EXTERNAL REVIEW DRAFT

BACKGROUND, STATUS, AND ISSUES RELATED TO THE REGULATION OF ADVANCED SPENT NUCLEAR FUEL RECYCLE FACILITIES

ACNW&M White Paper

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LIST OF ACRONYMS

ABR	Advanced burner reactor
ABWR	Advanced boiling water reactor
ACRS	Advisory Committee on Reactor Safeguards
ACNW	Advisory Committee on Nuclear Waste
AFCI	Advanced Fuel Cycle Initiative
AFCF	Advanced Fuel Cycle Facility
AHA	Acetohydroxamic acid
ALARA	As Low as reasonably achievable
AVR	Arbeitsgemeinschaft versuchsreaktor (working group test reactor)
BNFP	Barnwell Nuclear Fuel Plant
BWR	Boiling water reactor
CANDU	Canada deuterium uranium
CCD-PEG	Chlorinated cobalt dicarbollide-polyethylene glycol
CEQ	Council on Environment Quality
CFR	Code of the Federal Register
CFRP	Consolidated Fuel Reprocessing Program
Ci	Curies
CMPO	octyl-(phenyl)-N,N’-disobutylcarbamylphosphine oxide
CNNC	China National Nuclear Corporation
COEX	Co-extraction
COL	Construction and operating license
CP	Construction permit
DBA	Design basis accident
DOE	Department of Energy
DTPA	Diethylenetriaminepentaacetic acid
DUPIC	Direct use of spent PWR fuel in CANDU reactors
EBR-II	Experimental breeder reactor II
EIS	Environmental Impact Statement
EOI	Expression of interest
FBR	Fast breeder reactor
FRSS	Fuel receiving and storage station
FS-13	Solvent for extractants in CCD-PEG process
GANEX	Global actinide extraction
CBZ	Glass-bonded zeolite
GCR	Gas-cooled reactor
GDC	General design criteria
GEN IV	Generation IV
GESMO	Generic Environmental Statement on Mixed Oxide Fuel
GIF	Generation IV International Forum
GNEP	Global Nuclear Energy Partnership
GNI	Global Nuclear Infrastructure
GTCC	Greater than Class C
GW	Giga-watt
GWd	Giga-watt day

HAN	Hydroxylamine nitrate
HDEHP	di-2-ethylhexyl phosphoric acid
HEPA	High-efficiency particulate air filter
HILC	High-intermediate-level cell
HLC	High-level cell
HLGPT	High-level general process trash
HLW	High-level waste
HTGR	High-temperature gas-cooled reactor
HTTR (Japan)	High-temperature engineering test reactor
IAEA	International Atomic Energy Agency
ILC	Intermediate-level cell
INFCE	International Nuclear Fuel Cycle Evaluation
INIE	Innovation in Nuclear Infrastructure and Education
INPRO	International Project on Innovative Nuclear Reactors and Fuel Cycles
IPS	International plutonium storage
IPyC	Inner pyrocarbon layer
ISA	Integrated Safety Analysis
ISFSI	Independent spent fuel storage installation
KARP	Kalpakkam reprocessing plant
LAW	Low-activity waste
LLGPT	Low-level general process trash
LLW	Low-level waste
LMFBR	Liquid metal fast breeder reactor
Ln	Lanthanide
LS-VHTR	Liquid salt VHTR
LWR	Light water reactor
MAA	Material access area
MFRP	Midwest Fuel Reprocessing Plant
MNA	Multilateral approaches to the nuclear fuel cycle
MOX	Mixed oxide
mR	Millirem
MSR	Molten salt reactor
MSRE	Molten salt reactor experiment
MTR	Materials test reactor
MTIHM	Metric tonnes initial heavy metal
MTU	Metric tonnes uranium
MWe	Megawatts electrical
NEPA	National Environmental Policy Act
NERAC	Nuclear Energy Research Advisory Committee
NFS	Nuclear Fuel Services
NPP	Nuclear power plant
NRC	Nuclear Regulatory Commission
NWPA	Nuclear Waste Policy Act
OPyC	Outer pyrocarbon layer
PHWR	Pressurized heavy water reactor

PIC	Polymer-impregnated cement
PNSL	Plutonium nitrate storage and load-out cell
PPC	Plutonium product cell
PPF	Plutonium product facility
ppmw	Part per million by weight
PRA	Probabilistic risk assessment
PREFRE	Power reactor fuel reprocessing facility
PWR	Pressurized water reactor
PyC	Pyro-carbon
RMSC	Remote maintenance scrap cell
ROP	Reactor oversight process
RPC	Remote process cell
SiC	Silicon carbide
SRM	Staff requirements memorandum
SSNM	Source and special nuclear material
SX	Solvent extraction
TALSPEAK	Trivalent actinide-lanthanide separation by phosphorous reagent extraction from aqueous complexes
TBP	Tri-n-butyl phosphate
THTR	Thorium high-temperature reactor
TRISO	Tristructural-isotropic
TRU	Transuranium
TRUEX	Transuranium extraction
VA	Vital area
WSP	Waste solidification plant
WTEG	Waste tank equipment gallery
WVDP	West Valley Demonstration Project
YM	Yucca Mountain

SUMMARY

[To be provided later]

I. INTRODUCTION

The U.S. currently has 103 commercial nuclear power reactors that produce more than 2000 tonnes of spent nuclear fuel each year. DOE estimates that the Congressionally mandated capacity limit of 70,000 tonnes of heavy metal equivalent imposed on the proposed Yucca Mountain (YM) repository as presently planned will be reached by accumulated spent commercial fuel and other types of waste by about 2010 leading to the need for additional disposal capacity beyond this time. The environmental impact statement (EIS) for the Yucca Mountain project analyzed acceptance of up to 120,000 metric tons of nuclear byproducts in the repository. Such an expansion could accommodate spent fuel from an additional 35 years of operating existing nuclear power plants but proportionately less if the anticipated growth in nuclear power occurs. Factors that may further increase the need for additional geologic disposal capacity include spent fuel from reactors undergoing license extensions, new reactors similar to those presently deployed, and new types from advanced reactors being designed in DOE's Generation IV[1] initiative.

In the conference report associated with the FY 2006 Energy and Water Appropriations bill [Congress, 2005] Congress directed DOE to select a site for an integrated nuclear fuel recycle[2] facility by FY 2007 and to initiate construction of one or more such facilities by FY 2010. DOE subsequently submitted a program plan [DOE, 2006] and a strategic plan [GNEP-167312, Rev 0] providing details of their path forward and has continued to refine these plans. Fuel recycle would have a profound impact on the nature and amount of nuclear materials handled in and released from various fuel cycle (including transportation) and waste disposal facilities. In particular, recycle would have the following direct effects on the nuclear fuel cycle:

- Involve handling nuclear materials that could include mixtures of plutonium, minor actinides (e.g., ^{237}Np), and fission products as well as relatively pure ^{99}Tc, ^{129}I, ^{90}Sr, and ^{137}Cs in substantial quantities,

- Release gaseous radionuclides (tritium, ^{14}C, ^{85}Kr, ^{129}I) from the spent fuel matrix and potentially to the atmosphere depending on the retention technologies applied,

- Change the nature and amount of wastes going to disposal facilities including YM.

Fuel recycle would substitute one or more high-level waste (HLW) forms for spent fuel, and substantially reduce the volume and radionuclide compostion of the HLW as compared to the original spent fuel. This in turn could result in increasing the equivalent amount of spent fuel stored per unit repository volume by factors estimated by DOE to range from around 4 for relatively modest separations to over 200 for intensive separations [Laidler, 2006] as well as reduce the radionuclide inventory available to constitute a repository source term. This increase in storage efficiency is achievable because key heat-producing radionuclides (i.e., actinides, ^{137}Cs, and ^{90}Sr) would not be present in the HLW. There is the potential for numerous new wastes requiring disposition such as cladding hulls, waste forms containing gaseous radionuclides, solid wastes containing significant concentrations of transuranic elements, and

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[1] The U.S. Department of Energy's Office of Nuclear Energy, Science and Technology has engaged governments, industry, and the nuclear research community worldwide in a wide-ranging discussion of the development of next generation nuclear energy reactors known as "Generation IV".

[2] For the purposes of this document, "recycle" involves (a) separation of the constituents of spent nuclear fuel, (b) refabrication of fresh fuels containing plutonium, minor actinides, and possibly some fission products, c) management of solid, liquid, and gaseous wastes, and (d) storage of spent fuel and wastes.

possibly recovered uranium containing significant amounts of transuranic elements.

These ramifications of fuel recycle have the potential to require changes in the NRC's existing regulatory framework and expertise which are now structured to license light-water reactors and their associated once-through fuel cycle facilities including direct disposal of spent fuel. In recognition of this potential the Commission suggested [NRC, 2006 a, b] that the Advisory Committee on Nuclear Waste (ACNW) become knowledgeable concerning developments in fuel recycle and help in defining the issues most important to the NRC concerning fuel recycle facilities. Toward these ends the ACNW revised its Action Plan to include such activities.

In FY 2006 the Committee received initial briefings by Committee consultants, NRC staff, and DOE staff on fuel recycle. Based on this input the Committee decided that the most efficient way to meet the potential needs of the Commission was to prepare a white paper on fuel recycle. A group of expert consultants was chartered to do so. This document is the result of that effort. The primary goal of this paper is to consolidate and integrate technical, regulatory, legal, and other background information. This paper is one important basis for a letter to the Commission concerning the Committee's initial insights on important issues that fuel recycle would raise and provide recommendations on what should be done and the timing to address them. Additionally, noting that decades have elapsed since the NRC attempted to license fuel recycle facilities, a secondary goal of the paper is "knowledge management": capturing the expertise of the relatively few remaining experts concerning recycle technology and what transpired decades ago for use by all elements of the NRC.

In attempting to meet the above goals this paper addresses the following topics:

- A historical overview of fuel recycle including recycle programs, reprocessing technology and facilities, and fuel refabrication technology and facilities.

- A historical overview of the siting, design, and operation of fuel recycle facilities that describes how recycle technologies were integrated into an operating facility designed to meet then-applicable (the late 1970s) regulations and some needed improvements that were evident even at that time.

- An overview of current recycle activities including ongoing U.S. and international fuel recycle programs, a brief discussion of advanced reactors and the spent fuel they would generate (which is the feedstock for recycle facilities), and discussion of the advanced fuel recycle processes that are being developed.

- Discussion of regulation and licensing of fuel recycle facilities including experience with licensing two such facilities in the 1970s and earlier; options for licensing contemporary fuel recycle facilities including existing and potential new regulations; and related topics concerning environmental protection requirements (primarily effluent controls) and other environmental impacts. This discussion also addresses recent proposals by the NRC staff on how fuel recycle facilities might be licensed [NRC, 2007].

- A discussion of issues relevant to licensing recycle facilities.

II. RECYCLE FACILITY FEEDSTOCK: SPENT NUCLEAR FUEL DESIGNS

The well-known uranium-plutonium and thorium-uranium fuel cycles are discussed very briefly below, followed by a discussion of potential areas of application of some new and novel proliferation resistant reprocessing technology flowsheets being studied.

A. Overview of generic fuel cycles

1. Uranium-Plutonium Fuel Cycle

The uranium-plutonium cycle starts with uranium ore. Generally, the uranium is enriched to about 3 to 4.5 % in ^{235}U and fabricated into UO_2 for use in reactor fuel. Plutonium is generated ("bred") by capture of neutrons in ^{238}U. Eventually enough plutonium is produced that it contributes substantially to the fission reaction and thus to power production in power reactors. The plutonium can be separated by reprocessing the spent fuel and converted to PuO_2 which is mixed with UO_2 to produce "MOX" (mixed oxide) fuel. The advantage of this approach to fuel manufacture is that it uses the relatively abundant ^{238}U (99.275%) in uranium ore to produce fissile plutonium to replace part of the much less abundant ^{235}U (0.71 %) in the fuel.

2. Thorium-Uranium Fuel Cycle

The thorium-uranium cycle starts with enriched uranium and thorium. Neutron capture in ^{232}Th produces ^{233}U, which is fissile. In principle, when enough ^{233}U is produced it can completely replace the enriched uranium. The MSBR was projected to be a net breeder. The Shippingport reactor[3] was operated on the thorium-uranium fuel cycle and attained a breeding ratio of about 1.01. The thorium-uranium cycle has the potential to substantially reduce the consumption of enriched uranium for a given amount of energy produced.

B. Fuel Designs

1. PWR

The most basic part of PWR fuel is a uranium oxide ceramic fuel pellet which is about 1 cm in diameter and 2-3 cm long. The pellets are inserted into Zircaloy cladding tubes and plugs are welded in the end thus constituting a fuel element. The tubes are about 1 cm in diameter and about 4.5 m long. The gap between the fuel pellets and the cladding is filled with helium gas to improve the conduction of heat from the fuel to the cladding and minimize pellet-cladding interaction which can lead to fuel element failure. The fuel elements are then grouped into a square array called a fuel assembly (see Figure //1//).

There are 179 to 264 fuel elements per fuel assembly and 121 to 193 fuel assemblies are loaded into a reactor core. The size of the fuel element array ranges from 14x14 to 17x17. Typical PWR fuel assemblies are about 4.5 meters in length and 21.4 cm^2 in cross section. Control rods are inserted through the top and into the body of the assembly. The fuel usually is enriched to between 3 to 4.5 percent in ^{235}U.

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[3] The Shippingport (Pennsylvania) breeder reactor design for Light Water Reactors (LWRs) operating on the thorium-uranium fuel cycle was developed in the 1950s by the Naval Reactors Division of the US Atomic Energy Commission (USAEC) under Admiral Rickover.

Fig. //1// PWR Fuel Assembly and Hardware

2. BWR

In a boiling water reactor (BWR) the fuel is similar to PWR fuel except that the assemblies are smaller and are “canned”. That is there is a thin metal sheath (also known as a shroud) surrounding each assembly. This is done primarily to prevent local water density variations from affecting neutronics and to control thermal hydraulics of the nuclear core. Each BWR fuel element is filled with helium to a pressure of about three atmospheres (300 kPa). A modern BWR fuel assembly is comprised of 74 to 100 fuel elements that are slightly larger in diameter than those in a PWR, and there are up to 800 assemblies in a reactor core, holding up to approximately 140 tonnes of uranium. The number of fuel assemblies in a specific reactor is based on considerations of desired reactor power output, reactor core size, and reactor power density. Figure //2// shows modern BWR fuel assemblies and a control rod module. The fuel element array is typically 6x6 to 8x9. The assemblies are 10 to 15 cm across and about 4.5 m long.

Fig. //2// BWR fuel assembly

3. Fast Reactor

Historically, the core of a fast reactor consisted of an array of canned fuel assemblies containing a hexagonal array of fuel elements. The cladding and can are both made of stainless steel which allows these reactors to operate at higher temperatures than LWRs. When such reactors were designed to produce more plutonium than they consumed the core was comprised of a central region of mixed-oxide fuel (called driver fuel) that could sustain a chain reaction. Above and below the driver fuel in the MOX assemblies were pellets of depleted uranium called a blanket. Additionally, surrounding the driver assemblies in the radial direction were fuel assemblies in which the fuel pellets were all depleted uranium. When these assemblies are placed together, the result is creation of a central cylindrical “driver” region surrounded on all sides by the blanket. The purpose of this configuration was to use neutrons that leaked from the driver fuel to produce plutonium in the blanket.

The fuel elements are kept apart by spacers or in some cases by wire wound helically along each element. Driver fuel elements are typically stainless steel tubes 6 or 7 mm in diameter. In early designs, the elements in the blanket were larger in diameter, about 1.5 cm, because they require less cooling than the fuel elements. Both fuel and blanket elements may be more tightly packed in liquid metal (e.g., sodium, Na/K, lead, bismuth) cooled fast reactors than in an LWRs because the heat transfer properties of the liquid metal are so much better than those of water. This may not be the case for gas-cooled fast reactors.

In the GNEP concept the objective of future fast reactors is to fission as many of the transuranic elements as possible while still producing electricity. Thus, instead of producing about 10% more plutonium as would have been the case with breeder reactors, DOE is seeking to have advanced burner reactors (ABRs) consume a net 25% to 75% of the transuranic elements in fresh fuel. One consequence of this is it is unlikely that there will be any blanket fuel in the ABR and it is possible some or all of the ^{238}U in the driver fuel may be replaced by another element that does not produce plutonium.

Fast reactor fuel may be made of several different materials. The principle materials are discussed below.

a. Oxide

Oxide fuel is made up with pellets composed of a mixture of oxides of plutonium and uranium. In the ABR other transuranic elements may be included. The equivalent enrichments[4] of the fuel range between 15 to 35% depending on the reactor in question.

b. Carbide

Although virtually all historical and current fast reactors operate with uranium-plutonium oxide fuel[5], there is some interest in the use of fuel composed of uranium/plutonium carbide, particularly in India. Carbide fuels have high thermal conductivity and, where plutonium breeding is of interest, can attain breeding ratios larger than those of oxide fuels. The increase in breeding radio is due to the fact that while there are two atoms of oxygen per atom of uranium in the oxide, there is only one atom of carbon per uranium atom in the carbide. Light atoms such as

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[4] Uranium and plutonium isotopes are both fissionable, so it is convenient to refer to the fissile content of fuel in terms of “equivalent enrichment,” i.e., with fissile characteristics as though it were all enriched uranium.

[5] An important exception is the fast reactor development program in India, which is based on carbide fuels.

carbon and oxygen tend to moderate fission neutrons, and since there are fewer atoms per fissile atom in the carbide than in the oxide, it follows that the energy distribution of neutrons in a carbide-fueled fast reactor is shifted to higher energies than in a comparable oxide-fueled fast reactor.

c. U/Pu/Zr

An alloy of uranium/plutonium/zirconium (U:71 %; Pu:19 %; Zr:10 %) in stainless steel cladding has shown considerable promise as a fast reactor fuel. It has been irradiated to burnups well over 100 MWd/te with no deleterious effects that preclude serious consideration of its use.

d. Nitride

There has been interest shown in using uranium and/or plutonium nitride in fast reactors for many of the same reasons that carbide is attractive as a fuel and DOE is developing such fuels. An important disadvantage of nitride fuels is that they can form significant amounts of ^{14}C by neutron capture in ^{14}N in the nitride component. In order to surmount this problem it would be necessary to perform a nitrogen isotope separation to remove the bulk of the ^{13}N.

A drawing of typical historical fast breeder reactor fuel assembly is shown in figure //3//. Fuel designs for the ABR are still evolving.

Fig. //3// LMFBR Fuel Assembly

4. HTGR

There are two types of HTGR fuel assemblies: spherical (called pebbles) and prismatic. The former were developed in Germany in connection with the AVR and the first German HTGR power plant, the THTR 300.[6] Currently pebble bed fuel assemblies are being used in the experimental reactors HTR-10 in China and the HTTR in Japan. The latter type, prismatic fuels, were developed in the U.S. by General Atomics and were used commercially in the 330 Mwe Fort St. Vrain reactor.

In both cases the fuel element is composed of compounds of uranium, thorium or plutonium in the form of a ceramic (usually oxides, oxycarbides, or carbides) contained within small spherical shells made of pyrolytic graphite and silicon carbide. These fuel elements are enclosed in graphite "pebbles" or prismatic blocks that act as the primary neutron moderator.

The basic fuel "element" in both cases is the "triso" fuel microsphere which is typically about one millimeter in diameter. TRISO fuel typically consists of a fuel kernel composed of UO_2 (sometimes UC_x or UCO) in the center, coated with four layers of material. The four layers are a porous graphite buffer layer followed by a dense inner layer of pyrolytic carbon (PyC), followed by a ceramic layer of SiC to retain fission products at elevated temperatures and to give the TRISO particle more structural integrity, followed by a dense outer layer of PyC. TRISO fuel particles are designed to not crack due to the stresses from processes (such as differential thermal expansion or fission gas pressure) at temperatures beyond 1600°C, and therefore can contain the fuel and fission products in the worst accident scenarios in a properly designed reactor. See section III.B.3 for a detailed discussion of HTGR fuels.

5. Molten Salt Reactor (MSR)

The MSR is a unique reactor concept. It does not use a solid fuel. Instead, it uses a molten fluoride salt fuel that circulates in a loop. The loop contains a heat exchanger to extract fission energy and a system that removes fission products, primarily lanthanides and noble gases, whose presence would "poison" the salt (i.e., would capture neutrons) and ultimately prevent fission from occurring. The fuel for the MSRE was $LiF\text{-}BeF_2\text{-}ZrF_4\text{-}UF_4$ (65-30-5-0.1). A graphite core moderated the neutrons. The secondary coolant was FLiBe ($2LiF\text{-}BeF_2$). The reactor operated at a peak temperature of 650°C and operated for the equivalent of about 1.5 years of full power operation.

The culmination of the Oak Ridge National Laboratory research during the 1970-76 time frame resulted in a MSR design that would use $LiF\text{-}BeF_2\text{-}ThF_4\text{-}UF_4$ (72-16-12-0.4) as fuel. It was to be moderated by graphite with a 4 year replacement schedule, to use $NaF\text{-}NaBF_4$ as the secondary coolant, and to have a peak operating temperature of 705°C. However, to date no commercial molten salt reactors have been built.

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[6] South Africa has a modular pebble bed reactor under active development.

III. OVERVIEW OF SPENT NUCLEAR FUEL RECYCLE

A. Reprocessing Programs and Evaluations

A great deal of the technical information needed for reprocessing spent nuclear fuel and for fuel recycle in general has been available for many years and may be found in the publicly available literature [see the General References]. The general references at the end of this paper are indicative of the amount of detailed information available and the very long time it has been available. Notwithstanding this wealth of information there is another component of knowledge that is related to operating experience that cannot be conveyed in any way other than operating actual fuel cycle facilities. Some information on early fuel cycle evaluations and the current or formerly operating recycle facilities is presented in what follows.

1. U.S. Defense

In the post-World War II years spent fuel reprocessing to recover plutonium for use in weapons and highly enriched uranium from naval reactor fuel was still carried out for many years in DOE operated government facilities.

a. Reprocessing for Weapons Plutonium Recovery

Large-scale reprocessing of irradiated nuclear reactor fuel to recover plutonium for use in nuclear weapons began in the United States immediately following the second World War and continued until the 1980s. Reprocessing was carried out in large government–owned plants located in Richland, WA and Savannah River, SC for plutonium production. A plant was also constructed at Idaho Falls, ID to recover uranium from spent naval reactor fuels. The earliest large-scale plutonium recovery process was the *bismuth phosphate process* which was a multi-step precipitation process developed by G. Seaborg and co-workers in very small-scale laboratory experiments and carried directly into large-scale production at the Hanford site in Richland, WA. It was soon replaced with a succession of solvent extraction processes that were much simpler to operate and more efficient. They did, however, produce copious amounts of waste, both radioactive and non-radioactive. Millions of gallons of highly radioactive liquid waste (HLW) was stored in large "single-shell" and "double-shell"[7] tanks on the Hanford and Savannah River sites. Most of this waste still resides in the tanks as sludge and caked salt.

Solvent extraction as practiced in the spent nuclear fuel reprocessing facilities of the 60s and 70s was a process wherein an acidic aqueous solution containing the dissolved spent nuclear fuel was contacted with an essentially immiscible organic solvent that preferentially removed uranium and plutonium (and, if desired, other actinides) from the aqueous phase. Many of the solvents employed early had significant drawbacks, such as high flammability, susceptibility to chemical and radiation damage, volatility, excessive solubility in water, high viscosity, and high cost. Solvents used in early large-scale reprocessing plants included methyl isobutyl ketone (Hexone) which was used at the Hanford plant in Richland, WA, and β,β'-dibutoxydiethylether (Butex) which was used by the British. Ethylhexyl phosphoric acid (HDEHP) has been used in smaller scale applications. These solvents were soon replaced by tributyl phosphate (TBP), a commercially available solvent without many of the drawbacks of the other solvents. In practice TBP is diluted about two-to-one (~30 % TBP) with long-chain hydrocarbons (e.g., purified kerosene or dodecane) to produce a solution with properties optimized for use in selectively extracting actinides. The aqueous phase in the extraction process typically is a nitric acid

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[7] The terms single-shell and double-shell refer to whether the tanks had only one wall and bottom or whether they were, in effect, a tank within a tank. Many of the single-shell tanks have developed leaks to the sub-soil.

solution containing uranium, plutonium, neptunium, americium, curium, and fission products, most notably, cesium, strontium, iodine, technetium and the rare earth elements (lanthanides). The plutonium and uranium (and if desired, other actinides) extract selectively into the TBP phase as complex chemical species containing nitrate ions and TBP. Adjustments of the acidity of the solution and of the valence of plutonium [from Pu(IV) to Pu(III)] make possible its subsequent separation (in a process called "stripping") from uranium.

i. Bismuth Phosphate Process

The Bismuth Phosphate Process for extracting plutonium from irradiated uranium was demonstrated in a pilot plant built beside the Oak Ridge X-10 Reactor in 1944. The process produced a large amount of highly radioactive waste, and was replaced by a solvent extraction process. The process was designed to extract plutonium from aluminum-clad uranium metal fuel. The aluminum fuel cladding was removed by dissolving it in a hot solution of caustic (sodium hydroxide). After de-cladding, the uranium metal was dissolved in nitric acid. The plutonium at this point was in the +4 oxidation state. It was then carried by a precipitate of bismuth phosphate formed by the addition of bismuth nitrate and phosphoric acid. The supernatant liquid (containing many of the fission products) was separated from the precipitate which was then dissolved in nitric acid. An oxidant such as potassium permanganate was added to convert the plutonium to soluble PuO_2^{2+} (Pu VI). A dichromate salt was added to maintain the plutonium in the +6 oxidation state. The bismuth phosphate was then re-precipitated, leaving the plutonium in solution. Then an iron salt such as ferrous sulfamate[8] was added and the plutonium re-precipitated again using a bismuth phosphate carrier precipitate as before. Then lanthanum and fluoride salts were added to create a lanthanum fluoride precipitate which acted as a carrier for the Pu. Repeated precipitations and dissolutions were used to remove as many impurities as practical from the plutonium. The precipitate was converted to oxide by the addition of a chemical base and subsequent calcination. The lanthanum-plutonium oxide was then collected and plutonium was extracted from it with nitric acid to produce a purified plutonium nitrate solution.

ii. Redox (Hexone)

The REDOX process, which is a solvent extraction process that uses methyl isobutyl ketone (Hexone) as the extractant, was developed at Hanford in the late 1940s to replace the bismuth phosphate process and was used in the site's REDOX plant (also known as the S Plant) from 1951 through June 1967. The REDOX Plant processed over 19,000 metric tons of irradiated fuel. Hexone has the disadvantages of requiring the use of a salting reagent (aluminum nitrate) to increase the nitrate concentration in the aqueous phase and thus promote plutonium extraction into the Hexone phase, and of employing a volatile, flammable extractant. The aluminum nitrate salting agent substantially increased the volume of HLW. The Hexone, besides presenting a hazard, is degraded by concentrated nitric acid, leading to more waste as well as decreasing extraction efficiency. The REDOX process was replaced by the PUREX process.

iii. PUREX

A highly simplified flowsheet for the PUREX process is shown in Figure //4//. The PUREX process is the only large-scale process now used commercially for spent nuclear fuel reprocessing, and has many years of demonstrated excellent performance. However, the PUREX

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[8] Ferrous sulfamate was chosen because the ferrous ion reduced the plutonium to inextractable Pu (III) and the sulfamate ion reacted to destroy any nitrous acid present. Nitrous acid had a deleterious effect on the uranium-plutonium separation process.

process produces a pure plutonium stream. This may be considered to be a major drawback because of the nuclear weapons proliferation potential presented by separated and purified plutonium. This drawback is a major impetus for development and adoption of new processes such as the proposed U.S's UREX processes and the French GANEX process discussed in Section VI below. Another drawback to the PUREX process is that it can produce a relatively large amount of radioactive waste if plutonium reducing agents contained inorganic materials such as iron compounds are used, and because the tributylphosphate (TBP) extractant contains the phosphate radical whose radiolytic and chemical decomposition products are significant waste formers. Despite these drawbacks, which have attracted greater attention in recent years, PUREX with its modifications and improvements, is a major improvement over all preceding processes and remains the current process of choice for spent commercial nuclear fuel reprocessing. The PUREX process is described in some detail below based on the last attempt to build and operate a reprocessing plant (BNFP) in the U.S.

Fig. //4// Purex process flowsheet

Spent Fuel Receiving and Storage

The irradiated fuel assemblies would arrive at the reprocessing plant on a carrier in shielded casks. The cask and carrier would be monitored for external contamination and washed to remove external dirt. After the cask had been removed from the carrier, the condition of the fuel and cask would be determined. The cask would be vented, cooled, and prepared for entry into the cask unloading pool. The cooled cask would be moved by the cask handling crane to the cask unloading pool, where it would be lowered to the bottom of the pool. The top of the cask would be opened and the contained fuel would be removed. The identity of each fuel assembly would be established and compared against shipping documentation. The fuel would be placed in storage canisters, which would be moved to the fuel storage pool for retention until the fuel was scheduled for reprocessing. All operations would be performed under water.

Spent Fuel Inventory

A typical 1,500 MTU/year reprocessing/recycling facility will generally have a spent fuel storage capacity of approximately 2,000 fuel elements, which, depending on the burnup, will represent approximately one-fourth of the annual plant capacity (e.g., the BNFP could store 360 MTU at any one time). Initial BNFP specifications for spent fuel in the mid-70s are shown in Table //1//.

Table //1// Spent Fuel Specifications (mid-70's)

Characteristic	Value
Burnup, maximum	40,000 Mwd/MteU
Specific power, maximum	50 Mw/MteU
Enrichment	Initial: 3.5 - 5.0% U-235 or equivalent Final: 1.9 - 3.5% U-235 + Pu content
Plutonium yield, total	10 Kg Pu/MTU
Age of spent fuel, as shipped	90-day cooled, minimum
Age of spent fuel at start of reprocessing	90-day cooled, minimum
Cladding	Zircaloy or stainless steel
Maximum dimensions	11-3/8" sq. by 20' long

At the current time, however, initial feed spent fuel will be aged for years (some for as many as 40 years) since the fuel continues to be stored by the electric utilities.

Shearing and Dissolving

An individual spent fuel assembly container would be remotely transferred from the storage pool and the individual fuel assemblies would be removed and moved to the feed mechanism of the mechanical shear. Generally, a full batch or a lot of fuel from a single source would be processed at a time. The fuel assemblies would be chopped into small segments (approximately 2" to 5" long) to expose the fuel to the nitric acid dissolver solution.

The chopped fuel assemblies would fall into one of three dissolvers that contain hot 3 $\underline{M}$ HNO_3 to dissolve virtually all uranium, plutonium, other actinides, and most of the fission products. During dissolution, a soluble poison (gadolinium nitrate) would be added to the dissolver as a precaution to prevent a criticality. After the initial dissolution, a digestion cycle would be used (8 $\underline{M}$ HNO_3) to dissolve any remaining fuel (plutonium oxide is sometimes refractory and requires more aggressive dissolution conditions). Following digestion in nitric acid any remaining insoluble material would be rinsed with dilute nitric acid and these materials plus the undissolved cladding hulls of stainless steel or Zircaloy would remain in the dissolver basket. Gases released from the spent fuel during dissolution (primarily ^{85}Kr, tritium, ^{129}I and $^{14}CO_2$ with the possibility of some $^{106}RuO_4$) and nitrogen oxides would be directed to the off-gas treatment system to remove particulates, radioiodine, and nitrogen oxides. The cladding hulls would be rinsed, monitored for fissile material, packaged, and transferred to the solid waste storage area. The nitrogen oxides would be reconstituted to nitric acid.

Product Separation and Purification

After acidity and concentration adjustment the dissolver solution would become the solvent extraction process feed solution. It would be clarified by centrifugation, and then sent to the first solvent extraction decontamination cycle. In this cycle, the feed solution is contacted counter-currently in a ten-stage centrifugal contactor with an organic solution of 30% tributyl phosphate (TBP) in a kerosene or normal paraffin hydrocarbon diluent (primarily dodecane). The organic solution preferentially would extract the tetravalent plutonium and hexavalent uranium, leaving about 99% of the fission products in the aqueous raffinate (waste) nitric acid solution. The organic solution from the centrifugal contactor then would pass through a pulsed scrub column where aqueous 3 $\underline{M}$ HNO_3 solution scrubs (back-extracts) about 96% of the small amount of extracted fission products from the product-bearing organic solution. This scrub solution subsequently would be recycled to the centrifugal contactor for additional uranium and plutonium recovery to reduce the potential for product losses. The combined aqueous stream leaving the centrifugal contactor would contain approximately 99.6% (or more) of the fission products and would be sent to a HLW concentrator.

The organic solution from the scrub column (joined by organic raffinates from down-stream plutonium purification columns) would pass through a partitioning column where tetravalent plutonium would be electrochemically reduced[9] to the less extractable trivalent state. This would enable the plutonium to be stripped quantitatively into an aqueous nitric acid solution within the electrochemical unit. A substantial amount of uranium would follow the plutonium in the aqueous stream [some uranium is also electrolytically reduced from U(VI) to U(IV), and may in fact be the ultimate Pu reductant]. The aqueous stream, which is approximately 35% plutonium and 65% uranium, would flow to the plutonium purification cycles. The organic solution, now stripped of plutonium, would pass through another pulsed column where the residual uranium would be stripped into a weakly acidified aqueous solution (approximately 0.01 $\underline{M}$ HNO_3).

The aqueous strip solution containing the residual uranium would be concentrated by evaporation from 0.3 $\underline{M}$ uranium to 1.5 $\underline{M}$ uranium and adjusted with nitric acid to approximately 2.5 $\underline{M}$ HNO_3. This uranium would be preferentially extracted again by 30% TBP organic solution in another pulsed column. Before leaving the column, the organic solution would be scrubbed with dilute nitric acid solution, which would remove traces of extracted ruthenium and zirconium-niobium fission products, which are among the fission products most difficult to remove. Hydroxylamine hydrogen nitrate or hydrazine also would be added to the scrub solution to remove residual plutonium by its chemical reduction to the inextractable trivalent state. Uranium subsequently would be stripped from the organic solution in another pulsed column, using an acidified aqueous solution (0.01 $\underline{M}$ HNO_3). This solution would be concentrated, by evaporation, from 0.4 $\underline{M}$ uranium to 1.5 $\underline{M}$ uranium. Finally, the concentrated aqueous uranium solution would be passed through silica gel beds to remove residual traces of zirconium-niobium fission products, and the uranyl nitrate product solution would be analyzed and transferred to the UF_6 facility for storage or conversion to UF_6 and subsequent shipment. Uranium recovery was

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[9] Electrochemical reduction of plutonium was unique to the Barnwell plant. Plutonium is conventionally reduced chemically.

expected to be at least 99 % . Removal of fission products was to be 99.99 %.

Plutonium in the aqueous stream leaving the partitioning column would be re-oxidized to the organic-extractable tetravalent state by sparging the solution with di-nitrogen tetroxide (N_2O_4) and would be preferentially extracted into an organic solution in the first pulsed extraction column of the second plutonium cycle. In the top portion of this column, the organic stream would be scrubbed with 10 M HNO_3 solution to remove traces of extracted ruthenium and zirconium-niobium fission products. The organic stream then would pass through a strip column where tetravalent plutonium would be transferred to an aqueous stream of dilute (0.3 M) nitric acid. This cycle would also partition plutonium from the accompanying uranium, with the uranium being recycled. The extraction-scrubbing sequence would be repeated in a third plutonium cycle for further decontamination from fission products and uranium. To effect a higher plutonium product concentration, the plutonium would be reduced in the third-cycle strip column by hydroxylamine hydrogen nitrate to the more hydrophilic trivalent state. A TBP organic scrub solution would be added to remove any residual uranium from the plutonium aqueous stream as it leaves the third-cycle strip column. Following the third plutonium cycle, the plutonium nitrate solution would be washed with a stream of organic diluent in a final column to remove traces of organic solvent (TBP). Final plutonium concentration would be established in a critically-safe-geometry evaporator made of titanium. The plutonium product solution would be analyzed and stored in critically safe tanks. The plutonium recovery was expected to be 98.75 %.

The contaminated organic solvent stream from the co-decontamination and partition cycles would be washed successively with dilute aqueous solutions of sodium carbonate, nitric acid, and sodium carbonate to remove organic degradation products (primarily dibutyl- and monobutyl phosphate) generated by radiation damage to TBP. This step would produce waste solids formed from the sodium salts and organic phosphates.

The precipitated solids would be removed by filtration following the first carbonate wash. Fresh TBP and/or diluent would be added, as required, to maintain the 30% TBP concentration and the total solvent inventory at the desired level. The contaminated organic solvent stream from the second uranium cycle would be treated similarly in a separate system, except that the second sodium carbonate wash would be omitted.

The aqueous raffinate streams from the plutonium and uranium cycles, except the last product-bearing raffinate, would be treated with N_2O_4 for adjustment of the plutonium oxidation state to Pu(IV) and U(VI) and would be passed through a pulse column where residual uranium and plutonium would be recovered by extraction into a 30% TBP organic solution. The recovered uranium and plutonium would be recycled back to the decontamination cycle for recovery. The aqueous raffinate stream would be concentrated in a low-activity process waste evaporator.

Liquid Waste Streams

The radioactive aqueous waste streams from all the solvent extraction cycles would be concentrated in the high- or low-activity waste evaporators, depending on the relative radioactivity content. The acidic concentrated HLLW bottoms would be stored in a cooled stainless steel waste tank. The evaporator overheads would be passed through a distillation column to recover the nitric acid as a 12 M solution. The distillation column overhead (primarily water) then would be recycled as process water, or sampled and released to the stack from a vaporizer provided it met release specifications. The recovered 12 M HNO_3 would be used in parts of the process where the residual radioactivity could be tolerated.

Miscellaneous aqueous streams containing salts and fission products (approximately 1 Ci/liter) but no appreciable uranium or plutonium would be acidified and concentrated to approximately 50 Ci/liter in the general purpose evaporator. These evaporator bottoms would be stored in an uncooled stainless steel waste tank. The condensed overheads would be vaporized to the stack.

Process Off-gas Streams

Off-gases from the dissolver would be scrubbed with a mercuric nitrate solution to reduce levels of radioactive iodine in the effluent, and then treated in an absorber to convert nitrogen oxides to nitric acid suitable for recycling. The dissolver off-gas and vessel off-gas streams would be combined and passed successively through a second iodine scrubber containing mercuric nitrate, silver zeolite beds for iodine sorption, and high-efficiency filters before release to the stack.

Facilities for the retention of other radionuclides such as ^{85}Kr, tritium, and ^{14}C (as CO_2) were not in place in the 1970s reprocessing plant although there were plans to recover ^{85}Kr.

UF_6 Preparation

The UF_6 plant was designed with an annual capacity of 1,500 MTU and assumed to operate 24 hours per day for up to 300 days a year. Scrap from the plant operations would be stored until processed in the appropriate facility after which it would be shipped off site for either re-use or for disposal as contaminated waste, as determined by analysis.

The individual process steps for the conversion of uranyl nitrate to uranium hexafluoride in a UF_6 conversion plant co-located with a reprocessing/recycling facility are:

- Receipt of purified uranyl nitrate solution from a reprocessing plant;
- Concentration of the uranyl nitrate feed solution via evaporation;
- Conversion of the uranyl nitrate to UO_3 by heating to de-nitrate it;
- Hydrogen reduction of UO_3 to UO_2;
- Hydrofluorination of UO_2 to UF_4, using gaseous HF;
- Fluorination of UF_4 to UF_6, using electrolytically generated F_2;
- Freezing and then resubliming UF_6 in a series of cold traps to purify it[10]; and
- Packaging of the UF_6 product into standard transport cylinders.

All processing steps which involve radioactive materials would be performed inside equipment maintained at negative pressure relative to the adjacent, less radioactive, areas of the conversion building. The pressure differences would be maintained so that flow of air is from uncontaminated areas into areas of potentially higher contamination levels, thus limiting the spread of radioactivity.

The equipment forms the first level of confinement; the conversion building forms the second level. Pressure differences would be maintained by automatically controlled, zoned ventilation systems. Spare ventilation fans and required controls, which are provided, would be connected to independent or installed emergency power systems in the event of loss of normal plant power, to ensure that the required pressure differences would be maintained.

Plutonium Precipitation and Conversion

The feed material for the Plutonium Product Facility (PPF) would be separated plutonium nitrate

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[10] Small amounts of some radionuclides having volatile fluorides, most notably tellurium, neptunium, and technetium, follow the uranium all the way to the UF_6 plant, and must be removed by fractional sublimation.

solution from the plutonium nitrate storage tanks in the Separations Facility. Its typical characteristics are given in the Table //2//. The alpha, neutron, and gamma emissions require special features for confinement and shielding. The radioactive decay heat and potential criticality of concentrated plutonium solutions and products require special design constraints for the processing equipment within the PPF.

Table //2// Characteristics of plutonium nitrate feed to the BNFP plutonium product facility

Characteristic	**Value**
Plutonium concentration, g/liter[a]	100-360
Nitric acid concentration, M	2 - 10
Uranium concentration, ppm	Less than 10,000
Radioactive decay heat, Btu hr^{-1} (kg $Pu)^{-1}$	Less than 60
Radioactive hydrogen generation, scfh/kg Pu	Less than $5x10^{-4}$
Gamma emission, Ci/g Pu	80
Pu-238, % of total Pu	2.5
Pu-239, % of total Pu	50
Pu-240, % of total Pu	25
Pu-241, % of total Pu	15
Pu-242, % of total Pu	7.5

[a] Plutonium concentrations in excess of 250 g/liter may be processed if the heat generation rate is less than 60 Btu hr^{-1} (kg $Pu)^{-1}$.

The plutonium nitrate solution would be transferred from the storage tanks to one of two feed preparation tanks on a batch basis. The nitric acid concentration would be adjusted to 3.0 M to provide a constant feed for the conversion process. It is essential that the concentration be maintained at more than 2 M to ensure the prevention of plutonium hydrolysis to form plutonium colloid (polymer formation) and oxide precipitation. Hydroxylamine hydrogen nitrate (HAN) also would be added at the feed adjustment tank to reduce any Pu(VI) to Pu(IV) prior to the precipitation step.[11] After completion of the feed adjustment step, the plutonium nitrate solution would be heated to 60°C in an in-line heater and fed continuously into a precipitator equipped with a mechanical stirrer. A solution of 1.0 M oxalic acid would be added to the precipitator, and the resulting plutonium oxalate slurry would be allowed to overflow to the digester whose role was to grow large, well-formed crystals. The digester would consist of three in-line mechanically stirred vessels (identical to the precipitator) that would be arranged to permit the overflow of one unit to cascade into the next. The precipitation and digestion vessels would be sized such that the residence time is approximately one hour.

The slurry would be fed into a rotary-drum vacuum filter for liquid-solid separation. The oxalate cake would be rinsed on the filter drum and scraped off with a “doctor blade.” The filtrate would be transferred to a filtrate surge tank prior to further processing. The plutonium oxalate cake from the drum filter would be discharged directly into a rotary screw dryer-calciner. The oxalate anion would be destroyed by heating in air to form the desired plutonium dioxide product. The oxalate-cake feed rate, residence time, heating rate, and final calcining temperature are all critical to the production of a plutonium dioxide feed material with the proper characteristics for manufacturing into satisfactory fuel pellets during subsequent mixed-oxide (MOX) fuel fabrication operations. *Note: It should be recognized that this process was optimized for the anticipated MOX fuel specifications of the mid-seventies. The final product specifications required for a 2010+ fuel will establish the ultimate plutonium conversion process.*

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[11] The electrochemical potentials of the various Pu valence states are such that Pu(III), Pu(IV) and Pu(VI) can co-exist in solution at equilibrium. Consequently, it is necessary to chemically produce the sought valence state.

The calciner would discharge directly onto a continuously moving screen. The powder passing through the screen would be collected in a geometrically-safe blender body which has a maximum capacity of 40 kg of plutonium as plutonium oxide. The over-size product would pass off the top of the screen into a collection hopper. This hopper periodically would be emptied into a grinder which would reduce the particle size to meet the product specification. The grinder would empty into an identical 40-kg blender. The ground plutonium oxide would be recycled to either the top of the screen or to the dryer-calciner. These operations are especially "dirty" in that they produce a plutonium dioxide dust that is difficult to contain and handle.

Plutonium Sampling and Storage

A blender would receive nominally 32 kg of plutonium oxide, as indicated by a weighing element beneath the blender. To change the vessel, it would be remotely valved-off and transferred to the blending stand. The full blender body would be rotated about its radial center until completion of blending. The powder would be sampled and the samples analyzed to determine properties and insure homogeneity. The plutonium would be held in the blender body until the analytical results were received. Plutonium dioxide not meeting the product specifications would be either recycled or loaded-out and held for future re-work.

The blended powder in the blender body would be transferred to the powder load-out stand where the contents of the blender would be discharged into four product canisters, each holding nominally 8 kg of plutonium oxide. The canister covers would be installed, each canister would be sealed, and the outer surface would be decontaminated. Four product canisters would be loaded into a pressure vessel that would double as a storage container and primary containment vessel during shipment. The pressure vessels (which were never built) were to be vented through a 3-stage HEPA filter. The loaded pressure vessel would be placed either in the storage vault or into a shipping container for off-site shipment (if the MOX fuel fabrication plant were co-located with the reprocessing/recycling facility, off-site shipment would not be necessary).

Recycle Streams

Filtrate from the vacuum drum would be collected in the filtrate surge tank where gas and liquid would be separated. The gas would be routed to the vacuum pump. The majority of the gaseous output of the vacuum pump would be recycled to the vacuum drum filter. A small amount of the gas would be bled to the vessel off-gas system.

The liquid from the filtrate surge tanks would be pumped through cartridge-type secondary filters into the filtrate evaporator feed tank. The filtrate would be transferred from the filtrate evaporator feed tank by air lift into the filtrate evaporator. In the evaporator, the filtrate would be distilled sufficiently for destruction of the oxalic acid and to reduce the volume of solution containing plutonium.

Bottoms from the evaporator would be sequentially cooled, passed through another secondary cartridge-type filtration step to remove any possible solid (normally not expected), and then collected in the concentrate catch tank. The filtrate concentrate then would be transferred by jet to the concentrate sample tank where it would be sampled. If analyses indicated the presence of oxalic acid, it could be destroyed by returning it to the filtrate evaporator feed tank for reprocessing or by addition of acidified potassium permanganate in the sample tank. The contents of the sample tank also would be returned to the evaporator feed tank if the presence of solids containing plutonium was detected. When sampling indicates the Pu content/mixture is satisfactory, the concentrate would be transferred to a storage tank from which it would be pumped to the Separations Facility for plutonium recovery.

The evaporator overhead would be condensed, combined with condensate from the off-gas system, and filtered with cartridge-type filters to remove any possible solids. The distillate would be collected in the distillate catch tank from which it would be transferred batch-wise to the distillate sample tank. Depending on analyses, the distillate could be transferred to the evaporator feed tank for reprocessing, the concentrate storage tank when containing recoverable plutonium, or the distillate storage tank. From the distillate storage tank, the distillate could be transferred to the Separations Facility for acid recovery.

Waste Treatment

A typical commercial reprocessing/recycling plant of the 1970s generated gaseous, liquid and solid waste, as would any modern day plant. Continuing with the example of the 1500 MTU/y designed separations capacity of the BNFP, the waste treatment specifications were as follows.

<u>Low-level Liquid Wastes</u>

At the BNFP low-level aqueous liquid waste was planned to be released into local area streams at the rate of about 2,000 gallons per minute[12] (at full nominal rated operation). Maximum release temperature was 85^0F with essentially no radioactivity and only water treatment chemicals in the water.

<u>High-level Liquid Wastes</u>

High-level liquid waste was to be solidified after a minimum of five years of tank storage and transported to a federal repository within 10 years of generation. The BNFP initially constructed two 300,000 gallon storage tanks, manufactured of 304L stainless steel, double-walled and designed with internal stainless steel cooling coils. Relevant design data on tank contents are noted below:

Activity: 1.80×10^4 Ci/gallon
Acid concentration: 1 - 5<u>M</u> HNO_3
Temperature: 140^0 F
Heat generation rate: 72,000 BTU/hr•MTU

Each cylindrical high-level liquid waste tank was 16.5 m in diameter by 6.1 m high and was contained within an underground cylindrical concrete vault lined with stainless steel. Each vault was 18.3 m in diameter and 7.6 m high. The vault floor, walls and top were 1.2 m, 0.9 m and 1.7 m thick, respectively.

It was anticipated that three additional 300,000 gallon tanks would need to be constructed for a total capacity of 1,500,000 gallons. This was expected to allow for ample storage of liquid waste prior to solidification and off-site shipment to the federal repository (not identified at that time). Each high-level liquid waste tank contained the following equipment:

1. 48 5-cm-diameter cooling coils
2. 18 air-operated ballast tanks around the perimeter of the tank
3 9 air-operated ballast tanks in the main part of the tanks
4. 22 air-lift circulators
5. 5 steam-operated ejector pumps (empty-out jets)

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[12] It should not be inferred that this rate of liquid waste release into local streams would necessarily be practiced in a future reprocessing plant operation.

6. water-seal type pressure/vacuum relief system
7. Multiple external temperature sensing points
8. 10 instrument dip tubes to measure liquid level and specific activity.

The Waste Solidification Plant (WSP) would contain the waste vitrification equipment, canister sealing, inspection and decontamination equipment, off-gas treatment equipment, and remote maintenance facilities in four process cells. The primary process functions performed in each of the cells are presented in the Table //3//. All process cells in the WSP would be completely lined with stainless steel. The cells were to be surrounded by limited access areas for operating and controlling the processes in the cells. All operational and maintenance facilities in the process cells would be performed remotely using viewing windows, manipulators and cranes.

Table //3// Functions of cells in the BNFP Waste Solidification Plant

Cell Description	Cell Function
Waste vitrification	Calcine liquid waste; vitrify calcined waste; weld canisters closed
Canister decontamination	Remove external radioactivity from the canister
Off-gas treatment	Treat off-gas from WSP process vessels
Hot maintenance	Perform remote maintenance on contaminated equipment

Solid Waste Disposal

Solidified high-level waste, hulls, and alpha wastes were to be stored on-site in an interim storage area with eventual transport to a federal HLW repository. Spent fuel hull treatment was to be optimized (e.g., hulls would be compacted or melted) in order to minimize overall capital and/or operating costs. Because of the BNFP site location, transport may have been by truck, rail or inter-modal (including barge from site to port and thence by rail or truck to the repository).

Low-level solid waste would be disposed of at a licensed low-level waste facility. At the BNFP facility, such disposal was simplified as the Chem-Nuclear Barnwell low-level waste site was immediately adjacent to the facility. While minimizing transportation costs all other relevant regulatory requirements needed to be met.

Off-Gas System

For the principal plant off-gases the initially projected release rates were:

1. Iodine
   ^{129}I: 1.4 x 10^{-6} Ci/sec (99.9%+ % retained in plant)
   ^{131}I: 1.1 x 10^{-5} Ci/sec (99.9%+ % retained in plant)
2. Krypton
   ^{85}Kr: 4.3 x 10^{-1} Ci/sec (no recovery facilities were planned in the design being initially licensed)
3. Tritium
   ^{3}H: 1.8 x 10^{-2} Ci/sec (no recovery facilities were planned in the design being initially licensed)
4. NOx: 200 pounds/hr. [release concentration less than 150 ppm (at top of stack)]

At the time, these releases were acceptable. However, as these earlier designs proceeded through their review, agreement was reached with the CEQ[13] that an effort would be made to minimize krypton and tritium releases, even though, at the time capturing theses gases was not required. Cryogenic systems were considered and were being evaluated until INFCE started and the concomitant ban on reprocessing was invoked which halted further commercial reprocessing development activity.

b. U.S. Naval Fuel Reprocessing

The Idaho Chemical Processing Plant (ICPP) at the Idaho National Engineering Laboratory (now called Idaho National Laboratory, INL) began using variants of PUREX to process spent Naval and experimental reactor fuel for recovery and recycling of HEU in 1953. A new "head end" dissolving facility using the fluorinel[14] dissolution process, was built at ICPP in the mid-1980s. The ICPP shut down in November 1992. During its operation it recovered a total of 31.5 metric tons of uranium from research and test reactor fuel and spent Naval fuel, of which 5.1 metric tons was Naval fuel.

2. U.S. Commercial

Early in the development of nuclear power in the United States the U.S. encouraged commercial involvement, in the U.S. and abroad, in all aspects of the nuclear fuel cycle, including nuclear fuel reprocessing. The U.S. reprocessing initiative was directed principally at commercial reactor spent fuel; however, some irradiated fuel from the United States Atomic Energy Commission's (a predecessor to the Department of Energy) reactor research and development activities was also made available to provide a reprocessing base load at the Nuclear Fuel Services (NFS) West Valley reprocessing plant to encourage industrial participation.

a. Nuclear Fuel Services (NFS - West Valley Plant) – operated and being decommissioned

The Nuclear Fuel Services West Valley reprocessing plant was a 300 metric tons of heavy metal/yr plant that operated in upstate New York from 1966 until 1972 [West Valley, 1981]. Using the PUREX Process the West Valley plant reprocessed about 650 metric tonnes of initial metal (MTIHM), about 390 tons of which was metallic fuel from the Hanford plutonium production reactors. Consequently the fuel had a very low burnup of around 2000 MWd/tonne[15]. The remainder of the fuel reprocessed at the West Valley plant was uranium oxide fuel and fuel containing thorium. Because of seismic concerns and other issues that would have resulted in greatly increased cost, a planned expansion of the capacity of the West Valley plant was abandoned and the plant was closed. It is now undergoing decommissioning.

b. GE Morris, Il Plant – never operated

In 1967 the Atomic Energy Commission (AEC) authorized General Electric Co. to build a reprocessing plant in Morris, IL. It was to employ a novel reprocessing method based on the volatility of uranium hexafluoride to separate uranium from fission products and actinides.

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[13] CEQ stands for Council on Environmental Quality.

[14] The fluorinel process was based on the use of acid containing fluoride ions to aid in the dissolution of naval reactor fuels. While effective with naval fuels, fluorinel required the use of expensive process equipment that was resistant to fluoride corrosion.

[15] Plutonium production reactor fuel was irradiated to very low burnups to produce ^{239}Pu with a minimum production of higher mass isotopes of plutonium that diminish its value in weapons.

However, design and operational problems caused GE to halt construction of the plant before it processed any spent fuel. Fortunately it never became radioactive, which would have required costly decommissioning. Its storage pond is currently used as an independent spent fuel storage installation (ISFSI) to store commercial spent nuclear reactor fuel.

c. Barnwell Nuclear Fuel Plant – never operated

Construction of the BNFP in Barnwell, SC, near the DOE Savannah River site, began in 1970. The projected plant capacity was to be 1500 MTIHM/yr. The plant design, which incorporated redundant cross piping to accommodate possible piping failures, was based on the PUREX process and was discussed in detail in a preceding section. In 1976 President Ford announced that "...reprocessing and recycling plutonium should not proceed unless there is a sound reason..." Presidents Carter's veto in 1978 of S.1811, the Energy Research and Development Administration (ERDA) Authorization Act of 1978, and his decision to defer indefinitely commercial spent fuel reprocessing effectively ended any chance for commercial operation of the plant, and it was abandoned before operating with spent fuel (thus avoiding costly decommissioning).

3. International

Although the U.S. discontinued attempts at commercial spent fuel reprocessing in the mid-1970s this did not deter construction and operation of reprocessing facilities worldwide. Table //4// summarizes the capacity of civil reprocessing plants that are operating or planned.

Table //4// Civil Reprocessing Plants Operating and Planned in Other Nations [ISIS, 2007]

Country	Location	Scale	Rated Capacity, te HM/yr	Feed Material
China	Lanzhou*	Pilot Plant	0.1	PWR, HWRR
France	1. LaHague UP2-800	Commercial	800	LWR
France	2. LaHague UP3	Commercial	800	LWR
India	1. Kalpakkam Reprocessing Plant (KARP)	Demonstration	100	PHWR
India	2. Lead Minnicell Facility (LMF)	Pilot Plant	n/a	FBTR
India	3. Power Reactor Fuel Reprocessing Plant (PREFRE)	Demonstration	100	PHWR, LWR
India	4. Fast Reactor Fuel Reprocessing Plant*	Commercial	n/a	FBTR

Japan	1. Rokkasho Reprocessing Plant	Commercial	800	LWR
Japan	2. JNC Tokai Reprocessing Plant	Demonstration	210	LWR
Russia	1. Research Institute of Atomic Reactors (RIAR)	Pilot Plant	1	n/a
Russia	2. RT-1, Combined Mayak	Commercial	400	VVER-440
U.K.	1. BNFL B205	Commercial	1500	U Metal (Magnox)
U.K.	2. BNFL Thorp	Commercial	900	LWR, AGR Oxide

* Under commissioning or construction.

Table //5// lists the civil reprocessing plants that have operated in the past and which have been or are being decommissioned. Note the relatively large number of pilot plants built by the major reprocessors before proceeding to large-scale reprocessing plants which indicates the desirability of such facilities to test integrated flowsheets before plant construction and optimize large-scale plant operations.

Table //5// Decommissioned Civil Reprocessing Plants [ISIS, 2007]

Country	**Plant**	**Scale**	**Design Capacity, te/yr**	**Feed Material**
France	1. Experimental Reprocessing Facility	Pilot Plant	5	
France	2. LaHague - AT1	Pilot Plant	0.365	
France	3. Laboratory RM2	Laboratory	0	
France	4. Marcoule - UP1	Commercial	600	
Germany	Weideraufarbeitungsanlage (WAK)	Pilot Plant	35	MOX, LWR

Italy	Eurex SFRE* Pu Nitrate Line)	Pilot Plant	0.1	$Pu(NO_3)_4$
Japan	JAERI's Reprocessing Test Facility (JRTF)	Laboratory	0	
U.K.	BNFL B204 Reprocessing Plant	Commercial	0	
U.K.	BNFL B207 Uranium Purification plant	Commercial	0	
U.K.	BNFL THORP Miniature Pilot Plant (TMPP)	Pilot Plant	0	
U.K.	UKAEA Reprocessing Plant, MTR	Commercial	0.02	MTR
U.K.	UKAEA Reprocessing Plant, MOX*	Commercial		

* Standby. Plants are in decommissioned status unless otherwise noted. Not all decommissioned facilities are listed, e.g., Eurochemic in Belgium and U.S. commercial facilities discontinued in the 1970s (NFS, GE Morris, BNFP) are omitted.

a. France

France has the largest spent fuel reprocessing enterprise in the world. Commercial reprocessing is carried out at La Hague on the English Channel. LaHague reprocesses spent nuclear fuel from reactors belonging to French, European and Asian electricity companies. AREVA NC La Hague (formerly COGEMA) has a commercial reprocessing capacity of 2,000 metric tons of spent fuel per year (equivalent to annual spent fuel discharges from 90 to 100 light water reactors). For more than 10 years La Hague reprocessing has been split between the requirements of the French nuclear program (France's 58 nuclear power plants, generating 76% of the country's electricity) and those of the 29 European and Japanese power companies that have reprocessing agreements with AREVA NC. Power companies from seven countries have sent or are sending spent fuel to AREVA NC La Hague (France, Japan, Germany, Belgium, Switzerland, Italy, and the Netherlands). From 1990 to 2005, close to 20,000 metric tons of fuel were reprocessed at the La Hague site.

The UP1 reprocessing plant at Marcoule, commissioned in 1958, handled 18,600 metric tons of spent fuel from gas-cooled reactors and research reactors to recover the reusable nuclear materials (uranium and plutonium). The site, located in southern France close to the Rhone river reprocessed spent fuels for Commissariat à l'Ènergie Atomique (CEA) needs (G1, G2, G3 reactors and Chinon 1). France's commercial activities were initiated on the site in 1976, when UP1 began reprocessing spent fuel from the French GCRs natural uranium-fueled, which were graphite moderated, gas-cooled reactors. COGEMA was created the same year, and took over the operation of the UP1 plant. Production in the UP1 plant was terminated at the end of 1997 after 40 years of operation. Since the beginning of 1998, the plant has been undergoing final shutdown operations, to be followed by retrieval and repackaging of waste, then by dismantling and decommissioning the plant.

b. United Kingdom

Great Britain is the second largest reprocessor of power reactor spent fuel in the world. Reprocessing is carried out at the Windscale/Sellafield plant in the north-west of England on the Irish Sea. Civilian reprocessing began at Windscale in 1964, and is expected to continue until at least 2010. Magnox power-reactor fuel has been reprocessed at Windscale/Sellafield since 1964. Magnox fuel reprocessing is expected to continue until 2015, about five years after the shut down of the last Magnox reactor in Britain. Oxide fuel reprocessing began in 1969. Large scale oxide fuel reprocessing began with the commissioning in 1994 of the Thermal Oxide Reprocessing Plant (THORP) (nominal capacity: 700 metric tons fuel per year). About 70 percent of the first ten years' reprocessing at THORP was dedicated to foreign fuel. The British utility, British Energy, holds contracts to reprocess about 2600 metric tons of fuel, while additional contracts for 700 metric tons of fuel were signed by German utilities in 1990. These contracts would secure production at THORP until about 2011, after which it has been indicated that the plant might be shut down.

Fast reactor and materials test reactor (MTR) fuel has been reprocessed at Dounreay in northern Scotland since July 1958. This small reprocessing facility is now shut down and is undergoing decommissioning.

c. Japan

Japan has a small reprocessing plant at Tokai-mura, with a design capacity of about 270 metric tons per year (0.7 te/day). (The actual annual reprocessing rate has been about 100 te/yr.) Construction of Japan's first commercial reprocessing plant has been completed at Rokkasho-mura and testing for commercial start-up is underway. The plant, which is of French design, includes a number of buildings for the head-end process, separation and purification, uranium and plutonium co-denitration, high-level radioactive waste vitrification, and other processes related to spent fuel recycle. The nominal reprocessing capacity of the plant is 800 ton-U/year, enough to reprocess the spent fuel produced by about thirty1,000 Mwe nuclear power stations.

d. Russia

The primary Russian reprocessing activity is at Mayak. The Mayak nuclear fuel reprocessing plant is between the towns of Kasli and Kyshtym (also transliterated *Kishtym* or *Kishtim*) 150 km northwest of Chelyabinsk in Siberia. The plant is in the Ozersk central administrative territorial unit, formerly known as Chelyabinsk-40, later as Chelyabinsk-65, and is part of the Chelyabinsk Oblast.

In 1948 reprocessing irradiated fuel from the Russian plutonium production reactors began at the Mayak plant. The plant underwent several modernizations and continued operation until the early 1960s. Reprocessing irradiated fuel from the production reactors was continued at a second plant located next to the first. (The second plant subsequently was combined into a single industrial area called 235.) The second plant was adapted to extract isotopes from irradiated targets from the Chelyabinsk-65's isotope production reactors. In 1987, after two out of five production reactors were shut down, and the second reprocessing plant was shut down.

Plant RT-1 was commissioned in 1977 to reprocess spent fuel from VVER-440, BN-350, BN-600, research, and naval propulsion reactors. Most of the feed is from VVER-440 reactors. This is the only Russian facility that reprocesses spent power reactor fuel. The plant's nominal reprocessing capacity (based on spent fuel from the VVER-440 reactors) is 400 tons of spent fuel per year. The historical average throughput of spent fuel at RT-1 is estimated to be 200 MT of heavy metal per year. Since 1991 reprocessing of foreign spent fuel has become the main source of revenue for Mayak, and has served to cover the cost of domestic spent fuel reprocessing. Until 1996, Mayak Production Association had contracts with nuclear utilities from Finland, Germany, Hungary, Ukraine, and Bulgaria. By 1996, however, Bulgaria, Germany, and Finland had stopped using Mayak's services.

e. India

Three reprocessing plants with a total design capacity of about 200 metric tons, none of which is safeguarded by IAEA, are operated by the Indian Department of Atomic Energy (DAE). The first Indian reprocessing plant, at the Bhabha Atomic Research Centre (BARC) at Trombay, began operating in 1964 and has processed fuel from the Cirus and Dhruva research reactors. It was decommissioned in 1973 due to excessive corrosion, then refurbished and put back into service in 1982.

A second reprocessing plant, the Power Reactor Fuel Reprocessing (PREFRE) facility, dedicated to reprocessing CANDU power reactor fuel, was brought into operation at Tarapur in 1982. The design capacity of PREFRE is 100 metric tons of fuel per year. However, production at the plant has been constrained by logistical and technical problems. Furthermore, India has sought to avoid building plutonium stockpiles. In 1995, there was a serious leak of radioactivity at the Waste Immobilization Plant associated with the Tarapur plant.

In March 1996 cold commissioning (operation without actual spent fuel) began at the Kalpakkam Reprocessing Plant (KARP) located at the Indira Gandhi Centre for Atomic Research (IGCAR) near Madras. 'Hot' commissioning, with the introduction of spent fuel, was planned for the end of 1996. Originally, this site was planned to have 1,000 tons/year of reprocessing capacity by the year 2000, but these plans are now in limbo. The facility is currently designed to have a capacity of 100 metric tons of CANDU fuel per year, for an annual output of about 350 kg of plutonium.

f. China

China plans to reprocess spent nuclear fuel, stating: "China will follow Japan's lead and use the separated plutonium to fuel fast-breeder reactors." China also plans to recycle mixed-oxide (MOX) fuel for use in its pressurized water reactors (PWRs) and fast reactors. The China National Nuclear Corporation (CNNC) has announced plans to construct a facility to reprocess spent fuel with a capacity of 400-800 tons per year, and China has pledged that its new plutonium extraction facilities will be open to international inspections. At present China operates a 0.1 tonne/yr pilot plant at Lanzhou for commercial spent fuel reprocessing.

g. South Korea

It is not anticipated that South Korea will actually reprocess spent fuel or produce separated plutonium. However, South Korea has a collaborative reprocessing program with Canada to develop the DUPIC process. The DUPIC program is the subject of South Korea's national case study for the IAEA's INPRO[16] project, evaluating new fuel cycle technologies. The DUPIC process involves taking spent fuel from light water reactors such as PWRs, crushing it, heating it in oxygen to oxidize the UO_2 to U_3O_8 (thus changing its crystal structure and pulverizing it) and drive off about 40% of the fission products, (principally I, noble gases, tritium, Cs and Tc) and re-fabricating it into pressurized heavy water reactor (PHWR) fuel. The recycled fuel still contains all the actinides, including a plutonium content of nearly 1% and about 96% of the initial uranium, which contains about 1% U-235. Thus, the fissile content (^{239}Pu plus ^{235}U) is around 1.5% - more than double that of natural uranium (0.71 % ^{235}U), and suitable for use in today's PHWRs.

4. Consolidated Fuel Reprocessing Program

One of the earliest attempts by the U.S. government to address fuel recycle was The Consolidated Fuel Reprocessing Program (CFRP). CFRP was initiated in 1974 at Oak Ridge National Laboratory primarily to advance the technology of fast reactor fuel reprocessing, although in a broad sense many aspects of the technology were applicable in general to all conventional fuel reprocessing. The program emphasis was primarily on process automation technology, robotics, process computerization, and head end process steps to improve gaseous effluent control.

Automation technology has been widely adopted in the manufacturing industry and in the chemical processing industries, but only to a very limited extent in nuclear fuel reprocessing. Its effective use in reprocessing had been limited by the lack of diverse and reliable process instrumentation and the general unavailability of sophisticated computer software designed specifically for reprocessing plant process control.

A new facility, the Integrated Equipment Test (IET) facility, was developed by the CFRP in part to demonstrate new concepts for control of advanced nuclear fuel reprocessing plants using advanced instrumentation and a modern, microprocessor-based control system. This facility provided for testing of all chemical process features of a prototypical fuel reprocessing plant that can be demonstrated with unirradiated uranium-bearing feed materials. The goal was demonstration of the plant automation concept and development of techniques for similar applications in a full-scale plant. It was hoped that the automation work in the IET facility would be useful to others in reprocessing by helping to avoid costly mistakes because of the underutilization or misapplication of process automation. Eventually the CFRP became reliant on the infusion of money from the Japanese nuclear enterprise and on-site Japanese technical personnel for survival. Because of the moratorium imposed by the Carter administration on U.S. reprocessing, much of the U.S.-supported CFRP technology that was developed has to date found more application in Japan than it has found in the U.S.

5. International Nuclear Fuel Cycle Evaluation (INFCE)

President Carter's April 1977 statement on nuclear policy that made a commitment to defer indefinitely the commercial reprocessing and recycling of plutonium – coupled with low prices for fossil fuels and uranium – effectively ended consideration of non-defense reprocessing and recycle activities in the U.S. for decades. However, the immediate result of the deferral was the

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[16] INPRO is an IAEA program whose goal is to provide a "Methodology for Assessment of Innovative Nuclear Energy Systems as based on a defined set of Basic Principles, User Requirements and Criteria in the areas of Economics, Sustainability and Environment, Safety, Waste Management, Proliferation Resistance and recommendations on Cross Cutting Issues."

initiation of a series of studies to evaluate the need for reprocessing and plutonium recycle. The largest of these was the International Nuclear Fuel Cycle Evaluation (INFCE) which is described below.

INFCE addressed essentially all the important technical issues related to fuel recycle. In October 1977 the International Nuclear Fuel Cycle Evaluation Committee was initiated, mainly at the urging of the U.S., to investigate opportunities to safely internationalize the nuclear fuel cycle. INFCE participants met between 1977 and 1980 to address ways to use the nuclear fuel cycle to produce nuclear energy with a reduced risk of nuclear proliferation by modifying the fuel cycle technological base. INFCE highlighted a number of measures to counter the dangers of nuclear proliferation, including institutional and technical measures, as well as improvement and further development of International Atomic Energy Agency (IAEA) safeguards. Subsequent to INFCE, reprocessing of spent nuclear fuel and recycle of the resulting nuclear materials was virtually ignored in the U.S. until the turn of the century.

a. Content of the Study

INFCE focused on: 1) an overall assessment of the nuclear fuel cycle, 2) measures to improve assurances of availability of plutonium supply for reactor fuels to developing states, 3) spent nuclear fuel storage, 4) improvements to nuclear safeguards, and 5) alternatives to an international nuclear economy based on plutonium and highly-enriched uranium fuels. Concern about introduction of an international plutonium economy led the IAEA in 1978 to establish a Committee on International Plutonium Storage (IPS). Establishment of the IPS was the principle recommendation of INFCE.

b. Principle Conclusions

The report of INFCE Working Group 4 [INFCE, 1980], one of eight INFCE Working Groups, discussed reprocessing, plutonium handling, and recycle of plutonium to thermal reactors. Fast reactor recycle to was left to Working Group 5, and other fuel recycle concepts, e.g., thorium-based and research reactor fuels, were left to Working Group 8. The basic recommendation of INFCE was to deposit plutonium surplus to national needs with the IAEA. In this strategy for controlling plutonium it was envisioned that excess plutonium would be placed under international inspection and control until needed for use in civil nuclear power applications.

The proposed Global Nuclear Energy Partnership (GNEP) and closely related Advanced Fuel Cycle Initiative (AFCI) has as one of its cornerstones the development and reduction to practice of spent nuclear fuel separations processes that leave Pu primarily with actinides other than uranium. This necessitates equipment and methods for tracking, assay and accountability of the fissile material content of separations process streams that have not been faced heretofore in this country. The processes proposed above will require equipment, processes and detectors for real-time tracking and monitoring and fissile content assay of materials used in fabrication of fuels from fissile material from the separations processes.

B. Refabrication

Refabrication of fuel from reprocessed spent nuclear fuel is a large international industry. A recent IAEA publication [IAEA, 2007] gives information on both the characteristics (e.g., the ^{99}Tc concentration) and the specifications of reprocessed UO_3.

1. Fuel Refabrication Technology

LWR reactor fuels are of two types: 1) low-enriched uranium oxide and 2) mixed uranium-plutonium oxides (MOX). The uranium oxide fuels are much more common, but as more plutonium becomes available MOX fuels, in combination with uranium oxide fuel, are becoming more common. Both fuel types are made from what is essentially the dioxides of the two fissile metallic components.

For UO_2 pellet material fabrication uranyl nitrate solution is denitrated in a fluidized bed to form UO_2. For MOX fuel material preparation uranium and plutonium oxide powders are blended or uranium and plutonium solutions are mixed, concentrated, and denitrated (by microwave heating) to produce a mixed U/PU oxide (MOX). Plutonium nitrate solutions are treated in a manner similar to uranyl nitrate solutions if PuO_2 is sought. UO_2, PuO_2 and MOX are then treated by the following steps.

a. They are calcined in air at 800 º C., producing a partially reduced product.

b. The calcined product is heated in a reduction furnace in H_2/N_2 at 800 º C. to produce UO_2, PuO_2 or MOX fuel material suitable for pellet fabrication. (This two-step reduction saves hydrogen.)

c. LWR fuel fabrication is carried out using Zircaloy cladding and hardware. (The alloy of zirconium is used for neutron economy. It has a low cross section for capture of neutrons in the neutron energy spectrum found in LWR cores.)

d. Fast reactor (LMFBR) fuel is fabricated using stainless steel cladding and hardware. Stainless steel is suitable for use with liquid metal coolants and where the temperature is high. (Neutron economy is not as important in fast reactors where the neutron energy is higher than in LWRs resulting in smaller neutron absorption cross sections.)

The steps in MOX fuel fabrication are shown diagrammatically in Figure //5//.

Fig. //5// Diagram of MOX fuel fabrication process

2. MOX Fuel Fabrication Facilities

Some of the recovered plutonium is being fabricated into mixed oxide (MOX) fuel. Table //6// lists the capacity and status of MOX fuel fabrication plants worldwide.

Table //6// Capacity and status of operating MOX fuel fabrication plants [ISIS, 2007]

Country	**Plant**	**Scale**	**Design Capacity, te HM/yr**	**Product Material**
France	Melox	Commercial	195	MOX for LWRs
India	Advanced Fuel Fabrication Facility (AFFF)	Commercial	100 (nominal)	MOX for BWR, PFBR
India	Kalpakkam MOX Breeder Fuel Fabrication (under construction)	Commercial	?	MOX for PFBR
Japan	JNC Tokai (PFDF-MOX)	Laboratory	0.03	MOX fuel element
Japan	JNC Tokai (PFFF-ATR)	Pilot Plant	10	MOX fuel assembly
Japan	JNC Tokai (PFPF-FBR)	Pilot Plant	5	MOX fuel assembly
Japan	Rokkasho MOX Plant (planned)	Commercial	120	MOX for LWRs
Russia	Mayak- Paket	Pilot Plant	0.5	FB, RR MOX fuel
Russia	Research Institute of Atomic Reactors	Pilot Plant	1	FBR (Vibropack)
U.K.	Sellafield MOX Plant (SMP)	Pilot Plant	Likely 40	MOX for LWRs

In addition to the above MOX plants the U.S. DOE has a MOX plant under construction at the Savannah River site in South Carolina. The current plan is for the facility to be built as part of the national strategy to dispose of excess weapons-grade plutonium by using it for commercial power production. According to the plan the facility would be used only for the purpose of disposition of surplus plutonium and is subject to Nuclear Regulatory Commission (NRC) licensing. The current plan is for the facility to be shut down when the weapons plutonium disposition is completed. However, this plan may be reviewed at a later date.

A recent IAEA document provides details of MOX fuel fabrication world-wide [IAEA, 2003a].

3. HTGR Fuel Fabrication

HTGR fuel is very different from other types of solid reactor fuels, and fabricating HTGR fuel is entirely different from fabricating LWR fuel. Both Germany and the U.S. have developed HTGR fuel fabrication processes for HTGR TRISO fuel particle[17] preparation that consist of a number of similar steps. In both countries kernels containing the fissile material are made via a sol-gel process[18], followed by washing, drying and calcining to produce spherical UO_2 kernels in Germany and UCO kernels in the US. The major difference consists of a sintering step using CO in the U.S. process to ensure the requisite C/O stoichiometry in the kernel. The coating processes for the buffer layer are similar, based on chemical vapor deposition from a mixture of Ar and acetylene in a fluidized coater operating between 1250 and 1300 °C. A 5-micron seal coat is added in the U.S. process to seal the porous buffer coating. This step does not occur in the German process. Table //7// gives typical properties of coated fuel particles and pebbles. Figure //6// is a schematic diagram and photograph of TRISO fuel particles.

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[17] Triso fuel particles are so-named because they consist of a spherical fuel kernel coated with two layers of graphite and one layer of silicon carbide (SiC) to make a tiny "fuel element." It takes millions of them to fuel an HTGR.

[18] In sol-gel processes a colloidal suspension (sol) is "gelled" to form a solid by extraction of water and addition of a mild base. When carried out using droplets of sol, spherical gelled particles are formed.

Fig. //6// Schematic and photograph of TRISO particle

Table //7// Typical Coated Particle Composition and Dimensions for Pebble Bed Fuel

Microspheres

Kernel composition: UO_2
Kernel diameter: 501 µm
Enrichment (U-235 wt.%): 16.75
Thickness of coatings (µm)

Buffer	92
Inner PyC	38
SiC	33
Outer PyC	41

Particle diameter: 909 µm

Pebble

Heavy metal loading (g/pebble): 6.0
U-235 content (g/pebble) 1.00 ± 1%
Number of coated particles per pebble: 9560
Volume packing fraction (%): 6.2
Defective SiC layers (U/Utot): 7.8E-06

A photograph of a “pebble” of the type used in the pebble bed reactor is shown in Figure //7//.

Fig. //7// Photograph of German HTGR pebble fuel element

A prismatic fuel assembly of the type developed by General Atomic and used in the commercial Fort St. Vrain Power Reactor is shown in Figure //8//. These fuel assemblies are 14 inches from one flat vertical face to the opposing face and about a meter high. Fuel “sticks” of pyrolyzed carbon containing TRISO fuel particles are inserted into holes in the fuel block. There are also channels that go completely through the prismatic block for coolant gas flow. Larger channels provide openings into which boron carbide control rods may be inserted.

Fig. //8// Prismatic HTGR fuel element

A major difference in the production of the TRISO coating is that all three layers are coated in a continuous manner in the German process, whereas in the U.S. process the fuel particles are unloaded from the coater after each coating layer to perform quality control (QC) measurements. The inner pyrocarbon layer in both cases is deposited from a mixture of acetylene, propylene, and argon. The temperature in the U.S. process is somewhat lower than in the German process and the coating gas concentration is different, producing a different microstructure and density for the inner pyrocarbon (IPyC) layer. The SiC layer is deposited from a mixture of hydrogen and methyltrichlorosilane at similar coating rates, although the temperature for U.S. coating is about 150 °C higher than that used in the German process. The outer pyrocarbon layer (OPyC) layer is coated in a manner similar to the IPyC layer. In the U.S. a seal coat and protective pyrocarbon layer (PPyC) are added. These layers are not counted in determining that the particle is a "TRISO" particle.

The fuel pebble in Germany uses graphite powder and organic binders to produce a powder matrix that is used to contain the particles and to create the fuel pebble. In the US, a liquid matrix composed of petroleum pitch, graphite flour and graphite shim mixed with organic binders is used to make the fuel compact. Both fuel forms are pressed and carbonized at high temperature (800-900 °C).

Ultra high purity systems and feedstock are used in the manufacture of pebbles in Germany to ensure adequate control of impurities. Both fuel forms undergo a final heat treatment, with the U.S. compact heated at 1650 °C and the German pebble at 1800 to 1950 °C in vacuum.

IV. RECYCLE FACILITY SITING AND DESIGN

The purpose of a reprocessing plant is to chemically separate the fissile content of irradiated nuclear material from fission products and other actinide elements to recover fissile (^{235}U, ^{239,240}Pu, ^{233}U) and fertile (^{238}U, ^{232}Th) radionuclides. There are four major steps involved in building and operating a reprocessing plant: (1) site selection, (2) plant design, (3) plant construction, and (4) plant operation.

A. Site selection

Siting of a reprocessing plant is determined by a large number of considerations. These include proximity to reactors providing the spent fuel, geology, hydrology, seismology, climatology, flooding potential, topography, demographics, and uses to which the surrounding land is put, e.g., agriculture, industry, and transportation.

Proximity to reactors producing the spent fuel is important from the point of view of transportation and possible terrorists attack during shipment, but under some circumstances may not be of paramount importance. This would be true for example if the spent fuel were of foreign origin. In any case, shipment of the spent fuel to the reprocessing plant may be cause for concern by the public.

Geology of the site is important if radioactive liquid effluents are released because the rate of transport of radionuclides through the environment is strongly influenced by the nature and conformation of the soil. For example, clay has an affinity for important radioisotopes such as ^{137}Cs and rare earths and is likely to be self-healing if fractured, whereas granite has little such affinity, and there is no tendency for cracks to heal. Additionally, it is desirable to build a reprocessing plant where background radiation is low and not highly variable because it is difficult to establish an environmental monitoring radioactivity baseline where radiation levels are high or fluctuate widely. This problem can occur where uranium or thorium levels in the soil are high, leading to high radon levels that may produce large radiation background variations during climatic inversions.

Hydrology is important if radioactive liquid effluents are released because the predominant mechanism for transport of radionuclides is carrying by groundwater. (In the case of mal-operation transport by air can become of great importance, as for example the Chernobyl accident.) This transport pathway may be by the mechanism of carrying dissolved ions of radionuclides or by carrying colloids or pseudocolloids, e.g., colloids of plutonium or pseudocolloids of iron or clay to which radionuclides are sorbed. This pathway is the most likely source of non-natural radiation dose to the public through direct ingestion of radionuclides or through contamination of agricultural products by irrigation using contaminated groundwater obtained from wells or streams.

Seismology has a major impact on licensing plant sites and on plant construction. The plant must be sited where it is practical, both economically and physically, to ensure and demonstrate that its integrity can be retained during a projected earthquake. Those parts of the reprocessing plant that contain heavy shielding and contain the highest levels of radioactivity must be capable of withstanding earthquakes with no loss of containment integrity.

Climatology plays a role in plant siting because some areas are prone to seasonal weather extremes, such as hurricanes, tornadoes, snow and ice storms, and fires in dry weather.

Flooding potential is an important consideration if the site is located in a flood plane, near rivers or streams, or is in the path of seasonal snow-melt runoff.

Topography plays a role because the cost of plant construction may be high if grades are too steep, too much soil removal is required, or water drainage is inadequate and poses construction and subsequent operational problems.

Demographics play a major role in gaining public acceptance of a site. Whenever practicable it is desirable to site a reprocessing plant distant from large population centers. This consideration may be at odds with locating the reprocessing plant near reactors where transportation problems are minimized and is an example of the often conflicting siting considerations.

Agriculture and Industry in the neighborhood of a potential plant site can be of considerable importance. The presence of a facility that handles large amounts of radioactivity can be claimed to diminish the value of the crops, the land, or the products produced. Additionally, release of radioactivity and concomitant contamination of expensive crops or industrial buildings and machinery can lead to very large financial obligations.

Transportation activities, such as commercial air or truck traffic need to be considered. This applies both to the transport of radioactive materials and to ordinary commercial traffic. Heavily traveled highways such as interstates in the immediate vicinity of the plant may cause concern to the public or the Departments of Transportation, both federal and local. Intermodal spent fuel transport, including use of navigable waterways, may cause concern with sportsmen as well as health departments if the waterways are the source of drinking water. These transportation issues are especially nettlesome because of the need to balance negative public perception with the desirability for the plant to be reasonably close to the source of the spent fuel, whether generated in the U.S. or coming into ports from overseas.

B. Design and Construction

A typical spent fuel reprocessing facility is designed and constructed to minimize the release of radioactive materials both during routine operation and under unusual or accident conditions. Specifically, the current 10 CFR Part 20 Subpart E §1406 "Minimization of contamination" states the following: "Applicants for licenses, other than renewals, after August 20, 1997, shall describe in the application how facility design and procedures for operation will minimize, to the extent practicable, contamination of the facility and the environment, facilitate eventual decommissioning, and minimize, to the extent practicable, the generation of radioactive waste." At least two physical barriers (frequently more than two) contain the radioactive materials within the facility during operation. These barriers are typically the process equipment (vessels, pipes, etc.) and the building around the processing equipment. In most cases, the building itself provides two barriers – the hot cell or room where the process equipment is located and the outer building shell.

1. Design

Historically, recycle plants have consisted of the following four major processing facilities plus a fuel receiving and storage area:

(1) *The Separations Facility*, in which the spent fuel assemblies are processed to recover uranium and plutonium as nitrate solutions and where the bulk of radioactive by-product wastes are separated as a concentrated nitrate solution of HLW.

(2) *Uranium Hexaflouride Facility* in which the recovered purified uranyl nitrate solution is

converted to UF_6 suitable as a feed material for isotopic re-enrichment if desired.

(3) *Plutonium Product Facility* in which the recovered plutonium nitrate solution is converted to PuO_2, suitable for use in the production of mixed oxide fuel (MOX).

(4) *Waste Management Facilities* for the handling, solidification (presumably by vitrification in borosilicate glass), packaging, assaying, inspection, and interim storage of waste prior to shipment to the federal HLW repository (currently planned by DOE for location at Yucca Mountain, Nevada).

The actual design of these major facilities will be directly related to the regulations effective at the time of licensing and the desired/required form of both the fissile as well as the waste material discharged. Proliferation and safeguards are of national and international concern when considering the construction of a recycling plant, as are attacks by terrorists. Beyond these overriding considerations there are very important practical matters that must be taken into account in the design, construction and operation of a plant.

It is necessary to optimize the plant configuration for reprocessing to minimize the overall facility capital and operating costs. This is done by considering the interplay of a large number of factors. Initially, it must be decided if the plant is to be designed with a single, multiple step, process line or whether it will have parallel process lines. If the plant is to process a variety of fuel types or a very large throughput is required then parallel lines will facilitate processing dissimilar fuel types, allow maintenance of one line when the other line is in operation, or allow practical equipment sizes while achieving high throughput.

Another aspect of plant optimization concerns approaches for waste treatment (e.g., concentration of liquid wastes by evaporation and compaction or melting of spent fuel cladding hulls), and its storage and disposal. Design optimization also addresses radiation protection of workers through use of the minimum shielding thickness consistent with meeting ALARA and radiation dose and radioactivity confinement requirements; appropriate selection of the processes carried out in the plant; and careful choice of the equipment used to carry out those processes. Simple, reliable equipment, continuous operation where possible, and ease of remote removal and replacement of equipment all contribute to minimizing capital and operating costs. In addition to being able to achieve the desired throughput, each equipment piece in the high radiation areas of the plant must be capable of being replaced remotely, be critically safe, be matched to the characteristics of the fuel assemblies to be reprocessed, and chosen insofar as possible to be of standard sizes.

Inoperability of a reprocessing plant is likely to result from either (1) structural weakness in an inaccessible area as a result of corrosion or mechanical failure, (2) failure of essential inter-cell piping or (3) process failure.

Some general guidelines are useful to keep in mind in plant design: It is desirable for radiation protection and ease of operation to put equipment for receiving the spent fuel, spent fuel pool and HLW storage, the fuel segment storage, and the reprocessing product storage in separate cells interconnected through transfer channels to the processing area. Ventilation and waste treatment capabilities may be provided separately for each segment. However, some facilities such as those used in maintenance may be shared.

Experience has shown that improved technologies may be needed in the following areas:

1. Cask handling and cooling techniques for dissimilar transport systems associated

with different fuel types;

2. Techniques for safely storing different fuel types in a common storage pool;
3. Mechanical handling systems capable of handling dissimilar fuel assembly sizes and shapes.

After many years of successfully operating the DOE Savannah River Site reprocessing plant the duPont Company carried out and supported research and development by others of a conceptual design for what would be an NRC licensed fuel recycle complex based on duPont's reprocessing experience and lessons learned from that experience and the experience of others. The design studies were completed and reports issued in November 1978 [Behran, 2000]. This facility design was not considered in White House reviews of reprocessing during the Ford, Carter, or Reagan administrations.

Special features of the duPont facility design included:

- Canyon structures for containing process equipment that could be installed, maintained and replaced remotely using overhead cranes,
- Use of the best technology available, including centrifugal contactors for the first cycle of solvent extraction, and storage of solutions between process steps,
- Product recoveries greater than 99.8 %,
- Reprocessing one-year cooled spent fuel,
- Personnel access to operating areas, with close control of entry and exit,
- Vitrification of high-level wastes for ultimate storage,
- Flexibility to allow changes, additions, or upgrades of equipment, flowsheets, instruments, etc.,
- No accumulation of separated plutonium except in secure surge storage between reprocessing and fuel fabrication,
- Tritium and krypton capture,
- Sand filters,
- Opportunities for lowering cost through research and development as a result of longer cooling time before reprocessing.

2. Construction

Process equipment is to be fabricated from materials that are resistant to corrosive failure. Process equipment designed to prevent major releases of radionuclides under conditions assumed to be credible was designated as being of "Q" design. These "Q" systems must provide confinement integrity for design basis accidents and naturally occurring events such as earthquakes and tornadoes. In other less critical areas, the design membrane stress of the equipment had been established at 80% to 90% of the yield stress during a design basis earthquake. Structural barriers are designed to contain process materials if primary equipment barriers are breached. The principal structural barriers are constructed of heavily reinforced concrete.

The structural barriers for process equipment are generally termed "radioactive process cells" and are usually surrounded by maintenance or operating areas. The process cells where the spent fuel is chopped and dissolved and where high level liquid wastes are concentrated are very high radiation levels cells. At BNFP these cells were designed for remote maintenance (i.e., maintenance from outside the cell by the use of in-cell cranes, shielding windows and manipulators). Similarly, a cell for remote packaging of radioactive wastes and for performing remote decontamination and maintenance of equipment removed from other process cells was also provided. The rest of the process cells are designed to permit direct personnel entry and contact maintenance, but only after appropriate remote decontamination has been completed to allow safe entry. These cells were designed to minimize maintenance requirements.

The process and support equipment used in handling radioactive materials is contained in cells or glove boxes. Spent fuel bundles are stored and transported under water in pools. The cells, glove boxes, and pools provide a barrier between the highly contaminated or radioactive environment within and the outside the habitable environment. Cells with thick concrete shielding walls or pools with deep water cover are provided where protection is required against penetrating (gamma) radiation. Glove boxes are used to isolate radioactive material when radiation levels are low and contact operations are permitted. In the last commercial reprocessing plant constructed in the U.S. (BNFP) the portions of the building allowing personnel access were divided into the radiation zones shown in Table //8//.

Table //8// Radiation Zones and Permissible Radiation Fields at BNFP

Zone	Radiation Field (maximum)
Normal access, non-radiation zone (area)	0.1 mR/hr
Normal access, work zone (station)	1.0 mR/hr
Normal access, above work zone (station)	1.0 mR/hr (at 1 foot from shield)
Limited access, work zone (gallery)	10 mR/hr
Limited access, above work zone (gallery)	100 mR/hr (at 1 foot from shield)

The shielding design and designation of each room within the separations facility building are based on the functions to be carried on in the room, the expected occupancy and the anticipated exposure rate. Personnel access to cells is possible but is allowed only when absolutely necessary and only then with adequate protection and health physics coverage. Cell entry is possible only through heavily shielded doors or hatches, which are normally sealed.

The process equipment, piping, building and structures, casks, storage tanks, and fuel element cladding (prior to shearing) provide barriers for the confinement of radioactive materials. Essential confinement systems are designed to maintain their function under normal operating conditions, abnormal operations, upper limit accident conditions, and adverse environmental conditions throughout the life of the facility. Hatches and penetrations, which are an integral part of the structure, are designed so as not to compromise the confinement and shielding functions.

The floors of all cells in the facility are to be covered with continuous (welded) stainless steel liners. These liners serve to contain all liquids within the cells in the event of a primary vessel leak. The walls of the cells are covered with either stainless steel or a radiation-resistant paint. The choice of cell wall covering depends on the nature of the material to be processed within the particular cell and the need for decontamination. The wall covering serves to seal the concrete structural material from the corrosive atmosphere and radionuclides and, hence, facilitate decontamination. Figure //9// is a picture of the BNFP hot cell operating area.

Fig. //9// BNFP fuel reprocessing plant operating area in front of hot cells

Glove boxes are used to provide confinement when operational requirements and radiation levels permit hands-on operation. The penetrating radiation produced by the radionuclides within the glove box is to be sufficiently low such that personnel might operate and maintain the equipment without receiving exposure above approved standards. Therefore, the type of operation performed within glove boxes typically involves only small quantities of radionuclides with penetrating radiation. Generally, glove boxes are used for laboratory, sampling, inspections, or clean plutonium operations. Figure //10// shows a typical glove box setup for handling radioactive material having low levels of penetrating radiation.

Fig. //10// Glove boxes used for handling nuclear materials having low levels of penetrating radiation

3. Equipment Modules

The following major equipment modules are required: (a) spent fuel receiving and storage, (b) main process cells, HLW solidification plant; (d) uranium hexafluoride conversion plant; (e) plutonium product facility; (f) auxiliary process systems and service areas. These modules are discussed below.

a. Spent Fuel Receiving And Storage

In a typical reprocessing/recycling plant the Fuel Receiving and Storage Station (FRSS) is designed to receive and store irradiated fuel assemblies from LWRs. The spent fuel assemblies are received in shielding casks transported by either truck or rail, and unloaded under water. The fuel assemblies are stored under water to provide cooling and shielding. Facilities for decontaminating the shipping casks before they leave the plant, and equipment to circulate, filter, deionize, and cool the spent fuel storage pool water are provided. The major areas of the FRSS and their primary functions are summarized in Table //9//.

Table //9// Primary Functions of Areas in Spent Fuel Receiving and Storage Station

Area	Primary Process Functions	Remarks
Two vehicle loading bays	Receive rail and truck casks; unload casks from transport vehicle; load empty casks onto transport vehicles	
Test and decontamination pit	Prepare casks for unloading in cask unloading pool	Stainless steel floor pan
Cask unloading pools	Remove fuel from casks; place solidified waste canisters in cask	Stainless steel liner
Decontamination pit	Decontaminate casks after removal from cask unloading pool	Stainless steel floor
Fuel storage pool; waste canister racks	Store fuel assemblies and solidified waste	Stainless steel liner
Fuel transfer pool	Transfer fuel assemblies to main process building	Stainless steel liner
Deionization area	Circulate, filter, de-ionize and cool pool water	

The FRSS is connected to the Main Process Building by the fuel transfer conveyor tunnel and is connected to the waste solidification plant by an underwater transfer aisle. The pool walls and liners are designed to maintain their containment integrity during a design-basis earthquake or tornado. Building walls above the pools are non-Q structures.

b. Main Process Cells

The Main Process Cells are the functional center of the reprocessing/recycling plant. The uranium and plutonium are chemically separated from the other actinides and fission products in these cells in the Main Process Building. This processing is carried out in a series of process cells that occupy a major portion of the building. The Main Process Building also contains a wide variety of facilities and equipment that are used to monitor and control the process, maintain the equipment, carry out auxiliary operations, and treat gaseous effluents from the processes carried out in the building.

The primary functions of the main process cells are listed in Table //10//. Most of the building is constructed of reinforced concrete designed to remain intact during a design-basis earthquake or tornado. Process cell walls are rebar-reinforced and up to two meters thick to provide personnel shielding from radioactivity.

The areas outside the main process cells are generally divided into regions called ‘galleries’, ‘areas’, or ‘stations’. These regions enclose and protect service piping, process support equipment, instrumentation components, and some operating areas. Radioactivity levels range from essentially background to fairly modest levels.

Table //10// Primary Functions of Main Process Cells

Cell	Primary Process Function	Remarks
Remote process cell (RPC)	Shear and dissolve fuel; concentrate high-level liquid waste	Stainless steel floor pan; remote maintenance
Remote maintenance and scrap cell (RMSC)	Package leached hulls and other solid waste; remotely maintain contaminated equipment	Stainless steel walls and floor
High-level cell (HLC)	Accountability for dissolver solution; chemically adjust dissolver solution; centrifuge dissolver solution	Stainless steel floor pan
High-intermediate level cell (HILC)	Separate uranium and plutonium from high-level waste; separate uranium from plutonium; treat dissolver off-gas; solvent cleanup; concentrate intermediate-level waste	Stainless steel floor pan; contact maintenance
Intermediate level cell (ILC)	Treat vessel off-gas; recover nitric acid; concentrate low-level waste; burn used solvent	Stainless steel floor pan; contact maintenance
Uranium product cell (UPC)	Purify uranium stream; clean up solvent	Stainless steel pan; contact maintenance
Plutonium product cell (PPC)	Purify plutonium stream	Stainless steel pan; contact maintenance

Plutonium nitrate storage and load-out (PNSL)	Store plutonium nitrate solutions; transfer plutonium nitrate to Plutonium Oxide Conversion Facility	Stainless steel pan; contact maintenance

c. Waste Solidification Plant

The Waste Solidification Plant (WSP) is located adjacent to the Main Process Building. It receives high- and intermediate-level liquid waste solutions from the waste tank farm complex, converts the liquids to a vitrified solid, and facilitates the transfer of solidified high-level wastes to the FRSS for storage and eventual shipment offsite.

The WSP contains the waste vitrification and canister sealing equipment, inspection and decontamination equipment, off-gas treatment equipment, and remote maintenance facilities in four process cells. The primary process functions performed in each of the cells are presented in Table //11//. All process cells in the WSP are completely lined with stainless steel. The cells were to be surrounded by limited access areas for operating and controlling the processes in the cells. All operational and maintenance facilities in the process cells would be performed remotely using viewing windows, manipulators and cranes.

Table //11// Primary Functions of Major Process Cells in the Waste Solidification Plant

Area	Function
Waste vitrification cell	Calcine liquid waste; vitrify calcined waste
Canister decontamination cell	Decontaminate outer surfaces of canisters; transfer filled canisters to FRSS
Off-gas treatment cell	Treat off-gas from WSP process vessels
Hot maintenance cell	Perform remote maintenance on contaminated equipment

d. Uranium Hexafluoride Conversion Plant

In the BNFP described above, the UF_6 conversion facility consisted of two buildings, both of standard chemical plant construction. The main building was a multi-story structure containing the principal process areas. A second building located near the main process area was used for fluorine generation. The UF_6 facility was located near the Main Process Building to eliminate the need for shipping uranyl nitrate to a distant conversion plant. Elimination of the uranyl nitrate shipping requirement saved time, reduced the costs to the nuclear power industry, and lessened the chances of a radiological hazard to the public. Typical UF_6 plants are designed such that there is sufficient surge capacity between process stages to continue operation of adjacent stages when one section is down.

e. Plutonium Product Facility

A Plutonium Product Facility (PPF) was contemplated in the BNFP. Its purpose was to: (1) convert aqueous plutonium nitrate solutions to plutonium oxide, and (2) provide storage for plutonium oxide. The PPF process facilities were to be located in a separate building immediately adjacent to the Main Process Building.

The Plutonium Product Facility was to have a nominal design capacity of 100 kg of plutonium product (as plutonium dioxide) per day with an on-stream time of 250 days per year to give an annual conversion capacity of 25,000 kg of plutonium (1134 g of PuO_2 contains 1,000g of plutonium). This capacity would be divided between two identical production lines, each with a capacity of 50 kg per day. This design basis was selected to allow a 1,500 MTU/year reprocessing/recycling facility to process mixed-oxide feed material for fuels having plutonium contents higher than LWR recycled fuels. Although the design capacity was 25,000 kg of plutonium per year, it could be operated at a lower capacity.

Most of the operations and maintenance in the facility were to be carried out in glove boxes. Leaded gloves were planned to be used to protect against low-level gamma radiation, and relatively thin concrete and other hydrogenous shields would be used to protect against the neutron radiation from the ^{238}Pu [from (α, n) radiation produced when high-energy neutrons strike low-atomic number elements].

f. Auxiliary Process Systems and Service Areas

The Auxiliary Process Systems and Service Areas provide necessary services to the main process functions of the Main Process Building. The major areas are described below.

i. Ventilation System

The ventilation system consists primarily of supply and exhaust subsystems. The ventilation system was designed to provide once-through airflow by pressure controls from non-contaminated areas through potentially contaminated or low-contaminated areas to highly contaminated areas (i.e., process cells), then to treatment systems before being pumped by blowers out the stack. Three blowers were to provide exhaust for the main ventilation system. Each blower was to be capable of supplying fifty (50%) percent of the required capacity and was to be connected to emergency power sources.

Exhaust gases from the radioactive processing cells were to pass through at least two stages of high-efficiency particulate air (HEPA) filters. Off-gases from areas with high plutonium concentrations were to pass through three stages of HEPA filters. These extra stages of filtration were designed to provide for a minimum of one stage of filtration in the event of failure of the upstream filters by mechanisms such as fire. Exhaust gases from the main process and building ventilation systems exited through the main stack (100 meters high). Non-condensed gases from the concentrator vented through the service concentrator stack (30.5 meters high). The chemical makeup and addition tank vented through the chemical off-gas stack (29 meters high).

A major feature of the ventilation system was the ventilation filter station. This housed the primary supply and exhaust blowers and the final stage of HEPA filters through which the exhaust air passed before exhausting through the 100 meter stack. In this late twentieth century design, a cryogenic krypton capture and recovery system was planned to be incorporated into the design, but neither a krypton recovery system nor a system for capture of tritium and ^{14}C was built.

ii. Electrical Power

Normal electrical power was provided to the facility from a commercial substation by two transformers, each feeding a 2,000 amp main breaker. The main breakers distributed power through twelve 120 amp feeder breakers.

The emergency electric power system was designed to handle essential electrical loads in emergency situations. Emergency power was supplied by two independent diesel engine-driven generators. Each generator had a 2,200 kw continuous rating. An emergency battery supply was provided for instrumentation in the main control and the waste tank equipment gallery area.

iii. Fire Protection System

Fire detection and protection systems at the facility were designed to provide early warning and rapid control of fire. Automatic fire detection devices and audible alarms were installed in all areas of the facility. The process cells had dual detection systems. The remotely maintained process cells used manually operated non-combustible purges and water spray mist systems. The contact-maintained cells were served by automatically operated Halon[19] systems. The filter stations were designed with automatic mist suppression systems, and the FRSS had manually operated fire hoses. Most other areas used a manually operated water sprinkler system.

iv. Hot and Cold Laboratory Area

The laboratories provided analytical services for all non-radioactive and most radioactive process samples. At the BNFP, the laboratory building was a two-story complex adjacent to the Main Process Building. It was composed of thirteen individual laboratories equipped to provide specific types of analysis or services. Radioactive materials in these laboratories were handled in glove boxes. The sample and analytical cells are in a shielded facility designed to facilitate radiochemical analyses of samples from the more highly radioactive portions of the process. The cells provide a shielded area for remote sampling and analysis of these materials, and for preparation of samples to be analyzed in the plant analytical laboratories. Operation is through the use of either shielded cells with viewing windows and manipulators or glove boxes.

g. Control Room Area

The control room area houses the process-control and safety-related instrumentation for the plant. It serves as the communications center from which operators can be directed to perform manual functions. The control room area is not expected to be contaminated under normal operating conditions, since the only process connections to other facility areas are electrical.

h. Liquid Waste Storage Areas

High- and intermediate-level liquid waste[20] from the reprocessing operation would be concentrated and stored in large underground tanks until the wastes could be solidified and shipped off-site for disposal. The BNFP had a Liquid Waste Storage complex composed of two high-level liquid waste (HLLW) tanks, one intermediate-level liquid waste (ILLW) tank, and a waste tank equipment gallery (WTEG) that provides services for the tanks. One equivalent HLLW tank volume was to remain available at all times for use as a spare if difficulties should arise with any tank of HLLW or ILLW. Additional HLLW tanks were to be added to handle the continued generation of wastes. The WTEG is a concrete building located near the Main Process Building housing the control room, heat exchangers, coolant circulating pumps, off-gas treatment equipment, and ventilation filters for the waste storage tanks. These tanks were connected

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[19] Halon is a liquefied, compressed halogenated hydrocarbon gas that stops the spread of fire by physically preventing (suffocating) combustion. Although the production of Halon in the U.S. ceased on January 1, 1994 under the Clean Air Act, it is still legal to purchase and use recycled Halon and Halon fire extinguishers.

[20] NRC does not recognize an “intermediate-level” waste classification. The term is used in this report to designate wastes that are not very radioactive relative to high-level wastes, but are higher in radioactivity than Class C wastes.

through a small diverter cell beneath the WTEG and through underground pipe vaults to the Main Process Building and the waste solidification plant. Figure //11// is a picture of the BNFP plant HLW storage tanks under construction showing the extensive internal cooling piping required to remove radioactive decay heat. This large amount of coolant piping in storage tanks at plants undergoing decommissioning poses significant problems when solid salts and sludges must be removed, as is the case at some DOE sites. However, at BNFP all of the tanks were made of stainless steel which allowed storage of acidic wastes and essentially eliminated the presence of solids.

Fig. //11// Tanks for liquid HLW storage under construction at the BNFP facility

i. Solid Waste Storage

The BNFP design included a solid waste storage area of approximately twenty (20) acres -- an area deemed sufficient to store the solid waste generated during the first three years of operation. The solid wastes to be stored in this area were divided into three major categories: (1) spent fuel cladding hulls; (2) high-level general process trash (HLGPT); and (3) low-level general process trash (LLGPT)[21]. Hulls and HLGPT were to be stored in caissons mounted in an engineered berm or in concrete vaults. The LLGPT would be stored in earth-covered cargo containers.

4. Criticality Control Factors

Whenever enriched uranium or plutonium are present criticality control becomes an important consideration. The method used to control criticality depends on the physical and chemical nature of the fissile material, its mass and purity, and its geometry. Several control methods have been employed whose use depends on the above factors.

a. Physical and chemical nature

It is important to know if the physical form is such that fissile material can be compacted to increase its density. It is also very important to know what the chemical composition is. If it chemically conjoined with elements that absorb neutrons it is less likely to present a criticality risk.

b. Mass

For criticality to occur it is essential that the amount of fissile material equal or exceed the minimum critical mass. A common approach to preventing criticality is limiting the allowable amount of fissile material in any one location to less than a critical mass.

c. Purity

Certain chemicals mixed with the fissile material can prevent criticality by absorbing neutrons. Elements with isotopes having large neutron absorption cross sections such as boron, cadmium, or gadolinium, are commonly added to fissile materials. Usually these elements are in a form permitting their easy removal when desired.

d. Geometry

Vessels having geometries that allow for loss of neutrons through their surfaces in amounts such that a chain reaction cannot be sustained in the vessels are universally used. The vessels may be of many differing configurations, but cylindrical or flat “slab” configurations are common. Typically one dimension such as diameter in the case of cylinders or thickness in the case of slabs is limited to the order of 13 cm.

C. Operator Licensing and Training

The operation of a reprocessing/recycling facility entails all of the operational skills and safety requirements associated with a reasonably complex chemical processing plant overlain with the radiation safety, security and safeguards requirements of a significant radioactive nuclear facility.

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[21] NRC does not have a category of waste called “low-level general process trash. The term was used by BNFP as a descriptive identifier of a type of radioactive waste rather than as formal waste classification.

However, other than as noted in the preceding sections of this report, there is little current commercial experience remaining in the NRC regulated world and that which does exist resides mostly in people who have retired. Therefore, the training and qualification of the operating staff takes on major significance in the absence of the ability to rely on a pool of fully trained, experienced, and licensed personnel.

In the past, the general criteria was for operators, technicians and supervisors to have received at least a two year certificate from an established technical school. Applicants with this background normally have sufficient understanding of the physical, chemical, and engineering technologies to adequately undertake the necessary specific plant training.

The importance of qualified operators to the safety of a reprocessing plant can hardly be over emphasized. The contents of an application for licensing individuals who manipulate the controls of a properly licensed facility (at the time of BNFP) are set forth in 10 CFR Part 55, Subpart D, §55.31.

1. Experience at NFS

Experience gained from the past licensing of reprocessing plant operators at other commercial reprocessing plants may be of some benefit to this study. This historical experience may be used as an indicator of the validity of the requirement for training and the evolution of training programs over the years, as well as the possible direction of future training efforts.

During the planning stages of NFS, their management and the regulatory staff of the U.S. Atomic Energy Commission established four major operator categories:

(1) manipulator operators,
(2) chemical operators,
(3) control room operators, and
(4) senior operators.

These categories were similar, in most respects, to those presented in the USAEC Licensing Guide, which was used at that time for nuclear reactor operators.

The results of the original examining program in 1966 were disappointing. Of the total number of senior operator applicants taking the examination, seventy-eight (78%) percent were successful in obtaining licenses; however, only fifty-nine (59%) percent of the chemical operations personnel applying for licenses were successful. Only nine (9%) percent were initially awarded licenses. Some of the reasons for the excessive failure rate were as follows:

- At the time of testing, the head-end system had not been completed, and very little practical operating experience could be included in the training program.
- Most of the applicants were young, and, therefore, had little or no industrial experience.
- As is usually the case in a new plant, the inadequacies of the first training program were not apparent until the program had been completed.
- The first set of tests was, to some extent, experimental.

The disappointing results, and the underlying reasons, were similar to those experienced in the early phases of the program for examining power reactor operators.

Later, a pre-testing program was conducted at ORNL to establish the validity of future examination procedures. In this program, the same tests were administered to process foremen, chemical operators, and technicians who had considerable experience in the reprocessing field and to new employees with little or no experience. The questions posed were basically those to be used for examining NFS operators. The results obtained in this program verified that the questions proposed for the NFS tests were reasonable and confirmed that adequate training was a prerequisite for passing the licensing examination.

As the training methods improved at the NFS and new testing methods were developed, the number of successful applicants at the facility increased. A summary of the NFS licensing experience during the period 1966 - 1970 is presented in Table //12//.

Table //12// Experience in Applications Made By, and Licenses Awarded to, NFS Plant Personnel

				First Re-examination	
Year	# of Applicants	# of Licenses Awarded	# of Successful Applicants, %	# of Applicants	# of Licenses Awarded
1966	98	43	44	51	34
1967	30	23	77	2	2
1968	18	16	89	0	0
1969	49	32	65	6	4
1970	23	15	65	6	4
Total	218	129	Avg. 59	65	44

2. Experience at the MFRP

During 1971 - 1972, the operators of the General Electric Company's fuel reprocessing plant at Morris, Illinois, underwent formal training to prepare them for licensing. Of the 65 persons included in the program, only two failed to qualify for licensing. Many of the candidates for training in the MFRP plant had been licensed previously in the NFS plant and had obtained employment with General Electric when the NFS facility at West Valley, New York ceased operation. Operators in two general categories were trained for operation in the plant (mechanical processes and remote process equipment). In addition, several senior operators were trained for supervisory roles. It was estimated that more than 220 man-days of effort were expended for each candidate in the training program. The estimated cost for this undertaking, including salary, overhead, and training, was established at $25,000 per individual. This cost would be much higher today, of course, because of inflation.

3. Experience at BNFP

The pre-startup staff of the BNFP included a cadre of operators who had been involved in training and retraining over the previous one-to-four year period. In addition to the operator training program, programs for others such as analytical laboratory technicians and security patrol officers were also conducted. These programs were necessary to ensure that all operations would be carried out correctly, not only for safety related reasons, but also for safeguards- related and physical security-related reasons.

The operations personnel and analytical technicians at the BNFP were cross-trained. Security officers were also cross-trained in various areas of physical security. As a result, the personnel were considered to be highly trained and knowledgeable in BNFP operations but would have required retraining at that time consistent with any systems modifications required to generate a more proliferation resistant fuel cycle operation.

Operators, technicians, and patrol officers in the various categories were not given the necessary licensing examination to permit operation because BNFP licensing was terminated before completion. However, because of the extensive training and retraining taking place during checkout and “cold”-run operation, it was expected that the failure rate during the licensing examination would be low. The presence of more experienced personnel in any type of operating facility helps reduce the mistakes made by those who, although well trained, remain inexperienced.

4. Typical Reprocessing Plant Operator Training Program

The programs necessary to train reprocessing plant operators are far more rigorous than those employed in conventional industrial chemical facilities. Further, the process of choosing candidates with the necessary educational, psychological, and medical requirements to receive this training is a prime concern. The selection of candidates who cannot pass the required certification of licensing examinations results in a financial burden to the enterprise; in addition, unsatisfactorily trained individuals tend to jeopardize safety and hamper efficient operation of the plant under normal as well as abnormal conditions.

The great importance of training was recognized as a consequence of the experience gained in licensing reprocessing plant operators. Therefore, the ERDA Division of Operational Safety (ERDA followed the AEC and was the precursor to DOE) expended $146,000 during 1973 - 1976 to establish guidelines for selecting, training, licensing, and certifying reprocessing plant operators. These guidelines were published for use in both commercial and government operated plants.

The time required to adequately train an operator was found to be approximately 1 to 1.5 years.

The qualifications of applicants for operator licenses are determined through two methods of testing: (1) written examinations covering categories such as physics, chemistry, mechanical processing systems, chemical processing systems, equipment and instrumentation, power and auxiliary systems, administrative and procedural rules, and radiological safety; and (2) an oral examination.

The qualifications of the initial operators for future reprocessing/recycling plants is yet to be established, as the role of the DOE and the level of its interaction with the NRC and potential commercial owner/operators has yet to be established.

D. Needed Improvements

The proposed Global Nuclear Energy Partnership and closely related Advanced Fuel Cycle Initiative has as one of its cornerstones the development and reduction to practice of spent nuclear fuel separations processes that leave Pu primarily with actinides other than uranium. This necessitates equipment and methods for tracking, assay and accountability of the fissile material content of separations process streams that have not been faced heretofore in this country. The processes proposed above will require equipment, processes and detectors for real-time tracking and monitoring and fissile content assay of materials used in fabrication of fuels from fissile material from the separations processes.

1. Improved Processes

Precise and accurate tracking, detecting, monitoring and assaying the Pu/HEU content of product and waste streams from separations and fabrication processes will be required for any nuclear fuel recycle plants with improved proliferation resistance.

Computer programs to record, evaluate, interpret and provide real-time output from process equipment and fissile material monitors to local and central monitoring stations are requirements for the integrated, large-scale data handling programs for management of data from all parts of the fuel cycle plant (process control, process monitoring, material transfer, material inventory, portal monitoring) to improve plant proliferation resistance by interrelating and cross checking disparate sources of information, as well as to improve plant efficiency. Plant operating parameters should be compared on a continuous basis with computer simulated normal plant operating parameters to detect, evaluate and report off-normal operation both locally and remotely as a check on possible illicit operations and improper plant operation. These factors are enlarged upon below.

2. Improved Equipment

Process equipment for real-time monitoring and assay of fissile materials present in reprocessing and fabrication plants for fuels from separations processes such as those envisioned by GNEP are required. The proliferation-resistant separations processes proposed in GNEP produce a product of intentionally co-mingled fissile actinides that is to be fabricated without further purification for use in fast-neutron-spectrum reactors. Equipment for real-time monitoring of spent fuel separations processes based on recent advances in instrumentation and controls and adaptations of equipment and computerized analysis of data already in use can make possible improved tracking of fissile material through the processing steps. Flow rates through pipes and process equipment, e.g., centrifugal contactors, pumps, pulse columns, mixer-settlers, and centrifuges, can be better measured and controlled than in the past. Volume and concentration measurements can be made with greater precision and accuracy in feed and product tanks, thus improving material accountability. Fissile material concentrations and amounts can be measured through the use of better sampling and analysis techniques and subsequent computerized analysis of the data. For example, more and better energy spectra measurements and spectrum resolution can be obtained using technology and tools already available. Flow rates of UF_6 can now be measured accurately. These types of improved measurements make possible location, identification and quantification of chemical and isotopic species of interest.

3. Security and Safeguards

In addition to the normal industrial fences and barriers, nuclear facilities have additional requirements for both physical security and nuclear material safeguarding. These two requirements often, but not always, overlap. In light of potential terrorist threats security and safeguards activities are being stressed, and additional measures are being put into place. Physical, psychological and mental requirements of the guard and security forces are specified. These are under continuous review as threat levels are reassessed. Entry portals, coded badges, and other measures are used to control and monitor both personnel and equipment egress and ingress. Internal and external portal monitors are required. Periodic physical inventories of objects containing fissile material are to be performed.

4. Detectors

The proposed separations processes will require equipment, processes and detectors for real-time tracking and monitoring and fissile content assay of materials used in fabrication of fuels and fissile material from the low-decontamination separations processes.

Improvements in the proliferation resistance of nuclear fuel reprocessing plants through use of improved detectors are possible in a variety of areas. Personnel monitoring and record keeping of movements and activities of personnel is easily achieved through a variety of methods to ensure there are no illicit activities. Speciation technology (e.g., radiochemical methods for trace concentrations; laser spectroscopy; X-ray absorption fine structure spectroscopy; magnetic resonance techniques, redox speciation; ion-selective electrodes) for materials of interest has improved greatly in recent years. Computerized recording and analysis of data from the sensing and measuring equipment, conducted both locally and at remote locations, permits detection of off-normal operating conditions. This capability is useful both for monitoring plant operations and for maintaining accountability of fissile material. Potential areas of application of some of these new and novel technologies are discussed below.

5. Material Accountability

As already noted, all nuclear material separations and fuel fabrication processes produce products and wastes that contain fissile material. The amounts of fissile material going to waste can be quite large for high-throughput processes that operate over relatively long periods of time, and when purified and accumulated, potentially can exceed a critical mass. Highly sensitive detection and measurement equipment is now available to monitor and assay the plutonium and enriched uranium content of waste streams from separations plants and from both enriched uranium and MOX fuel fabrication processes.

Computerized, integrated, large-scale data handling programs for management of data from all parts of the fuel cycle plant (process control, process monitoring, material transfer, material inventory, portal monitoring) will be a necessary adjunct to any modern reprocessing or fuel fabrication plant. These programs can greatly improve plant proliferation resistance by interrelating and cross checking disparate sources of information.

Considerable effort, both nationally and internationally, is required between the groups responsible for establishing the permissible significant (SIGMA) Pu inventory differences (ID).

As shown in the Table //13//, there is a large difference between the IAEA, the NRC and the DOE with regard to the Sigma ID requirements, and the frequency of both long-term shutdown inventory and interim frequency requirements.

Table //13// Sigma ID Requirements Based on the Current Most Stringent, Category 1 Requirements for NRC and DOE[a]

Agency	Goal/Requirement Terms	Sigma ID	Frequency of long-term inventory (shutdown required)	Frequency of interim inventory (shutdown not required)
IAEA	Material Unaccounted for (MUF): - 8 kg Pu abruptly in one month - 8 kg Pu protracted in one year	Sigma ID ≤ 2.42 kg Pu	Annual	Monthly

NRC	Standard Error of Inventory Difference (SEID)	Sigma ID ≤0.1% of active inventory	Semi-annual	Monthly
DOE	Limit of Error (LOE)	Sigma ID ≤1% of active inventory of nuclear material	At least annually	Bi-monthly

[a] Based on a July 20, 2006 presentation to the ACNW by Kemal Q. Pasamehmetoglu, DOE, AFCI Fuels Development National Technical Director.

For a plant that processed 1 MTIHM/year, assuming 30% Pu content, the NRC Sigma ID would be 25 g, compared to the IAEA's 2.42 kg. For recycle facilities with the expected throughput to be commercially viable, attaining the NRC Sigma ID is a political, diplomatic and technological challenge.

In 1978, an NRC draft report discussed a then-reference safeguards system intended to meet seven design objectives for fixed sites and three for transport.

For fixed sites the objectives were:

(1) Ensure that only authorized personnel and materials are admitted into material access areas (MAA's) and vital areas (VA's).
(2) Ensure that only authorized activities and conditions occur within protected areas, MAA's and VA's.
(3) Ensure that only authorized movement and placement of SSNM occur within MAA's.
(4) Ensure that only authorized and confirmed forms and amounts of SSNM are removed from MAA's.
(5) Ensure timely detection of unauthorized entry into protected areas.
(6) Ensure that the response to any unauthorized activity is timely, effective, and appropriate to the particular contingency.
(7) Ensure the presence of all SSNM in the plant by location and quantity.

For transport the three design objectives were:

(1) Restrict access to and personnel activity in the vicinity of transports.
(2) Prevent unauthorized entry into transports or unauthorized removal of SSNM from transports.
(3) Ensure that the response to any unauthorized attempt to enter vehicles and remove materials is timely, effective, and appropriate for the particular contingency."

It was the belief at the time that the referenced system, based upon supporting studies and analyses, "was consistent with the safeguards requirements imposed by 10 CFR Parts 70 and 73." A generally conservative approach was adopted to further increase confidence in that finding.

Five alternative safeguards options were also examined, and, in general, these measures offered the potential for improving overall safeguards performance or reducing the overall societal impacts attributable to safeguards. None significantly outweighed any other in general efficacy.

It should be noted that the NRC's safeguards program for commercial licensees is part of a national safeguards structure introduced initially to protect defense-related SSNM. That structure included three primary components: 1) intelligence gathering, 2) fixed site and transportation security, and 3) recovery of lost material. Only the second component, fixed site and transportation security, which involves physical security and material control, would fall primarily within NRC's field of responsibility, The other two, intelligence and recovery operations, would remain the responsibility of other agencies such as the FBI, the National Security Council, DOE and State and local law enforcement agencies. NRC collaborates with these other agencies in developing contingency plans for reacting to and dealing with theft or diversion, but does not participate in intelligence operations or physically take part in recovery operations.

V. OVERVIEW OF ADVANCE SPENT NUCLEAR FUEL RECYCLE INITIATIVES

The National Energy Policy (NEP) [NEP, 2001] issued by the President Bush in May, 2001 recommended expanded use of nuclear energy in the U.S., including development of advanced nuclear fuel cycles, reprocessing and fuel treatment technologies. Consistent with the President's Policy, DOE adopted an "integrated strategy" with four initiatives: Nuclear Power 2010 program; the Advanced Fuel Cycle Initiative (AFCI); the Generation IV Nuclear Energy Systems initiative; and the Nuclear Hydrogen initiative. Additionally, on February 6, 2006, the Secretary of Energy launched GNEP, a comprehensive international strategy to expand the safe use of nuclear power around the world.

A. Advanced Fuel Cycle Initiative (AFCI)

The purpose of the DOE AFCI program is to develop fuel systems and enabling fuel cycle technologies for GEN IV reactors and future reactors. DOE anticipates that AFCI will provide options for the management of spent nuclear fuel through treatment and transmutation of radionuclides that will reduce the cost, hazards and volume of HLW disposal in repositories, reduce the amount of plutonium accumulating in the nuclear fuel cycle, and recover for beneficial use the energy potential remaining in spent fuel. DOE plans call for systems analysis to be an important part of the ongoing AFCI program and have an increased role during the next few years. The planned systems analysis will investigate key issues such as the required rate of introduction of advanced burner reactors (ABRs) and actinide separations facilities to avoid the need for a second HLW repository early in this century, and a detailed study of the technical requirements for the facilities and how the facilities might support the top level goals of the program. DOE plans to use the results of these analyses to establish the basis for each key decision in the AFCI program and for GNEP program planning.

AFCI is organized into the following program elements:

- Separations
- Fuels
- Transmutation
- University Programs

The purpose of each element is summarized below.

1. Separations

Separation processes will be devised to recover plutonium in a way such that it is never separated from at least some transuranic actinides and possibly some fission products. Essentially all of the transuranic elements in addition to the ^{137}Cs and ^{90}Sr will also be removed from the waste going to the geologic repository, thus, by reducing the heat load in the repository, greatly increasing the number of fuel assemblies whose wastes go to the repository, and consequently obviating the need for additional repository space for many decades. These separation technologies are not alternatives to a geologic repository but could help reduce the cost and optimize the use of a geologic repository.

2. Fuels

Fuel forms for advanced fast-spectrum reactors (ABRs) that will transmute transuranic actinides (i.e., Np, Pu, Am, and Cm) to fission products are being developed. Oxide, nitride, carbide, and metallic fuels are being considered. The AFCI is also developing fuels for GEN-IV power reactors.

3. Transmutation

Transmutation is a process by which long-lived radioactive isotopes, especially actinides such as plutonium and neptunium, but also selected fission products such as ^{99}Tc and ^{129}I, are converted to shorter-lived fission products or stable isotopes by fission and/or neutron capture from neutrons generated in a reactor or by the interaction of high-energy ions from a particle accelerator with a metal target such as mercury, tungsten, or bismuth. Theoretically, the preferred neutron source to fission actinides is one of high average neutron energy (yielding a high neutron fission-to-capture ratio), high flux (to which the transmutation rate is proportional), and large core volume (to accommodate more actinides). This has lead to a preference for fast reactors as the neutron source. Transmutation of fission products is usually more efficient in the low-energy neutron spectrum typical of thermal reactors such as LWRs, but DOE is currently focusing on actinide transmutation and, thus, on development of fast reactors with the lead candidate being a sodium-cooled reactor with stainless-steel-clad fuel.

4. University Programs

The goal of the AFCI University Programs element is to foster education of the next generation of scientists and engineers who will support the growth of nuclear power. This goal is to be achieved primarily by funding infrastructure upgrades at universities and education and research.

a. University Nuclear Infrastructure (UNI)

This program brings together several program elements supporting the increasingly vital university nuclear engineering infrastructure. Program elements include:

Innovations in Nuclear Infrastructure and Education (INIE)

This program strengthens the Nation's university nuclear engineering education programs through innovative use of the university research and training reactors and encouraging strategic partnerships among the universities, the DOE national laboratories, and U.S. industry. Currently there are 6 university consortia providing support for 38 universities in 26 states under the INIE program.

Reactor Fuel Assistance

The DOE provides fresh fuel to, and takes back spent fuel from, university research reactors. There are currently 27 operating university research reactors at 26 institutions in the United States. Many of these facilities have permanent fuel cores and therefore do not require regular fuel shipments. However, DOE supplies approximately a dozen universities with fresh fuel and shipments of spent fuel as needed. These reactors are unique assets for technical education and are used for a variety of research, educational and training purposes. Currently, a program is underway cooperating with the National Nuclear Security Administration to convert the reactors with highly enriched uranium to low enriched uranium. This activity will continue to be funded in FY 2007 under the Research Reactor Infrastructure program in the Radiological Facilities Management budget.

Reactor Upgrades

The DOE provides assistance to universities to improve the operational and experimental capabilities of their research reactors. Grants are provided to the universities to purchase equipment and services necessary to upgrade the reactor facilities, such as reactor instrumentation and control equipment, data recording devices, radiation, security, and air monitoring equipment, and gamma spectroscopy hardware and software. Each year up to 26 universities are eligible to receive this assistance. In October 2006, the Department issued a Solicitation for the FY 2007 Reactor Upgrades program.

Reactor Sharing

Through this assistance effort, the Department enables universities with reactors to "share" access to their facilities with students and faculty at other institutions who lack such a facility. The reactors are made available for use in research, experiments, material irradiations, neutron activation analysis and training, and for facility tours and other educational activities.

b. Nuclear Engineering Education Research (NEER) Grants

This highly competitive peer-reviewed program provides grants to nuclear engineering faculty and students for innovative research in nuclear engineering and related areas. The awards run from one to three years and are granted in nine separate technical areas related to nuclear engineering: reactor physics, reactor engineering, reactor materials research, radiological engineering, radioactive waste management, applied radiation science, nuclear safety and risk analysis, innovative technologies, and health physics.

c. Other University Support Activities

DOE/Industry Matching Grants

The Department of Energy and participating companies provide matching funds of up to $60,000 each to universities for use in funding scholarships, improving nuclear engineering and science curricula and modernizing experimental and instructional facilities. Typically 20-25 universities receive funding each year with approximately 35 private sponsors participating.

Nuclear Engineering/Health Physics Fellowships and Scholarships to Nuclear Science and Engineering Programs at Universities are provided.

Radiochemistry

DOE awards three-year grants to support education activities in the field of radiochemistry in the United States. Radiochemistry is linked to several national priorities including medicine, energy and national defense.

Nuclear Engineering and Science Education Recruitment Program

This program is designed to increase the number of students entering a university nuclear engineering course of study by developing a core curriculum to instruct high school science teachers in nuclear science and engineering topics through the use of teaching modules, teacher workshops and other outreach activities.

Summer Internships at National Laboratories

The Office of Nuclear Energy offers summer internships in technical areas related to nuclear engineering to undergraduate and graduate students at the Idaho National Laboratory, the Argonne National Laboratory, and the Oak Ridge National Laboratory . Each student works with a mentor and receives living expenses and a stipend for the 10-12 week program.

International Student Exchange Program (ISEP)

The ISEP sponsors U.S. students studying nuclear engineering for 3-4 months abroad doing research at nuclear facilities in Germany, France, and Japan. These three countries send their students to the U.S. for reciprocal internships at DOE national laboratories.

B. Global Nuclear Energy Partnership

GNEP is a broadly scoped DOE program the goal of which is promoting beneficial international uses of nuclear energy through a multi-faceted approach. Many of the ideas explored earlier by INFCE are embodied in GNEP, which is essentially an updated expression and extension of those ideas.

DOE has entered a Notice of Intent (NOI) to Prepare a Programmatic Environmental Impact Statement (PEIS) for the Global Nuclear Partnership in the Federal Register [DOE, 2007]. This NOI gives a detailed statement of what is intended to be provided in the GNEP PEIS as well as considerable information on what DOE's concept of what GNEP itself is.

1. GNEP Goals

GNEP is still embryonic and is changing as new information, new international alliances, and new program leadership come into being. The general goals of GNEP as expressed by DOE in its strategic plan [GNEP, 2007] are as follows:

> "The United States will build the Global Nuclear Energy Partnership to work with other nations to develop and deploy advanced nuclear recycling and reactor technologies. This initiative will help provide reliable, emission-free energy with less of the waste burden of older technologies and without making available separated plutonium that could be used by rogue states or terrorists for nuclear weapons. These new technologies will make possible a dramatic expansion of safe, clean nuclear energy to help meet the growing global energy demand."

DOE plans three facilities to implement GNEP:

(1) an industrial-scale nuclear fuel recycling center (Consolidated Fuel Treatment Center, CFTC) to separate the components of spent fuel required by GNEP

(2) an reactor (Advanced Burner Reactor, ABR) to fission the actinides yielding fission products that are more readily managed while producing electricity

(3) an advanced fuel cycle research facility (Advanced Fuel Cycle Facility, AFCF) to serve as an R&D center of excellence for developing transmutation fuels and improving fuel cycle technology.

The pursuit of these three facilities has two complementary components. The CFTC and the ABR would be led by industry with technology support from laboratories, international partners, and universities. The AFCF would be located at a government site and research therein would be funded by DOE and led by the national laboratories. The two components would work closely together to move GNEP forward by integrating research results into the industrial-scale activities.

DOE expects that the components of GNEP would provide the following benefits:

- Expand domestic use of nuclear power and reduce dependence on fossil fuels;
- Demonstrate more proliferation-resistant fuel recycle processes;
- Minimize high-heat-output nuclear waste and thus obviate the need for additional U.S. geologic repositories before 2100;
- Develop and demonstrate advanced burner reactors (ABRs) to produce energy from recycled fuel;
- Establish reliable fuel services to participating nations by providing fuel on a lease-and-return basis;
- Demonstrate small-scale reactors;
- Develop enhanced nuclear safeguards by designing safeguards directly into nuclear facilities and reactors and by enhancing IAEA safeguards capabilities.

2. GNEP Timetable – phased approach

On August 3, 2006 DOE announced $20 Million for GNEP siting studies and sought further cooperation with industry through issuance of a request for Expressions of Interest (EOI) in licensing and building a CFTC and a ABR.

The GNEP program is phased. Each phase would begin after a decision based on the results of the previous phase and an assessment of the risks associated with proceeding to the next phase. DOE has stated that it will proceed to detailed design and construction of one of the GNEP facilities after the it is confident that the cost and schedules are understood and after the project management framework that will allow these projects to succeed was in place. DOE plans a major decision on the future of GNEP in June, 2008 and has not been willing to specify potential schedules beyond this decision.

It is anticipated that NRC will regulate CFTC and ABR. The AFCF will be built on a DOE site and is not be expected to be licensed by the NRC.

C. Russian “Equivalent” Proposal (Global Nuclear Infrastructure - GNI)

Russian President Putin put forward in 2006 a broad non-proliferation initiative called the Global Nuclear Infrastructure (GNI) envisioning the establishment of International Nuclear Centers, and offered to host the first such center in Russia. The proposed Centers would provide participating nations with full “nuclear fuel cycle services,” including enriching uranium, fabricating fresh nuclear fuel, and storing and reprocessing spent nuclear fuel.

In states not possessing nuclear weapons, uranium enrichment and spent fuel reprocessing are activities permitted under the terms of the Nuclear Non-Proliferation Treaty (NPT), but they are considered to pose significant proliferation risks because they can provide access to weapons-usable nuclear material. The Russian Nuclear Center proposal would concentrate such activities in states already possessing nuclear weapons and limit the introduction of enrichment and reprocessing facilities in non-nuclear weapons states.

Russia has stated that it would be ready to set up a pilot international enrichment center by the end of 2006. This center would provide non-weapons nuclear power states with assured supplies of low-enriched uranium for power reactors, giving them equity in the project, but without allowing them access to the enrichment technology. The existing uranium enrichment plant at Angarsk - the smallest of three Siberian plants - will feed the international center which will be under IAEA supervision. The material will be under safeguards. Russian legislation is needed to separate the facility from the defense sector and open it to international inspection, as well as to provide for a shareholding structure for other countries involved with the center.

GNI will be the first expression of President Putin's initiative which is in line with IAEA's 2003 proposal for Multilateral Approaches to the Nuclear Fuel Cycle (MNA). GNEP proposals involving such centers are very similar [WNA, 2006], and collaboration with the Russian initiative is anticipated.

D. Generation IV Nuclear Reactors

The Generation IV International Forum (GIF), was chartered in May 2001 to lead the collaborative efforts of the world's leading nuclear technology nations to develop next generation nuclear energy systems (reactors) to meet the world's future energy needs. This international effort reached a major milestone on February 28, 2005, as five of the forum's member countries signed the world's first agreement aimed at the international development of advanced nuclear energy systems.

Five distinctly different reactor systems were identified for development. Initial emphasis was to be placed on those reactors whose next generation would be evolutionary improvements of PWRs and BWRs rather than radical departures from existing technology. All five of the reactors have had either substantial operating experience (PWR, BWR, Sodium Fast Reactors, and HTGR) or extensive research and development (MSR). Following is a list of advanced reactors with their expected principle uses:

GFR: Gas-cooled fast reactor – electricity production and actinide management (perhaps hydrogen production)

LFR: Lead-cooled fast reactor – electricity production, hydrogen production, and actinide management; good proliferation resistance

SFR: Sodium-cooled fast reactor – electricity production and actinide management

MSR: Molten salt reactor – electricity production and waste burn-down

SCWR: Supercritical-water-cooled reactor – electricity production and actinide management

VHTR (HTGR): Very-high-temperature reactor – hydrogen production and other process heat applications; electricity production

E. Nuclear Power 2010

The technology focus of the Nuclear Power 2010 program is on Generation III+ advanced light water reactor designs which offer advancements in safety and economics over the Generation III designs certified by the Nuclear Regulatory Commission (NRC) in the 1990's. To enable the deployment of new Generation III+ nuclear power plants in the United States in the relatively near-term, it is essential to complete the first-of-a-kind Generation III+ reactor technology development and to demonstrate the use of untested Federal regulatory and licensing processes for the siting, construction, and operation of new nuclear plants. DOE has initiated cooperative projects with industry to obtain NRC approval of three sites for construction of new nuclear power plants under the Early Site Permit (ESP) process, to develop application preparation guidance for the combined Construction and Operating License (COL) and to resolve generic

COL regulatory issues, to obtain NRC approval of COL applications. The COL process is a "one-step" licensing process by which nuclear plant public health and safety concerns are resolved prior to commencement of construction, and NRC approves and issues a license to build and operate a new nuclear power plant.

Although DOE is supporting industrial development of improved and advanced reactor designs there will be few if any new reactor construction starts before 2010. However, there has been a substantial number of operating license renewal applications. As of 2005 the NRC had received license renewal applications for 48 reactor units and had approved 20-year license extensions for 30 reactor units. Utilities have indicated they intend to apply for new reactor construction licenses in the near future.

VI. ADVANCED FUEL REPROCESSING TECHNOLOGY

In the early years of reprocessing in the U.S. the goal was to separate pure plutonium with optimized recovery of ^{239}Pu for use in nuclear weapons. Irradiations in the plutonium production reactors at Hanford and Savannah at low power densities and short irradiation times were carried out to minimize generation of undesirable higher mass number plutonium isotopes. As interest in commercial power-producing reactors grew, the emphasis changed from weapons plutonium production operating conditions to higher burnups and power densities of the fuel to maximize energy production and minimize cost. This emphasis led to a smaller proportion of weapons-grade ^{239}Pu and larger percentages of ^{240}Pu, ^{241}Pu, and ^{242}Pu in the spent fuel.

The present goal of reprocessing development includes a great interest in proliferation-resistant processes. One approach to this goal is to eliminate altogether the separation of a pure plutonium product. This type of process is known generically as co-extraction, or COEX. The U.S. UREX flowsheets, as well as the French GANEX flowsheet, are COEX processes designed to keep the plutonium with other actinides or with both actinides and fission products. Other important goals of present-day reprocessing methods and plants include minimizing the volume of radioactive waste, decreasing consumption and losses of fissile and fertile elements to waste, most notably Pu and U, and minimizing heat-producing radionuclides in the HLW (^{137}Cs and ^{90}Sr in the relatively short term and the actinides, primarily Pu, Np, Am and Cm, in the long term).

Figure //A1// in Appendix A shows the relative contributions of fission products and actinides to the heat produced by PWR fuel irradiated to 51 GWd/MTIHM as a function of time out of the reactor. Upon examination of radionuclide contributions it is apparent that removing actinides, most notably ^{241}Am and Pu isotopes, greatly reduces the heat production in the waste from spent fuel reprocessing. This fact is the impetus for actinide removal in the UREX processes.

It is important to know the efficiencies of the separations processes used in the flowsheets. This information is obtained as nearly as possible through laboratory experiments with non-radioactive materials, followed by experiments with radioactive tracers, then with small amounts of irradiated fuel, and finally by engineering-scale experiments with full-scale irradiated fuel. At the same time the various pieces of process equipment are tested individually and then as integrated systems to ensure process goals will be met. Data from the laboratory and engineering scale-up tests are used in equipment design and selection for pilot plant recycle facilities.

Out of these tests come data on separation factors[22], which are a measure of separations efficiencies for the suite of elements of interest. Besides uranium and plutonium, it is very important to have separations data on Cs, Sr, Tc, I, Np, Am, Cm and the lanthanide elements. The importance of this data derives from the fact that the extent of separation determines the distribution of these radionuclides among the products and waste streams and thus determines the need for additional cleanup or helps define disposal routes. Radionuclides that may be important in the future include tritium, ^{85}Kr and ^{14}C.

Because the power densities and fuel burnups of commercial power reactors have been increasing steadily as more and more reliable information on reactor and fuel performance has become available, and because the half lives of the radioisotopes cover an enormous range, it is very important to know how much of each of them is produced and how long they are permitted to decay before reprocessing. It is also essential to specify the process or processes to be used and the degree of separation needed for each of them from the others.

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[22] Separation factor is defined as the concentration of the species of interest in the feed to one stage of the separation process divided by its concentration in the product of that stage of the separation process.

All of this information goes into establishing mass balance and equipment flowsheets. With such a wide range of variables (fuel burnup, reactor power level, radioactive decay, a very large number of radionuclides, and degree of separation sought for individual radioisotopes or groups of radioisotopes, equipment options) the number of possible flowsheets becomes very large. Considerations such as degree of separations sought, process simplicity, ease of process operation, cost, volume of wastes generated, safety, regulations, and proliferation resistance of the processes are helpful in winnowing down the processes that are actually worthy of study and adoption.

The PUREX process can be modified to make the first step of any of the UREX (URanium EXtraction) processes (five variants have been considered in GNEP) that have the potential to be used to recycle spent nuclear reactor fuel, increase proliferation resistance of the separation process, and save space inside high-level nuclear waste disposal sites, e.g., the proposed Yucca Mountain site, by separating the relatively short-lived heat-producing ^{137}Cs and ^{90}Sr and the much longer-lived heat producing actinide elements from the remaining fission products, thus permitting closer spacing of waste packages.

A. UREX Processes

GNEP has conceived of a suite of UREX processes, each of which consists of a series of steps designed to remove successively specific groups of radionuclides to tailor products and compositions of desired product and waste streams. The first step in all the UREX processes is a modified PUREX process. This is followed by processes to remove major heat-producing radionuclides from wastes going to the repository and to aggregate transuranic actinides for recycle. In particular the UREX +1a process produces fissile material process streams that contain separated uranium in one stream and all the other actinides in another. The other actinides are to be fabricated into “target” rods for transmutation in an ABR.

One objective of this approach is to increase the proliferation resistance of fuel recycle by avoiding the production of a pure plutonium stream and to fission plutonium and the other actinides so that they produce energy and are not disposed of as a heat generating waste. A second objective is to remove the major sources of decay heat that would affect a geologic repository. Figure //A2// in Appendix A shows that the heat production rate of the actinides exceeds that of the fission products in the long term, and after about 400 years the predominance of the heat is produced by the actinides, illustrating the advantage of their removal from the waste sent to the repository. Because of the long half- lives of the actinides and their concomitant lower specific radioactivity, they are not the primary contributors to radioactivity (and thus to heat) for the first sixty or so years. This illustrates the potential advantage of keeping the lanthanides with the actinides from the point of view of proliferation resistance. Any potential disadvantages subsequently arising during irradiation in an ABR would need to be evaluated, but they are not expected to be large.

The UREX processes combine the modified PUREX process flowsheet with variants of solvent extraction processes that have many features in common. The PUREX process step is modified from the conventional PUREX process to prevent the plutonium from being extracted with the uranium in the first UREX process step. This can be done by adding a plutonium reductant to the first extraction cycle. This prevents the extraction of plutonium with the uranium by chemically reducing extractable Pu(IV) (using, for example, acetohydroxamic acid) to in-extractable Pu(III), thus providing somewhat greater proliferation resistance than the PUREX process, wherein the plutonium is extracted with the uranium and subsequently separated from uranium and further purified. *It should be observed that a relatively simple change in the first UREX process step [failure to add the Pu(IV) reductant] would result in co-extraction of U and Pu, which would be* *essentially the PUREX process.*

Several UREX process modifications have been considered. These are presented in Table //14//.

Table //14// Variants of the UREX Process

Process	Prod # 1	Prod # 2	Prod # 3	Prod # 4	Prod # 5	Prod # 6	Prod # 7
Urex +1	U	Tc	Cs/Sr	TRU+Ln	FP except Cs, Sr, Tc, Ln		
Urex+1a	U	Tc	Cs/Sr	TRU	FP except Cs, Sr, Tc		
Urex+2	U	Tc	Cs/Sr	Pu+Np	Am+Cm +Ln	FP except Cs, Sr, Tc, Ln	
Urex+3	U	Tc	Cs/Sr	Pu+Np	Am+Cm	FP except Cs, Sr, Tc	
Urex+4	U	Tc	Cs/Sr	Pu+Np	Am	Cm	FP except Cs, Sr, Tc

NOTES: TRU = Transuranic elements: Np, Pu, Am, Cm, Bk, Cf
FP = Fission products
Ln = Lanthanide fission products: elements 58 (cerium) through 71 (lutetium)

Of the processes DOE is supporting or planning to support detailed studies and laboratory- and engineering-scale evaluation on the UREX +1a process. This process separates the very long-lived fission product ^{99}Tc ($t_{1/2}$ = 2.12E05 yr), whose most common chemical species is the environmentally mobile pertechnetate anion (TcO_4^-), and the relatively short-lived, high-heat-producing fission products ^{137}Cs ($t_{1/2}$ = 30 yr) and ^{90}Sr ($t_{1/2}$ = 28 yr) from the high-level waste. In addition, the UREX +1a process puts all the actinides and some low-enriched uranium (LEU) into a single product stream for ultimate recycle to an ABR where they can be fissioned to produce energy and what is primarily a fission product waste, thus removing by transmutation the principal long-term heat producing actinides from the wastes.

Planning, experimentation, and evaluation of the UREX +1a process is in an early stage of development (as of early 2007). Some experiments with irradiated fuel have been carried out, but there have been no engineering-scale demonstrations. The difficulties associated with combining and operating continuously and in sequence the four distinctly different solvent extraction separations processes inherent in UREX +1a in one facility have not yet been addressed. These difficulties will pose serious operational problems on the commercial plant scale. This is in fact true of all the UREX processes, all of which require a reprocessing plant with multiple processes operating sequentially, using differing extractants, different equipment, different solvent cleanup and recycle processes, and producing different types of wastes and intermediate and final products. Such a plant would require extensive and expensive operator training, a very complex plant, and diverse equipment types. It would have the additional disadvantage that if one of the separations process steps became inoperable, in the absence of substantial inter-process surge capacity the entire plant would be shut down because the individual processes must operate sequentially.

Flowsheet and process development is underway at ANL, INEEL, SRS and ORNL both on the benchtop and in hot cells at the kilogram scale to establish the viability of the various separations processes. This work, especially integrated process operation in the hot cells, is very important for establishing the feasibility of the UREX +1a process.

UREX +3 is worthy of note because it separates Am and Cm as a product stream. This feature may be of importance, depending on how the ~55,000 tonnes of long-cooled spent fuel currently in storage at the reactor sites or spent fuel storage sites is phased into the reprocessing plant processing schedule along with the 2200 tonnes of spent fuel being generated annually from the existing 103 commercial power reactors (plus the fuel from any new reactors that come on line). Because of radioactive decay and their nuclear properties the Am and Cm from spent fuel aged 35 to 40 years is more efficiently burned in LWRs than in fast reactors [ORNL 2007], a fact that has the potential to reduce the number of or to eliminate the need for modular fast burner reactors currently planned for transmutation of actinides to fission products.

Figure //12// is a block diagram of the UREX +1a process which shows the process steps as the Head End, Central, and Tail End unit operations. The head- end operations include chopping the fuel elements into small pieces, fuel dissolution, and feed clarification to provide the input stream (H-5) to the central UREX +1a process. The head-end will likely also include trapping and immobilizing the gases ^{85}Kr, ^{129}I, ^{14}CO$_2$ and ^{3}H. In addition, the hardware and hulls are shown to be compacted and packaged for disposal. These head-end steps are for the most part current reprocessing practice.

Fig. //12// Diagram of Primary UREX +1a Process Unit Operations

The central unit operation steps are discussed in detail below and provide the various streams for separation and intermediate storage: U and Tc are separated in the UREX process step; the uranyl nitrate product stream undergoes denitration and solidification and packaging for storage; the Tc that reaches the Tc ion exchange step (some is lost in the dissolver) is converted to metal for disposal, presumably with the fuel cladding hulls; ^{137}Cs and ^{90}Sr are separated in the CCD-PEG process and stored as glass-bonded aluminosilicates after immobilization by steam reforming; the remaining fission products other than the lanthanides are separated by the TRUEX process, calcined, and sent to long term high-level waste storage; the TRU elements are separated from the lanthanides in the TALSPEAK process and blended with uranium for calcination, packaging and storage as the target material for the ABRs. The lanthanides are calcined and sent to long term high-level storage.

Figures //13// and //14// show more details on the four major processes in this flowsheet.

Fig. //13// Diagram of UREX +1a Steps 1 and 2

Fig. //14// UREX +1a Steps 3 and 4

1. Description of the UREX +1a Flowsheet

The purpose of this UREX +1a flowsheet discussion is to elaborate on the process steps, culminating in a description of the products, wastes, and separation efficiencies in the process steps. Major intermediate process stream compositions, recycle streams, and waste streams, and their purity, and impurities are presented and discussed insofar as they are available with the present state of knowledge of the separations (February 2007) and are in the publicly available literature.

The head-end and tail-end operations are for the most part based on well established technology. Although each of the four central solvent extraction separations steps has been studied separately experimentally and some have reached advanced stages of development, very little data on the efficiency and operability of the integrated separations is available. It must be borne in mind that except for the UREX separation step for uranium and technetium, whose features parallel the PUREX process, little or no large-scale operating experience is available in the U.S on the various steps of the UREX processes.

a. Head End

A representative reprocessing plant dissolver feed for a typical PWR fuel assembly is given in Table //15//.

Table //15// Typical PWR Assembly Composition

Fuel Assembly Component	Mass, kg
Fuel material	
Uranium (expressed as elemental U)	461.4
Uranium (expressed as the dioxide)	523.4
Hardware	
Zircaloy-4 (cladding, guide tubes)	108.4
Stainless steel 304 (end fittings)	17.1
Stainless steel 302 (plenum springs)	21.9
Inconel-718 (grid spacers)	5.9
Nicrobraze 50 (brazing alloy)	1.2
HARDWARE TOTAL:	154.5
FUEL ASSEMBLY TOTAL:	677.9

Note the large amount of hardware that must be disposed of as radioactive waste. In conventional reprocessing of spent LWR fuel in the head end step the spent fuel is removed from the storage area and segmented prior to dissolving it in nitric acid in the head-end hot cell. The assembly may be broken down into individual fuel elements or sheared as a whole. Segmentation is typically done with a remotely operable shear that cuts the spent fuel elements or assemblies into pieces one-to-two inches long. This permits ready access of the nitric acid dissolvent to the oxide fuel pellets.

During fuel segmentation and dissolution, gases or volatile fission products trapped in the fuel or present in the plenum space at the ends of the fuel elements are released into the hot cell off-gas system. The most important off-gas species from the point of view of waste management are ^{129}I, ^{85}Kr, ^{3}H, $^{14}CO_2$. The Zircaloy cladding hulls will contain an appreciable fraction of the tritium (as much as 41 %) as zirconium hydride. Some volatile RuO_4 may also be present in the off-gas.

Although its curie content in spent fuel irradiated to 33 GWd/te and cooled 25 years is small (^{99}Ru: 1.851E-02 Ci/ MTIHM), its mass is not negligible (8.691E+02 g/MTIHM), and it deserves attention because it may interfere with recovering the important off-gas species noted above.

Up to the present time only iodine has been trapped to remove it from the off-gas to prevent it from reaching the biosphere. Many of the candidate processes for trapping the other volatile fission products are in an early stage of engineering development and demonstration, with the exception of iodine trapping processes. Iodine trapping methods include scrubbing the dissolver off-gas in KOH solution, or concentrated nitric acid and mercuric nitrate solution, or trapping on solid sorbents, principally those coated with silver nitrate with which iodine reacts to form highly insoluble AgI or $AgIO_3$. Sorption on charcoal has been used, but charcoal has significant drawbacks primarily due to its flammability. Only the very long-lived ^{129}I iodine isotope ($t_{1/2}$ = 1.57E+07 yrs) is of consequence in spent fuel reprocessing because the other iodine isotopes are either very short-lived (^{131}I: $t_{1/2}$ = 8.02 days) or are present in very low yield. A small fraction of the iodine may remain in the dissolver solids as AgI and PdI_2. It may be put into solution and subsequently into the off-gas by the addition of KIO_3 to the dissolver, but this would require another process step.

Krypton removal has been studied using cryogenic distillation, sorption on zeolites and charcoal, and selective sorption in various liquids such as dichlorodifluoromethane (a refrigerant now out of favor because of its effect on the ozone layer). Diffusion through permselective membranes such as silicone rubber is also a candidate for krypton separation. Xe, which has negligible radioactivity in long-cooled fuel, has about 19 times the volume of krypton in the off-gas after 25 years decay. Both are chemically inert, and their physical properties are the basis of their separation from other gases. However, it is possible to separate krypton from xenon and thus reduce the volume of radioactive rare gas stored. It will be necessary to separate air (primarily nitrogen which is the major constituent of the off-gas) from krypton to keep the volume manageable. Because of the differences in molecular weights and atomic dimensions this is a relatively easy separation and could be performed, for example, with molecular sieves. Only the relatively short-lived ^{85}Kr isotope ($t_{1/2}$ = 10.72 yrs) is of consequence in spent fuel reprocessing. In 100 years of storage it will have decayed to a very low level of radioactivity.

Tritium, ($t_{1/2}$= 12.26 yrs) is a rare isotope in the natural environment. About two thirds of the tritium produced in LWR fuels is from ternary fission and one third from neutron activation. During aqueous reprocessing of spent LWR fuel any tritium that has not reacted with oxygen in the fuel or escaped as gas in the head end step can react with water in the dissolver producing tritiated water. A promising method for controlling tritium during fuel reprocessing is voloxidation. (See the short discussion of voloxidation in section IV.C.2 on pyroprocessing.) In voloxidation the tritium is vaporized from the spent fuel by heating in air or oxygen before spent fuel dissolution in acid. The T_2O thus formed may then be trapped in a dessicant such as silica gel or a zeolite. If tritium removal and containment is required for plant licensing, then voloxidation may be the method of choice. If tritium is not removed before acid dissolution of the fuel, then it exchanges with hydrogen in the acid in the dissolver solution to produce tritiated water whose disposal path would be through evaporation. This may not be an acceptable approach. In any case, the relatively short half-life of tritium means that after100 years of storage it will have decayed to a very low level of radioactivity.

Carbon in spent fuel contains ^{14}C ($t_{1/2}$ = 5.73E+03 years) produced from the ^{14}N (n,p)^{14}C reaction with the nitrogen that is always present in the fuel at the 10-60 parts per million level. ^{14}C is produced at a rate of about 10-20 Ci/GWe/year of reactor fuel irradiation [Chopin, 1987]. Its removal is a straightforward operation in principle because the carbon will be present as $^{14}CO_2$, which is readily sorbed in a large number of sorbents such as KOH, CaO and molecular sieves (zeolites).

From the above discussion it is apparent that because of their short half lives neither krypton nor tritium is a long term hazard. Their storage for one hundred years would suffice to remove them from further concern. On the other hand, if capture and storage are imposed requirements for iodine and carbon, they will remain as long-term concerns. A present there are no generally accepted methods for their permanent disposal.

b. Central Unit Operations

In the first step of DOE's UREX +1a process the uranium and technetium in solution[23] are separated by solvent extraction with tributylphosphate from the other actinides, the lanthanides and the fission products. The addition of the reducing agent acetohydroxamic acid (AHA) in the process prevents the extraction of plutonium by reducing it to in-extractable Pu(III). After being stripped into an aqueous stream with nitric acid the uranium is converted to oxide for storage and subsequent use or disposal. (As noted above, by simply omitting the AHA the first UREX +1A step becomes essentially the PUREX process because the uranium and plutonium would be co-extracted in purified form.)

The use of pulse columns for solvent extraction leads to process simplicity and reliability, but centrifugal contactors can process a given amount of spent fuel faster and in a much smaller space at the cost of increased complexity and decreased reliability. Specifically, centrifugal contactors cannot tolerate solids because of the close fit between the rotor and the wall. A small amount of solid noble metals precipitate slowly from the dissolver solution, and these could pose problems in a centrifugal contactor.

The volume of solid waste produced is directly related to the type of reagents used in reprocessing. For example, although the PUREX process has used tri-n-butyl phosphate (TBP) for many years, the TBP cannot be converted entirely to gaseous products because of the presence of the phosphorus atom in the molecule. This leads to an irreducible solid waste.

According to the UREX +1a process the technetium is removed from the acidic uranium product stream using an organic anion exchange resin (Tc is present as the pertechnetate anion). The pertechnetate anion is stripped from the resin and solidified by chemical precipitation as finely divided metal using an alkaline solution of sodium borohydride or by reduction to metal in a furnace. After multiple uses and stripping to remove residual Tc the anion exchange resin is carbonized, packaged, and shipped off-site for disposal. The technetium metal may be converted to a final waste form by combining it with the washed and compacted cladding hulls from the head-end dissolution step. Alternatively, it could be combined with the dissolver heal of noble metals sludge and disposed of with that waste. It is unlikely that all of the technetium will remain with the uranium through the first UREX +1a process step, both because part of the Tc will remain with dissolver solids, and because extraction of Tc into the uranium stream will not be quantitative

The raffinate from process step 1 contains the actinides Pu, Np, Am and Cm as well as the lanthanides, ^{137}Cs, ^{90}Sr, and other fission products. The raffinate becomes the feed to process step 2, the CCD-PEG process [CCD-PEG, 2003]], where the Cs and Sr are separated from the actinides, lanthanides and fission products using a chlorinated cobalt dicarbollide-propylene glycol (CCD-PEG) solvent as extractant. The separated Cs and Sr are to be converted to a stable

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[23] The pertechnetate anion, TcO_4^-, is thought to form an extractable complex with zirconium which upon extraction releases the pertechnetate ion which forms a complex with the uranyl ion (UO_2^{2+}) and remains largely, but not entirely, with the uranium stream.

alumino-silicate waste in a steam reforming process using an incorporated clay such as kaolin to reduce the solubility of the Cs and Sr. It is not clear whether the Cs and Sr will be packaged and stored separately or kept together.

The raffinate from process step 2 becomes the feed to process step 3, the TRUEX process [TRUEX, 1998], where the actinide (TRU) and lanthanide (Ln) elements are extracted from the remaining fission products using TBP- carboxylmethylphosphine oxide (CMPO) in *n*-dodecane extractant.

The raffinate from process step 3 containing the actinides and lanthanides becomes the feed to process step 4 where, after feed adjustment, the actinides are separated from the lanthanides using the TALSPEAK process [TALSPEAK, 1964], [TALSPEAK, 1999]. The TALSPEAK process performs the difficult separation of actinides and lanthanides, whose chemistries are very similar. This solvent extraction separation process is carried out using di-2-ethylhexylphosphoric acid in *n*-dodecane as extractant, with lactic acid and diethylenetriaminepentaacetic acid as wash stream complexants and nitric acid as a stripping agent. Very careful control of pH and careful control of organic-to-aqueous process stream phase ratios is required to effect the desired separation.

The actinides Np, Pu, Am and Cm are in the raffinate stream from the TALSPEAK extraction cycle. They are to be combined with some of the uranium and solidified, packaged, and stored until shipment off-site for subsequent fabrication into a target suitable for irradiation in a suitable reactor. At present the transmutation reactor is assumed to be a sodium-cooled fast reactor. The lanthanides and residual fission products are in the strip stream and are solidified, packaged, and stored until the time of final disposal.

The lanthanides (also called rare earths) are the radionuclides selected by both the UREX processes and the French GANEX [Bouchard, 2005] process for separation from the actinides because of their interference with efficient recycle and reuse of the actinides.

It is noted that a small amount of fluoride (~0.01 M) is used in the dissolution step because after fuel dissolution the acidity is reduced during feed adjustment to the point that fluoride ion is needed to prevent hydrolysis of some of the radionuclides. Although not listed in the flowsheets, fluoride ion appears in the feed and the raffinate streams in all the process steps. Fluoride ion can be of concern for corrosion issues, especially in equipment like the dissolver and the waste vitrifier.

2. Assumptions for modeling the UREX +1a flowsheet

In order to calculate the distribution of radionuclides among the waste and product streams it is necessary to make some assumptions about decontamination factors (DFs) achieved in the process steps. There has been considerable experience in reprocessing, and some DFs are known for common processes like PUREX. However, the major spent fuel reprocessors, e.g., France and the UK, consider the DFs to be proprietary information. There are, however, certain limits on radioisotopes in wastes that are specified in the U.S. in the Code of Federal Regulations, and in the absence of other data, these limits may be used as criteria that must be met, and thus as specifications for the wastes. Additionally, for some of the less common UREX +1a process steps, e.g., CCD-PEG, TRUEX, and TALSPEAK, there are publications that contain information from laboratory experiments or on limited plant experience that may be used to derive DFs. All of these sources of information, along with information from burnup calculations made with ORIGEN2 and the judgment of the authors, was used to obtain the information in Appendix F.

To put the discussion of waste streams on an internally consistent basis it is necessary to make some assumptions about the capacity and mode of operation of the reprocessing plant. There are currently about 55,000 MTIHM of spent fuel in storage from the operation of the 103 commercial nuclear power plants. These plants produce spent fuel at a rate of about 2200 MTIHM/year [ORNL, 2007]. A reprocessing plant (or plants) with a capacity of 2200 MTIHM/year (which is large relative to the capacity of all previous reprocessing plants) will reprocess the backlog of 55,000 MTIHM in 25 years. During that time another 55,000 MTIHM of spent fuel will have accumulated, the oldest fraction of which will have been out of the reactors for 25 years. (It is expected that new power reactors will be coming on line during that 25 years, so it is assumed that the reprocessing capacity will increase to keep pace with the increased production of spent fuel. This can be accommodated by building reprocessing capacity commensurate with the current and expected nuclear power growth.) Therefore, if the fuel is reprocessed on the oldest-first scheme, no fuel will ever be reprocessed that has aged less than 25 years. As a consequence, the ORIGEN2 calculations summarized in this paper assume the fuel has been decayed for 25y before it is reprocessed.

The following streams are important when considering managing the reprocessing plant wastes and products from a UREX +1a plant:

a. Off-gas stream

All plant operating areas have off-gas systems that capture the gases and vapors leaving the area and treat them before they are vented to the atmosphere. In general, air flows from areas of low radioactivity to areas of higher radioactivity, thus minimizing contamination. Each vented radionuclide has a different biological effect on the human body, and this must be taken into consideration when deciding what remediation action to take for that radionuclide. In general the radionuclides in the off-gas must retained at least to the level of retention required by the regulations. These limits and technologies proposed to meet them and to retain the radionuclides for storage and disposal have been discussed [ANL, 1983].

The most important separation process off-gas streams are those from the spent fuel shear and the dissolver. It is these streams that contain the bulk of the radioactive gases and vapors (krypton, iodine and carbon dioxide). It is anticipated that tritium will be removed from the spent fuel and zirconium cladding by a voloxidation step preceding the dissolver step. Other important off-gas streams are those from the waste calcination and vitrification steps, which are not examined in this paper.

b. Technetium stream

Because of the importance of technetium as a long-term contributor to dose if it reaches the biosphere it is important to decide how it should be managed. It is not clear how completely it will be recovered in the UREX processes because it does not extract quantitatively with the uranium in the first process step. Also, as much as 15 % of it may become part of a noble metal (e.g, Pd, Ru, Rh, Pt) sludge in the spent fuel dissolver, in which case that portion will remain with the washed hulls and will need to be managed differently than is shown in the UREX +1a flowsheet. The answer to these questions must await more definitive experimental information obtained from an integrated engineering flowsheet demonstration.

c. Uranium product stream

The uranium product stream produces 2097 tonnes of uranium (as uranyl nitrate) annually from a 2200 MTIHM/year reprocessing plant. Thus there will need to be a substantial uranyl nitrate denitration system to produce uranium oxide associated with the reprocessing plant. Denitration will produce nitrogen oxides which must be recovered to prevent escape of toxic NO_x gases to the atmosphere.

d. Solvent waste streams

There will be enough radioactivity in these wastes streams to require care in their disposal. As noted earlier, each process step has a different solvent, probably requiring a different solvent waste cleanup system for each solvent. As the solvents need to be replaced, solvent waste streams will be produced. It is possible that incineration may be an acceptable means for treatment of most of them because almost all the solvents are organic compounds. UREX and TALSPEAK process steps contain solvents (i.e., TBP and HDEHP) that cannot be completely oxidized to gaseous compounds.

e. Fission products stream

The fission product waste stream, as the term is defined in this paper, contains all the fission products except cesium, strontium, technetium, iodine, krypton, tritium, and carbon. These wastes are primarily the lanthanides and are the remaining wastes to be vitrified, packaged, stored and ultimately sent to a HLW geologic repository. They comprise a significant amount of the stored waste, but are not the major heat producers. It is assumed that vitrification will take place at the reprocessing plant.

f. $^{137}Cs/^{90}Sr$ stream

^{137}Cs and ^{90}Sr pose a special and significant waste management problem. Together they are a major short-term heat producer (see Appendix F and the graph in Appendix A), contributing more heat and more curies than all the remaining fission products combined. ^{137}Cs is a source of penetrating radiation[24] and merits special attention. It is apparent that they comprise a major waste management problem. They are to be fixed in a chemically stable waste form, packaged and stored together and held for up to 300 years for their radioactive decay to innocuous levels.

g. Actinide Stream

The actinides are the principal product of the reprocessing plant as well as being a principle heat source (see Appendix F and the graph in Appendix A.) About 27.7 tonnes per year of actinides from a 2200 MT/yr reprocessing plant (exclusive of any uranium that might be added) will need to be packaged, stored, and ultimately sent to a burner reactor for transmutation to fission products, which themselves will, after pyroprocessing according to the current GNEP plan, be added to the fission products already produced in the original irradiation that produced the spent fuel.

3. Quantitative Discussion of UREX +1a Waste and Product Streams

The bulk of the 55,000 tonnes of spent commercial fuel currently (early 2007) in storage will have aged for more than 25 years and will have burnups of about 33 GWd/te. This burnup will be about the same for the indefinite future. These values for age and burnup were chosen for the calculations to determine the amounts and compositions of the various waste streams.

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[24] Although the ^{137}Cs itself is not an important source of radioactivity (beta rays of <40 Kev), 92 % of it decays to ^{137m}Ba which decays with a half-life of 2.55 minutes; 90% of the ^{137m}Ba decays to yield a 0.662 Mev gamma ray.

In order to calculate the waste amounts and compositions it has been necessary to use values for separations of the various radionuclides in the process steps. In most cases good values from plant operating data are not available. These are usually considered to be proprietary by plant operators, although some data from early reprocessing have been published. There are also some data reported in the early literature and data from laboratory experiments using fully irradiated fuel for processes like CCD-PEG, TRUEX and TALSPEAK. By judicious selection of available data in Appendix F was assembled. Values in Appendix F were used to calculate the waste stream compositions amounts using ORIGEN2. The results of these calculations are given in Table //16//.

Table //16// Compositions and Amounts of Waste Streams
(Based on values in Appendix E and ORIGEN2 calculations)

	AMOUNT PER MTIHM FROM ORIGEN2				**FINAL OUTPUT STREAM CHARACTERISTICS**			
OUTPUT	Grams	Curies @ 25 yrs	TRU α curies @ 25 yrs	Watts @ 25 yrs	Density, g/cc	Grams nuclide/ Gram Waste	Waste vol. L/MTIHM	TRU nCi/g
Volatiles Released								
T	0	0	0	0				
C	0.0134	0.00598	0	0				
Kr	0.7	277	0	2.35				
I	0.9	0.0002	0	0				
Volatiles in Waste								
T[a]	0.0208	201	0	0.00677	2.2	0.079	0	0
C[b]	0.133	0.592	0	0.0002	1.6	0	1.81	0
Kr[c]	4	1570	0	2.35	0.005	0.011	77.4	0
I[d]	177	0.0313	0	0	2.1	0.0414	2.4	0
Cladding + Tc[e]	296000	1020	3.33	4.97	6.8	1	43.5	11300
U Product[f]	953000	8.22	0.00572	0.0876	3.5	1	272	6
TRU Product[g]	12600	44400	3970	222	10.8	1	1.17	3.15e+08
Cs/Sr Waste[h]	5150	154000	0	328	1	0.27	19.1	0
Fission Product waste[i]	19700	42300	1.41	235	2.65	0.38	19.6	71600

a. Tritiated water in polymer-impregnated cement
b. Calcium carbonate in cement
c. Compressed gas in cylinder
d. Grouted silver zeolite
e. Homogeneous alloy of structural material, dissolver solids, Tc, and some undissolved fuel

f. Uranium oxide having concentrations of other radionuclides suitably low to allow re-enrichment
g. Oxides of the various elements sintered to 95% of theoretical density
h. Glass-bonded aluminosilicate made by steam reforming
i. Vitrified into borosilicate glass logs

a. Volatiles in Waste

Although waste disposal forms for tritium, $^{14}CO_2$ and ^{85}Kr are shown here, these radionuclides have not been sequestered previously because no standards have been in place specifying their type of retention. Consequently, these waste forms are the authors best guesses of what they believe may be initially acceptable waste forms. At the very least, they provide some idea of the weights and volumes of the encapsulated wastes.

Because ^{129}I concentrates in the thyroid gland where, in sufficient amount, it may cause serious damage, especially in children, its sequestration has been required from the beginning of reprocessing. Care is required to ensure its complete release into the off-gas during sent fuel dissolution [CEA, 2007]. It is an especially troublesome radionuclide to dispose of as waste because there are few highly stable chemical compounds of it and because of the many valence states in which it can exist. Fixation on silver zeolite was chosen in the resent study because the system is inorganic and therefore less subject to radiation damage than organic materials and because AgI is insoluble under most conditions likely to be found in the environment and is quite stable to relatively high temperatures (It decomposes at its melting point of 552 °C). Because it is a halogen it is corrosive in the elemental form. Care will be required in its packaging and disposal.

b. Cladding + Tc

The cladding and Tc wastes shown in Appendix F may also contain the so-called noble metals Pt, Pd, Rh, Ru and Mo. These noble metals may or may not be combined with the cladding hulls. If they are not removed from the dissolver with the cladding hulls then they will be left in the dissolver and may carried into the UREX process step. Together they present a potential problem in that, being solids, they may cause hot spots in the dissolver and subsequently in the centrifuge used to clarify the feed to the solvent extraction equipment. If they get beyond the feed clarification step they may cause problems in the centrifugal solvent extraction contactors that operate with a very small gap between the rotor and the contactor wall. The noble metal problem is further exacerbated by the fact that small amounts of the noble metals continue to precipitate from the dissolver solution for up to two weeks. Further, if carried into the UREX process step they would add to the radiation damage to the solvent. The amount that slowly precipitates may be as much as 10 % of the amount that remains initially undissolved in the dissolver.

c. Uranium Product

The uranium may go any of several different disposal routes. It could be disposed of as low-level waste; it could be totally recycled by enrichment and fabrication into new power reactor fuel; and part of it could be combined with the actinide stream for use in the burner reactor.

d. TRU Product

The TRU product stream from the TALSPEAK process is destined for transmutation in an ABR. It produces about two-thirds as much heat as the Cs/Sr waste stream and thus requires packaging and storage in a way permitting cooling.

e. Cs/Sr Waste

^{137}Cs is a major-problem fission product. The radioactivity of its short-lived ^{137m}Ba daughter produces an energetic gamma ray that necessitates heavy shielding. While it is true that the relatively short half-life of ^{137}Cs means that it will be substantially gone in several hundred years, it will be a major heat producer for much of its existence because even after decaying for 25 years after removal from the reactor it is present in large amounts, as shown in Appendix F. Consequently, packaging, storing, shielding and cooling will be significant problems for decades. In addition, ^{135}Cs which has a very long half-life (2.3E+06 yrs), is present in amounts comparable to that of ^{137}Cs after 25 years of decay (see Appendix F) so the radioactive Cs package will require indefinitely long confinement. The above comment on heat production holds for ^{90}Sr too in the short term, although its radiation is softer, and there is no other long-lived Sr radionuclide present. The ^{90}Y daughter is quickly in secular equilibrium, and decays with a very short half-life to stable ^{90}Zr. Consequently, there may be merit to packaging the Sr separately from the Cs to reduce the volume of waste held in long-term disposal.

f. Fission Product Waste

The fission product waste, which in the present discussion does not include the gaseous and volatile fission products or the Cs/Sr fission product waste, is destined for vitrification in borosilicate glass and eventual permanent disposal. After 25 years of radioactive decay it produces about 30 % of the heat produced by the sum of it, the TRU products, and Cs/Sr stream. To get an appreciation of the total amount of fission products to be dealt that would be produced by the GNEP proposal it would be necessary to determine the amount of fission products produced by the ABR after the appropriate number of cycles and to add that amount to the amount given in Table 16 above.

4. Potentially Toxic and Reactive Materials

The solvents use in the four UREX +1a process steps are commercially available organic compounds and as such require the same handling procedures in a reprocessing plant as are required for safely handling these somewhat toxic chemicals in industrial operations. None is extraordinarily toxic, but all pose some danger to those who handle them. Other chemicals such as those used in solvent cleanup are inorganic compounds, and safe industrial practice should be observed. Nitric acid in a variety of concentrations is used throughout the process steps and because of its amounts and ubiquity it is probably the most significant toxic chemical. There is a significant potential chemical hazard in the form of “red oil.”

Red oil is a substance formed when an organic solution (in reprocessing the organic solution is typically TBP) comes in contact with concentrated nitric acid (> 10 M) at a temperature above 120 °C. Contributory chemicals can include diluents (e.g, hydrocarbons used to dilute TBP) and/or aqueous phase metal nitrates. Red oil can decompose explosively when its temperature is raised above 130°C. Three red oil explosions have occurred in the United States: one at the Hanford Site in 1953, and two at the Savannah River Site (SRS) in 1953 and 1975. A red oil explosion also occurred in 1993 at the Tomsk-7 site at Seversk, Russia. Equipment capable of producing red oil are evaporators and denitrators.

Controls for prevention or mitigation of a red oil explosion are generally temperature, pressure, mass, and reactant concentrations. Maintaining a temperature of less than 130 °C is generally accepted as a means to prevent red oil explosions. Vessel venting serves to keep pressure from destroying the process vessel in the case of an explosion, while also providing the means for evaporative cooling to keep red oil from reaching the runaway temperature. Mass controls utilize decanters or hydrocyclones to remove organics from feed streams entering process equipment capable of producing red oil. Limiting the total available TBP is another mass control that mitigates the consequence of a red oil explosion by limiting its maximum available explosive energy. Finally, concentration control can be utilized to keep the nitric acid below 10 M. A U.S. government study [DNFSB, 2003] concluded that none of the controls should be used alone; rather, they should be used together to provide effective defense in depth for prevention of a red oil explosion. The operator of French reprocessing plants (AREVA) recently stated [ACNW, 2007] that red oil has not been observed.

B. Pyroprocessing

Pyroprocessing is a generic term for high-temperature reprocessing using pyrometallurgical processes. In the current AFCI plan pyroprocessing would be adapted to reprocessing the actinide product from UREX +1a after it had been fabricated into metallic target fuel elements and irradiated in an ABR.

Pyroprocesses are often carried out in fused salt systems in conjunction with electrometallurgical steps that remove uranium, plutonium, and other elements from the fused salt by electrolytic reduction onto a cathode. This leaves the fission products in the fused salt, and is the pyroprocessing approach planned for GNEP. The fused salt ultimately becomes a process waste. Because most of the actinides will have been transmuted in an ABR the wastes consist primarily of ^{137}Cs/^{90}Sr, the lanthanides, and the remainder of the fission products.

Pyroprocesses are not currently in significant use worldwide, but a great deal of research and development has been carried out on them worldwide. An electrometallurgical spent fuel process has been studied and developed extensively at Argonne National Laboratory (ANL) for many years, and a fairly large-scale demonstration was carried out successfully at DOE's Idaho Falls facility using Experimental Breeder Reactor (EBR-II) spent fuel.

The principles behind the electrometallurgical process are well understood, and no apparent insurmountable technical barriers exist to their wider adoption [NAS, 2000]. An important obstacle to widespread adoption of pyroprocessing is that reprocessing is currently being carried out worldwide using aqueous processes and a very large experience base exists in large, well-established PUREX process plants. Consequently, there has been little demand for the relatively unproven pyrometallurgical systems, although a demand could develop if the Gen IV reactor development programs and the related AFCI become realities.

The feed to the (ANL) process was originally intended to be metallic spent fuel, and the process lends itself best to reprocessing this type of fuel. It is possible , however, to reprocess oxide fuels by first converting them to metal through a head end step that reduces the oxide to metal. This reduction is best accomplished using finely divided oxide, which can be prepared using voloxidation[25] to pulverize the oxide fuel. Process modifications are possible that separate uranium, plutonium and other actinides from the remainder of the radionuclides. Figures //15// and //16// are representations of two versions of the ANL pyroprocess. Note that in Figure //15// the cathodic product is uranium, whereas in Figure //16// the product is uranium plus the bulk of the actinides. Alterations in the details of operation of the system provide the capabilities to

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[25] Voloxidation is a process studied years ago at ORNL. As noted above, it is the basis of the DUPIC process being developed cooperatively by the S. Koreans and Canadians. Heating UO_2 in air or oxygen converts it to U_3O_8 which causes a phase change and consequent pulverization. These enhances release of volatile fission products.

tailor the product streams to the desired compositions.

Fig. //15// Schematic diagram of pyroprocessing with uranium recovery

Fig. //16// Pictorial representation of pyroprocessing operations

Pyroprocessing bears no resemblance to the more conventional aqueous processes such as PUREX . Pyroprocessing is inherently a batch process. The size of the batches is limited by the nature of the electrorefiner, although it is possible to incorporate more than one electrorefiner in a single high-temperature unit and thus increase the throughput of the system. However, in general, multiple electrorefiners would be required for commercial-scale reprocessing. After repeated batch processes the salt accumulates impurities and must be discarded.

There are two major waste streams from this process: 1) the cladding hulls and the noble metal fission products that accumulate in the electrorefiner, and 2) small amounts of residual TRU elements and fission products. The cladding hulls are a relatively low activity solid waste. The noble metal fission products occur mostly as a metallic sludge, and could present a number of problems, both from the point of view of handling them and because they could cause electrical short-circuiting problems. The residual TRU elements and fission products are sorbed on a glass-bonded zeolite (GBZ).

C. Reprocessing HTGR Fuels

High-Temperature Gas-Cooled Reactor (HTGR) fuels are distinctly different from other reactor fuels. This difference imposes a very different type of head-end processing. Unlike most other reactors, the HTGR fuel is not a ceramic oxide fuel clad in metal tubing. HTGR fuel is made mostly of graphite, and is in one of two geometric configurations, the spherical (pebble) form, and the prismatic form mentioned above, both of which are unlike any other reactor fuels. (See the discussion of the composition of HTGR fuels in Section IV. B. 2. d.) There has been no commercial reprocessing of HTGR fuels, although development work has been conducted at Oak Ridge National Laboratory and elsewhere. Some of the salient features of HTGR fuel reprocessing are discussed below.

1. Flowsheets

The first step in reprocessing HTGR fuels is removal of the bulk of the graphite, whether it is in the form of balls or prismatic blocks. Several removal approaches have been considered. The balls would be crushed or burned to release the TRISO particles, which contain the fuel material of interest. The crushed material would be sieved to recover the fuel particles and the inert graphite would become a waste stream. The separated fuel particle would then be put through a grinder to break the TRISO coatings and release the tiny fuel kernels that contain the uranium and actinides and fission products. Finally, the crushed fuel material and any residual graphite would be dissolved in nitric acid preparatory to solvent extraction. Alternatively, the residual graphite could be burned before crushing the fuel. The advantage to early removal of the graphite by crushing or burning is that it would remove the bulk of the graphite before dissolution in nitric acid. Nitric acid dissolution of finely ground graphite and carbides produces organic compounds that could interfere with the solvent extraction separation step, which is the next step in reprocessing. In any case the fragments of the silicon carbide inner coating would need to be removed before the solvent extraction step because their presence could interfere with the operation of the solvent extraction equipment, especially if centrifugal contactors were used.

For the prismatic fuel blocks it is desirable to separate the coated microspheres from the bulk of the graphite block as a first head-end step. This might be done by burning as described above or reaming the carbonized fuel sticks out of the blocks. In this way the bulk of the graphite could be physically removed, leaving the coated microspheres for treatment as outlined above for the fuel balls. The de-fueled prismatic blocks could then be disposed of in the same way as is done for managing graphite from reactors [IAEA, 2006].

2. Unusual Plant Features

The head-end of the HTGR spent fuel reprocessing plant would have unique features arising from the necessity to crush, grind, or burn the graphite fuels. These steps are to be contrasted with the relatively much simpler fuel shearing employed with LWR fuels. After these head-end steps, the remainder of the plant would be essentially conventional solvent extraction using some version of PUREX, assuming that interference from organic compounds formed by reaction of nitric acid with graphite could be kept acceptably low.

3. Reprocessing Wastes

The bulk of the graphite would become a moderately radioactive waste. The radioactivity would be due primarily to failed fuel particles that could release small amounts of radionuclides into the pebbles or the prismatic blocks but it would also contain amounts of ^{14}C that are large compared to what is in the fuel matrix. In the case where the graphite is burned there would be a CO_2 gaseous waste. Volatile radionuclides would be trapped in the off-gas filters or subsequent trapping systems. A HLW stream would be produced from the solvent extraction process just as in any solvent extraction process. The number and types of wastes from the separations processes would depend on the processes chosen, and on whether the fuel was based on the uranium-plutonium or the uranium-thorium fuel cycle. However, if the present UREX+1a flowsheet were used the wastes should be similar to those from processing LWR fuels with the exception of (a) producing much more ^{14}C in the form of CO_2 or a solid ^{14}C waste form and (b) generating a waste stream of SiC hulls in lieu of metal hardware.

D. French Proposals

The French have been especially active in pursuing a variety of proliferation resistant reprocessing methods [Boullis, 2006] other than PUREX. Areva has developed the GANEX process (Global Actinide Extraction) which co-separates lanthanides and actinides from fission products. It is designed to reduce the radiotoxicity and heat output of final wastes. It is envisaged for possible adoption by the La Hague plant in about 2040. It should be noted that the GANEX process makes no attempt to separate anything but the actinides and lanthanides as a group from most of the uranium. Cesium and strontium remain with the fission products.

In the GANEX process, shown very simplified form in Figure //17// below, uranium is separated in a preliminary step and the raffinate then undergoes three subsequent extractions which result in an actinide stream which is combined with the uranium product from the first step. The lanthanides and other fission products, including cesium, strontium and technetium, are formed into borosilicate glass for long term storage.

The GANEX process has the disadvantage of merging the high heat emitters cesium and strontium with the fission products into the glass for long term storage. The ^{137}Cs is reduced in activity to ten percent of its initial reactor discharge value in 100 years and the ^{90}Sr is reduced to about nine percent, so storage for 100 years before disposal in a geologic repository would greatly alleviate the short-term heat load in the repository, and along with actinide removal would permit substantially increasing the amount of waste stored per unit volume of repository.

Fig. //17// The French Ganex Process [Bouchard, 2005]

VII. ADVANCED FUEL REFABRICATION

Current preparation of conventional pelletized reactor fuels for LWRs and fast reactors requires grinding to achieve specified size and shape. This process produces finely divided fuel particles that must be recovered and recycled. A “dust-free” sol-gel microsphere pelletization process has been developed for fabrication of $(U,Pu)O_2$, $(U,Pu)C$ and $(U,Pu)N$ fuel pellets containing around 15% plutonium [Sol-Gel, 1997]. The microspheres can be pressed into pellets that can be sintered to 85% T.D. in Ar+8% H_2 at 1700 °C. The sintered oxide, monocarbide and mononitride pellets have an open pore microstructure with fine grain size.

Hydrated gel-microspheres of UO_3+PuO_2 and UO_3+PuO_2+C are prepared from nitrate solutions of uranium and plutonium by the “ammonia internal gelation” process, using hexamethylene tetramine that decomposes to ammonia in the presence of silicone oil at 90±1 °C in a gelation bath. For oxide fuel pellets, the hydrated UO_3+PuO_2 gel-microspheres are calcined at around 700 °C in Ar+8% H_2 atmosphere to produce “non-porous”, “free-flowing” and coarse (around 400 micron) microspheres which are directly pelletized at 550 MPa to green pellets. The mixed oxide pellets are subjected either to low temperature (~1100 °C) oxidative sintering in N_2+air containing ~1500 ppm O_2 or to high temperature (~1650 °C) sintering in Ar+8% H_2.

For monocarbide and mononitride pellets, hydrated gel-microspheres of UO_3+PuO_2+C were subjected to carbothermic synthesis in vacuum (~1 Pa) and flowing nitrogen (flow rate: 1.2 m^3/h) in the temperature range of 1450–1550 °C. The microspheres retain their individual identity in the sintered pellets because during sintering densification takes place mainly within and not between the microspheres.

VIII. REGULATION AND LICENSING OF FUEL RECYCLE FACILITIES

A. Licensing – An historical perspective

Application of NRC's regulatory process to commercial recycle facilities will not be simple. Deficiencies in regulations are known to exist that will require new rule making or many exemptions before a license can be approved, for example for reprocessing spent nuclear fuel at a commercial site. The body of regulations that currently establishes NRC's licensing and regulatory process for recycle facilities, associated waste streams, and effluents include at least the following 10 CFR requirements:

Licensing Process – Part 50/52 and Part 70
Radiation Protection – Part 20
Environmental Protection – Part 51
Fuel Fabrication – Part 70
HLW Vitrification and Storage – Part 70
Reprocessed Uranium Storage – Part 70
Low-Level Waste Disposal – Part 61
Disposal of HLW at Yucca Mountain – Part 63
Spent Nuclear Fuel Storage – Part 72
Cs/Sr and TRU Storage – Part 30 and Part 70
Transportation – Part 71
Physical Protection – Part 73
Material Control and Accountability – Part 74
Decommissioning – Part 50 and Part 51

For the purposes of this chapter of the report, spent commercial nuclear fuel reprocessing is the focus.

From an historical perspective, only limited regulatory experience exists, for example, with respect to licensing and regulating reprocessing facilities. Most of this experience occurred decades ago under the AEC and the then newly formed NRC.

1. Licensing experience at Nuclear Fuel Services

In 1966 the U.S. Atomic Energy Commission (AEC) used Part 50 to license Nuclear Fuel Services (NFS) reprocessing facility at West Valley. From 1966 to 1972 NFS reprocessed 640 metric tons of fuel, but in1972 the facility shut down to implement a number of improvements and never restarted. Since that time no other licenses were approved by the NRC for reprocessing spent nuclear fuel, although the BNFP had been undergoing a licensing review when President Carter terminated commercial reprocessing in 1976. Although some 30 years have passed since then, 10 CFR Part 50 still remains the default licensing basis for reprocessing spent nuclear fuel. Many changes have occurred to Part 50 over that time frame, but most relate to licensing utilization or power reactor facilities as opposed to reprocessing facilities like NFS.

2. Licensing experience at Barnwell

The Preliminary Safety Analysis Report for the BNFP Separations Facility was submitted on November 6, 1968. Following appearances before the Advisory Committee on Reactor Safeguards and a public hearing before an Atomic Safety and Licensing Board, a Construction Permit was issued on December 18, 1970. Subsequently, several substantial documents were submitted to the AEC, including: Environmental Report and Facility Safety Evaluation for the Uranium Hexaflouride Facility; Updated Environmental Report for the Separations Facility;

Final Safety Analysis Report for the Separations Facility (five volumes and several addenda); Technical Description in Support of Application for FRSS Operation; Preliminary Safety Analysis Report for Plutonium Product Facility; and Nuclear Materials Safeguards Supplement. In addition, the applicant submitted a large number of documents containing responses to questions by the AEC.

A public hearing was conducted before an Atomic Safety and Licensing Board to comply with the National Environmental Policy Act and preliminary to the issuance of an Operating License. The compliance with applicable regulations and the commitment implicit in issuing the Construction Permit were verified by the AEC who conducted more than 20 formal inspections prior to the cessation of commercial licensing activities (brought about by Presidents Carter and Ford.).

Some facilities and operations of the plant complied with 10 CFR Part 50 regulations, while other facilities such as the fuel fabrication plants complied with 10 CFR Part 70. In the past, the licensing process was complicated by the evolutionary character of regulations pertaining to reprocessing plants and waste management, and the inter-relation between the licensing of the facility and other regulatory actions taking place concurrently. Notable among those were the proceedings on the Generic Environmental Statement on Mixed Oxide Fuel (GESMO) and the Environmental Statement on the Management of Commercial High-Level and Transuranic Contaminated Radioactive Waste. These latter activities, however, were placed on hold when the International Nuclear Fuel Cycle Evaluation (INFCE) was invoked.

B. Current licensing process and alternatives

As it stands today, both production (reprocessing) and utilization facilities (power reactors) must comply with 10 CFR Part 50 to obtain a construction or operating license. There may be better options available given the appropriate time and resources. Five options including modifying Part 50 are described below, including their Pros and Cons.

1. Modify 10 CFR Part 50, *Domestic Licensing of Production and Utilization Facilities*, and 10 CFR Part 52, *Early Site Permits; Standard Design Certifications; and Combined Licenses for Nuclear Power Plants*

This approach would modify the general design criteria (GDCs) in Part 50 to accommodate technological differences between light-water reactors and reprocessing facilities. To ensure adequate protection of public health and safety, applicants must demonstrate that their designs meet the modified criteria, and a set of postulated accidents known as “design basis” accidents (DBAs) to within certain specified radiological release limits. A Probabilistic Risk Assessments (PRAs) would be developed and used to complement the accident selection process.

Pros: This approach provides an established licensing process that has been proven to be technically sound, risk-informed and performance based. Part 50 offers a structured process that is defensible based on previous experience. Non-technical process experience and lessons learned from licensing utilization facilities could be applied to production facilities. A one-step licensing process under Part 52 could also be modified to expedite the Part 50 licensing process for production facilities.

Cons: Part 50 was not written specifically for reprocessing spent nuclear fuel, and has some deficiencies for that use. For example, in Classification and Description of Licenses, §50.20, there is no specific acknowledgment of licensing reprocessing plants, and §50.34 (a) *Contents of applications; technical information* is directed solely to reactors. The NEPA process and required documentation for a such a facility has yet to be completely demonstrated. Earlier efforts in this regard for commercial reprocessing plants were subsequent to the submission of the safety analysis report and thus were very time consuming and contentious. The way Part 50 is written today, modification of or exemption to its requirements would need to be granted to accommodate the technical differences between licensing light-water reactors and reprocessing facilities. Modification could be extensive and public hearings on exemptions are likely to drag the process out.

2. Use 10 CFR Part 70, *Domestic Licensing of Special Nuclear Materials*

All fuel fabrication facilities are licensed under 10 CFR Part 70 Subpart H *Additional Requirements for Certain Licensees Authorized to Possess a Critical Mass of Special Nuclear Materials*. This regulation utilizes integrated safety analysis (ISA), sometimes known as a process hazards analysis, to assess the safety of the design and to identify the equipment relied on for safety. The rule could be expanded to encompass reprocessing facilities.

Pros: Part 70 would not need to be substantially revised to accommodate recycle facilities. Experience and lessons learned from licensing fuel fabrication facilities under Part 70 could be applied. Experience with application of Part 70 to licensing fuel fabrication facilities has been successful. Use of ISA is also an important step towards risk quantification.

Cons: Only limited deterministic criteria in the form of defense-in-depth exists in Part 70. Safety is ensured by relying extensively on ISA results. In a letter to the Commission dated January 14, 2002, the Joint Subcommittee of the Advisory Committee on Reactor Safeguards and the Advisory Committee on Nuclear Waste challenged the adoption of ISA methods in the regulatory process. Significant limitations were noted regarding its treatment of dependent failures, human reliability, treatment of uncertainties, and aggregation of event sequences. Such limitations can compromise the ability to defend regulatory decisions based on ISA results.

3. License under 10 CFR Part 53, (Risk-Informed, Performance-Based Framework)

Part 53 is expected to provide a risk-informed, performance based framework for licensing the next generation of reactor designs. The framework integrates safety, security, and emergency preparedness to establish a comprehensive set of requirements as the license condition. The approach focuses on the most risk-significant aspects of plant operations, and uses the Commission's safety goals (separate goals would need to be developed for recycle facilities) as top level regulatory criteria that designers must meet to ensure adequate safety. The approach eliminates the need for exemptions by implementing guidance to accommodate technological differences between designs.

Pros: The approach is advantageous because of its flexibility. Risk-informed technology neutral framework would be technology specific in its application and not require exemptions as would application of other rules developed for other technologies. Integration of safety and security on a common ground avoids tradeoff later in the process to accommodate specific aspects from one or the other.

Cons: It is primarily intended for new commercial power reactors. Safety goals would need to be developed for recycle facilities which could involve a long and tedious developmental process. Very little PRA experience exists for recycle facilities from which to build acceptance criteria. The current schedule for licensing recycle facilities may not support this approach.

4. Develop a new rule 10 CFR Part XX

A new rule could be designed specifically for licensing recycle facilities. The rule would avoid the need to modify existing regulations, and eliminate the need to write exemptions for rules already in place.

Pros: Would place all the regulations relevant to the recycle facilities under one part of the regulations, effectively leaving other parts of the regulations unchanged . The rule could be made to expedite the licensing processing by eliminating exemptions and long drawn-out hearings.

Cons: Extensive resources and time may be needed to develop a new rule. The rule may not be ready in time to process a license application.

5. Commission Order

Under this option, the Commission would direct the staff to develop a licensing basis document with technical criteria upon which the Commission would base a licensing decision. {How long before the LA submittal would the LBD need to be available?} The Commission would formally notice the receipt of the license application and offer stakeholders an opportunity to review and comment on the licensing basis document prior to its decision. That decision would determine whether to issue an Order to allow the facility to operate. This approach is likely to be considered only if time and resources do not allow for other alternatives.

Pros: Most expeditious approach, reduces the time and resources otherwise required for rule making.

Cons: Approach is outside the normal licensing process, and may undermine public confidence.

Whatever licensing approach is chosen, it is expected that implementation will be consistent with Commission policies including the Commission’s Probabilistic Risk Assessment (PRA) Policy Statement (60 FR 42622). This Policy Statement states in part: “The use of PRA technology should be increased in all regulatory matters to the extent supported by the state of the art in PRA methods and data, and in a matter that complements the NRC’s deterministic approach and supports the NRC’s traditional defense-in-depth philosophy.” The ACNW has gone on record repeatedly in letters to the NRC Commission about the use of risk-informed decision making, starting in October of 1997 and most recently in a May 2, 2006 letter. These letters are listed in Appendix C. Additionally, ALARA regulation for reprocessing facilities that establish design objectives and limiting conditions for radioactive material effluents, analogous to the current Appendix I for LWRs will need to be formulated.

C. Environmental Protection

Production and utilization facilities need to comply with environmental protection regulations. Protection is provided by both (1) designed-in barriers that block the release of radioactive material to the environment, and (2) operational performance and characteristics that limit the release of radioactive material to the environment.

Design Perspective

Prior to facility construction,10 CFR Part 51*, Environmental Protection Regulations for Domestic Licensing and Related Regulatory Functions,* requires that each applicant submit an environmental report that complies with Table S-3 §51.50 (Table of Uranium Fuel Cycle Environmental Data) as the basis for evaluating the contribution of its activity to the environment. Currently, Table S-3 only considers two fuel cycles, uranium-only recycle and no recycle. To accommodate other fuel cycles, for example, the UREX processes being proposed by GNEP, Table S-3 will need to be reconsidered to determine if it encompasses the releases of radioactivity to the environment from the facilities. Likewise, Table S-4 (Environmental Impact of Transportation of Fuel and Waste to and From One Light-Water-Cooled Nuclear Power Reactor) will also need to be reconsidered to determine if it encompasses the environmental impact of transportation of fuel and radioactive waste, taking into consideration conditions associated with recycle of spent nuclear fuel constituents and management of associated wastes.

Following the receipt of the environmental report, it will be necessary to prepare an environmental impact statement (EIS) for siting, construction and operation of the recycle plant(s). This requirement can be very time consuming and contentious and must be started well in advance of planned plant construction. The design of a spent fuel reprocessing plant, for example, is dictated to a large extent by the requirements to (a) protect the plant operators from radiation, to provide a safe working environment, and to prevent criticality and (b) limit routine and accidental releases of radionuclides to the public. These requirements necessitate remote operations of plant equipment and processes and impose limitations on both the size and location of process equipment.

The design must be such that the protection provided by the radiation shielding and confinement of radioactivity keep radiation doses as low as reasonably achievable (ALARA) and are consistent with the allowable limits of personnel dose and air and water contamination specified in the Code of Federal Regulations, Title 10, Part 20 (10 CFR Part 50) for occupational dose limits. Exceptions to the dose limits may be made in the case of Planned Special Exposures, but in any case the ALARA principle applies.

Operating Perspective

In 1974, the AEC initiated a study to examine the physical and social environmental impact from spent nuclear fuel reprocessing and widespread use of mixed-oxide fuel (MOX) fuel as a means to use reprocessed uranium and plutonium. The study, often referred to as GESMO, was published in August 1976 [NRC, 1976]. Five alternatives were considered:

- Prompt fuel reprocessing, prompt uranium recycle, delayed plutonium recycle
- Delayed fuel reprocessing, followed by uranium and plutonium recycle
- Prompt uranium and plutonium recycle
- Uranium recycle; no plutonium recycle
- No uranium or plutonium recycle

Findings from the study found no clear preference for any of the alternatives. Differences in health effects between the fuel cycles did not provide a basis for choosing one approach over the another. Any environmental benefit that did result arose from the conservation of uranium resources and not from differences in the way radiological waste was managed.

The study found, however, that for the various recycle alternatives (as opposed to the once-through fuel cycle) the collective dose increased by several orders of magnitude. Three gaseous effluents during operations were the source of this dose: ^{3}H, ^{14}C, and ^{85}Kr. Although the dose to any one individual was found to be small, the large integrated (world) population exposed to the gaseous effluents drove the results. While public hearings were being held on the GESMO study and BNFP license, the Carter Administration terminated reprocessing in the U.S. The public hearings were never completed, and the Commission postponed their decision on the widespread use of mixed oxide fuel and commercial reprocessing. This could become an issue once again should wide-scale reprocessing be considered as a mainstream activity.

About the same time period as the GESMO study (Jan 13, 1977), EPA released 40 CFR 190 *Environmental Radiation Protection Standards For Nuclear Power Operations, Subpart B* . This rule established the environmental standard for the complete (cradle to grave) uranium fuel cycle. Two criteria were prescribed:

Subpart B section 190.10 (a) - Annual dose equivalent to any member of the public for the entire fuel cycle,

Whole body	< 25 mrem
Thyroid	< 75 mrem
Any other organ	< 25 mrem

Subpart B section 190.10 (b) - Radioactive material released to the environment per gigawatt-year electrical energy produced,

^{85}Kr	< 50,000 curies
^{129}I	< 5 millicuries
Pu + TRU isotopes with half-lives greater than 1 year	<0.5 millicuries

Soon after the standards were released, stakeholders expressed concerns that the standards were overly conservative, costly, and that they would require technology that was considered to be beyond the state-of-the-art. In addition, industry believed that requirements should not be established until international agreements were reached that would restrict emissions from foreign sources. Nevertheless, EPA approved part of the standard (except for ^{85}Kr) on December 1, 1979, and a ^{85}Kr standard that was effective on January 1, 1983. By that time all reprocessing activities had ceased and interest in the new standard declined..

Today, the EPA standard for utilization (power reactors) facilities is being met through NRC's enforcement of 10 CFR 50, Appendix I, Numerical Guides for Design Objectives and Limiting Conditions for Operation to Meet the Criterion "As Low as Reasonably Achievable" for Radioactive Material in Light-Water-Cooled Nuclear Power Reactor Effluents . Part 50 Appendix I sets the following operating limits:

Liquid Effluents	< 3 mrem whole body, or < 10 mrem to any organ
Gaseous Effluents	< 5 mrem whole body, or < 15 mrem to the skin
Radioactive iodine and other material in	< 15 mrem to any organ

particulate form in
effluents to the atmosphere

These limits provide the basis for realistic implementation of EPA standards for single reactor units. The remaining (non-utilization) portion of 40 CFR 190 release is divided among the rest of the fuel cycle which up to now has not included reprocessing.

D. Decommissioning

Decommissioning commercial reprocessing plants can be very costly. There is not much information available based on decommissioning experience because so few reprocessing plants have been decommissioned.

In 1976 NFS withdrew from the reprocessing business and turned control over to the site owner, the New York State Energy Research and Development Authority (NYSERDA). In 1980, the NRC suspended West Valley's license to reprocess spent nuclear fuel, and the West Valley Demonstration Project (WVDP) Act was executed to clean up the site and its facilities. Under the Act, NRC maintained certain responsibilities including prescribing decontamination and decommissioning criteria.

Significant lessons learned and regulatory actions have resulted from West Valley decommissioning experience. The cost to clean up the site to date has exceeded $2 billion, although a fund of only $4 million had been set aside for decommissioning and decontamination. The English Sellafield reprocessing plant is currently undergoing decommissioning of their "first generation" reprocessing plants, including analytical laboratories, legacy wastes, and the "North Compound," a facility established to support Windscale pile operation and subsequently extended to include waste storage. The total estimated cost of this "interim" decommissioning is about £20 million [Sellafield, 2005]. The French UP1 reprocessing plant at Marcoule has an estimated decommissioning cost of EUR 5.6 billion, about half of which is for treating wastes stored on site [Hore-Lacey, 2007]. Thus, there is very wide range of real and anticipated reprocessing plant decommissioning costs. Additional actual decommissioning experience will be needed to better define the real costs for future reprocessing plant decommissioning.

Rule making to avoid future situations like those at West Valley resulted in 10 CFR Part 50 App. F *Policy Relating to the Siting of Fuel Reprocessing Plants and Related Waste Management Facilities*, Some sections of 10 CFR Part 50, appendix F, relevant to reprocessing plants are:

- Paragraph 3, which states "Disposal of high-level radioactive fission product waste material will not be permitted on any land other than that owned and controlled by the Federal Government."

- Paragraph 2, which states "...High level liquid radioactive wastes shall be converted to a dry solid as required to comply with this inventory limitation, and placed in a sealed container prior to transfer to a Federal repository in a shipping cask meeting the requirements of 10 CFR 71...Upon receipt, the Federal Repository will assume permanent custody of the waste materials although industry will pay the Federal Government a charge which together with interest on unexpended balances will be designed to defray all costs of disposal and perpetual surveillance."

- Paragraph 5, which states, "Applicants proposing to operate fuel reprocessing plants, in submitting information concerning financial qualifications as required

> by Section 50.33(f), shall include information enabling the Commission to determine whether the applicant is financially qualified, among other things, to provide for the removal and disposal of radioactive wastes, during operation and upon decommissioning of the facility.

Although Part 50, Appendix F might reduce the likelihood of future West Valley type issues, the Commission in an SRM dated February 7, 2006 [NRC, 2006c], directed that an important design criterion for any new reprocessing effort will be that decommissioning costs be manageable. NRC guidance under development should help designers address these concerns at the conceptual design stage.

Additional Nuclear Regulatory Commission regulatory requirements related to decommissioning include: 10CFR Part 20, Subpart E, Regulatory Criteria for License Termination, which gives Radiological criteria for unrestricted use (§20.1402), Criteria for license termination under restricted conditions (§20.1403), Alternate criteria for license termination (§20.1404), Public notification and public participation (§20.1405), and Minimization of contamination (§20.1406); 10CFR Part 72, Subpart B, §72.30, Financial assurance and recordkeeping for decommissioning; and 10 CFR Part 72, Subpart D, §72.54, Expiration and termination of licenses and decommissioning of sites and separate buildings or outdoor areas.

IX. ISSUES ASSOCIATED WITH LICENSING AND REGULATING FUEL RECYCLE FACILITIES

The focus of this chapter is on licensing and regulation of industrial-scale fuel fabrication and reprocessing facilities. In consideration of the foregoing information there are a number of licensing or regulatory issues that are likely to warrant consideration early in the licensing process. The following sections identify these issues and provides insight into their resolution.

A. Selection or development of licensing regulation(s) for recycle facilities

The most important issue facing the NRC is what regulatory framework(s) and regulation(s) should be used to license recycle facilities. For the purposes of this paper it is assumed that the framework and specific regulations used to license familiar fuel cycle facilities and operations (e.g., interim storage of spent fuel, radioactive material transportation, reactors) will not change. However, a comprehensive review of all fuel cycle facility licensing regulations would be worthwhile for the purpose of determining whether broader changes are needed or desirable to promote consistency or risk-informed, performance-based regulations.

The list of novel facilities for which decisions concerning the appropriate licensing framework and regulations will be required could include those for:

- Reprocessing fuels from LWRs and later for other advanced reactors
- Fabrication of fuels to recycle transuranic or fission product elements, or for some new reactor designs (e.g., graphite-moderated reactors)
- Disposal of new types of wastes such as cladding and transuranic (GTCC) waste,
- Extended interim storage of intermediate-lived radionuclides, Cs ans Sr, followed by *in situ* disposal.

The following sections discuss some of the factors that should be considered when making these decisions.

1. Multiple regulatory paths are available

As was discussed in Sect. IV, there are a number of existing regulations as well as the possibility of developing one or more entirely new regulations for licensing recycle facilities. None of the existing regulations is entirely suitable for the fuel recycle facilities. While detailed reasons for this conclusion are given in Sect. IV, the overarching physical reason is that existing regulations were designed for (a) reactors where maintaining heat removal capability is an important purpose of the regulations but where there are modest chemical hazards and few radionuclides in fluids, or (b) for facilities that handle relatively small amounts of radioactivity because they only process uranium. Maintaining heat removal capability is not particularly important in fuel recycle facilities, but there are substantial amounts of radioactivity in fluids and a higher likelihood of inadvertent criticality, in addition to a variety of toxic and potentially flammable or reactive chemicals in routine use. These differences lead to the need for substantial modification of regulations directed at specific types of facilities or major enhancement of “technology neutral” regulations to address the specifics of fuel recycle facilities. One or more new regulations could be developed to license fuel recycle facilities, but they would likely require an effort commensurate with that required for existing regulations.

2. Important factors in deciding on a regulatory approach

When deciding which existing regulation(s) or criteria to use for a new regulation there a number of factors that should be considered as follows:

- Probabilistic vs. Deterministic: Most regulations and license applications for fuel cycle facilities are based on deterministic criteria. In-plant safety issues are typically addressed using an Integrated Safety Analysis (ISA) approach common in the chemical industry and doses to the public are estimated using a scenario-based approach. The primary reason for this is the consequences of accidents in or routine releases from fuel cycle facilities is relatively small compared to consequences of accidents at reactors and the effort of doing probabilistic analyses has not been justified. However, the complexity and radionuclide inventory of the fuel reprocessing and minor actinide fuel fabrication facilities is sufficient that probabilistic approaches should be considered. It should be noted that the ACNW&M has continued to recommend probabilistic approaches.[ACNW, 2006].

- Best estimate vs conservative: A companion issue to that of probabilistic vs. deterministic approaches is whether analyses should be based on data and models that best represent the best estimate of what might really occur with an associated uncertainty analysis to explore the effects of incorrect data or models, or should be based on demonstrably conservative data and models. Most regulations and license applications for fuel cycle facilities have used a conservative, deterministic approach. The ACNW&M has letters on record pointing out problems with using this approach (see Appendix C) with some of the most important being that using very conservative assumptions can mask risk-significant items, and that most such analyses are not accompanied by a robust uncertainty analysis.

  In at least one recent instance DOE has used a dose assessment based on best estimates [DOE 2005]. This, when accompanied by a robust sensitivity and uncertainty analysis might be appropriate for less complex fuel cycle facilities. While a probabilistic analysis based on conservative data and models could be performed, there is no evident benefit to doing so and the conservatism would render the accompanying uncertainty analysis meaningless.

- Risk-informed, Performance-Based[32]: A risk-informed regulatory approach is one in which risk provides an important insight for licensing a facility but where other considerations such as cost, environmental impacts, etc. can be balanced against the required extent of risk reduction. The ALARA philosophy epitomizes a risk-informed approach. The ACNW&M has consistently advocated risk-informed regulations and licensing approaches for a wide range of situations and the opportunities for focusing scarce resources on the most-risk significant items in very complex facilities would indicate its appropriateness in this instance. It is prudent for regulations for licensing fuel recycle facilities to include provisions that allow the regulator to make exceptions on a case-by-case basis.

  A corollary factor to a regulation being risk-based is that it is performance-based. That is, the criteria for granting a license are expressed in terms of the requirements the applicant must meet but not the means by which the applicant meets the requirement. For example, a regulation that requires that a dose limit be met is performance based but one that requires use of a specific technology is not.

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[32] NRC Commission defined risk-informed regulation in their white paper, "Risk-Informed and Performance-Based Regulation" as “...a philosophy whereby risk insights are considered together with other factors to establish requirements that better focus licensee and regulatory attention on design and operational issues commensurate with their importance to public health and safety.”

- Single or multiple licensing approaches: The unique facilities that might be part of fuel recycle have a wide range of characteristics. Some are so different from what has been licensed in the last few decades (e.g., reprocessing) that substantial new regulatory language in an existing or new regulation would appear to be required. Others have many similarities with previously licensed facilities for which there are existing regulations. Examples might be the similarity between a potential engineered facility to store radiocesium and radiostrontium until it decays to innocuous levels, and interim storage facilities for spent nuclear fuel, and regulations for disposal of LLW even though the form and content of LLW from a fuel recycle facility may be very different from what is presently being generated. These differences and similarities lead to the possibility of a situational approach in which some recycle facilities would be licensed, at least initially, using existing regulations with no more than minor modifications while other facilities would be licensed using new or heavily modified regulations.

3. NRC's Proposed Options for Licensing GNEP (SECY-07-0081)

In SECY-07-0081, dated May 15, 2007, [Regulatory Options for Licensing Facilities Associated with the Global Nuclear Energy Partnership], the NRC staff identified four options for developing a regulatory framework to license advanced reprocessing and burner reactor facilities, which are summarized in Table //17// and recommended a path forward based on these options.

Table //17// Regulatory Options for advanced fuel recycle and burner reactor facilities

Option	**CFTC**	**ABR**
1	Revise Part 70 to include spent fuel reprocessing; consider additional safety analysis requirements for a reprocessing facility; and revise Part 50 as appropriate.	Use existing Part 50, with exemptions, as necessary, or a suitably modified and adapted Part 52 process, to address sodium-cooled fast reactor technology.
2	Same as Option 1.	Create a new regulation specific to advanced recycling reactors (Part 5X).
3	Develop a specific GNEP regulation applicable to both fuel reprocessing and recycle reactors (10 CFR Part XX).	
4	• Issue a Federal Register Notice (FRN) in FY 2007 soliciting public and stakeholder input on desirable attributes of the regulatory framework for GNEP, as well as comments on whether there are any major substantive technical issues relating to an accelerated schedule that may affect development of GNEP regulations and/or how such facilities should be regulated. • After consideration of public and stakeholder comments, decide on either issuing an Order or directing a rulemaking to establish specific requirements. • Concurrently, develop a licensing-basis document for fuel separations/fuel fabrication/advanced recycling reactor facilities to be used by the Commission in developing an Order or as the technical basis for the rulemaking process, as appropriate.	

The options are similar to those proposed in Section VII.B of this paper. After evaluating the pros and cons for each of the options, the staff recommended the Commission proceed with Option 1 in a phased approach. The first phase would involve development of the regulatory framework by preparing technical basis documents.to support rulemaking for Part 70 (for fuel recycle facilities) and potential rulemaking for sodium-cooled fast reactor. The first phase would also involve exploration of whether Part 52 could be modified to address sodium-cooled fast reactors and a gap analysis on Part 50 to identify what changes in regulatory requirements would be necessary to license recycle facilities and an advanced burner reactor.

In the second phase the NRC staff would shift to Option 3 and devleop a regulation covering advanced fuel recycle and reactor facilities. The analyses performed in the first phase would be used to evaluate whether ther are unique programmatic or technical interrelationships among all closed fuel cycle technologies as a basis for developing a new regulation for advanced fuel recycle and burner reactor facilities.

B. Impacts on related regulations

In addition to the need to make a decision on the licensing framework for fuel recycle facilities it will be necessary to evaluate the impact that recycle facilities and operations might have on other related regulations that might be invoked in the licensing framework or that might not yet exist and have to be developed. The following sections discuss various features of fuel recycle facilities and operations and how these features might impact other regulations.

1. New radioactive product, effluent, and waste materials

Fuel recycle facilities using any of the UREX process would produce a variety of new radioactive product, effluent, and waste materials. Because these wastes have not been generated by any facilities licensed in the last few decades, the current NRC regulatory system does not have provisions to address them. Examples of new materials are:

- Recovered uranium that will contain small amounts of contaminants such as transuranic actinides (e.g., ^{237}Np) and fission products (e.g., ^{99}Tc) that need to be considered when recycling the uranium to enrichment plants or disposing of it. These contaminants tend to accumulate in certain portions of enrichment equipment and to be concentrated into a waste stream by decontamination operations during maintenance. This requires that enrichment plants have features to (a) process wastes containing transuranic and fission product elements and (b) detect beta-emitting radionuclides and distinguish among alpha-emitting radionuclides.

  The amount of uranium recovered from fuel reprocessing may exceed its demand leading to the potential need to dispose of some of it. Determination of the acceptability of this uranium for near-surface disposal will need to consider the potential risks from species such as ^{237}Np and ^{99}Tc that are often more mobile than uranium under the geohydrological conditions that prevail in the near-surface at many sites.

- A gaseous effluent stream from the fuel reprocessing plant that initially contains most of the intermediate-to-long-lived volatile radionuclides such as ^{129}I, ^{85}Kr, ^{14}C, and ^{3}H in the fuel fed to the plant. Historically and to the present, most (~99%) of the ^{129}I has been removed from the effluent stream and managed as a solid waste before it is released. The other radionuclides have been released to the atmosphere. An existing EPA standard (40 CFR Part 190) and NRC regulation that cites the standard (10 CFR Part 20) require that about 99.5% of the ^{129}I and 85% of the ^{85}Kr be removed from the gaseous effluent before it is released (See Sect. V.C for more discussion of 40 CFR Part 190 and its implications to NRC regulations). However, Table S-3 in 10 CFR Part 51 is based on the assumption that all four volatile radionuclides are quantitatively released to the environment for the purposes of assessing environmental protection. If fuel recycle were to occur inconsistencies such as this would need to be reconciled.

- Implementation of the UREX +1a process would essentially fractionate spent fuel to yield numerous unique solid waste streams. Examples of such wastes are:
  - Spent fuel metal hardware containing small amounts of residual spent fuel, dissolver solids, and ^{99}Tc
  - Wastes containing the four volatile radionuclides mentioned above
  - Wastes containing ^{137}Cs and ^{90}Sr
  - Substantial volumes of materials and equipment contaminated with greater than 100 nCi/g of transuranic radionuclides
  - Surplus uranium product containing trace but potentially significant amounts of fission product and transuranic radionuclides
  - A "HLW" stream containing lanthanides and other fission products that is much less radioactive than what has been produced or planned in the past

Production of these wastes raises a number of issues. First, the appropriate classification of the wastes must be determined. Many of them would probably be classified as GTCC LLW under the present system because the concentration of intermediate (Cs, Sr) and long-lived (iodine, technetium, transuranic radionuclides) radionuclides would exceed the limits for Class C waste in 10 CFR Part 61. However, any uranium product deemed to be a waste and containing less than 100 nCi/g of transuranic radionuclides would be Class C LLW and possibly Class A LLW, and the waste containing ^{85}Kr would be Class A LLW under the present system. Such wastes were not contemplated when 10 CFR Part 61 was developed and the appropriateness of these classifications requires further evaluation.

The estimated volumes, masses, specific radionuclides and their curie amounts of wastes from UREX +1a processing are presented in Appendix F.

A second issue is what constitutes an appropriate waste form for wastes such as the volatile radionuclides, radiocesium, and radiostrontium. Options for the volatile radionuclides were studied in the 1970s and 1980s but development was not completed and a preferred waste form was not selected. Selection of a waste form for ^{85}Kr is particularly challenging because it is a non-reactive gas under all near-ambient conditions. Radiocesium and radiostrontium have been made into chloride and fluoride chemical forms, respectively, and stored by DOE in water pools for decades. However, these do not appear to be appropriate forms for near-surface disposal such as that being suggested by DOE and use of an aluminosilicate waste form has been proposed. 10 CFR Part 61 does not address waste forms or packaging for these materials even if they were to be classified as Class C or less, and there is no regulation addressing the form of GTCC LLW.

A third issue is the absence of a firm technical basis for predicting the distribution of some radionuclides circulating within and being routinely released from recycle facilities. This distribution is necessary for DOE to determine the process routing required by each stream (e.g., does a stream contain iodine that would be released during subsequent high-temperature processing so that the off-gas stream needs to go to iodine recovery). This distribution is also necessary for NRC to determine the adequacy of the design, estimate doses from release of effluents or disposal of wastes, and evaluate the consequences of accidents. Important radionuclides in this regard include:

- Tritium: To what extent is the tritide on cladding surface released during voloxidation, during acid dissolution of the SNF, and during the melting of the fuel assembly hardware to yield DOE's proposed waste form?

- Iodine: Do iodine species form that are not trapped by available technologies and which might exceed the ~0.5% allowable release? What fraction of the iodine is associated with dissolver solids and what fraction is released when the dissolver solids are included in the final waste form that involves high-temperature melting?
- Technetium and neptunium: What fraction of the technetium is associated with the dissolver solids? Of the neptunium and dissolved technetium, a small but potentially significant fraction can be found in various waste streams. What fraction is associated with the various waste streams and products from the reprocessing plant?
- Cladding: How much of the SNF is associated with the cladding? Is the radionuclide distribution the same as the SNF or are some elements preferentially associated with the cladding? This is somewhat important in a waste disposal situation but would be very important if DOE's concept of recycling the cladding becomes reality.

There are also other, more general radionuclide distribution issues including the separation efficiency of the four major separation processes in UREX+1a under large-scale conditions that are elaborated below.

A fourth issue is the appropriate disposal technology for the wastes listed above. For those wastes classified as GTCC the technology and possibly a specific site will apparently be identified as part of DOE's ongoing effort to prepare an EIS on this subject. The GTCC disposal facility will be licensed by the NRC using a regulatory framework that has not been decided. However, it is not evident that the EIS will consider potential GTCC wastes that are unique to recycle such as cladding waste, radiocesium and radiostrontium, and wastes containing ^{99}Tc, ^{129}I, and ^{14}C. Additionally, identification of an appropriate disposal technology (i.e., the acceptability of near-surface disposal) for mobile, intermediate-lived radionuclides such as ^{85}Kr and tritium may depend on the ability of the selected waste form to contain these radionuclides until they decay to innocuous levels.

Another issue is that use of the UREX +1a approach to recycle would change the fundamental nature of a geologic repository for HLW to the point that the term "HLW" is technically misleading[33]. By removing essentially all of the actinides (uranium and heavier), radiocesium, radiostrontium, radiotechnetium, and radioiodine, potentially the cladding, tritium, radiocarbon, and radiokrypton from the repository the result would be a compact waste generating little penetrating radiation, virtually no decay heat, and containing extremely small amounts of the long-lived radionuclides that dominate risks estimated from performance assessments. If some of the long-lived wastes mentioned above (Tc, I, C, cladding, and solid wastes containing some transuranic elements) were to be disposed of in a repository, the waste volume would increase somewhat but would still contain only small amounts of radionuclides important to risk. As a consequence of the foregoing, aspects of regulation that are driven by decay heat, penetrating radiation, the actinides, and degradation rates of the spent fuel cladding and matrix would become irrelevant. On the other hand, the performance of multiple waste forms tailored to specific radioelements over very long time periods would become important. The implications of this to the requirement to predict the performance of the repository to the time of peak dose is unknown.

2. Novel facilities

Fuel recycle using the UREX +1a process would involve a number of facility types that have not been licensed in decades. Regulatory issues concerning most of the major facilities were discussed in Sect. V. A and issues concerning licensing a GTCC disposal facility were discussed

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[33] The NWPA of 1982 amended the definition of HLW from solvent extraction wastes from reprocessing plants to include spent nuclear reactor fuel.

in Sect. V. B. 1 and will not be repeated here. However, DOE is considering a facility that involves a concept that has not been anticipated in existing regulations. Specifically, DOE is considering an engineered near-surface interim storage facility that will store radiocesium and radiostrontium waste forms for about 300 years at which time most of the radionuclides will have decayed to innocuous levels. At that time the facility will become a disposal facility with the waste forms remaining in place. This type of facility is crucial to achieving DOE's goal of greatly increasing the capacity of the repository because it removes a major source of decay heat from the repository. This approach raises interrelated issues of whether a licensable facility containing radionuclides emitting considerable amounts of heat and penetrating radiation can be reliably designed, built, and operated for 300 years; whether such a facility would be suitable for conversion to a permanent disposal facility at that time; and the technology to be used in such a conversion.

Construction and operation of a fuel reprocessing plant before advanced actinide burner reactors are available may result in the need to store significant quantities of transuranic actinide products containing Np, Pu, Am and Cm, possibly mixed with fission products emitting penetrating radiation to provide some degree of self protection until advanced burner reactors become available. Such a scenario would involve novel considerations such as the acceptable form and technology for storing such a product, and how best to safeguard it.

3. Novel process streams and paradigms for safeguards and security

A significant feature of DOE's UREX +1a approach is that fissile material (primarily plutonium) is never completely separated from other radionuclides. In particular, DOE's approach calls for the plutonium to remain mixed with other radionuclides (e.g., lanthanide fission products, some shorter-lived actinides) that impart self-protecting characteristics by releasing penetrating radiation. It is axiomatic that any two substances can be separated with enough effort although the magnitude of the effort can vary from trivial to impractical. Current levels defining what amount of radiation is self-protecting (e.g., 100 R/hr) were conceived with a spent fuel assembly (180 to 500 kg HM of spent fuel) in mind. It is not clear current values are applicable to or even achievable for amounts of plutonium and fission products on the order of 10 kg. The foregoing raises issues such as how much penetrating radiation from what amount of material is enough to be self protecting; how difficult does the separation of plutonium from other radionuclides have to be to be deemed self protecting; and how is the concept of a self-protecting material factored into the safeguards and security paradigms that will be used in the recycle facilities, if at all? These issues remain to be considered in regulations that will be used to support licensing.

4. Evaluation of integrated plant performance

The UREX+1a flowsheet is extraordinarily complex. In essence, this flowsheet includes four inter-connected processes operating in series. Each of these processes is as complex as the traditional PUREX process and some promise to be more difficult to control. Additionally, it is likely to include numerous unit operations beyond what have been included in PUREX plants to recover additional radionuclides from gaseous effluents, treat the many new waste streams mentioned previously, and to recycle various materials to reduce effluents and wastes. These complexities indicate that such a plant is likely to be difficult to operate, requiring extensive and expensive operator training and sophisticated control and monitoring systems. Of more relevance to a regulator is the difficulty and resource requirements to develop the technical capability (expertise, analytical tools) to evaluating whether such a complex system can be safely operated. This task is made even more difficult by factors such as the potential for various minor species unexpectedly appearing in a unit operation because of internal recycle and causing unanticipated hazardous reactions and the ramifications of a unit operations failure and quick shutdown on an entire inter-connected plant.

5. Design and operate with the end in mind

The NRC Commissioners have stated that an important goal in licensing nuclear facilities in general and fuel recycle facilities in particular is to include license conditions to minimize historical problems in decommissioning the facilities at the end of their operating life. This is a relatively new NRC requirement, but one that is very worthwhile. Residual site contamination, waste volumes, environmental problems, cleanup worker health and safety, and cost are all important issues affected by how decommissioning is carried out. How decommissioning is carried out is, in turn, determined by how facilities are designed and operated. So obtaining a license to construct fuel recycle facilities may be contingent upon paying attention to facility design and construction in the context of ultimate facility decommissioning. Specifying facility design considerations will be a delicate issue because the commercial plant designer and the ultimate plant operator will want freedom to build the plant to accomplish the principal plant mission, namely spent fuel recycle.

Consolidated general decommissioning guidance has been provided by the NRC [NUREG, 2007] and a MOU on decommissioning between the EPA and NRC has been signed [MOU, 2002]. Beyond this, the ACNW&M and NRC staff are presently working within their respective mandates to learn obtain ‘lessons learned’ related to decommissioning of fuel recycle facilities. This information will be used as a basis for recommending what additional requirements should be included in existing or new regulations concerning the design, construction, and operation of fuel recycle facilities to facilitate decommissioning and license termination. A separate white paper is being prepared on this issue.

C. NRC Test Facilities

As is evident from the foregoing, recycle facilities that are capable of meeting GNEP goals will involve many processes and pieces of equipment that have never been used at a commercial scale or in a licensed facilities. As a consequence, there is no established basis for assessing the performance and safety implications of these processes and equipment. It can be expected that DOE will base its assessments on information it obtains from lab-scale tests at its national laboratories plus possibly pilot-scale testing. When licensing facilities the NRC normally performs tests to validate key data and assumptions made by a licensee. In the case of recycle facilities, such tests require highly specialized facilities (e.g., hot cells) and equipment that is available only in a limited number of places none of which are part of the NRC community. The lack of NRC testing capability raises the issue of how the NRC will validate key data and assumptions.

D. Operator Licensing Examinations

It will be necessary to create and grade licensing examinations for fuel recycle facility operators at several levels of competence and responsibility. There are several levels of operator training for facilities such as reprocessing plants. In addition there is “cross training” in plant operations for other personnel such as guards and maintenance crews. Experience has shown that it is not easy to train and qualify plant operators. It will be challenging to find people qualified to prepare and administer examinations. The elapsed time since such examinations were administered and the likely requirements for new examination areas such as those related to proliferation prevention and detection and safeguards make this an important area for consideration.

E. Sigma ID Requirements

It is apparent from Table 13 that there are major differences among IAEA, NRC and DOE on the requirements for the permissible significant (SIGMA) Pu inventory differences (ID) with regard to the Sigma ID, and the frequency of both long-term shutdown inventory and interim frequency requirements. This disparity will need to be addressed and resolved to the extent practicable for any recycle facility licensed in the U.S.

F. Timing and urgency

As a practical matter, the number of fuel recycle facilities for which license applications are anticipated and when they are anticipated is an important factor in deciding the regulatory approach to be used. As this paper is written DOE's announced schedule for building recycle facilities goes no further than a major decision in June 2008 on whether and how to proceed based on the contents of a Programmatic Environmental Impact Statement now in preparation. Assuming DOE does decide to proceed with LWR fuel reprocessing in an orderly manner it is reasonable to expect a license application might be received as early as 2010 but more likely a few years later.

Even if DOE were to pursue such an orderly approach, the ~6 years left until the LA would be received to develop a licensing framework is, at best, barely adequate to address the many regulatory challenges described earlier. More likely, if the DOE schedule maintains, the NRC would have to undertake an urgent effort to develop/modify the necessary regulations, especially when it is recognized that some regulatory requirements will drive both R&D activities and design decisions that must precede a license application. It is very easy to state that all the NRC has to do is evaluate, select, and develop a preferred licensing approach for various types of recycle facilities. However, before doing so it must decide a number of policy issues such as whether the approach will be probabilistic, risk-informed, and performance based. Superimposed on this is the need to evaluate and modify the waste classification and disposal system, and the incomplete set of ALARA limits concerning effluent releases. All of this must be done in an environment that is likely to involve contentious litigation at many junctures and where the availability of qualified staff is likely to constrain the rate of progress.

If DOE were to embark on an urgent effort to build such facilities by scheduling a license applications for ~3 years hence the licensing approaches that could be used by the NRC would be severely limited. The time to develop new regulations, and probably to modify existing regulations, to be used for fuel recycle facilities is likely to be substantially greater than the three to four years until 2010 because (a) fuel recycle is a contentious topic, and standard approaches to public involvement (plus potential litigation) will take a substantial amount of time, and (b) many related regulations will require creation or modification. Additionally, external factors that can have a major impact on the NRC's regulations, such as EPA standards on effluent releases and a generic EIS on fuel recycle, are only at their beginning stages, if they are that far.

X. OTHER IMPORTANT ISSUES RELATED TO LICENSING

A. Completion of generic environmental documentation and standards

In the 1970s when nuclear fuel recycle was being aggressively pursued by AEC/ERDA/DOE efforts were undertaken to prepare a generic (programmatic) environmental impact statement (GEIS) on nuclear fuel recycle and by EPA to develop standards for radionuclide releases from recycle facilities. This effort was stimulated by and intertwined with the license application for the Barnwell Nuclear Fuel Plant. Some work continued on both fronts even after President Carter banned nuclear fuel reprocessing in the U.S. and the BNFP license application was withdrawn.

The GEIS and BNFP licensing efforts became the platform for a contentious debate over whether the U.S. should pursue fuel recycle. As discussed in IV. C, the GEIS effort ended with the publication of the GESMO document. The scenarios considered in that document do not conform to the GNEP scenario now being proposed and consquently this document is probably not useful. However, DOE has recently initiated preparation of what is essentially the follow-on to GESMO by issuing a Notice of Intent [DOE, 2007] to prepare a GNEP programmatic EIS.

The EPA undertook an effort to develop environmental radiation protection standards for the nuclear fuel cycle. Briefly, the approach used by the EPA was to assess the ability of existing and developing sequences of processes for removing various radionuclides from effluent streams as expressed in terms of the collective dose reduction that would result from each incremental process. The cost of each incremental processes was evaluated using then-standard cost-benefit techniques. At some point the cost per unit dose reduction ($/man-rem) from the last incremental process was deemed excessive and the extent of radionuclide removal without the last incremental process became the bases for the standard. The cost-benefit analyses were done for all major steps of the nuclear fuel cycle (e.g., uranium mining, reactors, reprocessing) based on technical studies supported by EPA and NRC. The result is codified in 40 CFR Part 190, *Environmental Radiation Protection Standard for Nuclear Power Operations*. Of particular relevance to fuel recycle is 40 CFR 190.10(b) which limits the release of ^{85}Kr and ^{129}I from normal operations of the uranium fuel cycle. Because fuel reprocessing is the only step of the nuclear fuel cycle that could release significant amounts of these radionuclides during normal operations, these limits are effectively release limits for the fuel reprocessing gaseous effluent. This standard was adopted by the NRC in 10 CFR 20.1301(e).

From the perspective of decades of hindsight, 40 CFR Part 190 raises a number of concerns as follows:

- The factors by which ^{85}Kr and ^{129}I must be reduced are approximately 7-fold and 300-fold, respectively. The evaluation which led to these factors was based on effluent control technologies that were under development at the time, but had not been demonstrated or deployed. Because fuel recycle was banned, development was never completed. Thus, meeting the standard with available technologies is probably not feasible.

- Background information accompanying the standard indicated that studies concerning limits on releases of ^{14}C and ^{3}H were underway. These studies remain to be completed and, thus, the standard is incomplete.

- The cost-benefit approach used in the analyses involved calculating the collective dose by integrating very small doses over very large populations and distances, and comparing them to then-common metrics such as a limit of $1000/man-rem to determine whether additional effluent controls were justified. As has been observed in multiple Committee letters and the ICRP, such a comparison is questionable and should not be used in favor of using dose to a maximally exposed individual or critical group.

- The scope of 40 CFR Part 190 does not include fabrication of fuels enriched with plutonium or actinides other than uranium. This addition would presumably be necessary for fuel recycle to proceed and, thus, the standard is not yet complete.

The implication of the above to the NRC is that the EPA standard on which effluent control requirements and other aspects of environmental radiation protection that may impose requirements that are infeasible in the near-term, is incomplete, and is based on analysis techniques that have become questionable over the years. This is a very fragile foundation (if not an inadequate one) for the NRC to develop implementing regulations and begin licensing a fuel recycle facility within a few years. It would appear that interagency discussions to evaluate the adequacy of what exists, what needs to be done, and who will meet the needs should be accorded a high priority.

B. Obtaining adequate numbers of qualified staff

Implementing fuel recycle will require a substantial number of staff knowledgeable about the technical and regulatory aspects of fuel recycle facility design and operation. The design and operation of the fuel reprocessing and recycle fuel fabrication facilities are particularly challenging because staff trained as nuclear chemical operators and engineers is required.

With the decline of work in the civilian nuclear fuel cycle in the 1976 - 1985 time frame to essentially nil and cessation of defense reprocessing activities in the following decade, older workers have moved into other areas or have now retired, and their expertise has not been replaced because there has been little demand. While the nuclear navy continues to offer a good supply of reactor operators, there is no parallel source for nuclear chemical operators, who are usually people that have an associate degree and are then trained on the job. As noted earlier, recycle facilities are very complex and the wash-out rate is high as evidenced by the experience at NFS and BNFP. Similarly, nuclear chemical engineers were historically people having a degree in another technical discipline that obtained graduate degrees in nuclear chemical engineering and then practical experience on the job. Unfortunately, nuclear chemical engineering programs have drastically reduced or eliminated, and the faculty that taught this subject are retired. This same expertise, especially nuclear chemical engineers, will be in demand by organizations performing fuel recycle R&D, designing and operating recycle facilities, and regulating recycle facilities thus exacerbating the demand for this very limited expertise base. The implications of the foregoing for the NRC is that it needs to develop a strategy for acquiring or developing the qualified technical staff it needs to fulfill its mission and to ensure that regulations include adequate provisions to ensure that people designing and operating fuel recycle facilities are qualified.

C. Potential international issues

The GNEP's goals include having once-through and recycle facilities in the U.S. providing services (fuel supply, fuel take-back) as a primary component. The relationship that must be established among the various countries is not yet clear. However, with substantial amounts of U.S. fuel going to many other countries and being returned to the U.S., there is the possibility that a more formal relationship between the NRC and regulators in other countries might be desirable or necessary.

D. Interface between NRC and DOE regulatory authorities

The DOE regulates its activities under its own authority while the NRC regulates licensees doing civilian and commercial nuclear activities. Decisions on whether a particular facility is regulated by DOE and NRC, especially if it is a relatively unique facility, is often done on a case-by-case basis. In the case of the projected fuel recycle facilities there is the potential for a very complex patchwork of regulations, with DOE regulating some facilities that interface with other NRC-regulated facilities (e.g., a fuel reprocessing plant and a recycle fuel fabrication plant). This could pose challenges concerning compatibility and consistency of regulatory requirements, especially as it concerns material that moves between facilities, and how it is moved.

Additional complexities could result if a decision is made to regulate closely coupled portions of a single facility under two regulators. For example, the NRC might license the separations and conversion portions of a reprocessing plant but the DOE might treat and dispose of reprocessing wastes using its facilities and disposal sites. This scenario is occurring at the MFF at SRS but it could be much more complex for a reprocessing plant with its myriad wastes and recycle streams.

Even if initial facilities are regulated under DOE's authority the design and regulation of such facilities provides an excellent opportunity to educate and train NRC staff for licensing subsequent facilities and to obtain insights useful in developing or modifying NRC regulations to license these facilities. Early and continuing interagency discussion of who will regulate which facilities or parts of these facilities and how NRC staff can get involved in some capacity sooner rather than later would appear to be beneficial to both parties.

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APPENDIX A: Decay Heat in Spent Fuel

There are advantages and disadvantages to reprocessing relatively short-cooled spent fuel. Advantages relate to reducing the amount of spent fuel stored. This reduces the need for spent fuel storage facilities and storage casks. It also reduces the potential risk of proliferation or of terrorist attack on the stored spent fuel.

The disadvantages of reprocessing relatively short-cooled spent fuel are related to the necessity to handle more highly radioactive fuel, which increases the potential hazards and adds to the complexity and cost of the reprocessing plant and processes. For example, reprocessing to remove plutonium ^{241}Pu (β decay; $t_{1/2}$ = 13.2 yrs) 4 years after removal of fuel from the reactor, i.e., before it decays extensively to ^{241}Am (α decay; t1/2 = 462 yrs), reduces the heat generation rate in the waste, assuming plutonium is recycled but americium is not. Additional advantages with respect to heat reduction in the waste are achieved as additional actinides and selected fission products are removed prior to storage of the waste. Figure //A1// shows the contributions of selected actinides and fission products to heat generation rate in waste as a function of decay time for fuel irradiated to 51 GWd/MTIHM and shows the advantage of removing the actinides ^{238}Pu and ^{241}Am with respect to decay heat reduction in waste as a function of decay time.

Fig. //A1// Contributions of selected actinides and fission products to heat generation rate
[OECD, 2006]

Fig. //A2// Process Waste Decay Heat without Pu and Am [OECD 2006]

APPENDIX B: Radiotoxicity of Reprocessed Spent PWR Fuel as a Function of Time
(MA: major actinides; FP: fission products; P&T: partitioning and transmutation)

Figure B1 shows the radiotoxicity of spent PWR fuel as a function of time. Radiotoxicity is a measure of the radioactivity of the sent fuel. The figure shows the beneficial effect of successively removing the plutonium and the major actinides and fissioning them to produce fission products. In the idealized case the fission products become the only radioactive material remaining for disposal. According to the diagram, by radioactive decay they reach the toxicity of uranium ore in about 300 year, after which they are less toxic than ore.

Fig. //B1// Effect of Recycling and Transmuting TRU Elements on Radiotoxicity of Waste from Spent Nuclear Fuel [RSC, 2006]

APPENDIX C: ACNW Letters Related to Risk-Informed Activities and Probabilistic Risk Assessment

- ACNW letter dated May 2, 2006, from Michael T. Ryan, Chairman, ACNW to Nils J. Diaz, Chairman USNRC, Subject: Risk-Informed Decision-Making for Nuclear Materials and Wastes.
- ACNW letter dated May 3, 2004 from B. John Garrick, Chairman, ACNW to Nils J. Diaz, Chairman USNRC, Subject: Risk Insights Baseline Report.
- ACNW letter dated August 13, 2003 from B. John Garrick, Chairman, ACNW to Nils J. Diaz, Chairman USNRC, Subject: High Level Waste: Risk-Significance Ranking of Agreements and the Use of Risk Information to Resolve Issues.
- ACNW letter dated July 2, 2002 from George M. Hornberger, Chairman, ACNW to Richard A. Meserve, Chairman USNRC, Subject: The High-Level Program Risk Insights Initiative.
- ACNW letter dated April 29, 2002 from George M. Hornberger, Chairman, ACNW to William D. Travers, Executive Director for Operations, USNRC, Subject: Response to Letter Dated March 6, 2002 Concerning Risk-Informed Activities in the Office of Nuclear Material Safety and Safeguards.
- ACNW letter dated January 14, 2002 from George M. Hornberger, Chairman, ACNW to Richard A. Meserve, Chairman USNRC, Subject: Risk-Informed Activities in the Office of Nuclear Material Safety and Safeguard
    - ACNW letter dated June 29, 2001 from B. John Garrick, Chairman, ACNW to Richard A. Meserve, Chairman USNRC, Subject: Risk-Informed, Performance-Based Regulation of Waste Management and Decommissioning.
    - ACNW letter dated July 27, 2000 from B. John Garrick, Chairman, ACNW to Richard A. Meserve, Chairman USNRC, Subject: Development of Risk-Informed Regulation in the Office of Nuclear Material Safety and Safeguards.
    - ACNW letter dated March 26, 1998 from B. John Garrick, Chairman, ACNW to Shirley Ann Jackson, Chairman USNRC, Subject: Risk-Informed, Performance-Based Regulation in Nuclear Waste Management.
    - ACNW letter dated October 31, 1997 from B. John Garrick, Chairman, ACNW to Shirley Ann Jackson, Chairman USNRC, Subject: Application of Probabilistic Risk Assessment Methods to Performance Assessment in the NRC High-Level Waste Program.

APPENDIX D: 10 CFR 55.31. Requirements for manipulating controls of licensed facility

(10 CFR Part 55 as originally written was not intended to apply to reprocessing plants.)

"(a) The applicant shall:
(1) Complete NRC form 398, "Personal Qualification Statement – Licensee," which can be obtained by writing the Office of Information Services, U.S. Nuclear Regulatory Commission, Washington, D.C. 20555-0001, by calling (301) 415-5877, or by visiting the NRC's Web site at http://www.nrc.gov and selecting forms from the index found on the home page;

(2) File an original of NRC Form 398, together with the information required in paragraphs (a) (3), (4), (5) and (6) of this section, with the appropriate Regional Administrator;

(3) Submit a written request from an authorized representative of the facility licensee by which the applicant will be employed that the written examination and operating test be administered to the applicant;

(4) Provide evidence that the applicant has successfully completed the facility licensee's requirements to be licensed as an operator or senior operator and of the facility licensee's need for an operator or a senior operator to perform assigned duties. An authorized representative of the facility licensee shall certify this evidence on Form NRC-398. This certification must include details of the applicant's qualifications, and details on courses of instruction administered by the facility licensee, and describe the nature of the training received at the facility, and the startup and shutdown experience received. In lieu of these details, the Commission may accept certification that the applicant has successfully completed a Commission approved training program that is based on a systems approach to training and that uses a simulation facility acceptable to the Commission under Section 55.45(b) of this part;

(5) Provide evidence that the applicant, as a trainee, has successfully manipulated the controls of either the facility for which a license is sought or a plant-referenced simulator that meets the requirements of 55.46 c. At a minimum, five significant control manipulations must be performed that affect reactivity or power level (this requirement is obviously directed to operating a nuclear reactor, not to a reprocessing plant). Control manipulations performed on the plant-referenced simulator may be chosen from a representative sampling of the control manipulations and plant evolutions described in 55.59 of this part, as applicable to the design of the plant for which the license application is submitted. For licensed operators applying for a senior operator license, certification that the operator has successfully operated the controls of the facility as a licensed operator shall be accepted; and

(6) Provide certification by the facility licensee of medical condition and general health on Form NRC-396, to comply with Sections 55.21, 55.23 and 55.3(a)(1)."

A copy of NRC Form-398 is to be attached for information. It should be noted that these requirements have evolved over the past several decades and are much more detailed than the10 CFR 55 rules in existence 30 years ago. It should also be noted that at this time there is no "simulation facility acceptable to the Commission" for a commercial reprocessing/recycling plant.

APPENDIX E: History of Fuel Cycle Issues

A. Historical View

1. Introduction

At the time when efforts were underway to establish a commercial recycling/reprocessing business in the United States (mid-1960s - 1970s), the industry identified a series of issues that it believed should be addressed if an efficient industry with predictable costs and profit margins was to exist.

The issue of what is "ALARA" (As Low As Reasonably Achievable) and hat is an acceptable value has yet to be determined for the reprocessing component of the cycle.

The conversion of uranyl nitrate to UF_6 and its subsequent enrichment has also been a constant which has permitted the development of its own unique regulatory protocol.

2. General

There are several issues that were identified in the past as generally applicable to the entire fuel cycle.

(a) NIMBY (Not In My Back Yard): an overriding non-technical issue was and remains NIMBY which has and can impact every step of the fuel cycle, from mining to waste disposal. NIMBY is subtle in that it affects not only the siting of facilities but also transportation routes, quantities as well as materials shipped, ALARA requirements of local jurisdictions (regardless of risk relevance vis-a-vis federal rules), responsiveness to political and special interest groups, etc.

(b) Choice of Fuel Cycle: While obvious, the choice of fuel cycle (LWR, HGTR, fast reactor, MSR) has an impact on the flexibility of fuel fabrication as well as reprocessing and waste disposal, and consequently cost and economics (major commercial considerations). Economic considerations such as initial investment requirements, return on investment and duration of payback significantly affect the ability and willingness of a commercial entity to invest in any aspect of a prospective business venture.

(c) Environmental Statements: In the mid-60s -70s time frame the level of detail required in environmental statements for fuel cycle facilities was evolving and varied considerably over time. There was, at that time, considerable backfitting - and backfilling - of additional required information and systems in order to comply with the constantly evolving requirements. Some of these additional requirements were due to regulatory changes and some were due to subsequent interpretations of previously acceptable positions.

(d) Import/Export: There were several aspects of this issue, e.g., from what could be imported and in what form and in what containers to what could be exported and to whom. Although this was the Cold War era - which automatically imposed certain restrictions - it was an evolutionary period for international cooperation. In addition to the IAEA, which was in a formative period, there were a large number of agreements between countries - both bilateral as well as multilateral, which further complicating free trade.

(e) Quality Assurance: As noted in the main text of this paper there was an uncertainly as to which QA requirements were to be followed, particularly with regard to non-reactor Part 50 licensed facilities. At the same time, the design, manufacturing and acceptance codes were in the process of being changed. Agreements were therefore made at the time of license application approval by the regulatory agencies that the decisions thus made would stand for the duration of the project. In some instances, these agreements were modified or nullified. There was also some confusion as to the role of EPA standards vis-a-vis the AEC's regulations, the then cognizant agency.

(f) Emergency Planning: This was an area with extreme uncertainties and impacted not only the necessary and proper interactions between federal agencies but also their interactions with state and local authorities. Each governmental jurisdiction had its own rules/regulations and thus there was a level of uncertainty as to future changes or interpretations. Regulatory stability was, to say the least, elusive. That time was one of extreme project-by-project judgement, changing regulations for the guard/security force, poor interagency communications, etc. It was also a time when some federal and state land was granted to commercial interests to foster development of the commercial nuclear fuel cycle. Occasionally the difference in regulatory practices between the federal facility and the adjacent private one was a cause of concern to both the federal and private operators as well as the nearby populace.

(g) Technology Shortfall: As the commercial industry gained a foothold and developed experience, the need for additional equipment with new or improved capabilities became apparent. Evolving regulations also prompted investigation into advanced systems for environmental protection, accident prevention and monitoring, actual "real-time" system measurements, et al. Technology that had functioned adequately for years and which had been improved mainly for process optimization, was no longer considered optimal or acceptable. However, even when the current instrumentation and technology were deemed borderline, there was still no well defined requirement or standard, other than the direction given to improve and utilize the constantly changing "best available technology".

(h) Safeguards: During this time, when the commercial recycling industry was in its infancy, previously acceptable safeguard-related processes and systems were deemed unacceptable; e.g., shipment of liquid uranium and plutonium mixtures, MUF (material unaccounted for) limits, LEMUFs (limit of error MUF), related security (and safeguards) guard force qualifications, radioactive material storage and shipping, container fabrication materials, standards, acceptance testing, etc.

The "bottom line" on these general issues was the creation of an environment that was unpredictable and unstable - conditions that are the very antithesis of a favorable investment climate for private funds - and this, as in other areas noted previously, lead to investment delay, cancellation or premature abandonment.

3. Fuel Fabrication

Fuel fabrication, although not the thrust of this paper, likewise had its concerns and issues 30 - 40 years ago. In addition to related ALARA definition concerns, investors were concerned with the following:

(a) Colocation: Colocation of a fuel fabrication facility was seriously considered for the BNFP. The thought at the time was that the plant had already constructed a UF_6 conversion facility, was prepared to incorporate a PuO_2 conversion facility, was actively pursuing with France a borosilicate solidification process for HLW and had entered into discussions with its sister company, Gulf United Nuclear Fuels (GUNF), to build a fuel fabrication plant on adjacent land. The GUNF plant was to be capable of providing both conventional enriched U fuel (which at that time was approximately 3.5%) as well as various blends of MOX fuel. It was believed by BNFP that this arrangement of two closely linked fuel cycle facilities (BNFP providing its product directly to GUNF) would result in a mutually more profitable and less controversial facility particularly since another related fuel cycle facility, the Chem-Nuclear LLW disposal facility, was also contiguous. Such a confluence of backend facilities would result in spent fuel delivery to the reprocessing plant and only fabricated fuel and HLW shipments from this "nuclear island" site. It was believed such a site would be more acceptable to the public as it minimizes transportation and facility siting concerns.

(b) Siting Criteria: In the 1960s-70s time period, it was anticipated that the number of commercial reactors generating electricity would be 4 to 8 times what became the actual 2006 number. Such a 500 - 1000 nuclear power plant generating capacity would necessitate a significant growth in all supporting facets in the nuclear fuel cycle. That growth could come about by an increase in the number of facilities, an expansion of the capacity of then-current facilities, or some combination of both. Any of these paths was not straight forward as considerable uncertainty existed as to acceptable regulatory criteria for siting new plants or expanding current ones.

(c) Backfitting: Resolution of this issue has always involved several considerations, e.g., what is needed in regulatory guidance to maintain the current safety atmosphere and what must be done to satisfy the regulatory structure that had evolved since the initial construction permit (CP). In some instances, dependent somewhat on facility age and regulations in existence at the time of CP issuance, little change in the nature of backfitting was required. In other instances, a major investment was required for a license change. As noted above, the lack of licensing agency predictability gave commercial investors reason to pause and reconsider. A case in point is the West Valley reprocessing plant. In this case the requirements for backfitting to accommodate evolving regulations to protect from seismic events and other events were judged to be so costly that the decision was made to close the plant. Current NRC regulations on backfitting relevant to recycle plants may be found in 10 CFR Part 70, Subpart H, §70.76; 10 CFR Part 72, Subpart C, §72.62; and 10 CFR Part 50.109.

(d) Decommissioning: In the 1960s-70s timeframe, decommissioning estimates were not initially required. However, as experience was gained with all types of facility decommissionings, it was found that the estimates were generally exceeded by a large amount. In some instances the overruns were expected by either the licensee and licensor and a considerable effort was made by both entities to better understand and control those costs. A whole new business arena developed for entrepreneurs who believed by virtue of their wide-ranging experience and improved analytic techniques that D&D costs could be reasonably understood - and estimated - and therefore remain predictively contained despite inflation. D&D considerations, when sufficiently analyzed, can provide insights into how design, construction and operating practices could result in cost savings while at the same time maintain or improve overall facility and public safety.

(e) Risk Assessment: The use of risk assessment was just attaining some level of acceptance by both the regulating entity as well as by those regulated. This was a significantly different concept from the then accepted sense of absolute safety associated with the maximum credible accident scenario. It was also a concept that has continued to evolve over time since its initial introduction and has gained widespread acceptance.

4. Transportation

Several of the issues noted under this topic were relevant in the mid 60s-70s timeframe, and while time has relieved somewhat their urgency, they are discussed below for historical completeness.

(a) Ad Hoc Task Group: This group was composed of companies interested in the reprocessing of spent fuel, cask design and fabrication and railroad and nuclear trade association representatives. The group tasked itself to optimize cask design and per unit fuel element transportation, storage and disposal costs - - while attaining regulatory approval for interstate transport. These casks ranged from truck and rail to intermodal designs (including river as well as ocean barges) and encompassed a spectrum of use, from single purpose (transport) to multi-purpose (storage / transport / disposal).

One of the major efforts of this group was to develop a mutually acceptable understanding of the size and composition of such a fleet of casks and associated vehicles. It should be noted that at this time the security and safeguards considerations were evolving which further complicated the resolution of the transportation issue. For example, vehicle / shipment tracking requirements were being instituted as was driver and security personnel training.

Initially, each of the then-three reprocessing entities (NFS, BNFP, GE-MFRP) had their own (although limited in both number and capability) licensing agency accepted cask designs. These casks, which had been licensed for spent fuel transportation by truck and rail and which had been in use, resulted in a reluctance by the nuclear utilities to change from a regulatory agency approved design. It was left to the ad hoc task group members to prove the efficacy of their innovative designs (which permitted higher payloads) and gain acceptance by the customer base. Nuclear utilities were reluctant to change their handling practices and, in some cases, use of large casks was prohibited by the physical capabilities of containment air locks.

Transportation of spent fuel became highly visible and one that evolved into a reasonably sophisticated undertaking, from both a technical as well as a political perspective. Significant funding was provided by the relevant commercial interests to resolve the engineering and logistical problems associated with the development of a mutually satisfactory resolution to this step in the nuclear fuel cycle. Unfortunately, the INFCE foreclosed these efforts and the existing momentum ebbed away.

(b) Pre-construction Approval: Although seemingly obvious, in light of the millions of dollars involved in the design, fabrication and production of multiple casks of the same design, relevant federal and state agency regulatory approval to proceed ahead without fabrication of a full size cask was considered essential. The establishment of approved scaling factors for cask acceptance testing was a significant hurdle to overcome. Such a decision was necessary as cask design and shielding technology was in a rapid state of development and a commitment to a design for fabrication and delivery of, for example, 20 rail casks, required a significant lead time to set up such a facility. That plant had to put in place a fabrication line, obtain the required materials, manufacture the casks, QA the product and meet the delivery schedule.. The constant question was "what was safe enough?" New analytical techniques and materials were frequently required due to the intense focus on this link in the fuel cycle. The large investment envisioned also required a step change in management decisiveness in this area which had been relatively tranquil (at least insofar as the aspect of commercial cask development) for several preceding decades.

(c) Recovery of Damaged Casks: Up to this time, although there had been several instances where truck casks had been involved in accidents, recovery had been relatively simple with only minor radioactive contamination. However, as the commercial nuclear industry leaned toward a closed fuel cycle, which included fuel reprocessing and recovered product refabrication, resulting in a significant increase in transportation mileage involving heavier (rail and intermodal) movement of larger quantities of irradiated fuel, increased awareness and concern by the local affected populace along the routes coupled with attention and coverage by the press and intervener groups. brought cask recovery to the forefront as an issue that must be addressed - - where it remains even today.

(d) Risk Assessment: Along with the development of cask design analytical techniques came an equally impressive development in the analytic techniques for radioactive releases from accidents. Heavier casks containing much larger quantities of irradiated fuels, were postulated as being involved in accidents where they were deposited in deep, steep ravines or navigable waterways. Such postulated accidents resulted in recognition that the risks must be placed in proper perspective and be capable of being both understood and accepted by the public. The maximum credible accident scenario was recognized as being subject to increasingly unlikely assumptions but the concept of probabilistic risk assessment was in its infancy and was applied only occasionally, as a secondary approach, to some nuclear reactor accident analysis.

(e) JCAE Recommendation: The Joint Committee on Atomic Energy, before its dissolution in 1975, took an active role for many years in all aspects of both commercial and government nuclear development. In the course of their routine review of federal expenditures on R&D, safety and regulatory issues and technology transfer to commercial interests, the Joint Committee fostered development of a better understanding of transportation issues since such concerns applied equally to both commercial and government transportation of spent nuclear fuels.

At that time the federal government had a much broader and deeper experience with the kind of problems the commercial nuclear industry could anticipate since frequent defense related shipments of radioactive materials and weapons were a reality. Through the auspices of the JCAE there was a significant transfer to private industry of both technology and R&D funding. With the termination of the JCAE in 1975 and the simultaneous demise of the AEC, private industry experienced a significant drop in federal support (which was one of the espoused objectives of the 1975 Energy Reorganization Act).

(f) Railroad Embargo: As a closed commercial fuel cycle (one that included reprocessing and HLW disposal) seemed about to become a reality, the railroads began to insist on (1) limiting train speeds to less than 30 mph and/or (2) requiring a special dedicated train with escorts and/or (3) limiting transportation routes and timing around more densely populated areas. Coincident with these potential requirements there was another societal movement that insisted that trains and trucks bypass metropolitan areas regardless of whether the routes were federally funded or not. Federal preemption, therefore, became another issue.

Nuclear industry trade associations established liaison with the American Association of Railroads (AAR) in an effort to remove/ameliorate these proposals as it was believed they failed to increase public safety while significantly increasing the costs associated with transportation (not only spent fuel transport costs but also costs for intra-facility product shipments). Such uncertainties caused significant concerns among potential fuel reprocessors insofar as their ability to quote firm prices - which was what the nuclear power generation utilities demanded. Without the ability to quote firm prices reprocessors could not only flounder but quite possibly fail - an unacceptable financial risk noted by several major companies (viz ARCO, Exxon) that early on had indicated their intention to enter the reprocessing business but subsequently delayed, then entirely canceled, their plans to invest in this market niche.

So, although there were many process facilities either in operation or planned in the nuclear fuel cycle: mining / milling, UF_6 conversion and enrichment, fuel fabrication, power generating reactors, fuel reprocessing and licensed waste disposal facilities, transporting relevant materials between each of these facilities could become either the unifying element between all or could disrupt the cycle between any two points. Resolution of the political and public emotional issues associated with transportation quickly became intertwined with the resolution of the technical, commercial and regulatory ones.

5. Fuel Reprocessing

Several of the issues associated with fuel reprocessing facilities have been touched on in the preceding discussions on fuel fabrication, namely colocation, siting criteria and decommissioning. The principal difference in the discussion of these issues as they relate to reprocessing is that while there were several fuel fabrication plants manufacturing commercial nuclear fuel, in contrast, the U.S. reprocessing industry (NFS, MFRP) was still in an embryonic stage from whence it never grew to commercial viability. The first truly dedicated commercial size facility (BNFP), although physically completed and preliminarily tested, never reprocessed irradiated spent nuclear fuel and never entered into commercial operation. Most of the mid-60s - 70s so-called commercial reprocessing industry was therefore based on extrapolation from pilot operations and conjecture as to how it would operate and the possible forward looking requirements of a fully functioning reprocessing/recycle industry.

Changing applicable codes (namely for piping) and the application of a reactor oriented 10 CFR Part 50 to a vastly different chemical reprocessing facility resulted in assumptions and decisions that hopefully would not be changed over time. Unfortunately, such hope did not become a reality. The uncertainty caused by potential backfits resulted in large costs and financing uncertainties which was unacceptable to an industry with a focus on a bottom line reasonable ROI perspective. These uncertainties resulted in numerous startup delays and consequential overruns with subsequent stop work orders and eventual cancellations. Issues identified at the time include:

(a) Decommissioning: Initially these costs were not estimated nor was eventual decommissioning even considered in either the costs or the design and construction of the plants and the auxiliary supporting systems. Further complicating such evaluations was the inherent nature of the reprocessing facilities themselves in that they handled and released unique radio chemicals (compared to the then current operating commercial nuclear facilities) such as Pu, Tc, Kr and H_3 - for which a final regulatory disposition was poorly defined if at all. While owner operators of these facilities could provide an extremely rough estimate for the costs associated to decommission a facility to a “green field” state, the margin for error was considerable. At that time there was yet to be developed a systematic acceptable approach to D&D estimating. There was also no clearly defined limit for an acceptable unconditional release nor was there an acceptable scenario for a conditional release under certain specified circumstances.

(b) Risk Assessment: As a 10 CFR Part 50 facility, the safety analysis for a reprocessing plant leaned toward a reactor type analysis, namely a maximum credible accident (MCA) scenario. However, the position generally taken in the safety analyses for reprocessing plants was that a reactor-type MCA was extremely unlikely and so much less energetic than a reactor as to pale in comparison. A probabilistic, performance-based risk assessment, as is known today, was essentially unknown in the mid 60s-70s. Rather, the comparison to a highly energetic reactor excursion and the subsequent possible core meltdown resulted in the conclusion that reprocessing plant accidents were relatively benign insofar as risk to the public.

Another consideration was that at least on the BNFP site, the owner-operator had experience with a 10 CFR Part 70 license and expected to use the same regulation for its fuel receipt and storage facility, UF_6 conversion facility and plutonium oxide conversion facility. Such Part 70 licensed plants were designed and constructed to different codes and accidents were generally analyzed consistent with practices for non-nuclear chemical facilities. Owner-operators such as the Allied Chemical Corporation were familiar with such analyses and facility operations and felt somewhat at ease with the prospect of operating plants similarly licensed. The transfer of that confidence gained from past successful Part 70 licensing experience to a reprocessing plant seemed acceptable as the implications of licensing that facility under Part 50 was not immediately apparent.

It is indeed interesting to note that these facility owner-operators recognized some more than 30 years ago that some form of a new risk assessment methodology should be developed for these plants if the risks associated with these facilities was to be understood in an acceptable context with other facilities involving hazardous materials.

6. Waste Disposal

For many years the disposition of high level waste was an issue that the federal government strove to solve after an abortive attempt at the Lyons, Kansas salt dome. Finally, with the passage of the Nuclear Waste Policy Act, HLW was to be disposed at the federal repository (wherever that might be - Yucca Mountain was not yet singularly selected). This was interpreted to mean for the reprocessing industry that not only the solidified waste from the reprocessing facility but also spent fuel hulls and TRU waste was to be sent to the repository.

Another concern arising in the 70s was that it was not until the early 70s that liquid shipments of waste (as well as the U and Pu nitrate process stream solutions) were banned. These decisions on the back end of the fuel cycle were felt throughout the industry as both the system process design and transportation related equipments were impacted. There was also much confusion, some of which still exists, as to the handling and disposal of mixed waste, the definition of which was unclear.

The last concern under this issue, but by no means the least important, was the disposal of LLW at commercial burial sites. In the mid 60s - 70s timeframe there were six commercial sites for LLW disposal. That number decreased over time due to several reasons: (1) local pressure being brought to bear on those sites that "leaked" off site, or (2) were forced to close for other reasons. Progress on potential sites envisioned at that time was also stopped and has yet to be rekindled. Had it not been for the site at Clive, Utah, LLW disposal in 2007 would have been extremely costly and quite problematic as no proposed site (e.g., California, Illinois, Texas, or North Carolina) was successful in navigating the licensing hearing process. The Compact process which seemed so promising at its inception eventually failed.

7. 2007 Status - and Beyond

For the past 30 years the status of reprocessing and the supportive and related fuel cycle activities has been moribund. While occasionally federal administrations have espoused a closed fuel cycle, for various reasons, decision makers at both the federal and private level lacked the motivation to move ahead. Among the reasons for industry failing to move ahead was a lack of predictability as to applicable regulations as well as final as-built operational costs, a lack of meaningful progress on the development of a HLW repository, and the apparent stalemate in new nuclear power plant orders. As each of these deterrents was addressed and with revived federal government interest, research and financial assistance, it appears that commercial reprocessing in the U.S. may once again be viewed as a possible viable business for commercial entities.

The list of difficulties envisioned by the industry as noted above still needs to be addressed. The paramount issues requiring a resolution satisfactory to the industry are:

(1) An operational HLW federal repository with approved transportation modes/routes;
(2) A licensing framework suitable for a reprocessing plant;
(3) An assurance that the political and regulatory infrastructure is not only in place, but an acceptance by all involved that there will not be arbitrary backfits.
(4) The development and acceptance of an applicable risk assessment methodology, that places relative risks in their proper relationship and facilitates public acceptance of facility siting.

The principal contribution to this rebirth must be recognition by the public that nuclear energy is one of the acceptable and reliable sources for resolving our nation's energy supply problem. Furthermore, it should be understood that it is an option that can and should be pursued as the technology is not only proven but is essentially available now.

APPENDIX F. Radionuclide Distribution Among UREX +1a Process Streams
BASIS: PWR fuel; 33 GWd/te; one MTIHM
Reference to grams and curie yields: ORIGEN 2 calculations

Head End Process Step

Noble Metal Fission Yields from one MTIHM @ 33 GWd/te

Ru, Mo, Pd, Rh, Tc, (Zr/Mo)

Tc: Grams, 774; Curies, 13.1
Ru: Grams, 2190; Curies, 0.0185
Pd: Grams, 1385; Curies, 0.113
Mo: Grams, 3351; Curies, Stable (HF dissolves ~ 0.98 of Zr/Mo, leaving ~ 67 g)
Rh: Grams, 468; Curies, 0.0216

TOTAL initial solids before dissolution: 8168 grams (or 4884 grams if Mo is partially dissolved as Zr/Mo); TOTAL Curies: 13.25

Solids amounts increase faster than linearly with fuel burnup.

461.4 Kg HM per 154.5 Kg assembly hardware: 335 Kg hardware/MTIHM. 2.874 Kg and 2.00 Ci noble metals in 335 Kg (per 1 MTIHM) hardware assuming all noble metals are combined with hardware.

Composition of solids in dissolver after dissolution

References: "X-Ray Diffraction Studies on Irradiated Nuclear Fuels," H. Kleykamp and R. Pejsa, Journal of Nuclear Materials 124 56-3, 1984; personal communication from D. O. Campbell (retired from ORNL); "Flowsheet and Source Terms for Radioactive Waste Projections," C. Forsberg et al., ORNL/TM-8462, p. 96, March 1985.

Element	Range, %
Tc	8 -12 (assume 15 %): 116 grams; 1.97 Ci
Ru	27-47 (assume 50 %): 1095 grams; 0.01 Ci
Pd	10-18 (assume 20 %): 277 grams; 0.023 Ci
Mo	16-41 (assume 40%): 1340 grams; 0.000 Ci
Rh	6-11 (assume 10 %): 46.8 grams; 0.002 Ci
U:	0.05 (assume 0.0005 fraction remains undissolved in sheared fuel); 478 Grams
	TOTAL: grams 3353; TOTAL Ci: 2.00

Fraction Tc in UREX Process feed stream: 0.85
Fraction Tc in dissolver residues combined with cladding hulls: 0.15
Fraction Tc in U product: 0.0001 (assumption)
Using three Reillex HPQ columns in series no Tc remained in the U product stream. All the Tc remained on the third resin column.
Fraction Tc in fission product waste: none (assumes all is combined with hulls)
Fraction Tc in TRU product: 0.0001 (assumption)

Volatiles and Gases

References: *Light Water Reactor Nuclear Fuel Cycle,* Eds. R. G. Wymer and B. L. Vondra, CRC Press, 1981; ORNL-/TM-5987, "LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period April 1 to June 30, 1977, B. L. Vondra; "Alternate Fuel Cycle Technologies Program Quarterly Report for Period July 1 to September 30, 1977," ORNL/TM-6076, B. L. Vondra; "Study on Gaseous Effluent Treatment for Dissolution Step Nuclear Fuel

Reprocessing," H. Mineo et al., WM'02 Conference, February 24-28, 2002, AZ

Voloxidation releases:

1.00 fraction ^{3}H from the fuel
0.06 fraction Kr
0.01 fraction I
0.5 fraction ^{14}C

Assume 1.000 fraction of all volatiles and gases left in fuel after voloxidation goes to dissolver off-gas.

^{3}H:

0.0208 grams (fission + activation); 201 Ci (fission + activation)
Fraction solidified as water in solid waste: 0.59 (fraction assuming no voloxidation)
Fraction in Zircaloy cladding: (0.41 if no voloxidation); 0.000 if voloxidation
39 cubic centimeters as gas at 1 atmosphere and 0 °C. (STP)

Kr

1.59 grams total of all isotopes (42.4 cubic centimeters at STP)
1847 curies ^{85}Kr (the only curie source of importance)
Fraction captured: 0.85 (assuming capture from voloxidation and from the dissolver off-gas)
Storage may be in cylinders, on solid sorbents, or by more exotic means such as ion implantation.

Xe

5.35 grams total of all isotopes (894 cubic centimeters at STP)
All isotopes are stable
Xe capture is determined by the removal process chosen, and can approach that for Kr, i.e., 0.85 fraction.
Storage may be in cylinders, or it may be released since it is essentially not radioactive.

I

"Alternate Fuel Cycle Technologies Program Quarterly Report for Period July 1 to September 30, 1977," ORNL/TM-6076, B. L. Vondra; "Environmental Radiation Requirements for Normal Operations in the Uranium Fuel Cycle," 40CFR190, Vol 1, p. 45

^{127}I: 5.594E+01 grams; stable
^{129}I: 1.800E+02 grams; 3.179E-02 Ci
TOTAL grams: 2.36E+02; TOTAL Ci: 3.179E-02

^{129}I

Fraction to off-gas from voloxidation: 0.01
Fraction in solids in dissolver: 0.022 (as AgI and PdI_2)
Fraction of I in PdI_2 (decomposes in vitrifier and goes to off-gas): 0.011
Fraction of I in AgI (stable) and goes with noble metals to hull wastes: 0.11
Fraction retained in dissolver solution: 0.0072
Fraction to off-gas from dissolver solution: 0.965
Fraction in off-gas captured and made into solid waste: 0.995
Bulk density of $AgNO_3$/silica gel: 0.719 (this absorbant will be used in Japan's Rokkasho reprocessing plant); the Iodox Process produces $Ba(IO_3)_2$ which is a potential waste form.

^{14}C

2.632E-05 g; 1.88E-06 moles (0.042 cubic centimeters at STP)
1.174E-04 Ci
Fraction from voloxidation to off-gas as CO_2: 0.05
Fraction from dissolver to off-gas as CO_2: 0.89
Fraction going to UREX process step: 0.06
Fraction going to CD-PEG: 1.00
Fraction going to TRUEX: 1.00
Fraction going to TALSPEAK: 1.00
Fraction going to F.P. waste stream: 1.00
Fraction released in F.P. waste vitrification step: 0.06
Fraction captured in off-gas trapping system: 0.96
CO_2 collected in $CaCO_3$ solution

UREX Process step

Fraction Tc in U product: 0.0001
Fraction U in U product: 0.997
Fraction U in raffinate: 0.003

Rare earths fraction to CCD-PEG: 0.9975

CCD-PEG Process step

Cs

^{133}Cs: 1.132E+3 grams; stable
^{134}Cs: 2.616E-2 grams; 3.386E+01 Ci
^{135}Cs: 3.013E+2 grams; 0.347 Ci
^{137}Cs: 6.713E+2 grams; 5.842 E+04
TOTAL grams Cs: 2.105E+03; Total Ci: 5.845E+04 Ci
Fraction Cs to product: 0.9984

^{137}Cs

Reference: "Lab-Scale Demonstration of the UREX+1a Process Using Spent Fuel," C. Pereira et al., WM'07 Symposium, February 25-March1, 2007

Fraction to U product: insignificant
Fraction to CCD-PEG: 1.00
Grams in CCD-PEG product: 670
Curies in CCD-PEG product: 5.83E+04
Fraction to TRUEX: 0.0016

Sr

^{86}Sr: 4.038E-01 grams; stable
^{88}Sr: 3.504E+02 grams; stable
^{90}Sr: 2.940E+02 grams; 4.012E+04 Ci
TOTAL grams Sr: 6.48E+02; TOTAL Ci: 4.012E+04

^{90}Sr

Reference: "Lab-Scale Demonstration of the UREX+1a Process Using Spent Fuel," C. Pereira et al., WM'07 Symposium, February 25-March1, 2007

Fraction to U product: insignificant
Grams in CCD-PEG product: 293
Curies in CCD-PEG Product: 3.98E+04
Fraction to CCD-PEG product: 0.9984
Fraction to TRUEX: 0.0016

U

U: Fraction to TRUEX: 1.000

TRUEX Process Step

Rare Earths

Reference: “Metal Recovery Plant Activities During FY 1956,” ORNL-2235; “Lab-Scale Demonstration of the UREX+1a Process Using Spent Fuel,” C. Pereira et al., WM’07, Symposium, February 25-March 1, 2007, Tucson, AZ

DF (total RE β): ~400 for uranium product; fraction in U product: 0.0025

Rare earths fraction to CCD-PEG: 0.9975
Rare earths fraction to TRUEX: 0.9993
Rare earth grams to TRUEX
Rare earths fraction to TALSPEAK: 0.9991
Rare earths fraction to waste: 0.99

U

U fraction to TALSPEAK: 1.000 (assumed)

TALSPEAK Process Step

Reference: “TRUEX/SREX Demonstration” Innovative Technology, OST Reference #347, Tanks Focus Area; “State of the Art in Nuclear Fuel Reprocessing,” Safe Waste 2000, B. Barre and H. Masson, October 2-4, 2000; “Partitioning and Transmutation: Radioactive Waste Management Option,” Workshop on Technology and Applications of Accelerator Driven Systems (ADS), ICTP Trieste, Italy, 17-28 October 2005, IAEA; *Light Water Reactor Nuclear Fuel Cycle,* Eds. R. G. Wymer and B. L. Vondra, CRC Press, 1981

Np

4.633E+02 g; 1.741+01 Ci

^{237}Np: 4.633E+02 g; 3.267E-01 Ci
^{238}Np: 3.236E-02 Ci
^{239}Np: 1.705E+01 Ci

TOTAL GRAMS: 4.633E+02; TOTAL Ci: 1.741E+01

^{237}Np

Fraction going to U product stream: (0.4 to) 0.003
Fraction going to CCD-PEG: (0.6 to) 0.997
Fraction going to TRUEX: 1.000
Fraction going to TALSPEAK: 0.9997
Fraction going to TRU product stream: 0.999
Fraction going to fission product waste: 0.001

Pu

Reference: "Lab-Scale Demonstration of the UREX+1a Process Using Spent Fuel," C. Pereira et al., WM'07, Symposium, February 25-March 1, 2007, Tucson, AZ; "Recent Advances in Reprocessing of Irradiated Fuel," Nuclear Engineering–part XX, No. 94, Vol. 65, 1969, Eds. W.A. Rodger and D. E. Fergson

^{238}Pu: 1.211E+02 g; 2.074E+03 Ci
^{239}Pu: 5.030E+03 g; 3.128E+02 Ci
^{240}Pu: 2.316E+03 g; 5.279E+02 Ci
^{241}Pu: 3.657E+02 g; 3.769E+04 Ci
^{242}Pu; 4.509E+02 g; 1.722E+00 Ci
TOTAL grams: 8.284E+03; TOTAL Ci: 4.061E+04

Fraction going to U product stream: 0.00002
Fraction going to CCD-PEG: 1.000
Fraction going to TRUEX: 1.000
Fraction going to TALSPEAK product stream: 0.9999
Fraction going to FP waste stream: 0.0001

U

Fraction to TRU product stream: 1.000

Am + Cm

Reference: "Lab-Scale Demonstration of the UREX+1a Process Using Spent Fuel," C. Pereira et al., WM'07, Symposium, February 25-March 1, 2007, Tucson, AZ; "TRUEX/SREX Demonstration" Innovative Technology, OST Reference #347, Tanks Focus Area; "State of the Art in Nuclear Fuel Reprocessing," Safe Waste 2000, B. Barre and H. Masson, October 2-4, 2000; "Solvent Extraction Separations of Trivalent Lanthanide and Actinide Ions using an Aqueous Aminomethanediphosphonic Acid," M. P. Jensen and K. L. Nash, Proceedings of ISEC '99, International Solvent Extraction Conference, July 11-16, 1999

Am: 9.49E+02 g; 2.996E+03 Ci Cm: 1.036 g; 7.602E+02 Ci

^{241}Am: 8.638E+02 g; 2.966E+03 Ci
^{243}Am: 8.550E+01 g; 1.705E+01 Ci

^{242}Cm: no value; 5.325 Ci
^{243}Cm: 2.226E-01 g; 1.150E+01 Ci
^{244}Cm: 9.182 g; 7.432E+02 Ci
^{245}Cm: 8.521E-01 g; no value
^{246}Cm: 1.014E-01 g: no value

Fraction going to U product stream: 0.00002
Fraction going to CCD-PEG: 1.000
Fraction going to TRUEX: 1.000
Fraction going to TALSPEAK product stream: 0.9997
Fraction going to FP waste stream: 0.0003

Notes on final output stream characteristics assumptions

Volatile Effluent

Not applicable; goes up the stack

Tritium Volatile Waste

- Tritium present after 25y decay assumed to be recovered by voloxidation in a closed system with zero external water present. 100% recovery assumed.

= ORNL/TM-3723 [GOODE, 1973] reported less than 0.1% of T remained in fuel marix after voloxidation

= T in the form of ZrT_2 should be dissociated because this occurs at ~300 C [OSHA web site] whereas voloxidation occurs at 450 C or higher and hardware melting occurs at 1450 C [www.azom.com] so the T should be evolved.

- Tritium assumed to be made into tritiated water by catalytic conversion [STI-DOC-010-421] and incorporated into polymer-impregnated cement based on studies showing at least 10x less leaching from PIC grout [Albenesius, 1983][DP-MS-83-114 (CONF-8311105-2)]
- 10% by weight of polymer replacing water [HTTP://irc.nrc-cnrc.gc.ca CDB-242]
- Although variable, 53 wt% water is optimal [www,cement.org FAQ]. The ratio can range down to about 45 wt %. Use larger value to account for higher density of water containing D and T.
- PIC grout density is 2.2 g/cc [http://irc.nec-cnrc.gc.ca CBD-242]
- Water density and tritium content

  = Hydrogen in water made from dissolver offgas (SNF water) is 84 wt % T, 1 wt % D, and 15 wt % H based on ORIGEN2 output and ratios of fission product yields for hydrogen isotopes.

  = Avg. molecular weight of hydrogen is 2.7 and avg. molecular weight of SNF water is 21.4.

  = Water density is 1.19 g/cc. Each g of SNF water contains o.12 g T.

- 2.2 g cement contains 2.2x0.53 = 1.17 g normal water or 1.17x1.19 = 1.39 g SNF water or 1.39x0.12 = 0.17 g T. Thus, 0.076 g T/g cement

C-14 Volatile Waste

- 99% of the C-14 assumed to be recovered from the dissolver off-gas using molecular sieves and scrubbed with calcium hydroxide slurry to yield calcium carbonate [DOE/ET-0028].
- Calcium carbonate is assumed to be fixed in grout [ORNL/TM-5171.

  = Grout density is 1.6 g/cc [ORNL/TM-5171]

  = Grout loading is 30 wt % [ORNL/TM-5171]

- Calcium carbonate is 12 wt % carbon
- Carbon is 0.08 wt % C-14 [DOE/ET-0028]
- Leads to 0.31X1.6X0.12X0.0008 = 4.6E-05 g C-14/g waste

Krypton Volatile Waste

- Kr assumed to be 100 % evolved in either voloxidation of dissolution
- Kr recovered using cryogenic distillation [DOE/ET-0028]
- Assume 85 % recovery of Kr based on requirements of 40CFR190

  = At 25y decay there is 351g/MT Kr (1.34 wt % Kr-85) and 5357 g/MT Xe based on ORIGEN2 calculation

    - Kr recovery is 0.85x351 =298 g/MT
    - Ratio of Xe in product to Kr in product ranges from 25 wt % [DOE/ET-0028] to 12.5 vol % (18 wt %) [STI/DOC/O1O/199]. Defer to IAEA values(18 wt %) that is based on pilot plant experience.
- Assumed to be stored in compressed gas cylinders at 1.5 atmospheres (Barnwell licence condition limiting pressure).
- Ignore cylinder volume
- Kr load factor is 0.0134x(1-0.18) = 0.011 g kr/g noble gas in cylinder
- Kr density in gas is 0.0047 g Kr/cc noble gas in cylinder at 1.5 atmospheres pressure

Iodine Volatile Waste

- Assume silver mordenite (AgZ) sorbent that is grouted to contain 34 wt % AgZ and use information in Table XI of STI/DOC/010/276
- Density of grouted AgZ is 2.1 g/cc
- From ORIGEN2 calculation iodine is 180 g ^{129}I /MT and 236 g total iodine/MT
- Iodine-129 loading in grout is 625 Kg Ix(180 kg ^{129}I/kg I)/11500 kg waste form + 0.0414 g ^{129}I/g waste

Cladding Waste plus Tc, Dissolver Solids, and a Fraction of Non-Volatile SNF

- Assume all cladding and other structural material (end pieces, grid spacers) will be melted into an alloy for disposal
- Include recovered Tc, dissolver solids, and fraction of non-volatile SNF. However, no tritium is included because ZrT_2 is assumed to be dissociated by voloxidation or melting.
- Radionuclide density is 1.0 because the entire waste form is composed of waste materials.
- Density is the mass-weighted average of Zr (for Zircaloy) and SS (for SS, Inconel, and Nicrobraze) which is 6.8 g/cc [ORNL/TM-6051]
- Continue to support use of 0.05% of non-volatile SNF being associated with the cladding
    - Historical reports use this value [ORNL/TM-5427, DOE/ET-0028]
    - Statement that after repeated leaching of Zr cladding with boiling nitric acid the Pu content was reduced to 0.0005%
    - Information from May 2007 AREVA presentation indicated 0.1% of Pu is in final waste forms (p 8 of presentation) and 0.04% of the alpha activity in the waste is in the cladding. This implies that 0.000004% of the SNF is associated with the cladding.

U Product

- Assume product is uranium oxide meeting ASTM [ASTM C 788-03] purity specs suitable for uranyl nitrate that is the direct product of rerocessing.
    - For ^{99}Tc C 788 refers to C 787 (specs for UF_6 for enrichment) and c 996 (specs for UF_6 product from enrichment). Adopt the C 787 value (0.5 ppmw) because the higher value in C 990 elects the effects of enrichment. Assuming 50% of the Tc in the U product from the first separation process remains with the U product after cleanup, this equates to a Tc DF of 0.997794 in the first separation process.
    - For TRU C 788 limits TRU alpha is 6.8 nCi/g U and Np is 3.4 nCi/g U. For Np this implies that 0.00875 of the soluble Np follows the U stream. Allowing the remainder of the allowance to the limit (i.e., 3.4 nCi/g) for the TRU elements other than Np yields a DF for Pu, Am, and Cm of 4.29E-07.
- Density of product can have a wide range because the degree of compaction is unknown, and the oxidation state is unknown; use a value of 3.5 g/cc.
    - UO_2 powder densities range from 2.0 to 5.9 [ORNL/TM-2000-161]. However, product is unlikely to have a high dioxide concentration because of the cost of oxide reduction.

= U_3O_8 densities range from 1.5 to 4.0 [ORNL/TM-2000-161].

= The product of the de-fluorination plants is a mix of the two oxides with more U_3O_8 than UO_2. Select the higher end of the U_3O_8 density range to account for the UO_2 component.

TRU Product

- Assume it's converted to an oxide (mainly dioxides) and fabricated into pellets.
- Calculate theoretical density of fuel based on values from CRC handbook and book on isotopic power sources weighted by mass in O2 TRU product
- Assume pellets are 95% of theoretical density

Cs/Sr Waste

- Assume Cs/Sr is made into an aluminosilicate waste form using steam reforming
- Bulk density of product is 1 g/cc [PNWD-3288]
- Waste loading is 27% [WSC-TR-2002-00317]

Fission Product Waste

- Base values on experience at DWPF
- Glass density 2.65 [WSRC-MS-2000-0053]
- Waste loading 38% [WSRC-MS-2004-00286]