INL/EXT-20-59106

Review – Nuclear Fuels and Reprocessing Technologies: A U.S. Perspective

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Prepared for the
U.S. Department of Energy
Office of Nuclear Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

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ABSTRACT

Reprocessing and/or waste management issues are of concern to the “back end” of the nuclear fuel cycle. Of course, there are a great many “nuclear fuel cycle” scenarios to consider; if not in practice, then at least in theory. The simplest conceptually is the “once through” fuel cycle in which the spent fuel is discarded. The more complex fuel cycle scenarios involve reprocessing spent nuclear fuels and a family of nuclear reactor technologies to accommodate burning and breeding for various military and commercial needs. Therefore, the selection of a specific “fuel cycle” is what ultimately imposes the engineering requirements of the reprocessing and waste management technologies. No one part is independent of the other parts in a fuel cycle flowsheet; all parts should be fully integrated.

This paper presents a summary of nuclear chemistry processes, nuclear reactor technologies, associated nuclear fuel types, and the reprocessing technologies that serve the different nuclear fuel types. Comprehending how this series of topics are related to each other is a prerequisite to understanding the requirements of any reprocessing strategy. The summary materials presented here are selective, as opposed to comprehensive. More detailed information on any one subject can be found in the reference materials.

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ACRONYMS

ABWR	Advanced Boiling Water Reactor
AEC	Atomic Energy Commission
AGR	Advanced Gas-Cooled Reactor
ALMR	Advanced Liquid Metal Reactor
ANL	Argonne National Laboratory
ANL-W	Argonne National Laboratory West
AHR	Aqueous Homogenous Reactor
BARC	Bhabha Atomic Research Centre
BISO	Bi-Structural Isotropic Fuel
BWR	Boiling Water Reactor
CANDU	Canada Deuterium Uranium
CORAL	Compact Reprocessing Facility for Advanced Fuels
CR	Conversion Ratio
CRBRP	Clinch River Breeder Reactor Project
CVD	Chemical Vapor Deposition
DFRP	Demonstration Fast Reactor Plant
DOE	Department of Energy
DU	Depleted Uranium
EBR-I	Experimental Breeder Reactor I
EBR-II	Experimental Breeder Reactor II
EFFBR	Enrico Fermi Fast Breeder Reactor
FBTR	Fast Breeder Test Reactor
FCF	Fuel Cycle Facility and Fuel Conditioning Facility
FFR	Fluid Flow Reactor
FFTF	Fast Flux Test Facility
FRFRP	Fast Reactor Fuel Reprocessing Plant
FRR	Foreign Research Reactor
GGR	Graphite Gas-Cooled Reactor
GTRI	Global Threat Reduction Initiative

HEU	High Enriched Uranium
HNPF	Hallam Nuclear Power Facility
HS	Hanford Site
HTI	High Temperature Isotropic
HTGR	High Temperature Gas-Cooled Reactor
HTR	High Temperature Reactor
HWR	Heavy Water Reactor
IFR	Integral Fast Reactor
IGCAR	Indira Gandhi Centre for Atomic Research
IMF	Inert Matrix Fuel
INL	Idaho National Laboratory
KARP	Kalpakkam Atomic Reprocessing Plant
kWth	Kilowatt Thermal
LAMPRE	Los Alamos Moten Plutonium Reactor Experiment
LANL	Los Alamos National Laboratory
LEU	Low Enriched Uranium
LFR	Lead-Cooled Fast Reactor
LLNL	Lawrence Livermore National Laboratory
LMFBR	Liquid Metal-Cooled Fast Breeder Reactor
LMFR	Liquid Metal Fueled Reactor
LMR	Liquid Metal-Cooled Reactor
LWR	Light Water Reactor
LTI	Low Temperature Isotropic
MA	Minor Actinides
MAGNOX	MAGnesium No OXidation Aluminum
MAPS	Madras Atomic Power Station
MOX	Mixed Oxide Fuel
MSBR	Molten Salt Breeder Reactor
MSR	Molten Salt Reactor
MSRE	Molten Salt Reactor Experiment

MT	Metric Ton (1000 kg)
MTHM	Metric Ton Heavy Metal
MWth	Megawatt Thermal
NRC	Nuclear Regulatory Commission
NU	Natural Uranium
ORNL	Oak Ridge National Laboratory
PDC	Pilot Demonstration Centre
PDPC	Pilot Demonstration Power Complex
PHWR	Pressurized Heavy Water Reactors
PRISM	Power Reactor Innovative Small Module
PRP	Plutonium Reprocessing Plant
PRTRF	Power Reactor Thoria Reprocessing Facility
PUREX	Plutonium Uranium Reduction by Extraction
PWR	Pressurized Water Reactor
RAPS	Rajasthan Atomic Power Station
RBMK	Reaktor Bolshoy Moshchnosty Kanalny (Russian Designation)
RCW	Radiochemical Works
REDOX	Reduction Oxidation
RERTR	Reduced Enrichment for Research and Test Reactors
RFP	Rocky Flats Plant
RIAR	Research Institute of Atomic Reactors
RR	Research Reactor
S1G, S2G	S = Submarine, 1 = First, 2 = Second, G = General Electric
S1W, S2W	S = Submarine, 1 = First, 2 = Second, W = Westinghouse
SAFR	Sodium Advanced Fast Reactor
SCE	Siberia Chemical Enterprise
SCP	Salt Cycle Process
SEFOR	Southwest Experimental Fast Oxide Reactor
SFR	Sodium-Cooled Fast Reactor
SFT	Spent Fuel Treatment

SGBR	Sodium-Cooled Graphite Breeder Reactor
SGR	Sodium-Cooled Graphite Reactor
SIR	Submarine Intermediate Reactor
SMR	Small Modular Reactor
SNF	Spent Nuclear Fuel
SRE	Sodium Reactor Experiment
SRS	Savannah River Site
SFL	Santa Susana Field Laboratory
SSN	U.S. Navy Designation for Attack Submarine
THORP	Thermal Oxide Reprocessing Plant
TRIGA	Training, Research, Isotopes, General Atomics
TRISO	Tri-structural isotropic
TVA	Tennessee Valley Authority
UNGG	Uranium Naturel Graphite Gaz-Cooled
UTSF	Uranium Thorium Separation Facility
VVER	Water-Water Energetic Reactor (Russian Designation)

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1. INTRODUCTION

In the U.S. since the 1940s research has pursued almost every imaginable avenue of nuclear technology development. Exploitation of natural resources involved the mining and extraction of uranium and thorium. Deployment of nuclear reactors required the development and testing of fuel and reactor designs. Military pursuits included weapons, propulsion, and remote power systems. And the requirements for supporting technologies introduced isotopic separations, spent fuel reprocessing, and nuclear waste management. Remarkable is the great number of options available to each of these engineering and scientific pursuits.

Nuclear energy has been proposed for explosives for military and civil engineering purposes, naval propulsion, airplane propulsion, rocket propulsion, radiation sources, electrical power generation, chemical process heat generation, and hydrogen generation. Nuclear fuel forms include metals, alloys, liquid metals, molten salts, aqueous solutions, organic solutions, oxides, hydrides, carbides, nitrides, and other ceramics.

The objective of this report is to provide a perspective on the history and body of knowledge associated with nuclear fuels and reprocessing technologies. Information in these areas is spread throughout the literature. Hopefully, the information consolidated and presented here will provide a convenient summary for those interested in these disciplines.

2. MATERIALS BACKGROUND

Nuclear reactors generate thermal energy (heat) by the processes of controlled fission. Fission occurs when a heavier element splits (fissions) into two lighter element fission products. There are only two fundamental sources of fissionable elements for use as nuclear fuel: i) recovery from the earth's crust and ii) transmutation in a nuclear reactor.[a] Terrestrially, ^{235}U is the only naturally occurring fissile isotope; natural uranium (NU) is nominally 0.73 wt% ^{235}U with the balance ^{238}U. As will be discussed later, reactors can operate with NU or uranium enriched in ^{235}U. ^{232}Th is the only naturally occurring thorium isotope, and ^{232}Th and ^{238}U are the only naturally occurring fertile isotopes. Reactors can be used to create fissile isotopes by exploiting the transmutation of ^{238}U to ^{239}Pu, and ^{232}Th to ^{233}U. ^{239}Pu and ^{233}U are, like ^{235}U, fissile isotopes.

Elements originate from cosmic stellar processes. It is theorized that the elements comprising earth formed during a supernova about 6.5E+9 years ago, while the earth itself formed into a planet about 4.5E+9 years ago. Therefore, the inventory of elements that comprised the earth at its formation have only been subject to the processes of radioactive decay. Consequently, all the actinide elements whose half-lives are considerably shorter than the age of the earth have, for all practical purposes, disappeared. For example, after a period of 20 half-lives, only one millionth of the original quantity remains. After 27 half-lives, about one billionth.

a. Reprocessing technologies are a means of recovering fissionable elements from spent nuclear fuels, and for the purpose of this discussion are not considered a fundamental source of these elements.

The heaviest stable isotope is ^{209}Bi. All elements heavier than bismuth are subject to radioactive decay and are in the process of disappearing. Consequently, elements heavier than uranium[b] do not exist in nature beyond trace quantities that are much too small to serve as resources. Table 1 lists the actinide isotopes with long half-lives. Note that ^{232}Th, ^{238}U, and ^{235}U have the longest half-lives, which explains the abundance of these isotopes in the earth's crust. Also, the relatively long half-lives of ^{233}U and ^{239}Pu, man-made fissile isotopes with transmutation of ^{232}Th and ^{238}U, render these isotopes useful for nuclear fission applications.

Table 1. Sorted Actinide Half-life.

Isotope	Half-life (year)	Half-lives Since Formation of Earth
^{232}Th	1.41E+10	0.32
^{238}U	4.51E+09	1.0
^{235}U	7.10E+08	6.4
^{244}Pu	8.28E+07	55
^{236}U	2.39E+07	190
^{247}Cm	1.64E+07	280
^{237}Np	2.14E+06	2100
^{242}Pu	3.79E+05	12000
^{248}Cm	3.52E+05	12906
^{234}U	2.47E+05	18393
^{233}U	1.62E+05	28043
^{230}Th	8.00E+04	56788
^{231}Pa	3.25E+04	139785
^{239}Pu	2.44E+04	186189

Fission cross section is a measure of the probability of a fissionable nucleus capturing an incident neutron and undergoing fission. There is a distinction made between fissionable and fissile isotopes. Fissile isotopes are a subset of fissionable isotopes; therefore, the number of fissionable isotopes is greater than the number of fissile isotopes. Fissile isotopes are readily fissionable in any spectrum. Whereas fissionable isotopes are only readily fissionable in a fast neutron spectrum. As stated earlier, ^{235}U, ^{233}U, ^{239}Pu, and ^{241}Pu are fissile isotopes. ^{232}Th and ^{238}U are not considered fissile isotopes because their fission cross section is very low in the thermal neutron spectrum. However, they are both readily fissionable in a fast neutron spectrum. Therefore, ^{232}Th and ^{238}U are fissionable isotopes.

Capture cross section is a measure of the probability of a nucleus capturing an incident neutron and undergoing transmutation. The isotopes of many elements exhibit this property and are capable of transmutation to heavier isotopes via the process of neutron capture. Fertile isotopes are those capable of transmutating into fissile isotopes by the process of neutron capture in a thermal neutron spectrum. As stated earlier, ^{232}Th and ^{238}U are the only two naturally occurring fertile isotopes.[1]

b. Actinide elements heavier than uranium are called transuranics. Elements heavier than the actinide series are called transactinides.

Transmutation by neutron capture also leads to the formation of activation products (radioisotopes) in the materials of construction of the reactor core. For example, ^{59}Co is the only naturally occurring cobalt isotope because all other cobalt isotopes have short half-lives and are not stable in a geologic sense, see Table 2. Cobalt is a common alloying element in stainless steels. A common phenomenon in nuclear reactor stainless steel hardware is ^{59}Co transmutation to ^{60}Co, which is subject to decay by beta and gamma emissions to stable ^{60}Ni. However, transmutation is not restricted to the process of neutron capture; transmutation involves any process by which an isotope can convert to another isotope of the same element or a different element. Radioactive decay is another process by which transmutation can occur.

Table 2. Cobalt Isotopes and their half-lives.

Isotope	Half-life
^{58}Co	7.13E+01 days
^{58m}Co	9.00E+00 hr
^{59}Co	Stable
^{60}Co	5.26E+00 yr
^{60m}Co	1.05E+01 min
^{61}Co	9.90E+01 min
^{62}Co	1.39E+01 min
^{72}Co	1.23E-01 sec
^{73}Co	1.16E-01 sec
^{74}Co	1.08E-01 sec
^{75}Co	8.02E-02 sec

All fissionable isotopes are radioisotopes, which means they are not stable isotopes. All radioisotopes are constantly subject to the processes of radioactive decay until they become stable isotopes. The decay chain is the path by which a radioisotope becomes a stable isotope. Mechanisms of radioactive decay include alpha decay (α or ^{4}He), beta decay (β), gamma decay (γ), electron capture, neutron emission, and spontaneous fission. Isotopes that undergo the processes of spontaneous fission are a subset of fissionable isotopes. This implies that as a material property, fissionable isotopes are always associated with an inherent, albeit small, neutron flux. However, the inherent neutron flux of fissionable isotopes can promote and accelerate nuclear fission when enough mass in accumulated under the right conditions of geometry, neutron moderation, and neutron reflection. Critical mass is the minimal mass required to sustain nuclear fission under a defined set of these conditions.

A reactor's core is designed to facilitate controlled fission. Control of a reactor's core is maintained by several mechanisms, from both external sources and internal sources. Control rods are an example of the former, and fuel reactivity is an example of the latter. Control rods work on the principle of inserting select isotopes into the reactor core that readily absorb neutrons. The reactor cannot achieve criticality with the control rods inserted into the core. An example is cadmium-bearing control rods in which ^{113}Cd (with its large capture cross section) absorbs thermal neutrons and transmutes to ^{114}Cd. However, capture cross section of cadmium drops precipitously in the fast neutron spectrum, meaning that cadmium is not an effective control mechanism in fast reactors. ^{10}B has applications in both thermal and fast reactors. Natural boron is nominally 19.9 wt% ^{10}B with the balance ^{11}B. Upon capturing a neutron, ^{10}B fissions into ^{7}Li and ^{4}He. Boron used for control applications is often enriched with respect to ^{10}B to levels greater than 90%.[2] A fueled control rod is the opposite approach. The reactor cannot achieve criticality unless the fueled control rods are inserted into the core.

The primary purpose of the fissile inventory is to support fission, and the primary purpose of the fertile inventory is to support transmutation leading ultimately to the creation of fissile isotopes. Fissile and fertile materials can be in two very distinct regions of the reactor, such as fuel and blanket regions, respectively; or they can be intermixed. Of course, even when partitioned into two very distinct regions some degree of transmutation always occurs in the fuel; just as some degree of fission always occurs in the blanket. The proportions and types of fissile and fertile materials, and how they are distributed within the reactor, are designed to control the reactor's breeding ratio. A reactor with a breeding ratio less than one is a net consumer of fissile material. A reactor with a breeding ratio equal to one is self-sustaining with respect to fissile material but will require a feed of fertile material. A reactor with a breeding ratio greater than one is a net producer of fissile material, but again will require a feed of fertile material. Nuclear fuel cycle reprocessing schemes are a requirement of the latter two.

Nuclear fuel cycle strategies are determined by government policy makers for both commercial and military applications. An "open fuel cycle" entails no form of reprocessing, the fresh fuels are fabricated from virgin reserves and the spent fuels are slated for interim storage and eventual geologic disposal. A "closed fuel cycle" utilizes reprocessing technologies for the combined purposes of natural resource conservation, carbon emission reduction, and waste minimization. As stated earlier, ^{235}U is the only naturally occurring fissile isotope and its concentration is only 0.73 wt% in NU. There are far greater reserves of the fertile isotopes ^{232}Th and ^{238}U, than the fissile isotope ^{235}U. Obviously, the most conservative utilization of ^{235}U necessitates a closed fuel cycle based on breeder reactor technologies.[c] The energy density differential between carbon fuels and nuclear fuels is many orders of magnitude, and due to its extremely high energy density, nuclear energy produces a correspondingly low mass of radionuclide waste. Waste minimization strategies are mostly concerned with the exclusion of long-lived radioisotopes and the stabilization of the waste forms designed for geologic repositories.

Nuclear fuel reprocessing technologies are designed to support nuclear fuel cycle objectives. Chemical separations lie at the heart of any reprocessing technology. The goal of chemical separations is to selectively separate the elements to be retained in the fuel cycle from the elements to be rejected from the fuel cycle as waste. Generally, all fissile elements are retained, all or a fraction of the fertile elements are retained, and all other fission product elements are rejected. However, the exact goal of chemical separations is largely dependent on the exact objectives of the fuel cycle. For example, in some fuel cycle scenarios the minor actinides are retained in the fuel cycle to lessen the radioisotope burden of these elements reporting to the process waste streams.

It is important to realize that there are a great variety of reactor designs and, consequently, an even greater variety of nuclear fuel types, as in some cases a single reactor design can utilize several different fuel types. The reasons for these great varieties of reactors and fuels are many fold and stem, fundamentally, from the great many varieties of size, application, location, and fuel cycle requirements. Sizes range from kWth to multiples of MWth. Applications include the production of neutrons, isotopes, heat, thrust, and electrical power. Locations are based on land, sea, air, and space. And fuel cycle objectives include open fuel cycles and many variants of closed fuel cycles, including breeding ^{239}Pu from ^{238}U, breeding ^{233}U from ^{232}Th, burning weapons grade plutonium, and burning minor actinides.

The public and governmental perception of nuclear energy was severely negatively impacted by the accidents at Three Mile Island (1979), Chernobyl (1986), and Fukushima (2011), and by the legacy nuclear wastes generated during the U.S./USSR cold war at sites like Hanford, Savannah River, and the Nevada Test Site. To make matters worse, nuclear wastes continue to be generated by the U.S. civilian energy reactor fleet and the U.S. military naval reactor fleet with no commercial reprocessing capabilities and no geologic repository to serve as the final destination for these materials. As a result of this history, safety and waste generation considerations are, more than ever before, the factors most significantly

c. During the early years of nuclear development in the 1940s to the 1960s, minable uranium reserves were considered to be very limited and mush attention was focused on breeder reactor technologies.

influencing reactor and fuel design requirements, particularly in the arena of civilian power production, which is the greatest application of nuclear energy.

2.1 Mining and Extraction of Uranium and Thorium

Uranium and thorium are recovered from the earth's crust via various mining and extraction methods.[3-8] In its raw form, refined uranium is recovered from ore as a mill concentrate called "yellow cake." The name is derived from its bright yellow color, and the fact that it is recovered as a vacuum-filter cake following precipitation during the final stages of the solvent extraction operations. Yellow cake has a process-dependent complex composition, but it is mostly ammonium diuranate ($(NH_4)_2{\bullet}U_2O_7$) with minor amounts of other uranium compounds. Subsequent purification and controlled calcination are required to convert yellow cake into nuclear-grade U_3O_8 or UO_2. Different process chemistries, but similar methodologies, are used to produce nuclear-grade ThO_2 from ores.

If the yellow cake, natural uranium oxide, or thorium oxide are to be used in any other forms, these materials must be converted by additional chemical processing such as reduction to metals, constitution into alloys, and conversion to carbides, nitrides, fluorides, chlorides, etc. When enriched uranium is required, the yellow cake is converted to uranium hexafluoride (UF_6) at a "conversion plant" by the action of hydrogen fluoride (HF). And the UF_6 is subsequently enriched at an "enrichment plant" by processes of diffusion-based or centrifuge-based isotopic enrichment.

2.2 Uranium, Lithium, Chlorine, and Nitrogen Enrichment

As stated earlier, NU is nominally 0.73 wt% ^{235}U with the balance ^{238}U. Although NU is used to fuel certain types of reactors, it is also the practice to use enriched uranium. The levels of uranium enrichment (with respect to wt% ^{235}U) are categorized as depleted uranium (DU $<$ 0.73), natural uranium (NU = 0.73), low enriched uranium (0.73 $<$ LEU $<$ 20), and high enriched uranium (HEU $\geq$ 20). High assay low enriched uranium (5 $<$ HALEU $<$ 20) is another category of interest for use in research reactors and the development of technologies like small modular reactors (SMRs).

The two most common technologies used for uranium enrichment are gaseous diffusion and centrifuging. In preparation for these processes, NU is purified and converted to uranium hexafluoride (UF_6), which is gaseous at moderate temperatures and pressures. Natural fluorine is isotopically pure ^{19}F. UF_6 from ^{238}U is 0.86 wt% heavier than UF_6 from ^{235}U. This small mass difference is exploited by the enrichment technologies to create two UF_6 product streams, one enriched with respect to ^{235}U and one depleted with respect to ^{235}U. The enriched UF_6 is converted to metallic uranium or uranium oxide depending on its intended use. These technologies can produce enrichment levels greater than 93 wt% ^{235}U. The U.S. has large stockpiles of depleted UF_6 stored in steel cylinders because, historically, the supply of DU is much greater than the demand for DU. The U.S. had gaseous diffusion plants at Paducah, Kentucky; Piketon, Ohio; and Oak Ridge, Tennessee. These plants are now shut down. Gaseous diffusion technologies have been supplanted by centrifuge technologies. A centrifuge cascade is shown in Figure 1. Variants of laser-based technologies, which are a third means of enriching uranium, are possibly gaining economic advantage.[9-11] The following is a list of historic enrichment plants in the U.S.

- K-25, K-27, K-29, K-31, and K-33 Plants: These were gaseous diffusion plants located at Oak Ridge National Laboratory (ORNL), Oak Ridge, Tennessee. They produced HEU and operated from 1944 to 1985. All plants were completely dismantled by 2017.
- Paducah Gaseous Diffusion Plant, Paducah, Kentucky. Produced LEU that was further refined to HEU at the Portsmouth and Oak Ridge plants and operated from 1952 to 2013.
- Portsmouth Gaseous Diffusion Plant (a.k.a., A-Plant), Piketon, Ohio. Produced HEU and operated from 1956 to 2001.
- S-50 Plant: Liquid Thermal Diffusion Plant. Produced HEU and operated from 1942 to 1946.

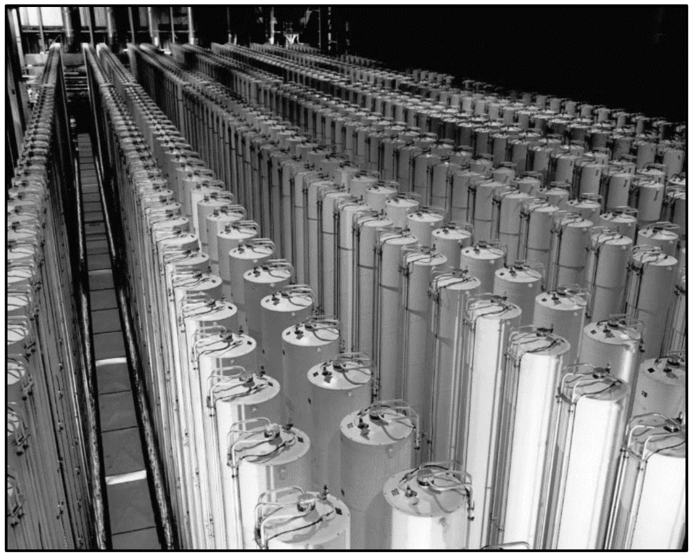

Figure 1. Cascade of gas centrifuges at Piketon, Ohio. DOE.

- Y-12 Plant: Used calutron technologies to perform isotopic separations based on electromagnetic principles. Produced HEU and other isotopes. Included 1152 calutrons arranged in nine Alpha and eight Beta *"racetracks."* The product of the Alpha process was feed to the Beta process. Operated from 1943 to 1946 when all Alpha and six Beta racetracks were dismantled. The Beta-3 racetrack still exists.

There are other elements whose enrichments are necessary to support molten salt reactor (MSR) technologies: lithium and chlorine. Lithium is proposed for use in fluoride salt MSRs as lithium fluoride (LiF). Natural lithium is 7.5 wt% ^{6}Li with the balance ^{7}Li. Interestingly, ^{6}Li is the only stable light element that can produce net energy through fission. The fission of ^{6}Li by neutron capture produces ^{4}He and ^{3}H. MSR applications desire lithium enriched with respect to ^{7}Li to suppress the production of tritium. For example, the ORNL molten salt reactor experiment (MSRE) used lithium that was 99.99 wt% ^{7}Li.[12,13] The ^{7}Li used in the MSRE was the biproduct of former ^{6}Li enrichment activities at ORNL for use in nuclear weapons.[14]

MSR concepts also include chloride salts. Natural chlorine is 75.8 wt% ^{35}Cl with the balance ^{37}Cl. The capture cross section of ^{35}Cl is 10 times greater than that of ^{37}Cl. Therefore, ^{35}Cl has a significant effect on the neutron economy within the core. Also, ^{35}Cl transmutes to ^{36}Cl by $^{35}Cl(n,\gamma) = {}^{36}Cl$, capturing a neutron and ejecting a gamma ray, which is a long-lived radionuclide with high energy beta emissions. The former issue must be considered in reactor design and application, and the latter in salt waste management. Therefore, chloride salt MSRs will desire chlorine enriched with respect to ^{37}Cl. However, this remains conceptual as there have been no chloride salt MSRs operated.

Nitride-based ceramic fuels require enrichment with respect to ^{15}N.[15-17] Natural nitrogen is 99.6 wt% ^{14}N and 0.4 wt% ^{15}N. In the presence of thermal neutrons, ^{14}N transmutes to ^{14}C by $^{14}N(n,p) \rightarrow {}^{14}C$ (capturing a neutron and ejecting a proton). The capture cross section of ^{14}N is five orders of magnitude greater than that of ^{15}N. This (n,p) reaction affects the neutron economy of the core and produces the undesirable radioisotope ^{14}C as a byproduct. The same (n,p) reaction occurs in earth's upper atmosphere and is a natural source of ^{14}C, which has a half-life of about 5,700 years; much too short to otherwise be a naturally occurring isotope if there was not a natural mechanism for its generation.

Isotopic enrichment of any kind is a costly process, which is one of the reasons why much consideration is given to fuel cycle scenarios involving the recovery of ^{235}U. Another reason is the conservation of ^{235}U as a natural resource. Since there are no natural resource shortages of lithium and chlorine, in MSR applications the decision to recycle ^{7}Li and ^{37}Cl is purely based on technical and economic considerations. Likewise, for the use of ^{15}N in nitride fuels.

2.3 Hydrogen

The three most significant hydrogen isotopes are ^{1}H (protium), ^{2}H (deuterium), and ^{3}H (tritium). Natural hydrogen is 99.99 wt% ^{1}H and 0.01 wt% ^{2}H. Heavy water is water enriched with respect to ^{2}H.[18-20] As a neutron moderator in nuclear reactor applications, heavy water can be enriched up to 99.75% ^{2}H and it is used in reactors that operate on NU. In contrast, light water is simply natural water.

Like ^{14}C, ^{3}H occurs naturally only in trace quantities from the transmutation of ^{14}N in earth's upper atmosphere ($^{14}N + n \rightarrow {}^{12}C + {}^{3}H$). The half-life of ^{3}H is about 12.3 years. There are many mechanisms, both intentional and unintentional, that produce ^{3}H in nuclear reactors.[21,22] If the purpose of the reactor is to generate electrical energy, then the goal is generally to minimize tritium production. However, a certain degree of tritium production is inevitable, and it is impossible to completely contain tritium within the reactor system. The most detrimental aspect of tritium release is its mobility in the natural environment, particularly when it exists as tritiated water.

The mechanisms for generating tritium in a reactor include the following transmutation reactions, which highlight the inevitability of experiencing some degree of tritium formation.

- Ternary Fission. ^{3}H is produced as a ternary fission product, i.e., when a nucleus fissions into three particles. The fission formation of ^{3}H is favored by heavy fissile isotopes and hard neutron spectrums. Therefore, this mechanism is more pronounced in fast reactors than thermal reactors.
- $^{2}H(n,\gamma) \rightarrow {}^{3}H$. Deuterium exists in heavy water moderated reactors and, to a lesser extent, in any light water reactor or water-cooled reactor.
- $^{3}He(n,{}^{1}H) \rightarrow {}^{3}H$. Helium is present in some gas-cooled reactors.
- $^{6}Li(n,{}^{4}He) \rightarrow {}^{3}H$ and $^{7}Li(n,{}^{4}He) \rightarrow {}^{3}H$. Lithium is present in MSRs and as an impurity in graphite.
- $^{10}B(n,{}^{4}He,{}^{4}He) \rightarrow {}^{3}H$ and $^{10}B(n,{}^{4}He) \rightarrow {}^{7}Li(n,{}^{4}He) \rightarrow {}^{3}H$. Boron is used in control rods, as a cooling system chemical additive in water-cooled reactors, and as an impurity in graphite.
- $^{12}C(n,{}^{4}He) \rightarrow {}^{9}Be(n,{}^{4}He) \rightarrow {}^{6}Li(n,{}^{4}He) \rightarrow {}^{3}H$. Carbon is used as a moderator and/or fuel matrix in some reactors.

An important fusion reaction involving hydrogen isotopes that generates both energy and neutrons for applications in fusion reactors and thermonuclear weapons is $^{2}H + {}^{3}H \rightarrow {}^{4}He(n)$. Therefore, ^{3}H production supports these research and military sectors. Furthermore, weapons physics utilize ^{6}Li (in the form of solid lithium deuteride) for the in situ generation of ^{3}H to enhance the $^{2}H/^{3}H$ fusion reaction for the purpose of generating additional fast neutrons capable of increasing the fission yields of ^{239}Pu and ^{238}U.[23]

2.4 Krypton, Xenon, and Iodine Fission Products

Krypton, iodine, and xenon are generated as fission products. And like ^{3}H, all are potentially highly mobile in the natural environment; krypton and xenon because they are noble gases, and iodine because it is an essential bio-nutrient (e.g., accumulated in the thyroid in humans). Krypton and xenon each have several stable isotopes; iodine has only one, ^{127}I. The longest-lived radioisotopes of these elements are ^{85}Kr (10.76y), ^{127}Xe (36.4d), and ^{129}I (1.57E7y). Radioisotopes with short half-lives cause few chronic environmental effects simply by virtue of their not remaining in the environment long enough to cause greater harm. In this regard, the long-lived radioisotopes are of greater concern and care must be taken to assure their containment. For example, the release of ^{129}I is likely to be a much more serious event than the equivalent release of ^{127}Xe.

Xenon is of particular interest in another way unrelated to spent fuels. ^{135}Xe has a very high neutron capture cross section and because of this xenon management is an important aspect of reactor operations with regards to neutron economy and core reactivity. ^{135}Xe is produced directly as a fission product, and also by the decay of other fission products such as ^{135}Te and ^{135}I according to the following decay series[24]:

$$^{135}\mathrm{Te}(\beta\text{-})11\mathrm{s} \rightarrow {}^{135}\mathrm{I}(\beta\text{-})6.7\mathrm{hr} \rightarrow {}^{135}\mathrm{Xe}(\beta\text{-})9.2\mathrm{hr} \rightarrow {}^{135}\mathrm{Cs}(\beta\text{-})2.3\mathrm{E}6\mathrm{yr} \rightarrow {}^{135}\mathrm{Ba}\ \mathrm{stable}$$

While the reactor is operating, two factors limit the ^{135}Xe equilibrium concentration: the natural decay of ^{135}Xe to ^{135}Cs (shown above), and the neutron capture of $^{135}Xe(n) = {}^{136}Xe$ stable. However, following reactor core shutdown, the ^{135}Xe concentration will temporarily spike as the latter mechanism is extinguished and the decay of ^{135}I continues and dominates. However, the short half-life of ^{135}Xe renders it much less of an environmental concern than its decay product ^{135}Cs.

Spent nuclear fuel contains many other long-lived radioisotopes, but these are less mobile in the natural environment than krypton, xenon, and iodine. Notable long-lived fission product radioisotopes include ^{79}Se (3.5E5y), ^{90}Sr (28.9y), ^{93}Zr (1.5E6y), ^{99}Tc (2.13E5y), ^{107}Pd (6.5E6y), ^{135}Cs (2.3E6y), and ^{137}Cs (30.2y). ^{131}I (8d) is of concern in, for example, fallout from a nuclear weapon detonation or a reactor core failure, but its half-life is too short to be of concern in typical spent nuclear fuel.

2.5 Burnable Poisons/Neutron Absorbers

The function of control rods was briefly discussed earlier and the isotopes that act as burnable poisons are related to this topic. Burnable poisons are simply neutron absorbers which transmute into isotopes that may or may not themselves be neutron absorbers. If not, the neutron absorption process ends; if so, it continues. In this scheme the neutron capture cross section of the poison should be large, and that of the resulting terminal transmutation product should be small. This transition from large to small capture cross sections does not necessarily occur in one transmutation cycle, as will be discussed shortly. If the reactor control scheme uses poisons, for example, in the discrete control rod assemblies or in the cooling water, then these materials are generally isolated from the fuel assemblies and the subsequent reprocessing operations. However, poisons are sometimes intimately incorporated into the fuel assemblies, in which case they become part of the separations burden of the reprocessing operations.

Generally, the function of burnable poisons is to control the reactivity of the core. However, in some reactors the core is refueled periodically and incrementally. In others the core is refueled all at once, and yet in others the core is never refueled because it is intended to last for the lifetime of the reactor. In each of these cases, burnable poisons can play an important role in managing the core reactivity. As fission continues, the fissile inventory of the core is depleted, and to maintain fission chain reactions, the reactivity of the core must be increased. Control rods can only provide so much reactivity control due to engineering and physics limitations of these mechanical systems. To compensate, burnable poisons can provide additional reactivity control via in situ placement in the core. As the fissile inventory is decreased, core reactivity is maintained by the consumption of the poisons, which in some regards is functionally equivalent to the slow removal of control rods from the core.

Elements are sometimes used as burnable poisons in their natural isotopic abundances, and sometimes burnable poisons are enriched with respect to select stable isotopes. Isotopes that are exploited as burnable poisons are listed below.[25] The first isotope in each series is naturally occurring, i.e., stable, or extremely long half-life. In each series transmutations and decays lead to isotopes with smaller neutron capture cross sections; in other words, the poison is consumed or "burned." Of course, the behaviors of burnable poisons are more nuanced than what is depicted here; but the principle of performance is adequately illustrated.

- $^{10}B(n,{}^{4}He) \rightarrow {}^{7}Li$
- $^{113}Cd(n,\gamma) \rightarrow {}^{114}Cd$
- $^{149}Sm(n,\gamma) \rightarrow {}^{150}Sm$

- $^{151}Eu(n,\gamma) \rightarrow {}^{152}Eu(n,\gamma) \rightarrow {}^{153}Eu$
- $^{153}Eu(n,\gamma) \rightarrow {}^{154}Eu(n,\gamma) \rightarrow {}^{155}Eu(n,\gamma) \rightarrow {}^{156}Eu(\beta\text{-})$ 15.2d $\rightarrow {}^{156}Gd$
- $^{155}Gd(n,\gamma) \rightarrow {}^{156}Gd$
- $^{157}Gd(n,\gamma) \rightarrow {}^{158}Gd$
- $^{164}Dy(n,\gamma) \rightarrow {}^{165}Dy$
- $^{167}Er(n,\gamma) \rightarrow {}^{168}Er$
- $^{175}Lu(n,\gamma) \rightarrow {}^{176}Lu$
- $^{176}Lu(n,\gamma) \rightarrow {}^{177}Lu(n,\gamma) \rightarrow {}^{178}Lu(n,\gamma) \rightarrow {}^{179}Lu(\beta\text{-})$ 4.6h $\rightarrow {}^{179}Hf$
- $^{177}Hf(n,\gamma) \rightarrow {}^{178}Hf$
- $^{180}Hf(n,\gamma) \rightarrow {}^{181}Hf(\beta\text{-})$ 42.4d $\rightarrow {}^{181}Ta$.

2.6 Neutron Moderators and Reflectors

Materials that are good moderators and reflectors will elastically interact with neutrons. That is, the materials are likely to absorb a fraction of the neutron's energy while not capturing it. In other words, these materials exhibit low neutron capture cross sections and high neutron scattering cross sections. Moderators are placed in the reactor to control the energy of the neutron spectrum within the core; reflectors are placed to keep neutrons in the core. There are several light elements that act as neutron moderators and reflectors. These include the hydrogen nuclides ^{1}H and ^{2}H (previously discussed), ^{9}Be (the only natural occurring beryllium isotope),[26] and carbon (naturally occurring carbon is 98.9 wt% ^{12}C with the balance ^{13}C). These materials can be present in their pure forms, or as oxides, hydrides, and carbides. Like burnable poisons, moderators and reflectors may or may not be intimately associated with the fuel assemblies. Stainless steels are also adopted as reflector materials.

3. NUCLEAR REACTORS

A unique feature of nuclear reactors is the great latitude of options available for their design. From a high-level perspective, the process of engineering design begins by defining the functional requirements of the engineered system, which, in turn, begins to constrain and limit the engineering design options. Nuclear reactors serve two overarching purposes: the production of useful heat and/or the production of useful radiation. The heat generated from a nuclear reactor can be used for the same purposes as the heat generated from the combustion of coal, fuel oil, natural gas, etc. There is nothing unique about the heat generated from a nuclear reactor, other than how the heat is generated, which is by nuclear reactions (e.g., fission) as opposed to chemical reactions (e.g., combustion). The radiation can be used for transmutation, medical and industrial radioisotope production, materials irradiation, materials interrogation, etc. Therefore, both the functional requirements and, consequently, the designs of nuclear reactors are varied and wide.

Another significant design factor is the selection of coolant. The processes of fission, transmutation, and radioactive decay release energy, manifested as heat, into the reactor core. This heat must be extracted from the core by a suitable coolant. Common coolants are water (e.g., light water and heavy water),[27] liquid metals (e.g., sodium, NaK-alloy, lead, and PbBi-alloy),[28] gases (e.g., air, carbon dioxide, and helium),[29] and molten salts (e.g., fluoride and chloride salts).[30,31] There are also design concepts incorporating water and carbon dioxide coolants as supercritical fluids. Some coolants serve multiple purposes, performing the function of moderator, fuel, blanket, or some combination of these.

Of the 1,000 different types of reactors proposed, 100 were built and tested, and 10 have found commercial success. Of course, this is only an anecdotal statement made for effect. However, it does highlight the fact that, of the seemingly limitless number of imaginable reactor designs, few have ever come to fruition, and fewer still have found commercial success. It is evident from the preceding discussions that design options for nuclear reactors are nearly unbounded. This is illustrated in Table 3, which lists several engineering options for different major design requirements. For example, *Function* is a major design feature, and several choices, such as *Electrical Power*, are listed in the adjoining column. The choices are not necessarily independent of each other, but even at this high-level it is apparent that there are a great many design options, and when the many underlying layers of technical detail are considered, the design options grow exponentially. That is why, during the early development of nuclear reactors, a great many distinct reactor concepts were pursued simultaneously. It was simply impossible at that time to predict which of the many avenues of research would prove successful.

Table 3. Summary of High-Level Nuclear Reactor Design Options.

Design Requirement	Engineering Options
Primary Function	Electrical Power Generation
	Military, Medical, and Industrial Isotope Production
	Naval Propulsion
	Irradiation Experiments
	Weapons Plutonium Production
	Industrial Process Heat
Secondary Function	Plutonium production reactors can generate electrical power
	Electrical power reactors can produce plutonium and isotopes
	Other Dual Use
Conversion Ratio (CR)	Breeder (CR > 1)
	Converter (CR = 1)
	Non-Breeder or Burner (CR < 1)

Design Requirement	Engineering Options
Conversion Cycle	$^{232}Th/^{233}U$
	$^{238}U/^{239}Pu$
	No Conversion Cycle
Neutron Spectrum	Thermal
	Epithermal
	Fast
	Variable Thermal or Fast Biased
Neutron Moderator	Graphite
	Water (e.g., heavy water or light water)
	Ceramic (e.g., beryllia)
	Organic Liquid
	No Moderator
Primary Coolant	Water (e.g., heavy water or light water)
	Gas (e.g., helium, air, carbon dioxide)
	Liquid Metal (e.g., sodium, lead, lead/bismuth alloy)
	Molten Salt (e.g., fluoride salt or chloride salt)
	Organic Liquid
	No Coolant (e.g., radiant energy release)
Secondary Coolant	Light Water
	Molten Salt
	No Secondary Coolant
Major Fissile Component	^{235}U as NU
	^{235}U as LEU
	^{235}U as HEU
	^{233}U
	^{239}Pu
	Minor Actinides
Fuel Form	Metallic
	Metallic Alloy
	Oxide
	Mixed Oxide
	Nitride
	Hydride
	Carbide/Oxy-carbide
	Molten Salt
Fuel Cladding	Ferro Alloy
	Zirconium Alloy
	Nickel Alloy
	Aluminum Alloy

Design Requirement	Engineering Options
	Magnesium Alloy
	Silicon Carbide
	No Cladding (e.g., liquid fuels)

A *breeder reactor*, on a net basis, produces more fissile inventory by transmutation than it consumes by fission. In other words, a breeder reactor produces excess fissile inventory, which can be recovered to fabricate new fuel for itself and for additional nuclear reactors. In contrast, a *burner reactor*, on a net basis, consumes more fissile inventory by fission than it produces by transmutation. The objective of a burner reactor is to consume, as much as possible, its initial fissile inventory and to minimize transmutation processes. There are only a few operating breeder reactors around the world; the majority of operating reactors are burner reactors. A *converter reactor*, on a net basis, also consumes more fissile inventory by fission than it produces by transmutation. However, the objective of a converter reactor is to exploit, as much as possible, transmutation processes to breed and burn fissile inventory simultaneously. *Conversion ratio* is the ratio of fissile inventory produced to fissile inventory consumed. The conversion ratios of breeder and burner reactors are greater than one and less than one, respectively. The conversion ratio of a converter reactor is also less than 1.

The major avenues pursued during early research and development into reactors for civilian power production included light water reactors (LWRs) (e.g., pressurized water reactors (PWRs) and boiling water reactors (BWRs)), heavy water reactors (HWRs) (e.g., pressurized heavy water reactors (PHWRs)), fluid flow reactors (FFRs) (e.g., MSRs, molten salt breeder reactors (MSBRs), aqueous homogeneous reactors (AHRs), and lead-cooled fast reactors (LFRs)), and liquid metal-cooled reactors (LMRs) (e.g., sodium-graphite reactors (SGRs), sodium-graphite breeder reactors (SGBRs)), liquid metal fueled reactors (LMFRs), and liquid metal fast breeder reactors (LMFBRs)). Today, PWRs dominate the application of civilian power production. But not because PWRs are the optimal design for this application, but because PWRs were viewed as the least-risk design for successful demonstration of commercial-scale civilian power production at a key point in U.S. history.[32] This perception was a consequence of the U.S. Navy's successful development and adoption of PWRs for naval propulsion.

Since the 1940s, a great many nuclear reactors have been operated in the U.S. A single comprehensive list of all U.S. reactors would include the following: over 130 commercial civilian power reactors; 52 research reactors at the Idaho National Laboratory (INL) Site;[33-35] 28 research reactors designed, built, and operated by Argonne National Laboratory (ANL);[33,34,36,37] 13 research reactors at ORNL;[38] 9 research reactors at Los Alamos National Laboratory (LANL);[39-41] additional commercial and university research reactors; 9 plutonium production reactors at Hanford Site (HS);[42-46] 5 plutonium production reactors at Savannah River Site (SRS);[43,47] and numerous military reactors for training, naval propulsion, and power production. A series of such lists were compiled between 1959 and 1996.[48,49]

4. NUCLEAR FUELS

As stated earlier, the process of fission generates fission products (lighter elements), transmutation products (lighter and heavier elements), and heat (mostly in the form of the kinetic energies of the fission fragments). Fission product elements and isotopes range across most of the periodic table. Exactly which fission products are generated is a complex function of the nucleonic physics within the core; but these relationships are understood and are well within the realm of computer simulation and modeling capabilities. Because there is a wide spectrum of fission product elements, there is an equally wide spectrum of fission product element physical-chemical properties. For example, there are noble gases, elements and compounds with low melting temperatures, and elements and compounds with high vapor pressures at the operating temperatures of the reactor. Managing this wide variety of physical-chemical properties is the source of one of the main challenges of fuel design.

In a broad sense, all fuels can be categorized into one of two groups: fuels that are contained within cladding, and fuels that are not contained with cladding. Examples of the former are LWR uranium oxide fuels contained in zirconium alloy cladding, and SFR metal fuels contained in stainless steel cladding. Certainly, the majority of operating reactors have employed contained fuels. Examples of the latter are molten salt fuels and liquid-media dispersion fuels; these are concepts that have been tested experimentally but have not yet been adopted for commercial or military applications. A reactor core must also serve as a heat exchanger. Typically, the heat generated within the core is transferred to a fluid that carries the heat from the core. Contained fuels are typically stationary within the core, as the heat exchange fluid flows through the core. Heat exchange fluids include water, heavy water, sodium, lead alloys, molten salt, air, carbon dioxide, and helium. The first two fluids serve the dual purpose of moderator. Uncontained fuels typically serve the dual purpose of fuel and heat exchange fluid. Heat is intrinsically generated as the fluid fuel flows through the core and carries the heat with it as it flows from the core. Additional requirements are that these materials must be chemically compatible. In the case of contained fuels, the inside of the cladding must be compatible with the fuel, and the outside of the cladding must be compatible with the heat exchange fluid. In the case of uncontained fuels, the fluid fuels must be compatible with the materials of construction of the reactor core, fluid pumps, and the subsequent primary heat exchangers.

Fuel systems experience extreme materials challenges during irradiation. For example, fission processes are not uniform throughout the fuel element, much less the entire reactor core. The compositional changes due to fission are spatially dependent and, consequently, create chemical compositional gradients. In addition, heat generated from within the fuel element creates temperature gradients which can be quite extreme in some cases. Based largely on fuel symmetry, the highest temperatures are experienced at the most central locations. For example, the highest temperature within a typical uranium oxide fuel pellet can reach in excess of 1,200°C in a PWR, and 1,800°C in a Canadian deuterium uranium (CANDU) reactor, while the outer Zircalloy cladding surface is in contact with pressurized water that is less than 400°C in most cases. Like fission, temperature gradients also cause chemical compositional gradients by processes such as chemical diffusion and phase segregation. However, these extreme temperatures are not always the case. The design thermal power and coolant temperature of the reactor, and the thermal conductivity of the fuel will influence the fuel temperature.

Fission gas generation and radiation damage also contribute to chemical and mechanical changes within the fuel. These effects are highly dependent on the fuel characteristics and the radiation history. For example, solid fuels can swell (due to fission gas generation) and develop chemical/physical properties gradients (due to thermal gradients) and interact with the cladding materials. The fission products in liquid fuels are free to migrate through the system and chemically interact with the materials of construction.

Thermal power and cooling are intimately related. Thermal power is a measure of the rate at which thermal energy is produced in the core, e.g., units of MW. At steady state, the thermal power is dissipated from the core by the flow of coolant at the same rate it is produced. The required flowrate of coolant is dictated by the thermal power, the efficiency of the core to function as a heat exchanger, the density and heat capacity of the coolant, and the design inlet and outlet temperatures of the coolant. These parameters are not all independent of each other. And the efficiency of the core to function as a heat exchanger is intimately related to fuel design. Coolants such as water, liquid sodium, molten salt, and helium all have unique densities and heat capacities that dictate their mass and volumetric flowrates for their intended applications.

From the preceding discussions it is evident that there are many factors affecting reactor, fuel, and fuel cycle designs. Other competing factors include resource conservation, societal acceptance, and public safety, which further complicates and sometimes dominates the fuel cycle design arguments.

The following sections provide brief descriptions of the major fuel types and the reactors that use them, of which there are many, and the reprocessing technologies that are applied to, or have been proposed for, their treatment. Instances where reprocessing considerations are absent are noted. Where such distinctions are possible, what is or has been done at the industrial scale to support commercial or military activities, what is or has been done at the laboratory scale in the course of research and development activities, and what is or has been merely conceptualized and proposed are noted. The order in which the following information is presented is not a reflection of the chronology in which the technologies were developed.

4.1 Nuclear Fuel Taxonomy

Why? There are many types of reactors for many types of applications developed at different stages of technical understanding during different socio-politico conditions. Therefore, there are many types of fuels. It is useful to have a system for categorizing fuels according to similarities in material characteristics.

The intent here is to provide a useful means of categorizing fuel types according to their as-fabricated material characteristics and subsequent incorporation into fuel forms. These are important considerations for selecting and designing applicable spent fuel reprocessing technologies. Although the details vary from case to case, the following categories are those that appear to have a shared agreement in the technical literature.

The three broadest categories are ceramic, metallic, and halide salt, which are summarized in Table 4 to Table 6, respectively. These summaries are selective, not comprehensive. Ceramics include oxides, carbides, nitrides, hydrides, silicides, and composites of these. Metallics include pure metals, alloys, and intermetallics, and halides include fluoride and chloride salts. These subcategories are summarized in Column 1 of the tables. Two other fuel systems worth mentioning, but not included in the tables, are aqueous and liquid metals.[50] Aqueous fuel systems relied on fertile and fissile materials dissolved in aqueous media, e.g., sulfates, nitrates, fluorides, phosphates, chromates, and carbonates. Liquid metal fuel systems were mostly based on dispersion fuel concepts in which insoluble compounds like UO_2, UC, UF_3, $ThBi_2$, and ThO_2 were dispersed in liquid metals like bismuth and NaK. Presently, there does not appear to be any active research in these two areas.

Column 2 of Table 4, Table 5, and Table 6, provides examples of the basic constituent material forms containing uranium, plutonium, thorium (fertile), and minor actinides (MA). Only the basic compositions are shown without the stoichiometries, which are quite variable in some cases. Furthermore, in many cases these material forms are not complete descriptions of the fuel forms. For example, uranium oxide (fuel material) is pressed and sintered into high-density ceramic pellets (fuel form) for use in LWRs. This is an example of a homogenous fuel; the ceramic pellet is made of uranium oxide. On the other hand, uranium oxide (fuel material) is formed into small spherical kernels, coated with layers of pyrolytic graphite and silicon carbide to form tri-structural isotropic (TRISO) fuel particles that are imbedded in graphite (fuel form) for use in high-temperature reactors (HTRs). This is an example of a heterogeneous fuel: the dispersed TRISO particles contain kernels of uranium oxide. With other fuel types the distinction between homogeneous and heterogeneous is less clear and not particularly important.

In Table 4 to Table 6, Column 3 (subcategorized into Columns A to K) provides more information about the fuel forms. The meanings of the letters in Column 3 are described in Table 7. This table provides examples of how the materials in Column 2 are incorporated into fuel forms. From the point of view of reprocessing, these tables indicate the material forms from which select elements must be separated. The final Columns in Table 4 to Table 6 indicate if the fuel forms are free of uranium. These uranium-free fuel forms contain only plutonium and minor actinides and require special considerations as fuels with additions of neutron absorbers and burnable poisons.

Table 4. Ceramic Fuels

		Fuel Form Designation											Uranium Free?
Subcategory	Example	A	B	C	D	E	F	G	H	I	J	K	
Oxide	(U)O	A			D				H	I			
	(Pu)O	A			D			G	H				Y
	(U,Pu)O	A							H				
	(Th)O	A							H				
	(U,Th)O	A							H				
	(Pu,Th)O	A											
	(U,Pu,Th)O	A											
	(Pu,Zr)O							G					Y
	(Pu,Zr,Y)O							G					Y
	(Pu,Ce)O							G					Y
	(U,Pu,MA)O	A											
	(Pu,MA)O							G					Y
Carbide	(U)C	A				E			H				
	(Pu)C	A				E							Y
	(U,Pu)C	A				E							
	(Th,U)C	A							H				
	(Th,Pu)C	A											
Oxy-carbide	(U)CO	A							H				
	(Pu)CO	A											Y
	(U,Pu)CO	A							H				
	(U,Th)CO	A							H				
Carbo-nitride	(U,Pu)CN	A											
Nitride	(U)N	A					F						
	(Pu)N	A					F						Y
	(Th)N	A											
	(U,Pu)N	A											
	(U,Th)N	A											

Subcategory	Example	Fuel Form Designation A	B	C	D	E	F	G	H	I	J	K	Uranium Free?
	(U,Zr)N	A											
	(Pu,Zr)N						F						Y
	(Pu,MA,Zr)N						F						Y
Oxy-nitride	(U)NO	A											
	(U,Pu)NO	A											
Hydride	(U)H	A											
	(Pu)H	A											Y
	(U,Pu)H	A											
	(U,Zr)H	A											
	(Pu,Zr)H	A											Y
	(U,Pu,Zr)H	A											
	(Th,U,Zr)H	A											
	(Th,Pu,Zr)H	A											
	(Th,Zr)H	A											
Silicide	(U)Si	A		C									
	(U,Al)Si	A		C									

Table 5. Metallic Fuels

Subcategory	Example	Fuel Type Designation A	B	C	D	E	F	G	H	I	J	K	Uranium Free?
Pure	U		B										
	Pu		B										
Aluminum	(U)Al			C									
	(Pu)Al			C									Y
Zirconium	(U)Zr		B										
	(Pu)Zr		B										Y
	(U,Pu)Zr		B										
	(Pu,MA)Zr		B										Y
Yttrium	(Pu)Y			C									Y
	(Pu,MA)Y			C									Y
Molybdenum	(U)Mo			C									
Iron	(U)Fe			C									

Table 6. Halide Salt Fuels

Subcategory	Example	Fuel Type Designation											Uranium Free?
		A	B	C	D	E	F	G	H	I	J	K	
Fluoride	(U)F										J		
	(Pu)F										J		Y
	(U,Pu)F										J		
	(Th)F										J		
	(Th,U)F										J		
	(Th,Pu)F										J		
	(Th,U,Pu)F										J		
Chloride	(U)Cl											K	
	(Pu)Cl											K	Y
	(U,Pu)Cl											K	

Table 7. Dispersion Matrices

Designation	Matrix Category	Examples of Matrix Materials
A	None	Used directly in ceramic form.
B	None	Used directly in metallic or alloy form.
C	Metal Matrix	Al, Zr, Mo, Mg, and stainless steel.
D	Ceramic Oxide Matrix	BeO, MgO, ZrO_2, and CeO_2.
E	Ceramic Carbide Matrix	ZrC and SiC.
F	Ceramic Nitride Matrix	TiN, ZrN, AlN, and Si_3N_4.
G	Ceramic Spinel Matrix	$MgAl_2O4$, $Y_3Al_5O_{12}$, yttria-stabilized zirconia (YSZ), with Er_2O_3, Gd_2O_3.
H	Coated Particle	Porous carbon, pyrolytic carbon, SiC, and ZrC.
I	Graphite Matrix	Graphite
J	Fluoride Molten Salt	LiF, NaF, ZrF, KF, and BeF_2.
K	Chloride Molten Salt	NaCl, KCl, LiCl, and $MgCl_2$.

The information provided in Table 4 to Table 7 demonstrates the complexity and vast variety of fuel types and fuel forms that have been or are being considered for use in a multitude of nuclear reactor types. In many ways it is the detailed characterization of the spent fuel that is the starting point for selecting and designing an applicable reprocessing technology. From strictly an engineering perspective, it is always possible to envision a technical means of recovery and purification of fissile materials from any type of spent fuel. However, a viable process requires more than technical merit, it also requires economic justification, public acceptance, and political support.

4.2 Natural Uranium Oxide Fuel

Why? The manufacture of natural uranium oxide fuel requires the least amount of processing. No uranium enrichment is required. Reactor grade NU oxide is pressed into dense cylinders and loaded into metal cladding. However, the reactors require heavy water or graphite for moderation.

CANDU reactors were developed in Canada for civilian energy production. These are pressurized, heavy water moderated, thermal spectrum reactors. Standard CANDU fuel is based on NU that is refined into high purity uranium oxide and manufactured into ceramic pellets. The pellets are loaded into Zircaloy fuel cladding and arranged into fuel bundles as shown in Figure 2.

High neutron economy and low fissile content requirement are salient features of the CANDU reactor. The reactor can operate with NU and slightly enriched uranium fuels. This feature can be exploited for several fuel cycle options opening the possibility to using spent LWR and mixed oxide (MOX) fuels as source materials for the fissile inventory. There are also concepts to use CANDU reactors to support Pu-Th and LEU-Th fuel cycles.[51]

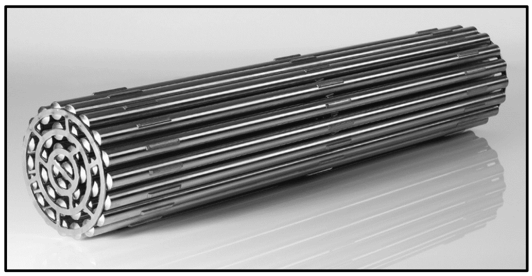

Figure 2. Photograph of CANDU reactor fuel bundle.[52] A typical CANDU fuel bundle is about 0.1-m-diameter and 0.5-m-length, and weighs about 24 kg. This bundle appears to contain 37 fuel elements in sequential layers of 18, 12, 6, and 1.

4.3 Low Enriched Uranium Oxide Fuel

Why? The manufacture of LEU oxide fuels requires a moderate degrees of uranium enrichment. Reactor grade LEU oxide is pressed into dense cylinders and loaded into metal cladding. However, these reactors require light water and graphite for moderation.

LWRs were developed in the U.S. for naval and civilian energy production. These are water-cooled, water moderated, thermal spectrum reactors. They are designed both as PWRs and BWRs. Standard LWR fuel is based on LEU uranium oxide fuel pellets clad in Zircaloy and arranged into fuel assemblies as shown in Figure 3. There are far more LWRs being used for civilian energy production than all other reactors for all other purposes combined.

Following the Fukushima Daiichi accident, research efforts have focused on development of accident tolerant fuels, which includes improving the safety performance of both the cladding and the fuel under abnormal conditions.[53- 58]

Figure 3. Photograph of PWR fuel assembly.[59] Typical PWR and BWR grids are square and contain 14 to 18 and 8 to 10 fuel elements per row, respectively. The square dimensions range from about 0.14 to 0.23 m, lengths from 3.9 to 4.8 m, and weights from 500 to 700 kg. Enrichment levels range up to about 5 wt% ^{235}U.[60]

4.4 Mixed Oxide Fuel

Why? There are over 2,000 MT of plutonium in the world, with the dominant inventories held by France, Japan, Russia, the United Kingdom, and the United States. MOX is a means of using these plutonium inventories for the production of civilian energy. MOX fuel does not require enriched uranium. MOX fuel is very versatile and can be used in a variety of reactor types.

MOX fuels are comprised of intimate mixtures of plutonium and uranium oxides. The MOX fuel is both manufactured as ceramic pellets and loaded into cladding much like LWR fuel and manufactured as powder and loaded directly into cladding.[61,62] The uranium oxide in MOX fuels is typically made from DU or NU, but there are exceptions. The plutonium content in MOX fuels is typically in the range of 5 to 8 wt% but can be as high as 12 wt%.[63]

MOX fuels are used in a variety of reactor types. Sometimes the fuel loading in the reactor core is a mixture of uranium oxide and MOX fuel, and sometimes the entire core is MOX fuel. MOX is suitable for use in both thermal spectrum reactors (e.g., PWR and LWR) and SFRs.

Figure 4 shows fuel assemblies from the BN-800 reactor, which is an SFR. Figure 5 shows a fuel assembly from the Ohma reactor, which is an advanced boiling water reactor (ABWR).

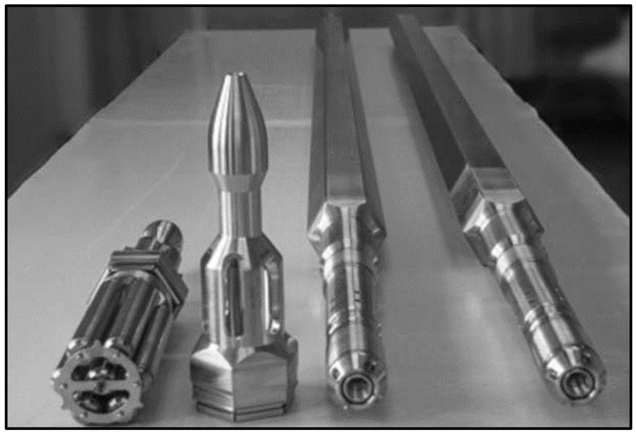

Figure 4. Photograph of BN-800 Reactor fuel assemblies, Beloyarsk Nuclear Power Station, Sverdlovsk Oblast, Russia.[64] MOX fuel clad in stainless steel. The BN-800 reactor is an SFR.

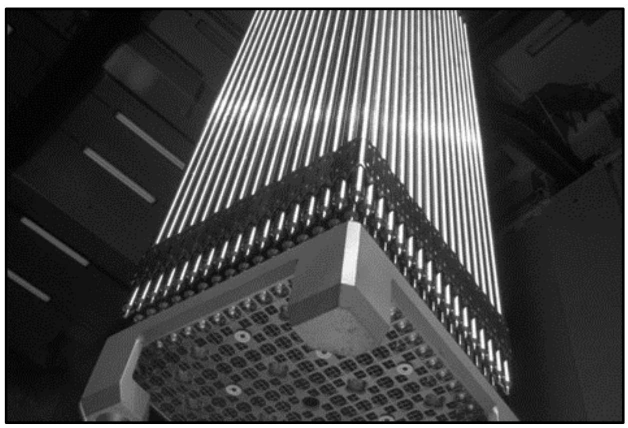

Figure 5. Photograph of Ohma Nuclear Power Plant fuel assembly, Aomori Prefecture, Japan.[65] MOX fuel clad in zirconium alloy. The Ohma reactor is an ABWR.

4.5 Natural Uranium Metal Fuel

Why? The manufacture of NU metal fuel requires the second least amount of processing; with natural uranium oxide fuel requiring the least. No uranium enrichment is required. Reactor grade NU metal is formed into cylinders and/or tubes loaded into metal cladding. However, these reactors require graphite or heavy water for moderation. This was the preferred method of producing weapons grade plutonium.

Fueled with natural uranium metals, one reactor at ORNL[d], nine at HS[e], and five at SRS [f] were developed for plutonium and tritium production.[66-68] The ORNL reactor was an early prototypic reactor. The two remaining families of reactors operated differently. The Hanford reactors were cooled by a combination of helium and water (single pass from the Columbia River) and moderated by graphite. The Savannah River reactors were cooled and moderated by heavy water, and the heavy water was, in turn,

d. The X-10 reactor. This was the second reactor to the Chicago Pile-1 reactor.

e. These were designated the B, D, F, H, DR, C, KW, KE, and N reactors.

f. These were designated the R, P, L, K, and C reactors.

cooled by water from the Savannah River. A great deal of fuel development was performed at both sites, so the fuels described here are only representative. The majority of fuel was simply metallic NU fuel clad in aluminum. However, the N-Reactor at HS was unique in this fleet of reactors. It was the only reactor to serve the dual purpose of plutonium and electrical energy production, use zirconium alloy cladding, and use slightly enriched uranium (1.25 wt% ^{235}U). The N-Reactor was the last to be decommissioned. N-Reactor also produced civilian electricity. Many of these reactors also produced tritium via the transmutation of ^{6}Li targets. Some of these fuel designs were more complex and involved enriched uranium metal and lithium targets. The plutonium uranium reduction by extraction (PUREX) process, described later, was developed specifically to recover weapons grade plutonium from spent fuels discharged from these families of reactors. Hanford also pioneered the bismuth phosphate and reduction/oxidation (REDOX) reprocessing technologies. The use of enriched uranium fuels required engineering modifications to the PUREX process equipment to accommodate the change in criticality issues associated with the higher enrichments.

Similarly, MAGNOX reactors using metallic uranium as fuels were developed in the United Kingdom initially for plutonium production and were later adapted for civilian energy production. These were carbon dioxide (CO_2) cooled, graphite moderated, reactors. Metallic NU fuel was clad in magnesium alloy “fuel cans.” Interestingly, the magnesium alloy developed for this application became the namesake of this family of reactors. The resulting alloy was called “MAGNOX AL80,” which stands for “MAGnesium No OXidation ALuminum 80 wt%.” Among other factors, this aluminum alloy was compatible with the CO_2 cooling gas, whereas aluminum alloys used at HS and Savannah River Site (SRS) were not.

Uranium naturel graphite gaz-cooled (UNGG) reactors were developed in France. These were similar to the MAGNOX reactors, but the metallic fuel was clad in a magnesium-zirconium alloy. Like the MAGNOX reactors, the UNGG reactors were initially used for plutonium production and were later adapted for civilian energy production.

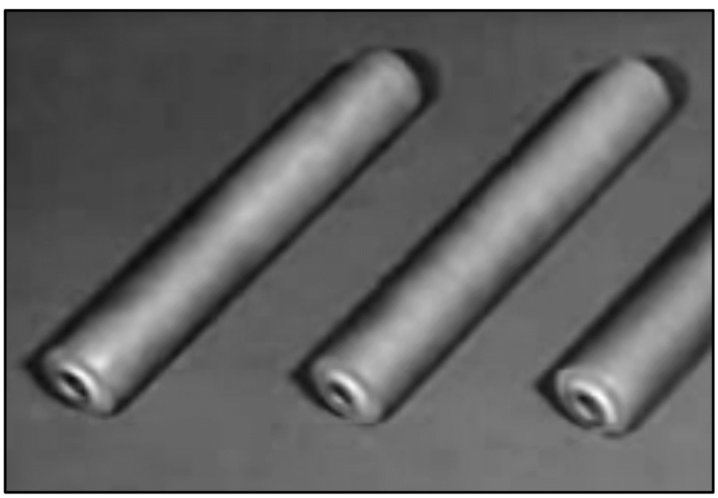

Figure 6. Photograph of typical Hanford Reactor fuel elements (single-pass-coolant reactor design). Single extruded tube of NU clad in aluminum alloy. 20-cm-length, 2.5-cm-outside-diameter, weighing 4 kg.

Figure 7. Photograph of Hanford N-Reactor fuel element (circulating-primary-coolant reactor design). Two coextruded concentric tubes (tube-in-tube design) of natural or slightly enriched (1.25 wt% ^{235}U) uranium clad in zirconium alloy. 66-cm-length, 5-cm-outside-diameter, weighing 24 kg.[69]

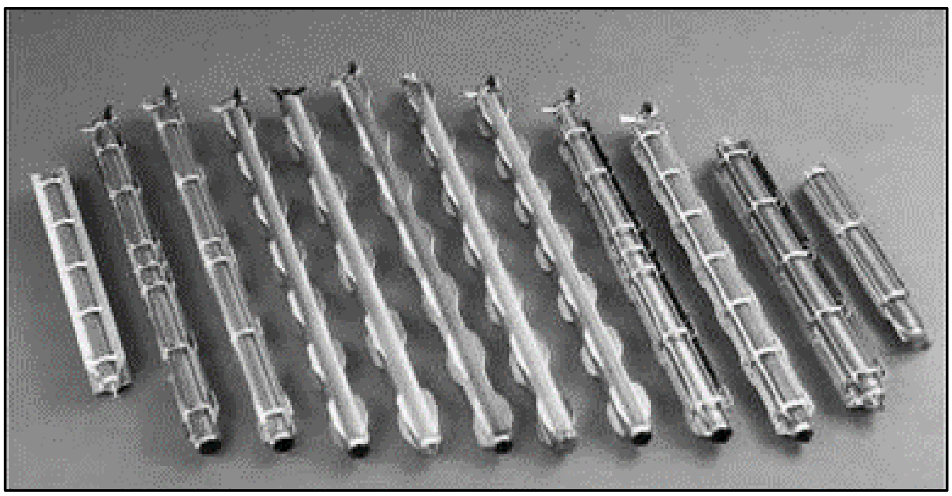

Figure 8. Photograph of examples of MAGNOX fuel cans. NU clad in magnesium alloy.[70] MAGNOX fuel slugs ranged in size from about 50 to 90-cm-length, weights 5 to 12 kg, and all were close to 2.8-cm-diameter.[71]

Technical details are scarce for the plutonium production reactors used in Russia and China.[g] At least some of the thirteen (or so) Russian reactors were similar in principle to the U.S. production reactors, in that they were water-cooled, graphite moderated, and fueled with metallic NU clad in aluminum alloy.[72-74] However, these reactors were equipped with additional cooling channels for nitrogen, not helium, flow through the graphite core. Other Russian plutonium or tritium production reactors may have been quite different utilizing heavy water moderation. The two plutonium production reactors in China followed suit.[75-77]

g. These include reactors at the Guangyuan Plutonium Production Complex, Sichuan Province, and the Jiuquan Atomic Energy Complex, Gansu Province.

If the intent is to produce so called “weapons grade” plutonium, the goal is then to optimize operations with respect to ^{239}Pu production. This means that the NU fuel is “short cycled” in the reactor, having a residence time of about 30 days. The longer the fuel remains in the reactor, the greater the production of undesirable plutonium isotopes such as ^{240}Pu and ^{241}Pu. If the intent is to produce electrical energy, such as with the Hanford N-Reactor and certain MAGNOX reactors, then the fuel remains in the reactor much longer and the resulting quality of the plutonium is diminished as it becomes so called “reactor grade” plutonium. Nevertheless, reactor grade plutonium is weaponizable, if not as desirable, for such applications.[78] At the time of this writing, all the plutonium production reactors mentioned above are no longer operational.

As a result of these plutonium production efforts, there are now large stockpiles of weapons grade plutonium in Russia, the United Kingdom, and the United States. And, as a consequence of reprocessing LWR fuels, there are large stockpiles of reactor grade plutonium in France, Japan, and the United Kingdom. There are also vast quantities of reactor grade plutonium in the spent nuclear fuel inventories residing in any country that has operated, or is operating, nuclear reactors. How best to manage these plutonium inventories from environmental and security perspectives is a matter of international concern. Some of the reactor types described here are specifically intended to “burn” these plutonium reserves in reactors as a means of mitigating these concerns.

4.6 Sodium-Bonded Metallic Fuel

Why? Sodium-bonded metal fuels were developed for liquid-metal-cooled, fast breeder reactor applications. These metallic fuels included HEU-based alloys and DU-plutonium-based alloys. Metallic fuels lend themselves well to non-aqueous reprocessing technologies capable of recovering purified metallic products from spent metallic fuels. However, these are not the only types of fuels that can be used in this family of reactors. For example, MOX fuels and carbide fuels (to be discussed) are also applicable.

Sodium-bonded metallic fuels gained prominence during the U.S. Sodium-Cooled Fast Reactor Program which culminated in development of the Experimental Breeder Reactor II (EBR-II) at Argonne National Laboratory – West (ANL-W) (now the INL Materials and Fuels Complex) and the Fast Flux Test Facility (FFTF) at Hanford.

In this fuel design, metal fuel pins are clad in stainless steel fuel elements that are arranged in fuel assemblies. Photographs of an EBR-II driver fuel assembly is shown in Figure 9 in sections from the bottom (upper left photograph) to the top (lower right photograph). Sodium metal is added along with the fuel pin to improve the thermal conductivity between the fuel and the cladding. The MOX fuel assembly shown in Figure 5 looks very similar to the EBR-II diver fuel subassembly shown in Figure 9. Many different fuel alloys were tested in EBR-II and FFTF. The most prominent alloys capable of high burnups were binary fuel of HEU with 10 wt% Zr, and ternary fuel of DU with 20 wt% Pu and 10 wt% Zr. A photograph of a U/Zr alloy fuel irradiated in FFTF is shown in Figure 10.

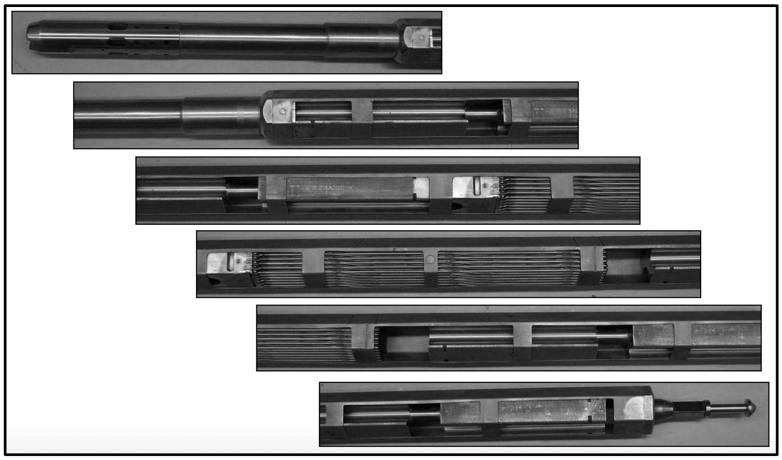

Figure 9. Photograph of a cut-away EBR-II driver fuel subassembly. (DOE photograph)

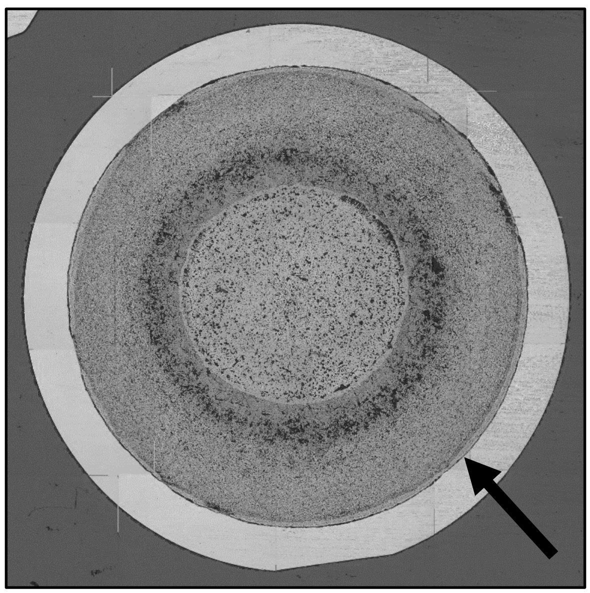

Figure 10. Mosaic microphotographs of a cross section of an EBR-II Mk-IIIA driver fuel element after 10% burnup. The outer grey ring is the stainless steel cladding that has an 0.23-in.-OD. The arrows mark the ID of the cladding wall. The fuel has swelled and is touching the cladding wall. (Luca Capriotto, INL)

4.7 Research Reactor Dispersion Fuel

Why? Research reactors serve a wide variety of applications. Aluminum-clad plate-type fuel is the most common. However, the enrichment levels vary widely from LEU to HEU. Recent efforts to convert research reactor fuels to LEU have led to the development of "high-density" uranium compounds such as uranium silicides to replace the more common uranium aluminides.

The category "research reactor" encompasses a very wide variety of reactor types, sizes, and functions. The purpose here is not to impose a strict and arbitrary definition of what entails a research reactor. However, compared to commercial-scale electrical utility reactors, research reactors are much smaller and used for purposes other than electricity generation. Presently, there are about 220 operational research reactors worldwide, with hundreds (>500) more in various states of decommissioning.[79] Thermal power ratings range from less than 1 kW to greater than 200 MW, with facilities in over 50 countries.

Providing neutrons is the one function that all research reactors share. Where they diversify is with the purpose of providing neutrons. Some of the many applications include the following:

- Education and Training
- Beamline Source
- Radiography and Tomography
- Isotope Production
- Materials Irradiation
- Nuclear Data
- Nuclear Fuels Testing
- Instrument Testing.

The status and disposition of research reactors are important topics because they operate on enriched uranium fuels spanning the full spectrum from LEU to HEU, including so called “weapons grade” uranium at 93 wt% ^{235}U. This means that the status of fresh and spent fuels from research reactors (that, remember, are spread globally) is a significant concern for the proliferations and safeguards of this material. In response, the U.S. Department of Energy (DOE) began the Global Threat Reduction Initiative (GTRI), which included the Reduced Enrichment for Research and Test Reactors (RERTR) Program and the U.S. Foreign Research Reactor Spend Nuclear Fuel (FRR SNF) Acceptance Program. The primary purposes of these programs are to convert research reactors from HEU to LEU fuels, and to return HEU fuels to the countries of origin. To accommodate these changes much research has been performed on new fuel designs with focus on increasing the uranium density and volume fraction to offset the undesirable performance characteristics of lower enriched fuels.

By far the most common dispersion fuel is the aluminum-clad plate-type. In this fuel design a uranium compound is formed into a thin plate and clad between two aluminum plates forming a fuel plate. Fuel and plate thicknesses are on the order of 0.5 to 1.5 mm, respectively. Several fuel plates comprise a fuel assembly as depicted in Figure 11. Cooling is provided by air or water, and fuel temperatures remain low. There are many hundreds of fuel designs and configurations used in research reactors. Widely used uranium compounds include aluminides, silicides, hydrides, and molybdenides of uranium. The stoichiometries of these compounds vary according to application. And burnable poisons, such as boron and gadolinium, may be added to the fuel compositions. Conversion of a reactor from HEU to LEU often accompanies changing the uranium aluminide fuel to the newer uranium silicide or uranium molybdenide fuels for their increased fuel densities.

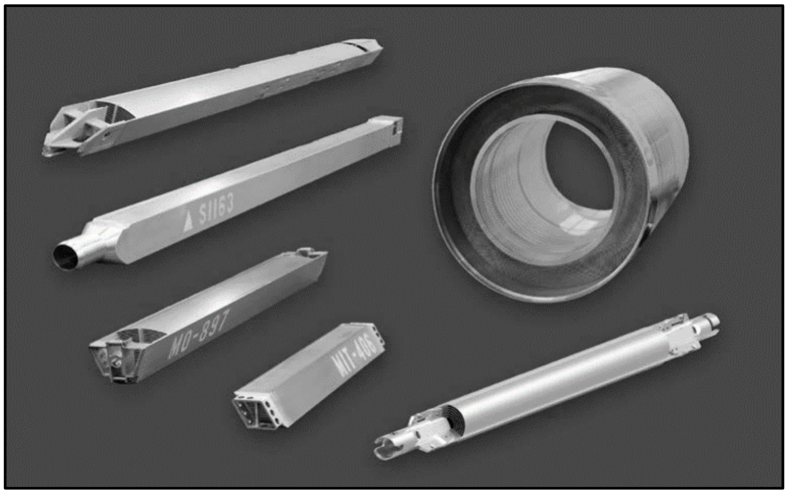

Figure 11. Photograph of typical research reactor plate fuel assemblies. (BWXT photograph)

The fuels described above are a type of "dispersion fuel." Dispersion fuels consist of fuel particles that are dispersed and suspended in a matrix. A wide variety of materials have been proposed to serve as fuel and matrix. The fundamental advantage of dispersion fuels over the monolithic fuels described earlier, is that the bulk of the matrix remains undamaged from radiation and fission product effects, allowing for higher burnups.[80] There are a great number of dispersion fuel systems proposed. Of these, TRISO fuel has received the most attention, and it is designed for advanced high-temperature gas-cooled reactors.

4.8 Coated-Particle Dispersion Fuel

Why? Developed for high temperature reactor applications. Higher coolant outlet temperatures translate to improved thermal-to-mechanical energy conversion efficiencies. And higher temperature coolants can be put to more uses than lower temperature coolants. Therefore, reactors capable of achieving 1000°C or higher coolant temperatures are desirable.

TRISO fuels are significantly different from any of the fuels previously discussed. There are many variants of this fuel type. This fuel design was developed for high temperature reactor applications, such as the high-temperature gas-cooled reactor (HTGR), but it is not limited in that regard. The basic structural components as seen in Figure 12 are as follows:

- Inner spherical fuel kernel: The fuel kernel is on the order of 0.5-mm-diameter. Several materials are candidate fuels. The materials most studied include uranium oxide (as seen in the Figure 12), uranium carbide, and uranium oxy-carbide.
- Low-density pyrolytic carbon layer: The porosity in this layer provides space for the accumulation of fission product gases.
- First high-density isotropic pyrolytic carbon layer: This layer protects the fuel kernel during silicon carbide deposition and aids in fission gas retention.
- High-density silicon carbide layer: This layer provides the primary means of mechanical strength and containment of fission products.
- Second high-density isotropic pyrolytic carbon layer: This layer protects the silicon carbide layer and provides surface bonding to the dispersion matrix.

- Together the various layers serve as the “cladding” surrounding the fuel kernel and barrier containing the fission products. And all layers must conduct the heat from the fuel kernel to the dispersion matrix.
- The dispersion matrix contains the TRISO particles and conducts the heat from the TRISO particles to the primary heat transfer fluid circulating through the core of the reactor.

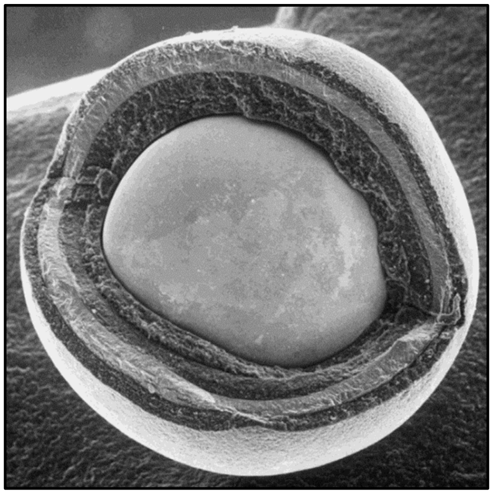

Figure 12. Photograph of breached TRISO fuel particle. The spherical uranium oxide kernel is encapsulated by successive layers of pyrolytic carbon and silicon carbide. (DOE photograph)

TRISO fuel particles dispersed in graphite cylinders, called “fuel compacts,” are shown in Figure 13. These compacts measure 12.5-mm-diameter by 50-mm-length. These were designed for a prismatic-core HTGR where they are loaded into hexagonal graphite elements fitted with channels for cooling gas flow and fuel compacts.

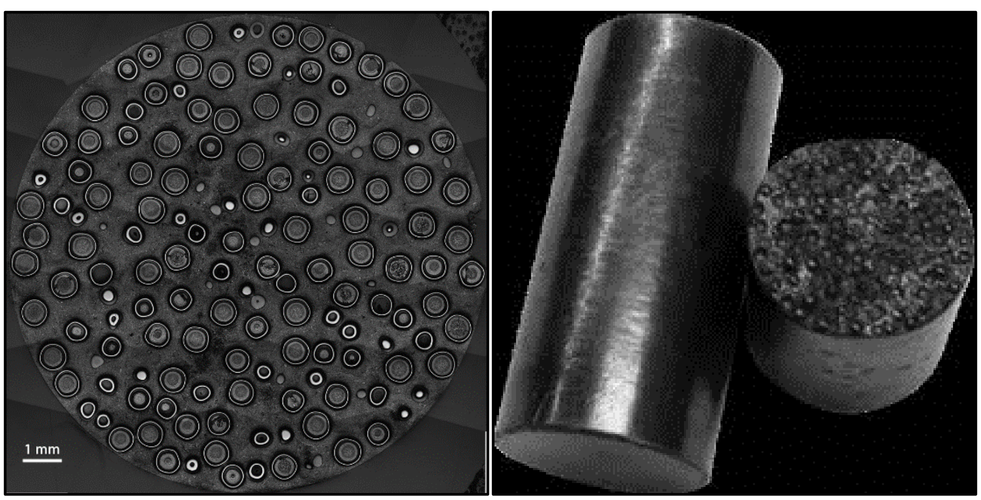

Figure 13. Photographs of graphite cylinder containing a dispersion of TRISO fuel particles. The image on the left is a cross section of the cylinder. (DOE photographs)

TRISO fuel particles dispersed in graphite spheres, called “pebbles,” are shown in Figure 14. These spheres measure 60-mm-diameter. The two-part design includes a 50-mm-diameter inner spherical fuel zone surrounded by 5-mm-thick fuel-free shell. These were designed for a pebble-bed HTGR.

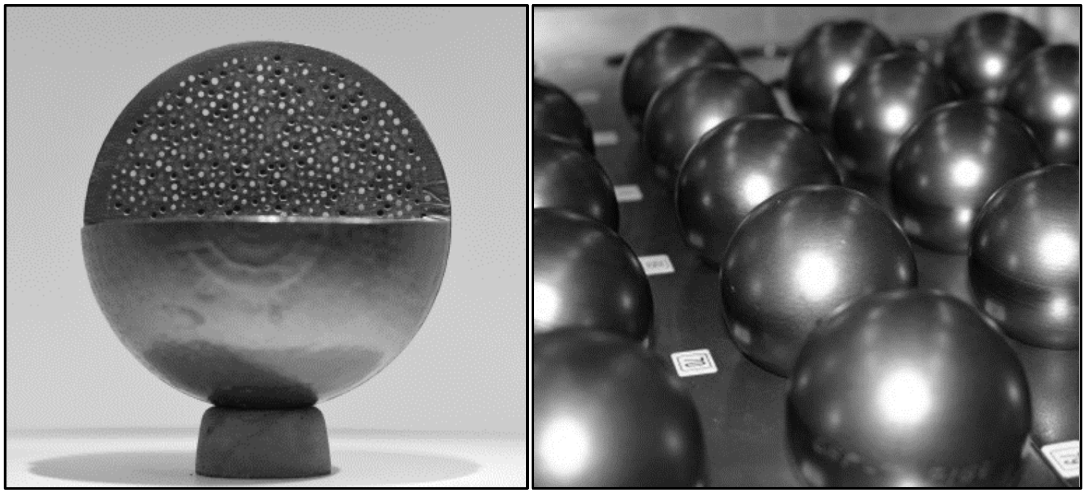

Figure 14. Photographs of a graphite sphere (“pebble”) containing a dispersion of TRISO fuel particles. The image on the left is a partial cross section of a graphite sphere. (DOE photographs)

There are structural variants of these particles as well. The bi-structural isotropic (BISO) fuel, and the quad-structural isotropic (QUADRISO) fuel. The silicon carbide layer is absent in BISO fuel, which is exploited in a particular reprocessing scheme as described later. A burnable poison layer is present in QUADRISO fuel. A further distinction is made by the means of depositing the high-density isotropic pyrolytic carbon layer. For example, there is a distinction between LTI-TRISO and HTI-TRISO. Low-temperature isotropic (LTI) pyrolytic carbon is derived from a mixture of propene and ethyne. High-temperature isotropic (HTI) pyrolytic carbon is derived from methane.

The silicon carbide layer is formed by chemical vapor deposition (CVD) from methyltrichlorosilane. However, silicon carbide is not the only choice; work has been performed on zirconium carbide coated particles.[81] There are many possible fuel combinations of DU, NU, LEU, HEU, and plutonium as oxides, carbides, and oxy-carbides. And it is also possible to combine fissile and fertile isotopes such as ^{232}Th.[82]

4.9 Naval Reactor Fuel

Why? Refueling a reactor aboard a ship or submarine is a difficult task. Fuel technologies have advanced to where refueling is no longer require because the initial fueled reactor core is expected to last the lifetime of the vessel.

Nuclear reactors for service aboard ships and submarines have special requirements not encountered elsewhere. The following list is cited directly.[83]

- Compactness: Reactor must be small enough to fit within space and weight constraints of a warship while still being able to provide adequate power to drive at necessary speeds for engagement or rapid transit.
- Crew Protection: The crew lives and works very close to the reactor for extended amounts of time.
- Public Safety: U.S. Navy ships use various ports around the world; it is a necessity that the safety of the general public at these ports be guaranteed so that ships are continued to be welcomed.
- Reliability: The reactor must be able to continuously provide power and electricity to the ship to ensure a self-sufficient operational status in the most demanding environments.

- Ruggedness: The reactors must be able to tolerate extreme conditions of being at sea as well as severe shocks during battle conditions.
- Maneuverability: The reactor must be able to provide rapid and frequent power changes to support the ships' tactical maneuvering.
- Endurance: It is crucial that the reactor to be able to operate for many years before refueling, the best-case scenario is a lifetime core. This will maximize ship availability, minimize occupational exposure, minimize life-cycle cost, and minimize demand on the support infrastructure.
- Quietness: This is especially important for submarines so to minimize the threat of acoustic detection.

There is not much information available to the public on the designs of fuels and reactors for naval applications. Submarines are fitted with a single reactor, while larger surface vessels – such as aircraft carriers – may be fitted with several reactors. Most modern naval reactors are PWR-type reactor. Enrichment levels range from 93 to 97% ^{235}U.[84] Therefore, spent naval reactor fuels could be an important source of HEU.

Other types of naval reactors were deployed in the past and are perhaps deployed presently. For example, the first two U.S. submarine reactors were water-cooled (designed by Westinghouse) and sodium-cooled (designed by General Electric).[85] The S1W was the first water-cooled naval propulsion reactor. It was tested on land by Westinghouse at the National Reactor Testing Station near Arco, Idaho. This facility is now on the INL site. The S2W was the second water-cooled naval propulsion reactor and the first nuclear reactor to be used on a submarine. It was tested on the U.S.S. Nautilus (SSN-571) from January 21, 1954 to March 2, 1980. The S1G (a.k.a., SIR Mk-A) was the first sodium-cooled naval propulsion reactor. It was tested on land by General Electric at the Knolls Atomic Power Laboratory, Kesselring Site, in Niskayuna, NY. The reactor containment dome was a 225-ft-dia. sphere, made of 1-in.-thick steel, weighing 3,850 ton; the dome itself was considered an engineering feat.[86] The S2G (a.k.a. SIR Mk-B) was the second sodium-cooled naval propulsion reactor and the second nuclear reactor to be used on a submarine. It was tested during sea trials on the U.S.S. Seawolf (SSN-575) submarine from July 21, 1955 to December 12, 1958, at which time it was replaced with a water-cooled S2Wa reactor. The S2G was also the final LMR tested by the Navy because PWR technologies were adopted for all future naval applications.[87,88]

Early Russian submarine reactors used uranium oxide fuel pellets clad in stainless steel and were cooled with a lead-bismuth alloy.[89] Countries with military nuclear naval fleets include China, France, India, the United Kingdom, and the United States. Nuclear reactors have also been considered for civilian commercial vessels.[90]

4.10 Inert Matrix Fuel

Why? Inert matrix fuels do not contain uranium. The fissile inventory is completely comprised of transuranic elements. This fuel type is intended to consume transuranics as efficiently as possible. Eliminating uranium from the fuel prevents the formation of additional transuranics via the transmutation of uranium.

The purpose of inert matrix fuels (IMFs) is to "burn" plutonium as efficiently as possible. This is done by providing a uranium-free fuel, which eliminates completely the transmutation processes that generate additional transuranics in the core. IMFs have been proposed for every kind of civilian power reactor, including CANDUs and LWRs. In these core conversion scenarios, the core is often shared by a mixture of standard fuel assemblies along with IMF assemblies.

"A major disadvantage of removing ^{238}U is the reduction or elimination of a prompt negative Doppler reactivity coefficient and a negative moderator temperature coefficient. A pure plutonium fuel type is not desirable in LWRs because of the low allowable mass loading per fuel rod (yielding short fuel cycles) and strong positive temperature coefficients. Any workable fuel composition must have a negative prompt temperature coefficient (i.e., reactor power decreases as temperature increases) for safety and control purposes. Plutonium cores in LWRs can have negative isothermal temperature coefficients if enough tungsten, erbium, or other resonance absorbers are added. The addition of burnable poison is also needed to hold down the core reactivity."[91]

Resonance absorbers affect the neutron spectrum and provide negative reactivity by interacting preferentially with fast neutrons. This effect of resonance absorbers is termed Doppler reactivity feedback. Effective resonance absorbers include iron, niobium, tungsten, molybdenum, and the oxides of these metals that perform a similar function as ^{238}U, which is absent in IMFs. But the options for resonance absorbers are not limited to these four elements.[92- 97]

4.11 Molten Salt Fuel

Why? MSRs have several promising, yet unrealized, features that make them attractive. These include high coolant temperatures, low pressure molten salt cooling systems, enhanced intrinsic safety features, and greater thermal-to-mechanical efficiencies than, for example, LWRs.

Molten salt fuels are significantly different from any of the fuels previously discussed. There are many variants of reactors designs that use molten salts, and many variants of the molten salt systems that can be used as fuel salts and cooling salts. There is a stark difference between molten salt fueled reactors and molten salt cooled reactors, even though both are MSRs. In most designs, MSRs have two molten salt loops: primary and secondary. The primary salt loop extracts heat from the core, and the secondary salt loop extracts heat from the primary salt loop. The most significant difference is the nature of the fuel. In a molten salt fueled reactor, the fissile inventory is contained in the primary molten salt. In a molten salt cooled reactor, the fissile inventory is stationary and contained in fuel elements in the core. Fluoride and, to a lesser extent, chloride salt systems have received the most attention. As discussed earlier, isotopic enrichment of lithium and chlorine may be necessary in order to preserve the neutron economy and to limit the formation of undesirable transmutation product such as ^{3}H and ^{36}Cl, respectively.

There are several technical requirements placed on molten salt fuels and coolants. Certainly, the physical/chemical properties that are important to basic engineering design aspects include heat capacity, thermal conductivity, viscosity, vapor pressure, density, and radiation performance. The more subtle aspects are related to the chemical and electrochemical properties of the molten salts, particularly regarding the chemical compatibilities of the molten salts with the materials of construction of the reactor components. Even in the simplest applications, such as a secondary cooling loop residing entirely outside the reactor core, the chemical interactions between the salt and systems are quite complex. The systems include heat exchangers, piping, valves, and fluid pumps. For example, the cooling salt flowing between two heat exchangers results in a substantial temperature gradient. The solubilities in the molten salt of certain elements in the alloys used for materials of construction may be greater at higher temperature than at lower temperature. This effect establishes a mass transport mechanism from the higher temperature region to the lower temperature region. Also, two dissimilar metals in electrical contact and exposed to a common electrolyte will establish a Galvanic cell. A multiplicity of such cells will exist in these complex mechanical systems, which can lead to significant sources of corrosion. The existence of these mechanisms is unavoidable; the goal is to manage their effects.

The situation with fuel salt is significantly more complex. All the same concerns addressed above apply, along with the additional complications associated with an ever changing and highly complex salt chemistry. Fuel salts contain the inventories of the fissile and fertile fuel components, as well as the fission and other transmutation products and radiolysis products. Metal fluorides and chlorides can be ranked according to their chemical stability relative to one another. Herein lies a concern. Many alloys considered for use in MSRs contain nickel and chromium. Fission product in the salt that are “more noble” than nickel and chromium – less stable in the salt than nickel or chromium – will form a redox couple with these alloy metals, if ionized. The noble metal fission products will be reduced from cations to metals, and the alloy metals will be oxidized from metals to cations. This mechanism is another form of unavoidable corrosion of the materials of construction. Furthermore, the plating of noble metals onto select surfaces of the materials of construction will establish additional mechanisms for Galvanic corrosion.

Research on MSRs began at ORNL in the late 1940s, with much attention focused on fluoride salts, and continued until the mid-1970s when these experimental programs were cancelled. Worldwide, only two MSRs have been operated. The Aircraft Reactor Experiment[98] during November 3 to 12, 1954. And the MSRE[99] during two campaigns between June 1, 1965 and December 12, 1969. The purpose of the MSRE was to gain experience on the ^{232}Th/^{233}U MSBR fuel cycle. The nominal composition of the fuel salt was LiF-BeF_2-UF_4-ZrF_4 (65-29.2-0.8-5 mole %). The first campaign used ^{235}U and the second campaign used ^{233}U. ^{232}Th was never added to the fuel salts. Reviews of MSR technologies were performed previously.[100,101,102] INL recently issued a report reviewing MSR technologies.[103]

5. REPROCESSING

Here *reprocessing* refers to any chemical process applied to the treatment of nuclear fuel, blanket material, or target material for the purpose of performing chemical separations. This is a broad interpretation of *reprocessing* that is meant to avoid the arbitrariness of a more exacting definition. Therefore, there are many diverse reasons why such materials would be reprocessed, which is an important realization. Selection of a specific reprocessing technology and, in finer resolution, selection of a specific reprocessing flowsheet requires answers to several key engineering and regulatory questions. All are classical engineering questions except the last two, which are unique to processes dealing with highly regulated materials including, but not limited to, materials such as these.

What are the characteristics of the materials to be reprocessed?

What chemical separations are required?

What separation efficiencies are required?

What recovery efficiencies are required?

What are the acceptance criteria of the recovered product materials?

What are the acceptance criteria of the recovered waste materials?

What are the environmental emissions standards?

What is the required reprocessing rate?

What is the required duration of operations?

What are the social requirements?

What are the economic requirements?

What are the materials control and accountancy requirements?

What are the safeguards requirements?

In a broad sense this all distills down to defining the task at hand. What follows are examples of reprocessing technologies that have been applied to, or conceived for, very specific applications both military and civilian.

5.1 Aqueous-Based Reprocessing Technologies

Just as the first large-scale nuclear reactors were built to produce plutonium, the first large-scale reprocessing plants were built to recover plutonium from these spent fuels. The early plutonium separations processes such as the bismuth phosphate precipitation process and the REDOX process eventually led to the development of the PUREX process. All were developed for the purpose of recovering ^{239}Pu for weapons production.

Hanford Site[104,105]

- T Plant: Operated from 1944 to 1956 for plutonium recovery from spent fuels via the bismuth phosphate precipitation process. This was the world's first large-scale plutonium separation plant.
- B Plant: Operated from 1945 to 1957 for plutonium recovery from spent fuels via the bismuth phosphate precipitation process. Then, following modification from 1968 to 1985 for cesium and strontium recoveries from tank wastes.
- U Plant (a.k.a., TBP Plant): Constructed in 1945 for the same purpose as the T Plant and B Plant, but never operated in that capacity. Following modifications, it was operated from 1952 to 1958 for uranium recovery from uranium-bearing tank wastes from the S Plant. During this second mission it was named TBP Plant; but also went by Metal Recovery Plant and Uranium Recovery Plant. The process used at the TBP Plant to recovery uranium was a modification of the PUREX process. Tributyl phosphate (TBP) was used as the extractant, hence the name TBP Plant.
- S Plant (a.k.a., REDOX Plant): Operated from 1952 to 1967 for plutonium recovery from spent fuels via the REDOX process.
- A-Plant (a.k.a., PUREX Plant): Operated from 1956 to 1972, and 1983 to 1988, and for a brief period in 1990, for plutonium and uranium recovery from spent fuels via the PUREX process. The plant was also used to recover ^{233}U from irradiated thorium oxide blanket in 1965, 1966, and 1970. These PUREX operations are described in the literature. ^{237}Np was also recovered as needed for production of ^{238}Pu.[106,107]
- UO_2 Plant: Operated on demand from 1956 to 1993 for the purpose of converting uranium nitrate hexahydrate from the U Plant to uranium oxide. The uranium oxide product was shipped to other locations for conversion to uranium hexafluoride and subsequent enrichment.

Savannah River Site[108]

- F Canyon: Operated from 1954 to 1957, shut down for upgrades, and 1959 to 2000, for plutonium and uranium recovery from spent fuels via the PUREX process.
- H Canyon: Operated from 1955 to 1959, shut down for three months for upgrades, and 1959 to present, for plutonium and uranium recovery from spent fuels via the PUREX process. Following the restart in 1959, the H Canyon used the "H-Modified" process, or simply the "HM" process, which was a modification to the PUREX process allowing for the processing of HEU fuels. The early PUREX processes were designed to process natural or DU fuels, not enriched uranium fuels. Modifications to the PUREX process equipment were needed to alleviate the criticality issues associated with enriched uranium. H Canyon has recently been used to downblend HEU research reactor fuels to LEU levels.[109]
- B-Lines: B-Lines were associated with both F Canyon and H Canyon. They were used to process the plutonium nitrate product from the PUREX process into plutonium metal.
- A-Lines: A-Lines were associated with both F Canyon and H Canyon. They were used to process the uranyl nitrate product from the PUREX process into uranium oxide.

Idaho National Laboratory[110]

- Idaho Chemical Processing Plant (ICPP, aka Idaho Nuclear Technology Engineering Center (INTEC)): Operated from 1953 to 1992. Designed as a hybrid REDOX/PUREX process to treat a variety of HEU fuels. The ICPP and H Canyon were the two PUREX plants capable of reprocessing HEU fuels.

U.S. Commercial Facilities[111]

- Nuclear Fuel Services Company, West Valley, NY: Operated by W.R. Grace Company from 1966 to 1971. It was the only commercial PUREX plant to be operated in the U.S. The plant reprocessed fuels from commercial LWRs and the Hanford N-Reactor. The nominal design capacity was 300 MTHM per year. The plant was shut down in December 1971 for reconstruction to nearly triple its capacity. Work on this effort continued until 1976 when the company suspended operations.
- Midwest Fuel Recovery Plant, Morris, Illinois: Built by General Electric between 1970 and 1974 but never operated with irradiated fuels. Early testing revealed a flowsheet design flaw, and the plant was declared inoperable in 1975. Reprocessing was based on the Aquafluor process, which is a combination of solvent extraction coupled with uranium fluoride volatility. The nominal design capacity was 300 MTHM per year.[112]
- Barnwell Nuclear Fuel Plant, Barnwell, South Carolina: Built by Allied General Nuclear Services between 1970 and 1975 but never operated with irradiated fuels. Reprocessing was based on PUREX with a nominal design capacity of 1500 MTHM per year.[113,114]
- Exxon Nuclear Fuel Recovery and Recycling Center, Oak Ridge Reservation, Ok Ridge, Tennessee: Construction permit application submitted to the Nuclear Regulatory Commission (NRC) by the Exxon Nuclear Company, Inc., in 1976. The nominal design capacity was 2100 MTHM per year. The project never came to fruition.[115] Getty Oil Company, Atlantic Richfield Company, and Allied-Gulf Corporation were developing similar plans to construct commercial reprocessing facilities at that time.

1976 saw the beginnings of a substantial policy shift that effectively ended commercial reprocessing in the U.S. This marked the beginning of the end to commercial reprocessing ventures in the U.S. Soon afterwards essentially all existing and planned projects were cancelled. It was an election year and President Ford was running against Mr. James Carter. In October 1976 during his campaign, President Ford announced,

> *"…the reprocessing and recycling of plutonium should not proceed unless there is sound reason to conclude that the world community can effectively overcome the associated risks of proliferation... that the United States should no longer regard reprocessing of used nuclear fuel to produce plutonium as a necessary and inevitable step in the nuclear fuel cycle, and that we should pursue reprocessing and recycling in the future only if they are found to be consistent with our international objectives."*[116]

And after the election, in April 1977, President Carter announced,

> *"We will defer indefinitely the commercial reprocessing and recycling of plutonium produced in the U.S. nuclear power programs." And later… "The plant at Barnwell, South Carolina, will receive neither federal encouragement nor funding for its completion as a reprocessing facility."*[116]

Other countries did not follow the U.S. lead, and instead developed vast infrastructures for commercial reprocessing of nuclear fuels since the 1970s. A summary of major international reprocessing facilities is given in Table 8. China and India are the only countries actively researching and expanding their reprocessing capabilities. Russia is faced with cold war legacy nuclear waste issues that are on par with, or worse than, those in the U.S. However, Russia is modernizing its reprocessing infrastructure and actively engaged in demonstrating new reprocessing technologies. Japan is modernizing its infrastructure with the construction of Rokkasho, which is expected to open in 2022 after significant delays. France continues to reprocess nuclear fuels for its own domestic needs and under contract to other countries. And the United Kingdom has recently terminated its reprocessing activities after a long history of nuclear development. Early on Germany chose not to develop its own reprocessing capabilities, and instead contracted with France and the United Kingdom to reprocess its spent LWR fuels up until 2005.

Table 8. Summary of Major International Reprocessing Facilities.

Country	Facility Name	Dates of Operation	Production Scale	Fuels Reprocessed
China	Lanzhou Nuclear Fuel Complex.	2010 to present.	Pilot Plant 10 to 20 MT/y.	
	Gansu Nuclear Technology Industrial Park.	Under development.	200 MT/y.	
	Reprocessing Plant. Based on Orano technology. Site location TBD.	Under development. Target early 2030s.	800 MT/y.	LWR fuels.
France	Marcoule UP1.	1958 to 1976.	900 MT/y Military.	GGR.
		1976 to 1993.	Military and civilian.	
		1993 to 1997.	Civilian.	
	LaHague UP2.	1966 to 1976.	800 MT/y.	GGR.
	LaHague UP2-400.	1976 to 2004.	400 MT/y.	LWR and GGR.
	LaHague UP2-800.	1994 to present.	800 MT/y.	LWR.
	LaHague UP3.	1989 to present.	800 MT/y.	LWR, MOX, RR.
Germany	Karlsruhe Reprocessing Plant.	1971 to 1990.	Pilot Plant 35 MT/y.	LWR.
	Wackersdorf Nuclear Reprocessing Plant.	Under development 1982 to 1988. Abandoned.	350 MT/y.	LWR.
India, Trombay, Bhabha Atomic Research Centre (BARC)	Uranium Thorium Separation Facility (UTSF).	2002 to present.	THOREX Pilot Plant.	ThO_2 irradiated in CIRUS reactor.
	Power Reactor Thoria Reprocessing Facility (PRTRF).	2015 to present.	THOREX Pilot Plant.	ThO_2 irradiated in Dhruva PHWR reactor.
	Plutonium Reprocessing Plant (PRP).	1964 to 1973.	PUREX 30 MT/y.	CIRUS reactor fuel. Al-clad metallic NU.

Country	Facility Name	Dates of Operation	Production Scale	Fuels Reprocessed
	PRP (refurbished).	1983 to present.	PUREX 60 MT/y.	Dhruva PHWR fuel. Al-clad metallic NU.
India, Kalpakkam, Indira Gandhi Centre for Atomic Research (IGCAR)	Kalpakkam Atomic Reprocessing Plant (KARP).	1996 to 2003 2009 to present.	PUREX 100 MT/y.	Madras Atomic Power Station (MAPS) PHWR fuel.
	KARP Expansion Project PReFRe-3A.	Under development.	PUREX.	
	Lead Mini Cell Facility.	2002 to present.	Pilot Plant.	Fast Breeder Test Reactor (FBTR) U/Pu carbide fuel.
	Compact Reprocessing Facility for Advanced Fuels (CORAL).	2003 to present.	Pilot Plant 12 kg/y.	FBTR U/Pu carbide fuel.
	Demonstration Fast Reactor Plant (DFRP).	Under development.	Demonstration Plant 100 to 500 kg/y.	FBTR and Prototype Fast Breeder Reactor (PFBR) fuels.
	Fast Reactor Fuel Reprocessing Plant (FRFRP).	Under development.	14 MT/y.	PFBR fuels.
India, Tarapur, Bhabha Atomic Research Centre (BARC)	Tarapur Plutonium Pant.	1964 to 1974.	PUREX 30 MT/y.	CIRUS reactor fuel. Al-clad metallic NU.
	Tarapur Plutonium Pant (Refurbished).	1984 to 1997.	PUREX 50 MT/y.	CIRUS and Dhruva reactor fuels. Al-clad metallic NU.
	Power Reactor Fuel Reprocessing Plant – 1 (PReFRe-1).	1979 to present.	100 MT/y.	MAPS and Rajasthan Atomic Power Station (RAPS) PHWR fuels.
	Power Reactor Fuel Reprocessing Plant – 2 (PReFRe-2).	2011 to present.	100 MT/y.	MAPS and RAPS PHWR fuels.
	Integrated Nuclear Recycle Plant (IP-1).	Under development.	600 MT/y.	

Country	Facility Name	Dates of Operation	Production Scale	Fuels Reprocessed
Japan	Tokai Reprocessing Plant.	1977 to 2009.	100 MT/y.	LWR, MOX.
	Rokkasho Nuclear Fuel Cycle Facility.	Target 2021.	800 MT/y.	LWR, MOX.
Russia, Production Association Mayak (PO Mayak), Ozersk (Formerly Chelyabinsk-65)	Defense Radiochemical Facility, Plant B (a.k.a., Plant 24).	1948 to early 1960s (Shutdown as Plant BB was brought online.).		Spent fuels from plutonium production reactors.
	Defense Radiochemical Facility, Plant BB (a.k.a., Plant 35).	1959 to 1987.		Spent fuels from plutonium production reactors.
	RT-1 Reprocessing Facility (Incorporating parts of Plant B).	1977 to 2016.	400 MT/y.	VVER-440, BN, and Naval fuels.
	RT-1 Reprocessing Facility (Refurbished).	2016 to early 2030s (To be shut down as RT-2 comes online.).	400 MT/y.	VVER, RBMK, BN, and Naval fuels.
Russia, Siberia Chemical Enterprise (SCE), Seversk, (Formerly Tomsk-7)	Radiochemical Works (RCW) Unit 15 (Contained two reprocessing lines.).	1961 to 1994 1962 to 1994.	6,000 MT/y.	Spent fuels from plutonium production reactors.
	Pilot Demonstration Power Complex (PDPC) Nitride Fuel Plant KEU-2.	Under development. Target 2024.	5 MT/y.	BREST-300 mixed nitride fuels.
Russia, Zheleznogorsk (Formerly Krasnoyarsk-26)	Defense Radiochemical Facility.	1953 to 1995.	3,000 MT/y.	Spent fuels from plutonium production reactors.
	RT-2 Reprocessing Facility.	Under development. Target 2025.	1,000 to 1,500 MT/y.	VVER, RBMK, BN and fuels.
	Pilot Demonstration Centre (PDC).	2015 to present.	10 to 250 MT/y as capacity is increased.	VVER and BN fuels.

Country	Facility Name	Dates of Operation	Production Scale	Fuels Reprocessed
United Kingdom, Sellafield	B204 Reprocessing Plant.	1952 to 1964.	300 to 750 MT/y.	Windscale Pile, MAGNOX.
	B204 Head-End Plant (Head-end to B205.).	1969 to 1972.		LWR.
	B205 (aka MAGNOX) Reprocessing Plant.	1964 to 2020.	Military and civilian. 1,500 MT/y.	MAGNOX.
	Thermal Oxide Reprocessing Plant (THORP).	1994 to 2018.	1,200 MT/y.	AGR, LWR.

From the preceding discussions it is evident that aqueous reprocessing technologies are mature and have a long history of being used at massive industrial scales for both military and commercial applications. No other reprocessing technologies come close to these levels of development and deployment. There are a great many variants of aqueous reprocessing technologies as described in contemporary literature.[117-125]

A universal feature of all aqueous reprocessing technologies is that uranium, plutonium, and minor actinides are, after separations and calcination, recovered as oxide materials. If any forms other than oxides are needed for fuel fabrication, then additional chemical conversions are necessary. It is predominantly this feature that provides opportunities for other non-aqueous reprocessing technologies to compete with aqueous technologies.

5.2 Non-aqueous Reprocessing Technologies

If the PUREX process is used as the benchmark of aqueous reprocessing technologies, against which non-aqueous reprocessing technologies are to be compared, then the following attributes have been offered as potential benefits of the latter over the former.

1. Metallic products: Non-aqueous reprocessing technologies can recover actinides as refined metals from both metallic and non-metallic spent fuels, thereby eliminating the need for subsequent oxide-to-metal chemical conversion.
2. Hotter fuels: Non-aqueous reprocessing technologies can process spent fuels directly from reactor operations without the need for interim storage and cooling. Non-aqueous separations media are not subject to radiation damage as are aqueous and organic separations media.
3. Decreased criticality susceptibility: Aqueous and organic media always acts as neutron moderators, whereas certain non-aqueous systems do not. Non-aqueous reprocessing technologies generally eliminate moderator materials, allowing for greater concentrations of actinides in the process fluids.
4. Smaller footprint: Due to the abilities to process fuels more quickly from the reactor and maintain higher actinide concentrations in process fluids, the footprint of a non-aqueous reprocessing facility is less than that of an equivalent-scale aqueous reprocessing facility.
5. Less process waste volume: Due to the higher actinide concentrations in process fluids and, in some cases, reduced number of flowsheet unit operations, a non-aqueous reprocessing facility will generate less waste volume than that of an equivalent-scale aqueous reprocessing facility.

What follows is an attempt to categorize and describe the major non-aqueous reprocessing technologies that have been researched and, in some limited capacities, used to reprocess or treat spent fuels. The order in which the information is presented is not chronological and the focus is on U.S. history.

5.2.1 Treatment Technologies Developed to Support EBR-II

Historically, eleven LMRs have been operated in the U.S. between 1949 and 1994 as shown chronologically in Figure 15. Since the 1940's, many other LMR concepts were proposed and advanced to various rigor levels of design without ever being built and operated. Of these, four additional reactors (LAMPRE-II, CRBRP, SAFR, and PRISM) are included in Figure 15 (grey arrows) because of their significance. Although never completed and operated, these four reactors underwent significant development before their projects were terminated. PRISM remains a viable contender for future LMFBR development as its design is actively updated and improved.

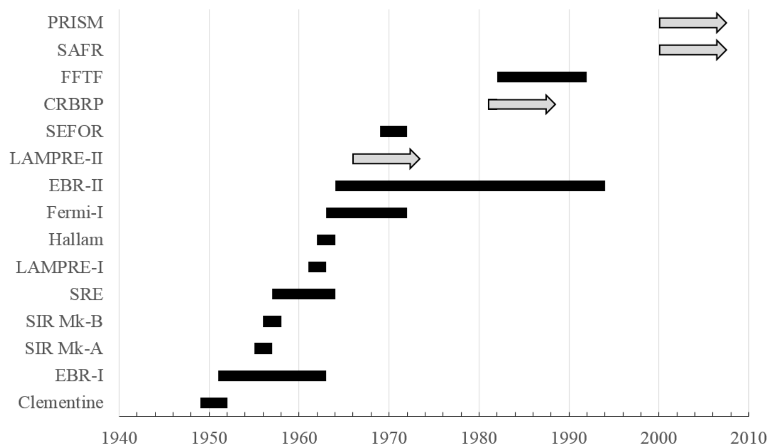

Figure 15. Operational chronology of U.S. LMRs.

Brief descriptions of the 15 LMRs presented in Figure 14 are presented below. And some technical specifications are given in Table 9.

- Clementine was the first fast-spectrum reactor. Its purpose was to study the viability of plutonium-based reactor fuels and provide basic nuclear physics data for the atomic weapons program. It was fueled by molten plutonium and cooled by liquid mercury. [126, 127, 128]
- Experimental Breeder Reactor I (EBR-I) was the first LMFBR to simultaneously breed plutonium and produce electrical power. Its purpose was to demonstrate the principle of breeding ^{239}Pu from ^{238}U and the feasibility of operating a LMFBR for future civilian power production. It also provided information on fast neutron physics and radiation-induced metallurgical damage that was useful in the design and development of Fermi-I and EBR-II. The HEU fuel region was cooled by NaK and the DU blanket region was cooled by air.[129-]
- SIR Mk-A (a.k.a. S1G) was the first sodium-cooled naval propulsion reactor. It was tested on land by General Electric at the Knolls Atomic Power Laboratory, Kesselring Site, in Niskayuna, NY. [137, 138]
- SIR Mk-B (a.k.a. S2G) was the second sodium-cooled naval propulsion reactor. It was tested during sea trials on the U.S.S. Seawolf (SSN-575) submarine. [139, 140]
- Sodium Reactor Experiment (SRE) was the first LMR designed for civilian power production. Its purpose was to demonstrate the technical and economic feasibility of a SGBR for this application. Engineering and physics data gained from the SRE were used to support the design of the Hallam Nuclear Power Facility (HNPF) reactor. It was operated by Atomics International at the Santa Susana Field Laboratory near Simi Valley, CA. [141-143]

- Los Alamos Moten Plutonium Reactor Experiment-I (LAMPRE-I) was used to test fast reactor fuels and materials. The engineering and physics data gained from LAMPRE-I was used to support the design of LAMPRE-II.[144- 148]
- Hallam Nuclear Power Facility (HNPF) was the first LMR commercial venture. It was part of the Atomic Energy Commission's (AEC's) Power Demonstration Reactor Program. The project was managed by the AEC, Consumers Public Powers District (CPPD), Atomics International, and Bechtel Corporation.[149- 154]
- Fermi-I (a.k.a., Enrico Fermi Fast Breeder Reactor (EFFBR)) was the first LMFBR commercial venture. It was operated under the Power Reactor Development Company, a consortium of more than thirty private companies. Its purpose was to demonstrate the operation of a LMFBR in the environment of a commercial utility power company.[155- 159]
- EBR-II was the first LMFBR to simultaneously breed plutonium, produce electrical power, and operate on reprocessed fuel taken from its core. Its purpose was to demonstrate a closed fuel cycle on an operating LMFBR. Engineering and physics data gained from EBR-II were used to support the design of the FFTF reactor.[160- 170] EBR-II was located at ANL-W and operated from 1963 to 1994. It was a sodium-cooled fast breeder reactor with driver and blanket regions. The driver fuel was sodium-bonded metallic HEU alloyed primarily with fissium (HEU-5Fs) or zirconium (HEU-10Zr), and the blanket was sodium-bonded metallic DU. EBR-II is important in the present context because it has served as one of the primary focal points for the development of non-aqueous reprocessing technologies in the U.S.
- Los Alamos Moten Plutonium Reactor Experiment-II (LAMPRE-II) was to be a higher energy version of LAMPRE-I. However, this reactor was never built.
- Southwest Experimental Fast Oxide Reactor (SEFOR) was a research reactor operated under a consortium that included the U.S. AEC, General Electric, and several electric power companies. Its purpose was to study the nuclear physics and safety of MOX cores. One way it achieved this was to demonstrate the ability of the Doppler coefficient to terminate a transient, which has to do with the relationship between absorption characteristics and temperature of fissile atoms.[171-173]
- Clinch River Breeder Reactor Project (CRBRP) was intended to be a LMFBR demonstration reactor to assess the economics of breeder reactor technology and attain experience and engineering data to design a larger cost competitive LMFBR commercial reactor. The project was managed by AEC, Tennessee Valley Authority (TVA), Commonwealth Edison, and Project Management Corporation. The project was authorized by congress in 1972, terminated by the Carter Administration in 1977, and resumed by the Reagan Administration in 1980. After expenditures exceeded $1B, and the cost of completion estimated to be an additional $2.5B, the project lost congressional support as was terminated in October 1983.[174- 183]
- Fast Flux Test Facility (FFTF) reactor was used as a testbed for LMFBR development. It was used for irradiation testing of fuels and materials under a fast-spectrum, and to develop procedures, components, and systems used to design future commercial LMFBRs.[184- 190]
- Sodium Advanced Fast Reactor (SAFR) was a design study performed by Rockwell International, Bechtel Corporation, and Combustion Engineering under the DOE Advanced Liquid Metal Reactor (ALMR) program. Development work on SAFR was terminated in 1988 when the DOE selected Power Reactor Innovative Small Module (PRISM) for further consideration as a demonstration reactor.[191-193]
- Power Reactor Innovative Small Module (PRISM) was a design study performed by General Electric under the DOE ALMR program. The PRISM design has undergone several iterations since its conception in 1981 and the termination of the ALMR program in 1994.[194- 201]

Table 9. Summary Technical Specifications and References of U.S. LMRs.

Reactor Name	Clementine	EBR-I	SIR Mk-A Prototype
Reactor Type	LMFR	LMFBR	SGR
Coolant	Mercury	Sodium/Potassium	Sodium
Design Type	Loop	Loop	Loop
Fuel Type	Molten Plutonium	Metallic HEU Alloy	Oxide HEU
Blanket Type	—	Metallic DU	—
Moderator	—	—	Graphite
Thermal Power, MW	0.025	1.2	—
Electrical Power, MW	Zero	0.2	—
Start Date	1949	12/1951	1955
End Date	1952	12/1963	1957
Location	LANL Site, NM	INL Site, ID	Niskayuna, NY
Reactor Name	**SIR Mk-B**	**SRE**	**LAMPRE-I**
Reactor Type	SGR	SGBR	LMFR
Coolant	Sodium	Sodium	Sodium
Design Type	Loop	Loop	Loop
Fuel Type	Oxide HEU	Metallic LEU Alloy	Molten Plutonium
Blanket Type	—	Thorium	—
Moderator	Beryllium	Beryllium	—
Thermal Power, MW	—	20	1
Electrical Power, MW	—	6	Zero
Start Date	1956	1957	1961
End Date	1958	1964	1963
Location	U.S.S. Seawolf (SSN-575)	SSFL Site, Semi Valley, CA	LANL Site, NM
Reactor Name	**HNPF**	**Fermi-I**	**EBR-II**
Reactor Type	SGR	LMFBR	LMFBR
Coolant	Sodium	Sodium	Sodium
Design Type	Loop	Loop	Pool
Fuel Type	Metallic LEU Alloys	Metallic LEU Alloys	Metallic HEU Alloys (Some MOX)
Blanket Type	—	Metallic DU	Metallic DU
Moderator	Graphite	—	—
Thermal Power, MW	254	430 Design, 200 Achieved	62.5
Electrical Power, MW	76	125 Design, 66 Achieved	20
Start Date	1962	8/1963	7/1964
End Date	1964	9/1972	10/1994
Location	Lancaster County, NE	Monroe County, MI	INL Site, ID

Reactor Name	LAMPRE-II	SEFOR	CRBRP
Reactor Type	LMFR	LMFR	LMFBR
Coolant	Sodium	Sodium	Sodium
Design Type	Loop	Loop	Pool
Fuel Type	Molten Plutonium	MOX	MOX
Blanket Type	—	—	DU Oxide
Moderator	—	—	—
Thermal Power, MW	20	20	975
Electrical Power, MW	Zero	Zero	350
Start Date	1966 Proposed	1969	1972 Proposed
End Date	—	1972	—
Location	LANL Site, NM	Washington County, AR	Roane County, TN
Reactor Name	**FFTF**	**SAFR**	**PRISM**
Reactor Type	LMFBR	LMFBR	LMFBR
Coolant	Sodium	Sodium	Sodium
Design Type	Loop	Pool	Pool
Fuel Type	MOX (Some Metallic HEU)	Metallic U/Pu/Zr Alloy	Metallic U/Pu/Zr Alloy
Blanket Type	DU Oxide	Metallic U/Zr Alloy	Metallic U/Zr Alloy
Moderator	—	—	—
Thermal Power, MW	400	900	471
Electrical Power, MW	Zero	350	155
Start Date	4/1982	—	—
End Date	4/1992	—	—
Location	Hanford Site, WA	Rockwell/ Westinghouse	General Electric

5.2.1.1 Melt Refining and Skull Reclamation Processes

Why? Fast breeder reactor technologies were a U.S. priority at the time EBR-II was commissioned. The melt refining process offered a simple process flowsheet for reprocessing metallic HEU fuels remotely. The skull reclamation process was developed to recover actinides from the disposable process crucibles used during melt refining.

From the very first days of operation, the EBR-II reactor was co-located with a hot cell reprocessing facility called the Fuel Cycle Facility (FCF). Spent driver fuels from EBR-II were reprocessed in FCF from 1964 to 1968 using the *melt refining process*. More than 400 subassemblies (34,000 fuel elements) were remotely fabricated from spent EBR-II driver fuels and recycled back to the reactor core. The melt refining process was intended to be minimalistic in terms of the number of process steps to facilitate remote hot cell operation. The EBR-II HEU-5Fs fuel elements were mechanically de-clad to liberate the fuel pins, which were loaded into one-time-use zirconia crucibles and melted. Separations partitioned materials to various locations based on volatility and reactivity with the crucible materials. The majority (90 to 95%) of the actinides and noble metal fission products (the fissium alloy elements) reported to the consolidated metal ingot product. Metals capable of reacting with zirconia formed a “skull” within the zirconia crucible. The balance of metals (5 to 10%) not reporting to the metal ingot product, reported to the skull along with some of the more reactive fission product metals.

The purpose of the *skull reclamation process* was to recover uranium losses to the skull. Unfortunately, the skull reclamation process was more complex and involved several steps of oxidation, salt fluxing, additions of zinc and magnesium, and decanting. And the process only recovered uranium from the skulls; it did not recover transuranics. The skull reclamation process was developed to pilot scale, but it was never installed and implemented in FCF before the end of the reprocessing campaign. Since that time, the complexity of the process and the unacceptable losses of actinides to the skull materials have rendered the melt refining and skull reclamation processes obsolete.[202-229]

5.2.1.2 Early Conceptual Integral Fast Reactor Pyroprocessing

Why? The IFR pyroprocessing flowsheet was meant to improve upon the deficiencies of the melt refining and skull reclamation processes. The electrorefining process would produce a higher purity HEU product and provide an improved means of transuranic recovery.

A goal of the Integral Fast Reactor (IFR) program was to demonstrate a closed fuel cycle around EBR-II that was more efficient than the earlier melt refining and skull reclamation processes of the 1960s. The IFR program required a new driver fuel capable of higher burnup levels, and one that was also compatible with the new reprocessing technologies. In preparation for the IFR program, the EBR-II core was converted from HEU-5Fs to HEU-10Zr. And the intention was that once reprocessing operations began, the core would be converted to a DU-20Pu-10Zr fuel as plutonium was recycled from the driver and blanket. ANL researchers began proposing IFR pyroprocessing flowsheets around 1984. These early flowsheets included unit operations of *fuel chopping, uranium electrorefining with liquid cadmium anode, halide slagging, cathode processing, casting furnace, and fuel fabrication*.

The purpose of fuel chopping was to cut the fuel elements into smaller pieces to expose the metallic fuel to the solvent cadmium and molten salt electrolyte. The mechanical de-cladding used for the earlier melt refining process was not adopted for the IFR program for two reasons: i) the higher burnup levels achieved during the IFR program resulted in greater metallurgical interactions between the fuel and cladding rendering mechanical de-cladding difficult, and ii) there was no longer a need to de-clad because the fuel could be effectively dissolved or oxidized away from the stainless steel cladding.

Electrorefining was the primary means of chemical separations and was a significant improvement over the former melt refining and skull reclamation processes in terms of both separation and recovery efficiencies of actinides and overall waste reduction. The electrorefining operations transported uranium from an impure metal anode to a purified metal cathode through a high-temperature molten salt. The salt system used was the LiCl-KCl eutectic with a nominal concentration of UCl_3. Electrorefining in the presence of a trichloride provided excellent separations. Metals more electronegative than uranium remained with the anode, while metals more electropositive than uranium accumulated in the salt. Cathode processing was a vacuum retort furnace used to treat the dendritic electrorefined uranium. It was designed to accommodate both salt distillation and uranium metal consolidation into an ingot. The purified HEU would then be processed and cast into fuel pins in the casting furnace for fabrication of new fuel elements.[230-241]

During these early stages of process development, the liquid cadmium anode was thought necessary as a means of dissolving the metallic fuel into a molten metal pool beneath the electrolyte. Uranium and plutonium would be oxidized from the cadmium pool into the salt, while metals more noble would remain in the cadmium pool as a sludge. In the next iteration of this technology, the liquid cadmium anode deep enough to accommodate a chopped fuel basket was replaced by a shallow liquid cadmium pool beneath the chopped fuel basket suspended in the electrolyte.

Halide slagging, like the skull reclamation process, was another unit operation that was proposed but never deployed during the EBR-II mission. It was originally proposed by ANL researchers as an improvement to the melt refining and skull reclamation processes for processing high plutonium containing fuels. During those early days of EBR-II operations, the intention was to convert the core from HEU-Fs fuel to DU-Pu-Fs fuel. Halide slagging was thought to be an improved process for purifying these future fuel types. However, the reprocessing mission was cancelled before the conversion was attempted. And much later, halide slagging was proposed for the IFR mission as a means of recovering plutonium from EBR-II blanket materials as plutonium chloride to be advanced to the electrorefiner.

5.2.1.3 Later Conceptual Integral Fast Reactor Pyroprocessing

Why? As research on an electrorefining based flowsheet continued, it was determined that a liquid cadmium anode and halide slagging were not necessary. The separations processes of the finalized IFR flowsheet included electrorefining and cathode processing to produce a HEU-Zr alloy ingot for fuel fabrication. And the liquid cadmium cathode technology was developed to recover plutonium from the electrorefiner salt.

The finalized IFR pyroprocessing flowsheet developed by ANL researchers between 1986 and 1994 included the unit operations of *fuel chopping, uranium electrorefining, liquid cadmium cathode*, *cathode processing, casting furnace, and fuel fabrication*. The liquid cadmium cathode was a means of co-collecting uranium and plutonium from the electrorefiner salt for use in fuel fabrication. It was again the intent to convert the EBR-II core from HEU fuel to plutonium alloy fuel during the reprocessing mission.[242-297]

Research was also underway on how to adapt the IFR pyroprocessing technologies developed for EBR-II metallic fuels, to the recovery of plutonium from LWR oxide fuels.[298-300]

Much work was performed readying FCF to accept this mission. For example, the hot cells were decontaminated to permit human entry into the cells. This facilitated refurbishment of the infrastructure and installation of the process equipment. However, the IFR was not to be realized.

5.2.1.4 EBR-II Spent Fuel Treatment

Why? On September 30, 1994, EBR-II was shut down. Subsequently, the driver and blanket fuels were removed from the reactor and the sodium-coolants drained from the system.[301,302] *On October 1, 1994, the IFR program was terminated by the DOE by order of the U.S. Congress.*[303] *The mission at ANL-W changed from one of reprocessing spent EBR-II fuel for fuel recycle research (the IFR program) to one of treating spent EBR-II fuel for disposition (the Spent Fuel Treatment Program).*

When EBR-II was shut down, the Yuca Mountain Nuclear Waste Repository was under development and thought to become the destination for EBR-II spent fuels and other high-level nuclear wastes. However, because EBR-II fuels and blankets contained bond-sodium, and because sodium metal is highly reactive with water liberating both heat and hydrogen, untreated these materials were not candidates for direct disposal to the Yuca Mountain repository. To meet the acceptance criteria of Yuca Mountain, it was necessary to neutralize the reactivity of the bond-sodium. The EBR-II spent fuel treatment (SFT) process was developed to meet these criteria in three distinct phases, which can be called process selection, process demonstration, and process operation.

Process selection proceeded from 1995 to 2000. The National Research Council issued a series of reports evaluating electrometallurgical techniques for treating the inventory of EBR-II sodium-bonded spent fuel and blanket materials.[304-311] And the DOE issued a series of reports evaluating the environmental impacts of managing and treating the sodium-bonded spent fuel inventories in Idaho.[312-324]

Process demonstration proceeded from 1996 to 1999 as the EBR-II Spent Nuclear Fuel Treatment Demonstration Project. Once electrometallurgical fuel treatment was selected as the disposition technology, select technologies developed for the IFR program were adapted to meet the requirements of this new mission.[325-378] There are several key differences between electrometallurgical processing as intended for the IFR program and electrometallurgical processing used for SFT.

- The IFR program was intended to process EBR-II driver fuels by electrochemical means, and to process EBR-II blanket materials by halide slagging. The SFT program processes both driver fuels and blanket materials by electrochemical means in two electrorefiners; one designed for driver fuels and one designed for blanket materials.
- The IFR program would recover plutonium from both driver fuels and blanket materials for the manufacture of U-Pu-Zr alloy fuels. The disposition path for plutonium in the SFT program is to leave the plutonium in the electrorefiner salts, which are converted into a salt waste form.
- The IFR program used the casting furnace to cast fuel alloy pins for the manufacture of new fuel elements. The SFT program uses the casting furnace to cast a single sample pin used to verify the uranium enrichment level of the final electrorefined uranium product.
- The IFR program included fuel fabrication equipment. The SFT program does not utilize such equipment.

Process operations proceeded from 1999 to January 2005 as ANL researchers continued to perform process improvements and treat spent nuclear fuel in FCF. In January 2005 there was change in contractor management. ANL-W that was managed by the University of Chicago became INL Materials and Fuels Complex managed by Battelle Energy Alliance. From that time forward the SFT program has been overseen by INL researchers who continue to perform process improvements.[379-486]

5.3 ORNL MSBR Salt Processing

Why? The ORNL MSBR concept required stringent ^{233}Pa management in the fuel salt. This was performed by treating a slipstream of salt in a chemical processing plant to recover and isolate ^{233}Pa until it decayed to ^{233}U. The resulting ^{233}U was harvested. A portion could be returned to the reactor and excess could be used to fuel other reactors. The ability to effectively manage this strategy was never fully demonstrated and remains conceptual.

MSR development began under the Aircraft Nuclear Propulsion Program (1946 to 1961), after which MSR development continued for civilian power production (1961 to 1976). A significant portion of this large body of work is related to the ^{232}Th – ^{233}U fuel cycle MSBR concept developed at ORNL. The MSBR concept being proposed required continuous chemical processing of molten fluoride fuel salt to control the breeding ratio via management of the ^{233}Pa inventory. Major unit operations for processing the salt include *fluorination, hydrofluorination, vacuum distillation, reductive extraction, metal transfer process, electrolytic oxidizer/reducer*, and others. This history was recently summarized in a report.[487]

5.4 Chloride Volatility Processes

Why? These processes exploit the high vapor pressures of zirconium chloride and aluminum chloride to affect separations. Chlorination processes are proposed to volatilize zirconium cladding from oxide fuels and volatilize the zirconium from zirconium matrix dispersion fuels. Similarly, chlorination processes are proposed to volatilize the aluminum from aluminum matrix dispersion fuels.

The *ZIRCEX process* is proposed as a head-end process to aqueous as well as non-aqueous reprocessing. It is discussed in this section because, after all, it is a high-temperature non-aqueous process. The ZIRCEX process has been proposed as a means of de-cladding Zircaloy-clad oxide fuels, volatilizing the bulk of the aluminum and zirconium from Training, Research, Isotopes, General Atomics (TRIGA) reactor fuels, and volatilizing the bulk of aluminum from research reactor plate fuels. The process exploits the differences in vapor pressures of metal chloride species to affect separations. The fuel is chlorinated in an atmosphere of chlorine, $Cl_2(g)$, hydrogen chloride, HCl(g), or carbon tetrachloride $CCl_4(g)$. Metals such as aluminum and zirconium form chlorides that have much higher vapor pressures than uranium chloride, allowing aluminum and zirconium to be separated from the uranium. These bulk separations significantly reduce the mass of fuel materials that are required to be dissolved into caustic or acidic solutions for aqueous reprocessing. And reducing the mass of material to the dissolvers reduces the volume of solution to be treated by the aqueous process and the volume of waste generated by the aqueous process.[488- 497]

5.5 Fluoride Volatility Processes

Why? These processes exploit the high vapor pressure of uranium hexafluoride to affect separations. Fluorination processes are widely used during uranium enrichment to convert purified uranium oxide to uranium hexafluoride prior to enrichment by diffusion or centrifuge technologies. And fluorination processes were proposed by ORNL for processing MSBR fuels for ^{233}Pa management.

Fluoride volatility processes have been proposed for several different applications. Only select applications are described here. The *Aquafluor process* was developed by General Electric for its Midwest Fuel Recovery Plant in Morris, Illinois. However, as discussed earlier, the plant never came to fruition. The Aquafluor process was predominantly a PUREX-based separations process designed to separate, recover, and purify uranium, neptunium, and plutonium from spent LWR fuels. A unique feature of this process was the conversion of uranium oxide (the calcined product of uranyl nitrate hexahydrate) to uranium hexafluoride in a fluidized bed reactor. Additional processing steps purified the uranium hexafluoride, which was intended to be packaged and transported to enrichment facilities.[112,493,498]

The *FLUOREX process* was proposed by Hitachi-GE as a means of reprocessing a variety of oxide fuels, over an extended period, as the reactor fleet transitions from LWRs to FBRs utilizing MOX fuels. Like the Aquafluor process, the FLUOREX process is a hybrid process using both aqueous and non-aqueous technologies. The Aquafluor process included fluoride volatility of uranium at the back end, to produce uranium hexafluoride for re-enrichment. The FLUOREX process includes fluoride volatility at the head-end for two purposes: to control the Pu:U ratio of the materials entering the PUREX process, and to produce purified uranium hexafluoride for re-enrichment or other disposition.[499- 503]

The *Nitrofluor process* was an entirely non-aqueous reprocess technology proposed by Brookhaven National Laboratory. The process was claimed to be applicable to variety of fuel types. The fuel was dissolved in a mixture of anhydrous nitrogen dioxide and HF at moderate temperatures between 100 and 200°C. The primary separation stage is based on which metals form soluble fluorides in the solvent, and which for insoluble oxides and oxy-nitrides. Subsequently, the solvent is decanted from the solids. Uranium and plutonium form soluble species and report with the solvent. Selective fluorination of the solvent will volatilize uranium as uranium hexafluoride (by the action of bromine fluoride) and plutonium as plutonium hexafluoride (by the action of fluorine gas). These two product streams would be purified further.[493,498,504-506]

The ORNL ^{232}Th-^{233}U fuel cycle MSBR concepts required processing of the fuel salts to manage the ^{233}Pa inventories and removing fission products. This subject was discussed earlier. In the fuel salt processing flowsheets proposed by ORNL, fluorination and hydrofluorination were major unit operations used primarily to volatilize uranium from the salts. There were two application for this operation. To remove the bulk of the uranium from the salt stream entering the chemical process ahead of ^{233}Pa extraction, and to recover ^{233}U from the process salt kept in storage while ^{233}Pa decayed to ^{233}U.

Fluoride volatility has been proposed for several other fuel reprocessing schemes. For metal fuel processing, there is a significant distinction between fluoride volatility and chloride volatility routes. In metal dispersion fuels, chloride volatility is proposed to volatilize the zirconium or aluminum matrix away from the uranium and plutonium. By contrast, the fluoride volatility is proposed to volatilize the uranium and plutonium away from the matrix metals.[507-510] And similar fluoride volatility concepts have been proposed for graphite matrix fuels.[511] Fluoride volatility processes have also been proposed for reprocessing LWR and FR oxide fuels with the absence of aqueous separations.[512-514]

5.6 Fluoride Salt Electrowinning (Hall-Héroult Analog)

Why? The Hall- Héroult process has been effective for over a century in manufacturing primary aluminum. Research was performed on adapting a similar process for reducing uranium oxide to uranium metal. Most of the research was focused on primary uranium production, but it has also been proposed as a reprocessing technology for spent fuels.

The process of “Bomb reduction” is the standard method of producing uranium metal via the thermal reduction of uranium tetrafluoride by magnesium metal in batch operations. The reduction reaction is highly exothermic resulting in a temperature and pressure spike in the reaction vessel, hence the expression “bomb reduction.” This process necessitates the conversion of uranium oxide to uranium tetrafluoride and produces significant quantities of process wastes. The electrowinning process was initially conceived as a means of continuous production of uranium metal from uranium oxide, which was attractive from a production perspective. At the time, large tonnages of uranium metal were needed to fuel the U.S. plutonium production reactor fleet. Later the concept was proposed as a reprocessing technology for uranium oxide fuel.

In this process uranium oxide is dissolved in a fluoride salt that has a solubility for uranium oxide and is suitable for operations above the melting temperature of uranium metal. The dissolved uranium is reduced on the surface of a molten pool of uranium metal, and the dissolved oxygen forms CO(g) and CO_2(g) on the surface of a graphite anode.[515-527]

5.7 Mercury Amalgamation Processes

Why? Mercury has a low melting temperature and forms an amalgam with many other metals. These properties were exploited to affect the reduction of uranium oxide fuels to metal and affect separations and purifications of both uranium and plutonium metallic fuels.

The *METALLEX process* was proposed as a means of reprocessing uranium oxide fuels. The uranium oxide is exposed to mercury containing a reductant such as magnesium. The magnesium reports to an oxide slag, along with other impurities, and the uranium reports to the mercury as an amalgam. The mercury amalgam is purified further by subsequent washing and filtering steps. The purified amalgam is oxidized with steam to form uranium oxide. The uranium oxide and mercury are separated in a retort furnace and the hydrogen partial pressure is controlled to produce a purified UO_2 product.

The *HERMEX process* was proposed as a means of purifying uranium and plutonium metals, either as unirradiated materials or as irradiated fuels. The HERMEX process is simpler than the METALLEX process because oxide reduction and reoxidation are not necessary. Variations of these processes can be used to convert and purify oxide-to-metal, and metal to oxide.[528- 543]

5.8 Salt Cycle Process

Why? Much attention was focused on breeder reactor technologies during the period of early development in the U.S. because uranium reserves were thought to be much scarcer than they were later determined to be. A molten salt process for reprocessing MOX fuel to support FBRs was developed at Hanford. Because MOX reactors were not greatly pursued in the U.S. for civilian power production yielding to LWR technologies, research into this reprocessing technology was short lived. However, Russia continues active research in this area in support of their MOX reactor fleet.

The *Salt Cycle Process* was "conceived by a Hanford chemist in 1959."[544] Development of the process culminated in the United States in 1966 with a demonstration using irradiated MOX fuel.[545] A few years later a comprehensive summary of the process was published in the open literature.[546]

As development of the Salt Cycle Process ended in the United States in the mid-1960s, development began in Russia in the late-1960s at a research facility in Sverdlovsk (now Ekaterinberg).[547] Today, Russian development continues at the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad where the process is called the *Dimitrovgrad Dry Process* or the *Russian Institute of Atomic Reactors (RIAR) Dry Process*. The U.S. "Salt Cycle Process" and the Russian "Dry Process" are essentially the same process, albeit with minor variations. Here the process will be called the Salt Cycle Process (SCP).

There are different variants of the process, each using a different set, or different configuration, of unit operations. The three primary variants are the MOX to MOX, PuO_2 and UO_2 to MOX, and MOX to PuO_2 and UO_2. The first variant represents the reprocessing of spent MOX fuel without the separations and recoveries of uranium and plutonium oxides. The second variant represents the utilization of plutonium and uranium oxide reserves to produce MOX fuel. For example, the utilization of weapons plutonium for the production of MOX fuel. And the third variant indicates that the process can be used to recover purified plutonium and uranium oxide from spent MOX fuel.

The first step of the process is chlorination of the feed oxides into a molten salt. The chlorine and oxygen chemical potentials within the salt, and the salt temperature, are controlled to affect the formation of chlorides, oxychlorides, and oxides in the salt. This control makes it possible to selectively deposit mixed oxides onto a graphite cathode (an electrochemical process utilizing a chlorine gas evolving anode) or to precipitate oxides from the salt. The purified oxide products are washed in water to remove the salt, thermally processed to affect the desired oxide stoichiometry, and blended to the desired MOX compositions. Fission product impurities are recovered by similar processes, but separately from the uranium and plutonium oxides.[548- 599]

5.9 Salt Transport Process

Why? The salt transport process was another attempt at a reprocessing technology for MOX fuels. However, where the Salt Cycle Process was designed to produce a purified oxide product, the salt transport process was designed to produce a purified metal product.

The *salt transport process* was proposed by ANL researchers as a means of reprocessing stainless-steel-clad LMFBR MOX fuels. There are several discreet steps in the process. The cladding is dissolved in a pool of liquid zinc at 850°C, which does not affect the oxide fuel. The bulk of the zinc solution is transferred away, and the residual zinc is removed by vacuum distillation leaving behind the oxide fuel and some residual stainless steel. During oxide reduction, the oxide fuel is contacted with salt and a calcium-containing alloy. The calcium reports to the salt as dissolved calcium oxide, while the reduced fuel reports to the alloy phase as metals. The salt from oxide reduction is a waste product and the metal alloy are advanced in the process. Plutonium and uranium are purified by successive transfers of these metals between different salt and metal phases. The final recovery of the purified metal products is achieved by distillation.[600- 617] The salt transport process has similarities to the halide slagging process and the ORNL metal transfer process.

5.10 Two Phase Exchange Processes

Why? Many processes have been proposed that exploit chemical separations across the interfaces between two high-temperature liquid phases. Examples are metal/salt and metal/oxide slag. These processes are usually proposed as one of several unit operations within a flowsheet, as opposed to being proposed as a stand-alone reprocessing technology.

Two phase exchange processes are a reoccurring theme in both aqueous and non-aqueous reprocessing schemes. They exploit the relative stabilities of species between the two phases to affect the desired separations. *Halide slagging* was discussed earlier as a unit operation proposed during the early development of the IFR program. The process exploits separations between metal/salt interfaces. Halide slagging has also been proposed as a means of purifying liquid metal plutonium fuels such as Pu-Fe alloys.[618- 621] Other types of slagging operations include *oxide slagging* and *carbide slagging*.[622] Generally, these terms imply that a molten metal phase is equilibrated with an oxide phase or a carbide phase. Oxide slagging was used in the EBR-II melt refining process described earlier.

The *lithium reduction* process is the reduction of oxide fuel to metal by the action of lithium metal. This is another form of a two phase exchange process. The process is based on the greater stability of lithium oxide compared to that of uranium and plutonium oxides.[623- 627] This lithium-based chemical oxide reduction process is similar in principle to lithium-based electrochemical oxide reduction process cited earlier. In the former process, lithium metal is directly introduced as the reducing agent. In the latter process, lithium metal is electrochemically produced on the surface of the oxide fuel in a $LiCl\text{-}LiO_2$ molten salt medium. And in the latter process, the electrochemical reduction of the uranium and plutonium oxides is a contributing factor not experienced in the former process.

5.11 Processes Applied to TRISO-Type Fuels

Why? TRISO-type fuels are highly refractory materials, meaning it is difficult to penetrate the coating materials to chemically access the fuel kernels. However, once the fuel kernels are liberated from the silicon carbide and pyrolytic carbon layers, various reprocessing routes can be applied. Therefore, the feature that most distinguishes these coated particle fuels from other fuel types is the head-end processing required to prepare the fuels for chemical reprocessing.

Many varieties of TRISO-type fuel particles have been under consideration and many avenues for reprocessing these spent fuels have been proposed. Reprocessing technologies face major challenges. For example, in the ^{232}Th-^{233}U fuel cycle application, the fertile materials are encapsulated in BISO particles, and the fissile materials are encapsulated in TRISO particles. These two particles' types are intimately mixed within the graphite fuel matrices. As a head-end to chemical separations, these BISO and TRISO fuel particles must be liberated from the matrix and separated from each other. The particles are liberated from the matrix by burning (oxidation of the graphite), and then separated from each other by physical classification based on the size distribution. In one proposed application, the fertile-BISO particles have greater than 355-µm-diameter and the fissile-TRISO particles have less than 355-µm-diameter. Separations of the two families of particles is accomplished by screening at 45 U.S. sieve size.

Another challenge common to any TRISO fuel application is liberating the fuel kernels from the encapsulating layers of silicon carbide. Once the fuel kernels are exposed, then both aqueous and non-aqueous means may be applied to extracting and recovering the fissile materials. Methods of breaching the silicon carbide lays include mechanical comminution, chemical conversion (e.g., chloride and fluoride volatility), and thermal shock. Once the fuel kernels are exposed, methods of extracting the fissile materials include traditional aqueous digestion, supercritical CO_2 solvent extraction (as the carrier for an appropriate extractant), and molten salt dissolution.[628- 638]

5.12 Weapons Plutonium Refining

Why? The first applications of nuclear technologies in the U.S., as well as several other countries, were weapons production. The first large-scale nuclear reactors were operated to produce plutonium and the first large-scale reprocessing plants were operated to recover that plutonium. Several metallurgical processes are needed to manage and use the stockpiles of metallic plutonium for weapons applications.

The primary sources of weapons plutonium are the plutonium production reactors described earlier. These reactors are designed and operated to produce *weapons grade plutonium*, which means maximizing the yield of ^{239}Pu and minimizing the yield of other plutonium isotopes, which generally requires that the ^{238}U target has a relatively short residence time within the reactor. This contrasts with *reactor grade plutonium*. For example, all uranium-based fuels and blankets produce plutonium by transmutation. However, if the reactor is designed and operated for electrical power production then, typically, the residence time of fuel within the reactor is maximized. Spent fuel produced under these conditions will contain plutonium, but the plutonium will contain significant quantities of isotopes in addition to ^{239}Pu making it undesirable for weapons use. However, this is not to imply that reactor grade plutonium is not weaponizable.

As described earlier, the PUREX and other separations processes are used to recover plutonium from the spent fuel. Purified plutonium oxide is converted to metallic plutonium via two primary methods. By fluorination to plutonium tetrafluoride followed by magnesium-thermal-reduction to plutonium metal in a magnesium fluoride-based salt, and by direct calcium-thermal-reduction to plutonium metal in a calcium chloride-calcium oxide-based salt. The latter process is called direct oxide reduction and eliminates the fluorination step. The resulting plutonium metal may or may not require additional purification prior to alloying and casting operations.

The desired plutonium isotope for use in weapons is ^{239}Pu. ^{240}Pu is undesirable because it has a significant rate of spontaneous fission that can interfere with weapons physics. ^{241}Pu is undesirable because it has a relatively short half-life and decays to ^{241}Am, which decays to ^{237}Np by alpha and gamma emissions that cause detrimental effects within weapons.

- $^{241}Pu(\beta\text{-})$ 14.35y = ^{241}Am
- $^{241}Am(^{4}He,\gamma)$ 432.2y = ^{237}Np

There are no means of chemically separating the isotopes of plutonium from each other, and plutonium is not subject to isotopic enrichment processes on large scales like uranium and some of the other metals discussed earlier. However, when the growth of ^{241}Am reaches a threshold, the ^{241}Am can be chemically separated from the plutonium. Americium separation from plutonium is performed by chlorination of the metallic americium from molten plutonium, which is a molten salt extraction process. Chlorination is performed by exchange reactions with magnesium chloride. The reduced magnesium reports to the molten plutonium; but is later separated from the plutonium during vacuum casting operations. Americium can also be removed by sparging the molten plutonium with chlorine gas to preferentially form americium chloride.

Purified plutonium metal has six allotropes (crystallographic phases) between ambient temperature and its melting temperature. These allotropes exhibit significant density variations. Alloying elements are added to plutonium to stabilize these phase-transition effects over the temperature ranges encountered during operations such as plutonium machining and weapons deployment. Gallium is an example of an alloying element used to stabilize plutonium for weapons production. Small amounts of gallium will stabilize the high-temperature plutonium δ-phase (normally stable between 310 and 415°C in purified plutonium) down to ambient temperatures.

Purified plutonium metal can be produced by electrorefining impure plutonium metal. The process is performed in a chloride salt at temperatures well above the melting temperature of plutonium metal. The process is performed in small batches, a few kilograms, and low amperages. Plutonium is oxidized from the impure phase and reduced to metal on a tantalum cathode. The molten plutonium drips from the cathode into a pool of purified plutonium metal.

Since the 1960s, researchers at LANL, Rocky Flats Plant (RFP), and Lawrence Livermore National Laboratory (LLNL) have been developing molten salt technologies to reduce plutonium oxide derived from the PUREX process to metal, and to subsequently electro-refine plutonium metal for purification and americium control.[639-691]

6. EFFECT OF REPROCESSING EFFICIENCY

The consequences of reprocessing efficiencies on the utilization of fissile materials in a fuel cycle scenario is profound. Two highly abstracted models of an integrated fuel reprocessing scheme are presented here that illustrate the importance of maximizing the retention of fissile materials, and maximizing the rejection of fission products, during reprocessing. The integrated fuel reprocessing scheme is illustrated in Figure 16.

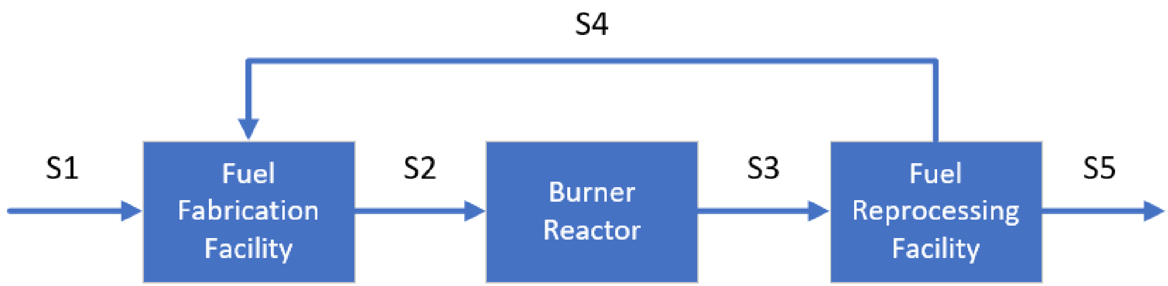

Figure 16. Illustration of the Integrated Fuel Reprocessing Scheme

The process streams shown in Figure 16 are identified as follows. As is the case with an integrated recycle system, the composition of any process stream is affected, to one degree or another, by the performances and efficiencies of all of the integrated processes.

S1: **Adjustment stream to fuel fabrication.** This stream is needed to maintain the fuel cycle. It could, for example, contain plutonium from weapons stockpiles or the fissile materials recovered from LWR fuels. It is also possible this stream could be net negative for fissile materials if the reactor is a breeder reactor generating more fissile inventory than its input.

S2: **Fresh fuel stream from fuel fabrication to the reactor.** This is the "new" fuel (i.e., preprocessed, or reconstituted fuel) entering the burner reactor. It will contain a complex mixture of plutonium, minor actinides, and fission products.

S3: **Spent fuel stream from the reactor to reprocessing.** This is the "spent" fuel entering the reprocessing facility. It will contain the post irradiation fissile and fission product inventories. Fissile inventory can be larger than its input, in the case of a breeder reactor. A burner reactor will generate lower fissile inventory as an output than its input. Fission product inventory output is always larger than its input.

S4: **Recovered stream from reprocessing to fuel fabrication.** This stream contains the fissile materials and fission products that are retained in the fuel cycle.

S5: **Discharged stream from reprocessing to waste.** This stream contains the fissile materials and fission products that are rejected from the fuel cycle to the waste streams.

In general terms, the goals of nuclear fuel reprocessing are to maximize the retention of fissile materials, and minimize the retention of fission products, in the fuel cycle process. However, the separations sciences embedded within these two goals do not behave independently of each other. For example, those technologies which are deployed to maximize the retention of fissile materials will, at the same time, tend to increase the retention of fission products. In other words, if the primary goal is to meet some established target threshold for the retention of fissile materials (e.g., 99.5 wt% of the fissile materials must be retained in the fuel cycle), then the secondary goal becomes optimization of rejection of fission products while meeting that target. This is only one of many ways in which this engineering challenge can be expressed.

6.1 The Importance of Maximizing the Retention of Fissile Materials

The integrated fuel reprocessing scheme illustrated in Figure 17 is considered here in regard to the retention of fissile materials in the fuel cycle. A simple process efficiency model for fissile material retention is illustrated in Figure 17.

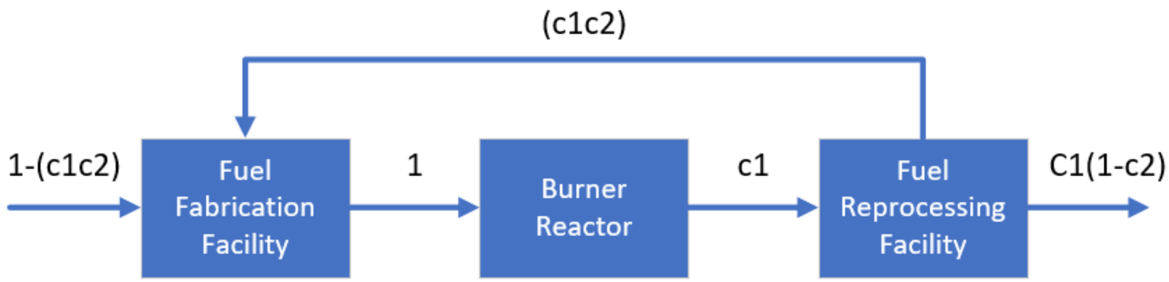

Figure 17. Process Model for Fissile Material Retention.

The mass distribution parameters shown in Figure 17 are identified as follows.

1: **Mass of Fissile Materials in Stream S_2.** This value is normalized to 1, which means that the "new" fuel stream contains one arbitrary mass unit of fissile materials. In engineering units, this value would equal the fissile materials demand of the reactor.

c_1: **Mass of Fissile Materials in Stream S_3.** This value is the mass of fissile materials in the "spent" fuel stream, expressed as a fraction of the mass of fissile materials in the "new" fuel stream. For a burner, c1 is less than 1 and for a breeder c1 is larger than 1.

c_2: **Reprocessing Efficiency**. This is the efficiency of the reprocessing facility to retain fissile materials in the fuel cycle and c2 is between 0 to 1 in that 1 means full retention of fissile materials while 0 means no retention of fissile materials.

$1-c_1$: **Reactor Efficiency.** This is the efficiency of the reactor to burn fissile materials. This value is in terms only of comparison of the fissile materials loadings of the "new" and "spent" fuel streams. This is not a rigorous definition of burn-up efficiency. For the case of a breeder, $1-c_1$ is negative value implying that more fission materials are generated than burned.

c_1c_2: **Mass of Fissile Materials in Stream S_4.** This value is the mass of fissile materials in the recycle stream, expressed as a fraction of the mass of fissile materials in the "new" fuel stream.

$c_1(1-c_2)$: **Mass of Fissile Materials in Stream S_5.** This value is the mass of fissile materials in the waste stream, expressed as a fraction of the mass of fissile materials in the "new" fuel stream.

$1-(c_1c_2)$: **Mass of Fissile Materials in Stream S_1.** This value is the mass of fissile materials needed to complement the mass of fissile materials in the recycle stream, in order to meet the demand of the reactor. For a breeder reactor (c1>1) and high retention reprocessing scheme for fissile materials (c2 ≈1), this term can be negative meaning surplus fissile inventory needs to be discharged from the system.

Results of the model are presented in Table 10. The loss of fissile materials to the waste stream ($c_1(1-c_2)$) from the reprocessing facility is shown as a percentage of the fresh fissile materials adjustment ($1-(c_1c_2)$) to the fuel fabrication, for various values of c_1 and c_2. This relationship is expressed in the following equation.

$$Fissile\ Material\ Loss\ (\%) = \frac{c_1(1-c_2)}{1-(c_1c_2)} \times 100$$

Typical prototype burner reactor designs give fissile materials burn-up efficiencies ranging between 0.10 and 0.15. Considering this range as an achievable "near term" performance from a burner reactor, demonstrating fissile materials recycling efficiency better than 0.99 (> 99%) is crucial to justifying the integrated burner/reprocessing cycle as an effective means of burning and minimizing fissile materials discharges to a long-term geological repository.

Table 10. Fissile Material Loss Results of Fissile Material Model.

Reactor Efficiency (1-c1)	Reprocessing Efficiency (c2)		
	0.9	0.99	0.999
0.05	65.52	15.97	1.86
0.10	47.37	8.26	0.89
0.15	36.17	5.36	0.56
0.20	28.57	3.85	0.40
0.25	23.08	2.91	0.30
0.30	18.92	2.28	0.23
0.35	15.66	1.82	0.19
0.40	13.04	1.48	0.15

Referring to Table 10, for the case in which the reactor efficiency is 0.10[h] and the reprocessing efficiency is 0.99,[i] 8.26 wt% of the fissile materials in Stream S1 are channeled to waste via Stream S5.

6.2 The Importance of Maximizing the Rejection of Fission Products

The integrated fuel reprocessing scheme illustrated in Figure 16 is considered here in regard to the rejection of fission products from the fuel cycle. An abstracted process efficiency model for fission product rejection is illustrated in Figure 18.

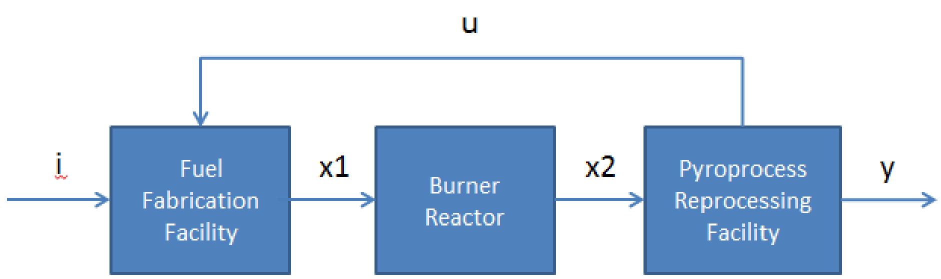

Figure 18. Process Model for Fission Product Rejection.

h. There is a net 10% reduction of the fissile materials inventory of the reactor fuel as a result of irradiation.

i. The reprocessing facility retains a net 99% of the fissile materials in the fuel cycle.

The mass distribution parameters shown in Figure 18 are identified as follows.

i: **Mass of Fission Products in Stream S_1.** This value considers the condition in which Stream S1 contains some mass fraction of fission products. For example, this could be the case if the source of burnable fissile materials is preprocessed LWR fuel. For the case of a breeder where some fissile materials are to be discharged from the system due to surplus of fissile inventory in the system, the term *i* can be negative as part of inseparable fission products from the recycled fissile materials shall be discharged along with fissile materials.

x_1: **Mass of Fission Products in Stream S_2.** This value is the mass of fission products in the "new" fuel stream.

x_2: **Mass of Fission Products in Stream S_2.** This value is the mass of fission products in the "spent" fuel stream.

u: **Mass of Fission Products in Stream S_4.** This value is the mass of fission products in the recycle stream.

y: **Mass of Fission Products in Stream S_5.** This value is the mass of fission products in the waste stream.

The following equations describe the model.

$$x_{1,n} = u_{n-1} + i_n$$

$$x_{2,n} = x_{1,n} + b_n$$

$$y_n = c(x_{1,n})$$

$$u_n = (1-c)(x_{2,n})$$

$$x_{2,n} = \frac{(i+b)(1-(1-c)^{n-1})}{c}$$

In the equations above, i_n is an impurity related constant associated with Stream S_1, b_n is a burn-up related constant associated with the reactor, and c is the rejection efficiency of fission products from the reprocessing facility. The involved mass terms reach steady states as the number of cycles (n) approaches infinity.

$$\lim_{n\to\infty} x_{2,n} = \frac{i+b}{c}$$

$$\lim_{n\to\infty} x_{1,n} = \frac{i+b}{c-b}$$

$$\lim_{n\to\infty} y_n = i+b$$

$$\lim_{n\to\infty} u_n = \frac{(1-c)(i+b)}{c}$$

Results of the model are presented in Table 11. This set of results reflects a case in which a burner reactor may be used, and no fission products are present in Stream S_1; which is to say that $i = 0$.

Table 11. Results of Fission Product Model.

Relative Burn-Up in Reactor	Fission Product Generation in the Reactor (wt% of total fuel)	Fission Product Rejection Fraction from Reprocessing	Fission Product to Fuel Fabrication (wt% of total fuel)	Fission Product to Reprocessing (wt% of total fuel)
	(x_2-x_1)	y/x_2	u	x_2
Low	2	0.9	0.22	2.22
	2	0.8	0.50	2.50
	2	0.7	0.86	2.86
	2	0.5	2.00	4.00
Medium	4	0.9	0.44	4.44
	4	0.8	1.00	5.00
	4	0.7	1.71	5.71
	4	0.5	4.00	8.00
High	8	0.9	0.89	8.89
	8	0.8	2.00	10.00
	8	0.7	3.43	11.43
	8	0.5	8.00	16.00

Table 11 is divided into three sections representing low, medium, and high burn-up of fuel in the reactor. This particular comparison is completely subjective, but for these purposes “low” burn-up converts 2 wt% of the incoming fissile materials mass to fission products; “medium” burn-up, 4 wt%; and “high” burn-up, 8 wt%.

For each of the three levels of burn-up, the table considered four values of reprocessing efficiencies: 0.9, 0.8, 0.7, and 0.5. A value of 0.9 means that 90 wt% of the fission products in Stream S3 are rejected to Stream S5.

Referring to Table 11, for the case in which the reactor is operated at a “medium” burn-up, and the reprocessing efficiency is 80%, the “new” fuel to the reactor will contain 1 wt% fission products (as a result of 1 wt% fission product returned to fuel fabrication in Stream S4) and the “spent” fuel from the reactor will contain 5 wt% fission products (as a result of an additional 4 wt% fission products generated in the reactor).

6.3 Remarks on the Abstracted Fuel Cycle Models

The fissile materials retention and fission products rejection models presented here are highly abstracted, but they reflect the consequences associated with these process effects concisely. What is not addressed in this development is the relationship between fissile materials retention and fission products rejection with regards to the engineering design and operations of the reactor and reprocessing facility. The exact nature of these relationships is very complex.

However, what can be stated with certainty is that fissile materials retention and fission products reject are not independent considerations. In general, there should be a tradeoff between fissile materials retention and fission product rejections. That is, as we seek to increase fissile materials retention, fission products rejection will decrease. The degree of this tradeoff should depend on the considered reprocessing technology.

Nevertheless, much information can be gleamed from the data in Table 10 and Table 11. The following are some of the observed relationships.

- Higher burn-up improves the consumption of fissile materials.
- Increased fissile materials retention improves the consumption of fissile materials.
- If burn-up is a limitation, then improved fissile materials retention can allow for higher consumption of fissile materials.
- Increased fission products rejection decreases fission products loading of the fuel.
- Higher burn-up increases the fission products loading of the fuel.
- If fissile materials loading of the fuel is a limitation, then improved fission products rejection can allow for higher burn-up.
- Improved purity of the fissile materials source material decreases the fission products loading of the fuel.

6.4 The Considerations of Fundamental Complexities

The abstracted model addresses the behaviors of fissile materials and fission products in only the broadest of terms. However, each category is comprised of a family of elements with unique separations behaviors. The consequences of these "uniquenesses" is that each fissile material will have an independent retention efficiency, and each fission product will have an independent rejection efficiency. And, as is already understood, there will be some degree of overlap between the separation behaviors of the two families.

For example, if, categorically, 99.9% is the minimum retention efficiency for any single fissile materials element during reprocessing, then there will be corresponding lower (i.e., lower than the mean) rejection efficiencies for those fission products that have separations characteristic similar to some of the minor actinides. A consequence of this dilemma is that the composition of the steady state loading of fission products in the fuel cycle will be shifted toward these particular fission products.

In general terms, the composition of the retained/rejected fissile materials (Streams S_4 and S_5) will not exactly reflect the composition of the bulk fissile materials (Stream S_3). And similarly, the composition of the rejected/retained fission products (Stream S_5 and S_4) will not exactly reflect the composition of the bulk fission products (Stream S_3).

7. CONCLUSIONS

Many different types of nuclear reactors using many different types of nuclear fuels serving many different applications are possible. The varieties of reactor and fuel types are seemingly endless. Consequently, the selection of reprocessing technologies and strategies are highly application specific. And in addition to these technical challenges, the engineering considerations are also influenced by economic, social, and political objectives.

A great amount of historical work has been performed on the study of reprocessing technologies, both aqueous and non-aqueous. This report provides a high-level summary of that body of work. Aqueous reprocessing technologies have been used at massive scales for plutonium production and civilian power production. Non-aqueous reprocessing technologies have not seen the same level of use as aqueous technologies, but there are certain fuel cycle scenarios that are better served by non-aqueous methods. These may include, for example, many of the fuel cycle concepts adapted to the production of metallic fuels and molten salt fuels.

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