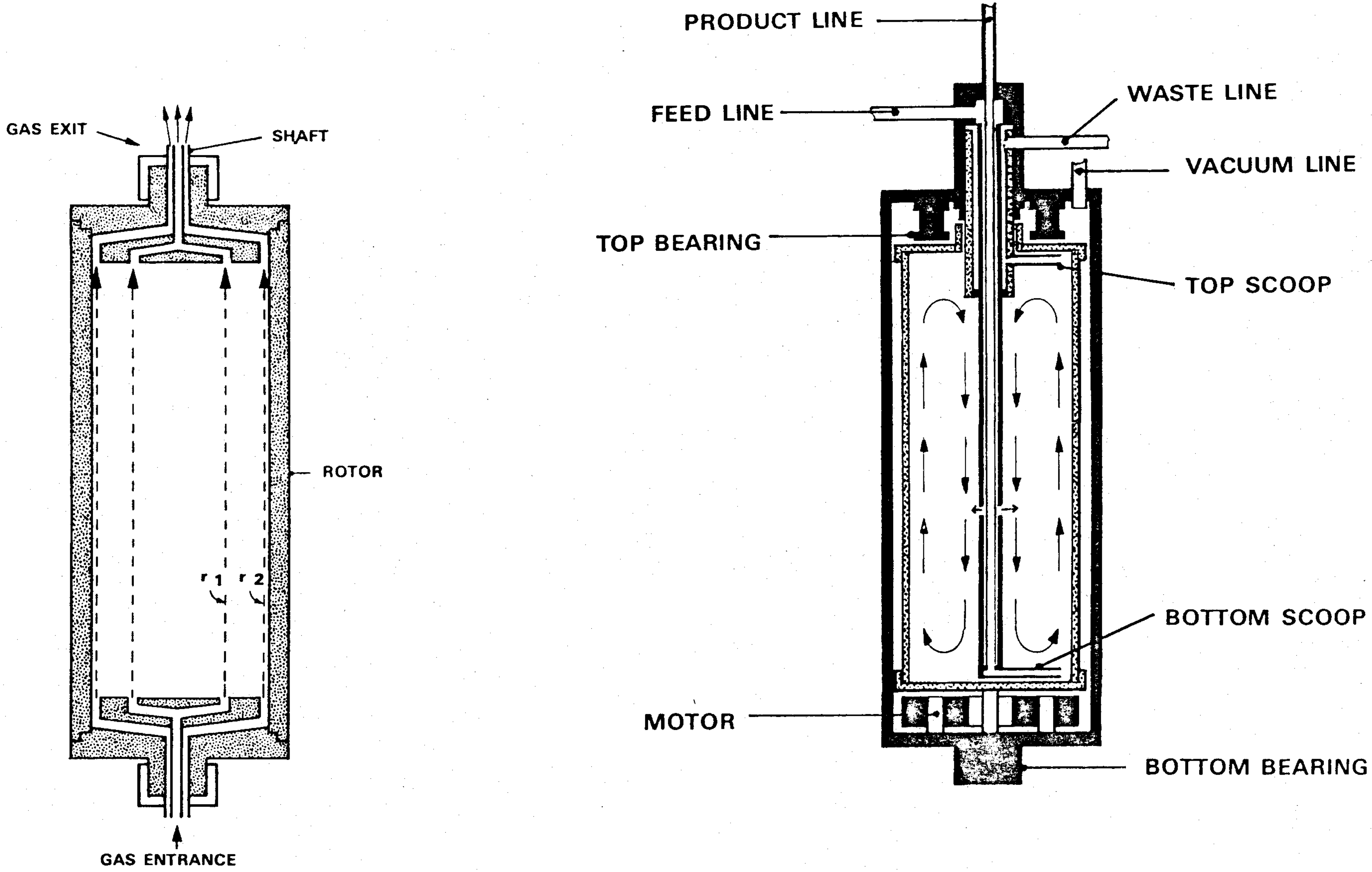
Centrifuge technology basically involves a cylinder containing the UF_6 gas rotating at a high rate of speed. The UF_6 gas within the cylinder has the same rotational speed as the cylinder. This rotational speed imposes a centrifugal force on the UF_6 molecules ($U^{235}F_6$ and $U^{238}F_6$) which modifies the density distribution resulting from random kinetic motion. The resulting density distribution for a gas of molecular weight M is:

$$\rho(r) = \rho(o) \exp\left(\frac{Mw^2r^2}{2RT}\right)$$

where $\rho(r)$ is the density at radius r and w the angular velocity of rotation. The key feature of this formula is that the molecular weight M appears in it. Thus, if two molecular species are present in the gas ($U^{235}F_6$ and $U^{238}F_6$), the concentration of one in relation to the other is different at the center from the rim of the cylinder.

In the early stages of development the simplest techniques were used to design a functioning centrifuge. The result of this early work was the Concurrent Centrifuge (Figure V-7). In this scheme, the gas (UF_6) flows in one end and takes up the rotational speed by collisions with the walls of the centrifuge. By the time the gas reaches the other end of the cylinder, the equilibrium radial density distribution should be set up making it relatively simple to draw off the heavy ($U^{238}F_6$) and light ($U^{235}F_6$) fractions.

From this early work, countercurrent centrifuges were designed offering better performance. Most of todays large scale centrifuge facilities utilize countercurrent machines (Figure V-7). In the countercurrent arrangement, the fed UF_6 is passed through an annular space and emerges into the center of the rotor. Pitot tubes, or 'scoops', are used to extract the gas from the top and bottom of the rotor which will be depleted and enriched, respectively, in the light isotope.



Concurrent Centrifuge

Countercurrent Centrifuge

Figure V-7

The ratio of the U^{235} concentration between product and waste is given theoretically by:

$$\frac{Cp}{Cw} = \exp\left(K_1 \frac{L}{d} \cdot \frac{\Delta M v^2}{RT}\right)$$

where L and d are the rotor length and diameter, v the peripheral velocity, K_1 a factor depending on throughput. At maximum separative work output, the ratio Cp/Cw is about half its maximum value.

The maximum separative work output is given by:

$$S = \left(\frac{K_2 \rho D \Delta M v^2}{2RT}\right)^2 \frac{\pi L}{2}$$

where ρ and D are the density and diffusion coefficient in the gas, and K_2 is a design constant. Note that the longer the centrifuge the more output, that for a given wall speed, rotor radius is immaterial, and finally and most importantly that on this simple theory, separative work output depends on the fourth power of wall speed. In reality, separative work output deviates significantly from its fourth power dependence on velocity but is still a very important factor.

So much for the physics of centrifuges. For practical outputs and enrichments they have to be operated in cascades, just as do diffusion plant stages. Some simplification results from the fact that the output of a centrifuge is measured in kilograms of separative work per annum, rather than metric tons per annum from a large diffusion plant stage. Thus, whilst diffusion plant stages of several different sizes have to be designed and installed, the optimum shaping of a centrifuge cascade can be achieved cheaply and simply by employing centrifuges of a single size and connecting them in a series arrangement.

Leap-frogging technology, we come to advanced isotope (enrichment) separation techniques. There are three advanced enrichment processes being investigated. These processes are the Plasma Separation Process (PSP), the Molecular Laser Isotope Separation (MLIS) Process, and the Atomic Vapor Laser Isotope Separation (AVLIS) Process. All of the processes work on physical principles which give rise to greater isotopic selectivity than either gaseous diffusion or gaseous centrifuge.

To start with, the PSP takes advantage of the difference in the gyrating frequency of different mass ions in a magnetic field. The fact that moving charged particles are deflected by a magnetic field is an old physical fact. This process exploits that fact. By applying an electromagnetic field whose frequency equals the frequency of the U^{235} ion, it is possible to accelerate the U^{235} ion over U^{238} which is not in resonance with the field. In this manner, the resonant U^{235} ion gains energy and increases its orbit size. As the U^{235} ion is heated it moves towards a collector array. The higher energy U^{235} ion has a larger probability of being collected as product than the lower energy U^{238}. At present, there are several areas requiring further resolution.

The next two processes are Laser Isotope Separation (LIS) processes. These processes also take advantage of different resonances in U^{235} and U^{238}, but these resonances are internal. Because these internal resonances (energy states) correspond to different degrees of internal energy, the resonant electromagnetic frequencies are considerably higher than that of PSP. In the case of a molecule, the internal vibrational and rotational levels of the molecule correspond to radiation in the infrared portion of the spectrum. For atoms, the electronic energy states correspond to radiation in the visible portion of the spectrum. Thus, these processes use lasers as their radiation source.

The Molecular Laser Isotope Separation (MLIS) Process involves the following. UF_6 gas is expanded through a nozzle along with an inert buffer gas. The established supersonic flow lowers the internal temperature of the molecule. This increases the spectral resolution (clarity) of the vibrational bands, and separates the U^{235} and U^{238} resonances to the point where a precisely tuned infrared laser can preferentially excite or heat the U^{235}. This increased heating makes it possible for a second laser (high power ultraviolet laser) to preferentially dissociate the $U^{235}F_6$ into UF_5+F. The new molecule UF_5 has a lower vapor pressure than UF_6 and can be made to condense on an appropriate collector.

The approaches to Atomic Vapor Laser Isotope Separation (AVLIS) utilize the reasonably large separation between the absorption bands of U^{235} and U^{238}. Using tunable dye lasers operating on excited levels of the atom, sufficient energy can be imparted to preferentially ionize only the U^{235}. Once the U^{235} is ionized (charged), one can use electric and/or magnetic fields to extract the ions onto a collector.

To conclude, while conceptually these advanced isotope separation processes appear to be relatively simple, the development beyond scientific feasibility are far more complicated and years away.

7. LWR Fuel Fabrication

Uranium in the form of uranium hexafluoride (UF_6) is enriched to the requirements of the fuel design, and shipped to a fabrication plant in UF_6 cylinders. The UF_6 is a solid at normal temperatures. It is sublimed in a steam chest and fed into the process. Water is mixed with the gaseous UF_6 to convert the gas into a manageable liquid form that permits precise process flow control. This reaction generates hydrofluoric acid and uranyl fluoride solution. This solution is pumped to a precipitation tank where it is reacted with ammonium hydroxide. There, the uranium precipitates out as ammonium diurante (ADU), a yellow solid. The slurry is pumped into a horizontal bowl centrifuge where the solid and liquid phases are separated. The overflow solution still contains a small amount of solid. Therefore, it is channelled into a high-frequency centrifuge where the balance of the solid is removed.

The wet solid stream, from the horizontal bowl centrifuge, is fed into a rotary calciner. In this step, the wet solid is dried and the ammonium diurante chemically is converted to uranium dioxide powder. When the uranium dioxide (UO_2) is discharged from the calciner, it is pulverized in a mill to obtain the desired particle size. To insure that the uranium dioxide powders have the same physical and chemical properties, cross blending is used. Blending is accomplished by mechanically mixing batches of powders of the same enrichment into one homogeneous mass.

Slugging (low pressure pressing) shapes the powder into a form resembling a coin about the size of a quarter. The pressed slugs are granulated, or crushed into small particles, and passed through a screening operation, which provides a properly balanced particle distribution. This improves the flowability of the powder, which is a requirement in obtaining proper feed for the pellet presses. The screened granules are fed into high-speed presses where the final fuel pellets are made. This pressing produces unsintered (green) density pellets. The green density determines the final size of the fuel pellets. The green pellets are now sintered at high temperature in electric furnaces. This process consolidates the aggregate of fine particles and results in shrinkage and densification of the pellet.

In the pressing and sintering operations, the fuel pellets are purposely formed slightly larger than the final size desired. To achieve the final size each pellet must be ground to the exact dimensions required. Grinding is performed on precision centerless grinders.

Fuel pellets, after inspection, are loaded into zircalloy tubes. The tubes are sealed by welding machined end plugs to each end of the tube. Prior to insertion of the top end plug, a spring is inserted into the fuel rod. The gap provides for fission gas accumulation and axial thermal expansion of the fuel column. Also, the spring prevents movement of the fuel column during handling and shipping.

The tubes are sealed by TIG welding the end plugs to the end of the tube and pressurizing the rods. After the rods are welded and leak tested, they are inspected for burrs, pits, gouges and discoloration. The rods are now ready for assembly into fuel assemblies.

The fuel assembly support structure consists of a top and bottom nozzle, rod cluster control guide thimbles and spring-clip grids. A skeleton assembly consisting of the bottom nozzle, thimble tubes, and grids provides the framework which receives the loaded fuel rods and later the top nozzle -

thus creating the final fuel assembly. Fuel rods are loaded by inserting them through the grids. After rod insertion, the top nozzle is welded to the control rod guide thimbles to complete the fuel assembly.

The completed fuel assemblies are inspected, cleaned, and packaged. Completed fuel assemblies are handled and stored in a supported upright manner until loaded into a shipping container for transportation to the reactor site. A core of a nuclear plant contains close to 10,000,000 UO_2 pellets and over 666,000 feet of zircalloy tubing. The fuel rods alone require about 80,000 welds.

There are variations to the fabrication process described above, a PWR fuel fabrication process, depending upon reactor type and even the fuel fabricator.

8. Magnox Fuel Fabrication

The uranium tetrafluoride (UF_4) is mixed with shredded magnesium and this mixture is compressed into pellets, each weighing about three (3) kilograms. These pellets are then stacked in a steel vessel lined with graphite. On heating of the vessel, a thermal reduction takes place and molten uranium and slag are produced. The molten uranium falls into a catch pot in the base of the vessel to form a billet upon solidifying.

The pure uranium billets are re-melted under vacuum, alloyed and cast into rods. The rods are subjected to heat treatment before being machined to size and finally inserted into Magnox cans. The cans are helium filled and closed by means of end caps which are welded into place. After this, the rods are pressurized. After cleaning, inspection and the fitting of various strengthening and stabilizing devices, the now complete fuel elements are packed into shock absorbing metal cans for transport.

Versatile production facilities are required to produce the varieties of Magnox fuel elements used today. The fuel elements range from simple helically finned cans through spiral polyzonais to the current herringbone designs with integral splitters. Furthermore, the uranium rods range from one (1) inch to 1.6 inches in diameter and 18 to 42 inches in length.

9. AGR Fuel Fabrication

The enriched uranium hexafluoride (UF_6) undergoes vapor phase pyrohydrolysis to UO_2F_2 powder in a fluidized bed reactor. Pyrohydrolysis of the UO_2F_2 to U_3O_8 is accomplished in a rotary kiln after which, the U_3O_8 is dissolved in nitric acid to produce a solution of uranyl nitrate. Precipitation of ADU, filtration and drying of the ADU slurry gives a free flowing powder. The powder (ADU) is calcined and reduced to UO_2 in a rotary kiln.

A shorter more economic dry process for the conversion of hexafluoride to ceramic grade UO_2 is now being used. This process is identical to that described (and used) in LWR fuel fabrication.

Once produced, the uranium dioxide pellets are oven dried, inspected for chips and formed into stacks which are length corrected and weighed. Simultaneously, AGR cans are scrubbed, vapor degreased, and bore plug gauged. The first stage of pin fabrication comprises loading the pellet stacks into the cans, brushing and suction cleaning the back end, helium filling, insertion of special insulation pellets and end cap roller spot welding at each end.

After mass spectrometer leak testing, the can ends are dressed off to the proper length and a second weld, an argon arch sealing weld, is applied to each end. Subsequently, pins are pressurized hydraulically, blast hot air dried and again leak tested. Some pin designs require that the pins be subjected to a closely defined heat treatment to produce the requisite can properties. After this, the pins are externally cleaned by electro-plating before water spray rinsing and hot air drying. After cleaning, component pre-assemblies comprising outer and inner graphite sleeves, retaining rings, grids and braces are prepared. For final assembly, 36 pins are inserted into the pre-assemblies and an extension piece extremity is spun over the bottom grid.

The stainless steel clad fuel pins range from 0.2 inches to 0.8 inches in diameter and from 36 inches to 54 inches in length.

10. CANDU Fuel Fabrication

For the CANDU system, since fluorination and enrichment are not required, the U_3O_8 is converted directly to UO_2 powder. This is accomplished via hydrogen reduction. The yellowcake is digested in a nitric acid solution. The saturated solution is fed through a solvent extraction circuit to obtain uranyl nitrate. The uranyl nitrate is then mixed with aqueous ammonia and the mixture is filtered. The precipitate - ammonium diuranate - is dried in ovens. The dried ammonium diurante then undergoes reduction to UO_2 in a reduction furnace with hydrogen.

The UO_2 powder is compacted into large diameter wafers or "slugs" under controlled pressure. The "slugs" are then broken up and passed through a sieve to produce a granular material suitable for feeding to the pill pressing operation.

The granulated material is then fed to a double action mechanical pill press where it is pressed into "green" compacts. The pressed "green" compacts are then sintered in a hydrogen atmosphere. The sintered pellets, after being discharged from the sintering furnace, are ground to the required diameter. Then, the ground pellets are washed, dried and inspected.

The UO_2 pellets are now ready for loading into zircalloy tubes. The pellets are loaded by hand into the tube sheaths. When the tubes have been loaded with pellets and end caps welded into place by a tungsten (or resistance) weld, the structure is called a fuel element (or rod). The end plugs of the finished element are profile machined to form a projection which will facilitate the welding of the elements to the end plate at the bundle assembly stage. The fuel elements are then vapor degreased.

The completed fuel elements are moved to the bundle assembly area. Here, the filled rods are assembled between zircalloy plates to form a bundle. The support plates are welded to the fuel elements at the proper angular position using a resistance projection weld. Structural fittings, such as wire wrap spacers, are added to complete the basic fuel design.

CANDU fuel is approximately 50 cm in length by 10.4 cm in diameter. Each bundle consists of 37 zirconium alloy tubes. Each of the fuel channels in a 600 MWe reactor contain 12 fuel bundles.

11. Heavy Water Production

For the production of heavy water (D_2O), large quantities of ordinary water are required. Although direct distillation could be used, an isotopic exchange process is more economical. By this method, the concentration of heavy hydrogen or deuterium is increased by successively raising and lowering the temperature difference between water and a gaseous hydrogen compound (H_2S). All naturally occuring hydrogen compounds contain some deuterium which can be extracted. The process is based on the fact that deuterium migrates to the water stream at low temperature and to the hydrogen sulphide gas at high temperature. By suitable arrangement of the flow in separating towers, deuterium can be extracted from a feed of ordinary water. In each tower the water flows down through a series of perforated plates, while the hydrogen sulphide bubbles up through the traps. This arrangement promotes efficient mixing.

In this manner, the hydrogen gas is enriched in deuterium and, leaving the top of the hot towers, passes into a cold tower where deuterium migrates to the water feed. A portion of the H_2S gas stream enriched in deuterium is extracted and passes to the next stage. This process is then repeated in the second stage and in a further stage. Enriched water from the third stage then passes to a finishing section where it is distilled to a reactor product that is 99.75% pure D_2O.

12. Reprocessing

The earliest known reprocessing occurred at the Hanford Laboratories in the early-to-mid 1940's. The process utilized bismuth phosphates and lanthanum fluorides to precipitate plutonium from the low burn-up, metallic fuel in a batch process (since the sole objective was to obtain plutonium for weapons, residual uranium was discharged with the fission product wastes).

Apparently, the process worked well, achieving acceptably high efficiencies for fission product removal and plutonium extraction. However, the waste volume was high.

Other than the very fact that it worked, the primary lasting technological achievements of the Hanford facility were the concepts of remote operation and maintenance.

The next technological steps took place in both the US and the UK in the late 1940's and early 1950's. In both countries, the objectives were to develop a process which could operate continuously and which would recover uranium as well as plutonium.

At Hanford, the basic solvent extraction process was refined and accepted. In this process, countercurrent flows of aqueous and organic solutions are made to move through some sort of mixing chamber (column, bowl, tank, etc.). The organic solvent strips both uranium and plutonium from the aqueous feed solution, leaving the fission products behind. Later, by adjusting valence, the plutonium can be made insoluble, thus separating the uranium and plutonium.

Hanford called this the Redox process and used Hexone (Methyl-isobutyl Getone) as the organic solvent. While the process indeed achieved the objectives of continuous operation and extraction of both uranium and plutonium, Hexone was expensive and flammable and the process generated very large quantities of waste.

The UK, constructing a separations plant at Windscale, developed a similar process but utilized a different solvent - Butex. Its major advantage over the US solvent, Hexone, was a significant reduction in waste volume.

Finally, during construction of the Savannah River Plant near Aiken, South Carolina in the early 1950's, the Purex process was developed. This process was tributylphosphate (TBP) dissolved in a kerosene-like solvent. TBP has advantages of being chemically stable in nitric acid, relatively cheap, generating low waste volumes, and superior separations capability.

The basic Purex process is now generally accepted as the fundamental operating process for all reprocessing plants (with the notable exception of the abortive GE Morris effort). Different head ends (the means of dissolving the fuel and chemically preparing it for entry to the Purex process) and different final conversions (the means of converting the recovered uranium and plutonium to the desired chemical/physical form) are used depending upon the type of fuel to be reprocessed and the end-use of the recovered products.

Virtually all development work from then on has focused upon the methods used to bring the aqueous and organic liquid streams together and then separate them. The apparatus to do this is called a contactor. This is, of course, the heart of the solvent-extraction process and the key to both the process efficiency and waste volume.

Simple vertical columns were used as the first and most obvious contactor. These extraction columns were packed with various metal or ceramic shapes to create a very long flow path. The heavy aqueous solution was introduced at the top of the column and allowed to flow downward under the influence of gravity. The lighter organic solution, introduced near the bottom of the column, was displaced by the aqueous solution and forced to flow upwards. Thus, a countercurrent flow could be established and because of the metal or ceramic shapes, intimate aqueous/organic contact occurred. However, since flow rates were low and aqueous/organic contact was gentle, the columns had to be very tall to achieve reasonable efficiency. (The process building at Windscale is said to be 20 stories tall.)

The first step in the evolution of more efficient contactors involved pulsed columns. Pulsed columns utilize multiple perforated plates and, by applying alternating positive/negative pressure "pulses", force the liquids to pass back and forth through the perforations. This imparts a vigorous mixing action of the two streams. Thus, even though the two liquid streams still moved only by gravity, the extraction efficiency was significantly improved and column height could be reduced. This type of pulsed extraction column was used at the Hanford, Idaho National Engineering Laboratory (for naval propulsion and research reactor fuel reprocessing), and Nuclear Fuel Services' plants.

The next evolution in contactors involved a device called a "mixer-settler". This was a horizontal device (i.e. - required much lower buildings) of multiple stages. In each stage, the aqueous and organic streams are first drawn together and vigorously "mixed" by an agitator. Then the mixed solutions are driven by the agitator into a long, horizontal "settling" chamber. In this chamber, gravity again takes effect and the lighter organic solvent rises to the top while the heavier aqueous solution settles to the bottom. Both solutions are then separately drawn off from the chamber and introduced to further mix/settle stages.

This contactor must be considered a major step in that the mixing action was very strong and - for the first time - the two streams were mechanically propelled through the stages. Since the device was horizontal it was very amenable to either remote maintenance (as in its first use at Savannah River) or to a design where the mixer motors were physically remote from the mixers (as at later Windscale facilities). Unfortunately, the design inherently involves large volumes of mixed solutions and thus, major inventories of dissolved uranium and plutonium. This in turn causes considerable chemical and radiolytic solvent degradation - involving both solvent makeup expense and somewhat increased waste volumes. This problem was the driving force to further improve contactor performance.

The centrifugal contactor, developed at Savannah River, was the next improvement. In this device, the mixed solutions are forced to flow from the motor driven agitator to a small centrifugal separator bowl mounted on the same shaft. In this bowl (which effectively replaces the long horizontal settler chamber), the aqueous and organic solutions are separated by centrifugal force.

In this device, for the first time, mechanical force was applied to all three primary solvent extraction sub-processes - stream movement, stream mixing, and stream separation. The results are impressive, including much lower in-process inventory (~25% of a mixer-settler), high separations efficiency, and low solvent degradation (and therefore, lower waste volume). Because of these advantages, Savannah River replaced their existing mixer-settler contactors with centrifugal settlers.

A derivative of the centrifugal contactor is the multi-stage centrifugal contactor developed by Saint-Gobain Techniques Nouvelles. This contactor, called Robatel, incorporates the equivalent of eight separate centrifugal contactors on a single motor driven shaft. This device is used in the first extraction stage at the AGNS Barnwell facility. Presumably, it may also be used in the planned new French plants and in any plants exported by the French.

Fuel reprocessing technology has been well established over the last twenty years in several countries, particularly for low burnup metallic fuels. Large scale experience has not yet been obtained in handling high-burnup oxide fuels.