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EXISTING AND PROJECTED PLANTS AND PROCESSES FOR THERMAL REACTOR FUEL RECOVERY: EXPERIENCE AND PLANS

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Introduction

This paper presents a summary of the status of industrial fuel reprocessing facilities in the U.S.A. It covers those plants which are built and operating, under construction, or in a final planning stage. In a very general manner it describes the facilities and technology utilized, innovations in process technology and equipment, and experience gained to date. Discussed briefly are recent U.S. policy developments in the siting of fuel reprocessing and waste management facilities. Possible future technological requirements are discussed.

Nuclear Fuel Services Plant

The Nuclear Fuel Services (NFS) plant at West Valley, New York was the first industrial fuel reprocessing plant built in the U.S. It is located on a 3300 acre site owned by the State of New York, about 32 miles southeast of the city of Buffalo. Plant construction was completed late in 1965. Hot operations were started in April 1966.

The NFS plant is a multipurpose plant principally designed to reprocess LWR fuels (UO₂ and UO₂-PuO₂ up to about 5% enrichment in U-235 or equivalent reactivity) at a design rate of 1 MTU/day. It also has a capability to reprocess ThO₂-UO₂ fuels, and certain high enrichment U-alloy fuels at a reduced rate. The process employed consists of a shear-leach mechanical head-end treatment followed by several cycles of tributyl phosphate (Purex) solvent extraction and ion exchange.

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Irradiated fuel is received at the plant either by truck or rail. The plant is capable of receiving and handling LWR fuel as long as 16 ft. and 6 in. square. Irradiated fuel is cooled for a minimum of 150 days from reactor discharge prior to reprocessing. When scheduled for reprocessing, fuel is transferred to a mechanical cell where the end hardware is removed by a remotely operated saw. The prepared fuel element is loaded into the shear magazine, where it is pushed, by a ram, into the shear. The shear is a 300-ton hydraulic unit in which the fuel can be sheared into pieces ranging in length from 1/2 to 2 in. The sheared pieces fall into a chute and are collected in chopped fuel canisters.

The chopped fuel canisters are perforated boron-stainless steel cylinders 8 in. ID by 7 ft. high. Because shearing disintegrates much of the fuel into powder, the canisters are fitted with consumable thin-wall mild steel liners.

The NFS dissolver design is somewhat unique. Its design was complicated by the fact that it had to be capable of handling fuel of widely varying composition and enrichment. It consists of a lower 3 in. annulus which contains six 10 in. ID barrels which accept the 8 in. fuel baskets. This portion of the vessel sits down over a boron-containing concrete core which serves to isolate each barrel position from the others. The top of the annulus connects with a full diameter surge tank which contains the amount of dissolvent necessary to produce a final fuel concentration which will be critically safe under all conditions.

Off-gas from the dissolver is treated by passage through a downdraft condenser, a secondary condenser, a scrubber, iodine removal by absorption on a chemical trap, and filtration through high-efficiency particulate air filters. The filtered discharge is exhausted through a 63-meter stack. This stream includes the chemically inert but radioactive species of krypton, and a portion of the tritium initially contained in the fuel.

Fuel solution from the dissolver is transferred to a feed adjustment and accountability tank. The leached hulls are rinsed with water, and this rinse liquid is added to the fuel solution. After the hulls have dried they are inspected and sampled to determine what losses may have been experienced, after which they are buried as solid waste.

The volume of the fuel solution in the accountability tank is measured accurately and sampled for analysis. This analysis represents the feed input to the system and establishes the financial accountability of the plant.

The recovery and decontamination of uranium and plutonium from the fuel solution is accomplished by a Purex solvent extraction process in which the solvent is tributyl phosphate dissolved in n-dodecane. Pulse column contactors are used. The first cycle is a partition cycle in which uranium and plutonium are separated from each other and from 99.9+% of the fission products. Two additional cycles of solvent extraction are used to complete the purification of the uranium product. The plutonium product is processed through one additional solvent extraction cycle, followed by anion exchange treatment to achieve final purification and concentration; it is then further concentrated in a titanium evaporator and transferred to bottles for shipment.

The acid waste streams are evaporated to concentrate them for storage, to decontaminate the excess liquid, and to recover nitric acid for recycle. The principle process off-gases from the operating vessels are contained in treatment systems designed to remove radioactivity and are subsequently discharged with process building exhaust gases to the atmosphere through a high efficiency filtration system.

The high-level radioactive waste concentrate is neutralized and stored in 750,000-gal. waste tanks that are buried underground. These liquid wastes will eventually be converted to stable solids for ultimate disposal.

The NFS plant has now been in operation for almost $5\frac{1}{2}$ years and has processed about 700 tonnes of irradiated fuel. The most highly irradiated fuel processed to date was irradiated to a burnup of 22,500 MWD/T and cooled 9 months.

In general, performance of the plant has been very good. Recovery of fissile materials has been in excess of 98.5%, decontamination of uranium and plutonium from fission products has been satisfactory, and in-specification products are produced routinely. The plant consistently has met all federal regulations for the release of gaseous and liquid radioactive wastes to the environs.

The performance of the shear has also been good. As the first unit in hot operation, some design modifications and repairs have been necessary, but these were accomplished remotely without excessive downtime.

The existence of some design deficiencies in the plant, mainly in the acid recovery and ventilation systems, was encountered. Two filters in the off-gas system failed, releasing some activity to the environs. In neither case was the release near the maximum permissible level. In addition, the routing of certain off-gas lines created radiation fields in certain work areas. Corrective measures have been taken to resolve these problems.

Modifications to the plant are currently in progress which will increase the capacity for reprocessing LWR fuels from the design rate of 1 MTU/day to 2-3 MTU/day. Future planned modifications include the addition of facilities to convert $\text{Pu}(\text{NO}_3)_4$ to PuO_2 , uranium nitrate hexahydrate (UNH) to UF_6 , and liquid waste (high level) to solid form.

Midwest Fuel Recovery Plant

Construction of General Electric's Midwest Fuel Recovery Plant (MFRP) at Morris, Illinois has been completed, and cold shakedown operations are in progress. The plant has an initial capacity of 1 MTU/day to recover uranium, plutonium and neptunium from LWR fuels originally enriched to about 5% in U-235 or equivalent plutonium reactivity. Figure 1 is a simplified block diagram of the patented Aquafluor⁽¹⁾ process which is summarized below.

(1) Service Mark of General Electric Company

FIGURE 1. AQUAFLUOR* SERVICE

*Service Mark of the General Electric Company

Feed preparation is by mechanical disassembly of the as-received fuel after which the fuel rods are sheared into short lengths and fed into a dissolver. The fuel, containing the plutonium, neptunium and fission products formed during irradiation as well as the unspent uranium is leached out of the cladding hulls with nitric acid. After fuel dissolution, the cladding hulls are conveyed to a storage area of capacity appropriate to long range plant needs.

Initial separation of the fuel constituents of economic interest from 99.9+% of the highly radioactive fission products is by a single, co-decontamination solvent extraction cycle of the Purex type. The high activity waste stream which results from this operation is reduced to a small volume of dry, inert solids for retention in cooling basins provided as part of the plant facilities. Solvent degradation products resulting from the highly radioactive environment are washed from the solvent and routed to the high activity waste disposal system provided for the plant.

Recovery and final decontamination of plutonium and neptunium are accomplished by anion exchange, after which the recovered products are concentrated and then routed to loading facilities in the form of nitrate solutions. Spent resins -- caused either by chemical or radiolytic degradation -- are sent to the high activity waste handling system for disposal.

The uranium process stream coming from the ion exchange operation is fed to an intercycle concentrator to form a UNH solution suitable for the subsequent calcination operation. Conversion of the UNH to UO_3 is accomplished by means of a fluidized bed calciner. Particulate UO_3 which is elutriated from the calciner is routed to a fines recovery tower wherein the UO_3 is dissolved in nitric acid and subsequently recycled to the UNH concentrator for uranium recovery. Finally, the UO_3 is reacted with elemental fluorine to form UF_6 in a fluidized bed fluorinator. The inert bed material used in the fluorinator (fused alumina) periodically will be disposed of to the low level waste disposal system. Final purification of the UF_6 from traces of plutonium, neptunium and certain fission products is accomplished by passing the gaseous UF_6 through NaF and MgF_2 beds followed by distillation of liquid UF_6 .

A disposal system is provided for high activity wastes (i.e., those wastes which are both radioactively and thermally "hot") in which the waste streams are concentrated, reduced to solid form, and packaged in high integrity containers so that they can be submerged in a water filled basin for on-site retention prior to shipment to an AEC repository.

A low activity waste disposal system is provided which includes liquid concentration equipment and an underground storage vault in which the radioactive material sets up as a salt cake, on cooling, for immobilized on-site retention. Gaseous waste streams from the process are all filtered, scrubbed, and monitored prior to release from the plant ventilation stack. There are no radioactive liquid effluents from the GE recovery plant.

Figure 2 is an artist's rendering of the MFRP. Seen in the foreground are the high activity waste pool, the cladding storage vault and the low activity waste pool. Indicated are the mechanical cell where the fuel is disassembled and chopped into small pieces, from which the core material is leached, and the main process canyon area. On the right hand side toward the center of the picture is the sand filter through which all the ventilation air is filtered prior to discharge through the stack.

The measurement techniques to be used by the reprocessor are important in accounting for the valuable materials which are being recovered. General Electric has chosen to base the uranium input to the recovery plant on the weight of uranium as originally fabricated and charged into the reactor. The fuel fabricator's certified weight of uranium is adjusted for burnup and transmutation based on mass ratio analysis. The uncertainty in this gravimetric method is about 0.22% as compared to the approximately 0.75% associated with the alternate volumetric technique. The plutonium input is determined by multiplying the uranium input by the plutonium to uranium ratio. The output uranium and output plutonium are based on direct measurements using weighing techniques.

General Electric considers transportation of the irradiated fuel from the reactor to the reprocessing plant to be a key component of fuel recovery service. General Electric has designed shipping casks for the spent fuel and their design and other transportation considerations are discussed by C. W. Smith, et al⁽²⁾.

Barnwell Nuclear Fuel Plant of Allied-Gulf Nuclear Services

The Barnwell Nuclear Fuel Plant (BNFP) of Allied-Gulf Nuclear Services is designed specifically to process irradiated fuels from LWR plants. The plant, with a design capacity of 5 MTU/day, is located on a 1700 acre site near Barnwell, South Carolina at the eastern boundary of the AEC Savannah River Plant. The location permits receipt of fuel by highway and rail as well as water shipments via the Savannah River. The Construction Permit for the plant was granted by the AEC in December 1970. Construction commenced early this year, and cold start-up will be initiated in 1973.

Paramount considerations in the design of the plant have been the safety of the workers, the safety of the general public and the protection of the environment. No radioactive materials will be added to the cooling water discharged from the plant to local water courses. Reliability of operation was also of key importance in selection of the process and design of equipment. To assure its customers of on-time delivery of recovered products as well as for the plant to be economical, it must be able to maintain a high on-stream capability, at an optimum capital cost, performing as designed. The plant is capable of receiving LWR fuel as long as 20 ft. overall and 11½ in. square; enriched to 5% U-235 prior to irradiation, or equivalent reactivity with recycle plutonium; burnups of up to 40,000 MWD/T; and exposure to power densities as high as 50 megawatts per ton. The plant will be able to recover uranium and plutonium as nitrates, or uranium as the hexafluoride and plutonium as the oxide. In addition, provisions have been

(2) Smith, C. W., et al "Shipment of Irradiated Power Reactor Fuel in the United States"

made to allow neptunium or other potentially valuable by-products to be recovered if desired. The high plant capacity will make possible timely availability of recovered products for use in recycle fuel.

Accountability of special nuclear material has been given a high priority in equipment design and in the establishment of operating techniques. To assure the highest level of special nuclear material safeguards, Allied-Gulf has elected to base input measurements on methods which are independent of fabrication.

While the process employed in the BNFP will be essentially the classical Purex solvent extraction process using a shear-leach headend system, certain unique features of process and plant design have been incorporated. These are briefly discussed as follows:

1. The plant was designed with considerable flexibility in the fuel receiving and handling areas. This was necessary to allow accommodation of the many fuel casks which must be handled every day to supply this large plant. In addition, the design incorporates a capability to safely handle any leaking fuel elements which might be received.
2. The shear used in the BNFP is of unusual design in that it has capability to shear fuel without prior disassembly of fuel elements and fittings. Grids and other items of hardware will be chopped directly into the dissolver. The shear will be able to accommodate the multiplicity of sizes which will come from reactors of varying generations and different manufacturers. It is designed to confine dust resulting from the shearing operation so that there will not be gross contamination of the process cell. It is also designed for ready maintenance. The shearing-dissolution system is sized for operation at a capacity substantially in excess of the plant's five tonne per day rate to allow for minor routine maintenance and for clean out and accountability between batches of fuel from different sources.
3. With a plant this size it was felt strongly that, for proper control of gaseous effluents, dissolution should be as continuous as is practical to minimize the peaking of any off-gas release which could not otherwise be contained. To accommodate this requirement, the BNFP design incorporates a unique dissolver which operates in a semi-continuous mode with dissolution taking place as fuel is chopped directly into the nitric acid solution. Provision is made to insure that dissolution is taking place at the desired rate. This prevents an excess of accumulated undissolved material and uneven off-gas generation.
4. A somewhat troublesome feature of Purex processes is the addition of chemicals in various stages of the solvent extraction cycles for valence control. This addition of chemicals results in increased volumes or higher potential corrosion rates of the acidic solutions. The most commonly used reductant, ferrous sulfamate, gives, in nitric acid, the undesirable products of ferric nitrate and sulfuric acid as well as an increase in volume of stored wastes through increased salt content. To improve this step in the process, an electrolytic

means of valence control has been developed for the BNFP. This is used both in the partitioning step, where uranium and plutonium are separated after the bulk of the fission products have been removed, and is also used in the subsequent cycles of plutonium purification. Allied-Gulf is enthusiastic about this means of valence control because of its applicability to future plants handling breeder reactor fuel with high plutonium content.

5. It is a requirement that high activity wastes be contained with no chance of dispersion to the environs. At the BNFP, interim storage of the high activity wastes, prior to their conversion to stable solids, will be in stainless steel tanks located in massive concrete vaults which are lined with stainless steel. The plant will be equipped with spare tankage so that the contents of any tank can be transferred to another in the unlikely event that a leak should develop.

Removal of fission product heat from the wastes will be accomplished by submerged coils through which water will be circulated in a secondary circuit, with the heat ultimately being removed via exchangers through which primary cooling water will flow. The maintenance of cooling capability will be assured by provision of multiple back-up cooling water supply and alternate full capacity diesel-driven pumps.

6. Additional significant features of the plant are: (a) The maintenance philosophy includes use of both remote and contact maintenance. Arrangement of the cells is such that the contact cells are aligned adjacent to the remotely maintained cells. (b) In addition to the cells provided for the process, the BNFP has had incorporated in it liberal areas for maintenance outside of the process cells. These facilities are shielded and equipped with master slave manipulators, power manipulators, cranes and decontamination equipment so that equipment worth salvaging can be repaired after it has been removed from the cells. (c) A series of sampling and analytical cells, adjacent to the contact cell system, house all of the remote sampling systems as well as a number of the in-line instruments which require shielding and have the likelihood of requiring reasonably frequent maintenance. Here high activity samples can also be taken and prepared for handling with minimal shielding in the laboratory areas. Considerable attention has been paid to the arrangement of the plant to provide for an efficient ventilation system as well as a system wherein personnel traffic can be accommodated without the need for frequent passages through areas of possible contamination.

The Barnwell Nuclear Fuel Plant, the first five tonne per day plant, designed solely for commercial light water reactor fuel processing, incorporates a unique combination of the Purex process and several contributing innovations. From its inception, the plant has been designed to be safe to both operator and public alike with minimal environmental impact.

Atlantic Richfield Reprocessing Center

The Atlantic Richfield Reprocessing Center (ARC Plant) is designed specifically to process irradiated fuels from LWR plants. It will be located on a site of approximately 2500 acres adjacent to the Broad River, approximately 13 miles southwest of Chester, South Carolina. The plant will have a nominal capacity of 6 MTU/day. Application for a Construction Permit was made on October 29, 1970, and plant start-up is scheduled for mid-1976.

The basic ARC Plant process functions will be conducted in a massive, high-integrity concrete structure (Process Building) especially designed to isolate operating personnel and the public from radioactivity by the provision of radiation shielding and strict radiological confinement. To provide additional isolation of the plant from the surrounding environs, the principal facilities will be located near the center of the site. Personnel access to the area and its facilities will be controlled.

Spent fuel assemblies from LWR-type reactors are received at the Process Building by rail or truck shipment. Typical fuels are expected to be enriched to a maximum average of about three percent U-235, but the plant will have the capability for processing mixed oxide ($(\text{UO}_2\text{-PuO}_2)$) fuels and higher enrichment fuels up to about five percent U-235, or equivalent reactivity. At the ARC Plant, the fuel assemblies are unloaded from the shipping casks in a water-filled pool, where they are also stored until scheduled for reprocessing.

When scheduled for reprocessing, fuel assemblies are transferred mechanically to a Remote Mechanical Cell, within the Process Building, where they are disassembled by shearing into suitable lengths, e.g., one to three inches long. As they are sheared, the pieces fall into a dissolver. In the dissolver, the fuel is leached from the insoluble cladding with nitric acid. The residual leached hulls are lifted in baskets from the dissolver and are transported to high-integrity Hull Disposal vaults for isolation from the surrounding environment.

The nitric acid solution of fuel is the feed for the solvent extraction separation of uranium and plutonium from each other and from fission products and other impurities. In the ARC Plant, a Purex solvent-extraction process is used with pulse column contactors. Three cycles of separation and purification are employed. The organic solvent is a 30 percent solution of tributyl phosphate in a normal paraffin hydrocarbon diluent. The product forms will be UNH , UF_6 , $\text{Pu}(\text{NO}_3)_4$, and/or PuO_2 as necessary for shipment and return to the nuclear fuel cycle. Provision has also been included for recovery of neptunium as a nitrate solution.

The spent organic solvent from the solvent-extraction systems is continuously reclaimed for reuse in specifically designed organic treatment systems. Nitric acid is continuously recovered by distillation and rectification from waste concentration operations and is recycled.

The treatment of liquid and gaseous streams containing radioactive waste products results in effluents meeting all regulatory requirements. Liquid effluent systems are engineered to effectively eliminate their radionuclide content through the recycle of process condensates to the extent practicable. The principal process off-gases from the operating vessels are contained in treatment systems designed to remove radioactivity (exclusive of the krypton and tritium components), and are then discharged along with the Process Building ventilation exhaust gases to the atmosphere through a high efficiency filtration system.

Essentially all of the fission products are contained in the high level waste generated during solvent-extraction operations. These liquid wastes are stored in vessels in a vault within the Process Building structure. Their ultimate solidification and disposal will be consistent with Atomic Energy Commission requirements.

Recent Developments In Siting and Waste Management Criteria

From the time it assumed responsibility for nuclear energy programs in 1946, the Atomic Energy Commission has been acutely aware of, and concerned about, hazards associated with the highly radioactive wastes produced in reprocessing nuclear fuels. While over 25 years of experience has been gained with the safe operation of reprocessing and related waste management facilities utilizing tank storage of liquids, there has been a continuing concern that these long-lived wastes ultimately be converted to a more stable form and isolated from man's environment for the many thousands of years required for their radioactive decay. To this end, the AEC has conducted extensive research and development programs to characterize the wastes and to determine the most suitable form and location for ultimate disposal. Results of this research and development have led to the conclusion that the wastes should be solidified for transportation to, and containment in, selected deep geologic formations, such as bedded salt, for long-term storage.

The increasing acceptance of nuclear reactors by the U.S. power utility industry, and the projected requirement for more and larger capacity fuel reprocessing plants led the AEC several years ago to consider the establishment of a policy for the siting of industrial fuel reprocessing plants and related waste management facilities. Primary concern in these deliberations was given to the development of a practicable policy based on the need to restrict, in the interest of public health and safety, the quantities and mobility of the high-level radioactive wastes stored on-site at fuel reprocessing plants. As a result of the studies which were conducted, and after discussion with the industry, a policy was established. Background information leading to establishment of this policy is discussed in detail in U.S. Federal Register Notice No. 35FR17530 dated November 14, 1970. Appended to it is a new regulation for incorporation into the Code of Federal Regulations. The new regulation, "Appendix F, 10CFR Part 50", became effective on February 12, 1971.

In addition to the previously existing regulations for the siting, licensing and operation of industrial nuclear facilities, the new regulation provides the following criteria for siting and operating licensed fuel reprocessing and related waste management facilities.

1. Reprocessing plants need not be located on federal land.
2. High-level liquid waste inventory must be limited to that produced in prior five years.
3. All high-level waste must be transferred in AEC-approved, dry, stable, solid form to a federal repository no later than ten years following separation of fission products from the irradiated fuel.
4. The Federal Government will have physical responsibility and industry financial responsibility for disposal of high-level waste.
5. Disposal of high-level fission product waste material will be permitted only on land owned and controlled by the Federal Government.
6. A design objective for reprocessing plants shall be to facilitate decontamination and removal of all significant radioactive waste from the plant, prior to decommissioning.

Industrial fuel reprocessing and related waste management facilities currently under construction and being planned will conform to the new regulatory criteria. Modification of equipment and procedures will be required at the NFS plant, since this plant was constructed prior to establishment of the policy discussed above.

Possible Future Technological Requirements

In support of our commitment to reduce the release of radioactivity from U.S. nuclear facilities to the minimum practicable, we look forward in the 1970's to increased research and development of technology for the removal and containment of radioactivity from plant effluents, particularly from the gaseous effluents. While release to the environment of fission gases which results from current operating practices is within a very small fraction of the internationally accepted environmental limits, we feel that an adequate technological base must be available in the not too distant future for the removal of these gases to high decontamination factors. As a result, the development of this technology has been defined as a firm objective of the AEC program for development of reprocessing technology for fast breeder reactor fuels. It is expected that this technology will find application in LWR fuel reprocessing plants as well.

Conclusion

In conclusion, we are convinced that the U.S.A. now has a technically sound and economically competitive base capability for reprocessing LWR fuels from the growing nuclear power utility industry. Improvements in process equipment and procedures are expected in these plants in the next decade as additional experience is gained and more efficient economic modes of operation become apparent. Looking further into the future, we expect this industry to rise to the greater challenge of reprocessing the more advanced breeder reactor fuels by incorporating in these plants the required new technology which we are developing, or by building new facilities specifically designed for that purpose.

EXISTING AND PROJECTED PLANTS AND PROCESSES FOR THERMAL
REACTOR FUEL RECOVERY: EXPERIENCE AND PLANS

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(see attachment 1)

The status of existing and planned industrial nuclear-fuel reprocessing facilities in the U.S.A. is summarized. The technology utilized, innovative modifications in process technology and equipment, experience gained, and potential future technological requirements are discussed.

The first commercial facility, the Nuclear Fuel Services Plant at West Valley, New York, has successfully operated since April 1966. The process employs a shear-leach head end, a technology first applied in this plant, followed by Purex decontamination and separation of uranium and plutonium, and a final ion-exchange cycle for plutonium. Current plans are to increase plant capacity from 1 MT/day to 2 to 3 MT/day of light-water-reactor (LWR) fuel.

Construction of the General Electric (GE) Midwest Fuel Recovery Plant at Morris, Illinois, has been completed. The design is based on the GE "Aquafluor" process and has the capability, at 1 MT/day, of recovering plutonium, uranium, and neptunium from LWR fuels. A mechanical head end provides for fuel disassembly, multirod shearing, and vibratory tray dissolution. A Purex cycle provides separation of uranium, plutonium, and neptunium from fission products. Partition is accomplished by anion exchange. The uranium stream is converted to UO_3 and UF_6 in fluidized-bed converters. A unique feature is the use of the fluorination portion of the fuel cycle to effect the final uranium decontamination. High-level wastes are converted to solids.

The Allied-Gulf Barnwell Nuclear Fuel Plant near Barnwell, South Carolina, is designed to reprocess 5 MT/day of LWR oxide fuel. It is expected to be in commercial operation early in 1974. A mechanical chop-leach head end is coupled with basic Purex technology. Novel features include a unique semicontinuous dissolver design and utilization of centrifugal contactors and electrolytic reduction of plutonium in the extraction system.

The Atlantic Richfield Company (ARCO) plans to build a reprocessing plant near Leeds, South Carolina. The ARCO plant, with a design capacity of 6 MT/day for LWR fuel, is expected to be in commercial operation by July 1976. Current plans are to use a chop-leach head end and Purex technology and include a capability for neptunium recovery.

A recently developed national policy on reprocessing-plant siting and waste disposal defines criteria for use in the U.S.A.

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Attachment 1 (Sinclair, Session 2.4)

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