

Sodium Cooled Fast Reactors and the Pyro-Process:
Conversion of Nuclear Waste into a Fuel Source

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Abstract

A review of the sodium cooled fast reactor(SFR) program in conjunction with a new non-aqueous reprocessing method, pyro-processing, was carried out. The purpose was to assess the viability of the program for future deployment with respect to the Generation IV Initiative criteria of: sustainability, proliferation resistance, safety, and economic competitiveness. It was found that the SFR fuel cycle excels in creating a resource efficient, proliferation resistant fuel cycle. However, reliability issues such as sodium leaks, in addition to high production costs make the SFR program highly unlikely for development in the near future ~50 years.

Contents

1. Introduction.....	6
1.1 Background.....	7
1.2 Generation IV Initiative.....	11
1.3 Fuel Cycle.....	14
1.3.1 Voloxidation.....	17
1.3.2 Electrolytic Reduction.....	18
1.3.3 Electrowinning.....	20
1.3.4 Fuel Fabrication.....	23
1.3.5 Reactor Startup.....	24
1.3.6 Plant Design.....	25
2. Analysis.....	27
2.1 Sustainability.....	27
2.1.1 Resource Utilization.....	27
2.1.2 Waste Minimization.....	28
2.2 Proliferation Resistance.....	30
2.3 Safety.....	32
2.4 Economic Competitiveness.....	37
3. Conclusion.....	38

I dedicate this thesis to my parents. I have yet to meet any other individuals whom handle hardship and adversity with such grace and poise.

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1. Introduction

Fast reactors have been around since the 1960s. Originally known as breeder reactors, this reactor type has received renewed interest amongst the nuclear science community as the debate over nuclear waste has increased. This growing interest is due to the ability of fast reactors to burn a greater variety of isotopes than current reactor designs; including those that are typically found in spent nuclear fuel rods. Fast reactors are able to utilize higher energy neutrons than the nuclear reactors in common use today. With funding for the Yucca Mountain repository cut, other options for nuclear waste disposal must be considered. In addition, rising concerns of climate change from the scientific community and the public have created the desire to pursue low emission forms of energy generation. Currently, the United States generates 45% of its electricity from coal.¹ The United States hopes to approach an eighty percent reduction in greenhouse gasses by 2050.² Public and environmental policy on greenhouse gasses has been changed over the years in the hopes of offsetting the emissions from industrializing nations. If the United States is to achieve an eighty percent reduction in greenhouse gas emissions by 2050 then a drastic shift towards low emission energy forms must take place.

There are many forms of alternative energy such as: wind, solar, tidal, geothermal and hydroelectric. These methods of generating electricity produce little to no carbon emissions. However, most of these methods are presently incapable of meeting the growing demand for electricity in the United States. Hydroelectric is the exception to this but it is limited by location, proximity to moving water, and quantity, as a river can only be dammed so much. One form of power that can fulfill the goal of reducing greenhouse gas emissions while meeting energy demand is nuclear power, which accounts for 20% of United States electricity.¹ Nuclear power creates electricity through a sustained nuclear chain reaction which creates heat which is in turn

used to boil water. The steam is passed through turbines just as in a coal plant and this generates electricity. If nuclear power were to pick up the electricity generation of coal, vast amounts of nuclear waste would be created in the form of spent fuel rods. This waste is currently stored in cooling ponds at the reactor site while it awaits final disposal at Yucca Mountain.

There exists another type of reactor which has the potential to produce energy from burning this spent fuel once it has been reprocessed. These reactors are known as fast reactors, named for their use of fast neutrons, which allow for a wider range of fuels that can be burned. In particular, the sodium cooled fast reactor(SFR) design and its fuel cycle will be discussed. This choice was made based on SFRs being the most researched and implemented of fast reactor designs.³ In addition, a new form of proliferation resistant reprocessing, pyroprocessing, will be covered. The SFR fuel cycle will be assessed with respect to the main goals of the Generation IV Initiative; sustainability, safety, proliferation resistance and economic competitiveness.³

1.1 Background

The United States current method of nuclear power generation is that of a Light Water Reactor (LWR). This design is referred to as a thermal reactor because the neutrons emitted during fission are slowed by a moderator. In this case it's ordinary water which also serves as a coolant. Other types of moderators in use are heavy water and graphite used by the Canadians and the Russians respectively. There are two types of LWR in use in the United States, the Pressurized Water Reactor(PWR) and the Boiling Water Reactor(BWR).⁴ The fuel and control rods are located in arrays within a steel pressure vessel, in which water circulates. The heat generated from fission heats up the water which evaporates and turns a turbine in a BWR. In a PWR the heat is transferred to a secondary water coolant loop which evaporates and turns a

turbine for electricity generation.⁵ The steam is then condensed in a condenser cooled by river water or cooling towers and re-circulated.

Natural uranium consists of 0.7% uranium-235, 99.3% uranium-238 and trace amounts of uranium-234 by weight. Fuel rods are composed of uranium dioxide pellets which have been enriched to increase the content of U-235 to 3-5%. Uranium 235 is the main source of energy in a LWR; however, plutonium-239 provides some energy once it is formed via U-238 neutron absorption. Uranium-235 and plutonium-239 are “fissile” elements, meaning they can sustain a nuclear chain reaction. Uranium 238 is a “fertile” element because it can become fissile via neutron absorption. However, U-238 can fission if the neutron energy is $> 1\text{MeV}$. Enrichment is necessary for a thermal reactor because the nuclear cross section of U-235 is two orders of magnitude larger for thermal neutrons as opposed to fast neutrons.⁶ The nuclear cross section of an element is the probability that a nuclear reaction will occur when nuclei collide. Thus by enriching the uranium to increase the U-235 content more nuclear reactions will take place thereby sustaining the chain reaction. The neutrons are slowed down via nuclei scattering with water molecules until they reach a kinetic energy level of 0.025 eV. At this energy level the neutrons are referred to as thermal neutrons as they achieve thermal equilibrium with their surroundings. Thermal neutrons have a larger effective neutron absorption cross-section meaning they are more likely to be absorbed by an atomic nucleus and create heavier and often unstable isotopes.⁶ Neutron absorption by U-235 results in the nucleus fragmenting into two fission products and the release of 202.5 MeV of energy along with two or three neutrons which continue the chain reaction.⁷ A nuclear reaction that has become self-sustaining is said to have reached a critical state.

Successive neutron absorption by U-238 produces a range of isotopes which are transuranic (TRU) elements, defined to be elements with an atomic number greater than uranium. All of these elements are synthetic or had long since decayed by the time man discovered the atom. They include Plutonium, Americium, Curium, and Neptunium. They are at the core of the debate over what needs to be done with our nuclear waste as they all possess half-lives of many thousands of years, Table 1. The radioactivity of spent fuel in the short term is dominated by the fission products, created from the splitting of heavy metal nuclei, such as Strontium-90 and Cesium-137. Those with medium to long-lived half-lives, created from successive absorption of nuclei, are a concern when considering a repository for spent fuel as it takes on average 130,000 years¹⁸ for them to reach the radioactivity of natural uranium.⁸ Strontium-90 and Cesium-137 are short lived isotopes with half-lives of about 30 years that release high energy beta particles and gamma radiation, respectively. Additional, fission products that are of concern are iodine-129 and technetium-99 due to their solubility in water.

Isotope	Half-Life (years)	Decay Mode	Isotope	Half-Life (years)	Decay Mode
Np-237	2.14 x 10 ⁶	$\alpha \rightarrow$ Pa-233	Am-241	432	$\alpha \rightarrow$ Np-237
Pu-238	87	$\alpha \rightarrow$ U-234	Am-242m	141	$\gamma \rightarrow$ Am-242(99.54%)
Pu-239	2.41 x 10 ⁴	$\alpha \rightarrow$ U-235	Am-243	7370	$\alpha \rightarrow$ Np-238(.46%) $\alpha \rightarrow$ Np-239
Pu-240	6560	$\alpha \rightarrow$ U-236	Cm-243	29.1	$\alpha \rightarrow$ Pu-239
Pu-241	14.4	$\beta \rightarrow$ Am-241	Cm-244	18.1	$\alpha \rightarrow$ Pu-240
Pu-242	3.76 x 10 ⁵	$\alpha \rightarrow$ U-238	Cm-245	8500	$\alpha \rightarrow$ Pu-241

Table 1: Half-lives and decay modes of various transuranium isotopes found in spent nuclear fuel. Concentrations vary based on reactor, and burn time. Pu generally accounts for ~1.3% of spent fuel while minor actinides(those that are not plutonium or uranium) account for ~.1%⁹

Currently nuclear power accounts for 5.9% of the world's energy supply and 13.8% of its electricity generation. With an installed capacity of 106 GW the United States produces 30.8% of the world's nuclear power.¹⁰ Within the U.S. 19.4% of our electricity comes from nuclear power.¹ Clearly the United States is a major player in the field of nuclear energy. However, there have been no new orders for nuclear power plants since the 1970s, and the last plant to be completed was the Watts Bar nuclear reactor in 1996. The expansion of nuclear power has effectively ground to a halt in the United States. However, worldwide energy demand is expected to increase by a factor of 2.5 within the next 50 years.¹¹ If this demand is to be met while lowering green house gasses, nuclear energy must play a part.

Arguably the largest problem with expanding nuclear power is the accumulation of radioactive waste, much of it plutonium which could potentially be used in the manufacture of nuclear weapons and requires many thousands of years before it is no longer a safety hazard.⁸ Other problems included proliferation of nuclear weapons via uranium enrichment, which is required for both fuel rods and nuclear weapons. However, the degree to which the uranium must be enriched is much greater for the latter, $\leq 85\%$.

In terms of conservation of resources, fuel consumption efficiency is the major problem with our current methods of nuclear power generation. In the once through cycle used by the United States only 1% of the energy content from the uranium ore is utilized.¹² Spent fuel reprocessing programs have been created in Britain, France, Russia and Japan to address this problem. The most common method utilized is the PUREX (Plutonium URanium EXtraction) process. PUREX was developed in the United States in 1947, and was used primarily for the extraction of nuclear weapons material. It is a complicated aqueous process that is well documented and thus is not addressed in detail in this paper; however, the basics are covered.

The spent nuclear fuel is first dissolved in a strong acid, typically nitric acid. It is then mixed with an organic solvent, tri-n-butyl phosphate (TBP). Pu^{4+} and UO_2^{2+} are extracted here because they form neutral nitrate compounds, $\text{Pu}(\text{NO}_3)_4$ (aq) and $\text{UO}_2(\text{NO}_3)_2$ (aq), which become soluble in the TBP solvent. The fission products remain in the aqueous phase. Pu and U are further separated by using a reducing agent to strip the Pu compound of its organic state, this redox reaction leaves the $\text{UO}_2(\text{NO}_3)_2$ (aq) unaffected.¹³

The separated fuels are then further refined and enriched, with the final product being a mix of plutonium and uranium oxides. These mixed oxide (MOX) fuels can be burned in a modified LWR or fast reactor. MOX fuels are not used within the U.S. due to a ban issued on civilian separation of plutonium in 1977, stemming from rising concerns on nuclear weapons proliferation.

MOX fuel is not without problems. MOX fuel rods have a higher content of plutonium, americium, and curium than conventional spent fuel rods. This causes spent MOX fuel to require three times as much space as low-enriched uranium spent fuel and an additional few hundred years of cooling.⁸ Additionally, the PUREX process does not capture all of the minor actinides, so some make their way into the environment, or into the MOX fuel, reducing its efficiency

1.2 Generation IV Initiative

In response to “concerns over energy resource availability, climate change, air quality and energy security”³ the Generation IV Nuclear Energy Systems Initiative was approved by President Bush in 2001. The goal of this initiative is to have a new generation of reactors to start deployment by 2030 to replace the United States aging fleet of reactors. The plants are to be designed with the following criteria³:

1. Enhanced Safety
2. Cost effectiveness
3. Proliferation resistance
4. Minimization of nuclear waste

An international forum of thirteen countries selected six design concepts to fund for research and development. The six designs are the Gas-Cooled Fast Reactor (GFR), the Very-High-Temperature Reactor (VHTR), the Lead-Cooled Fast Reactor (LFR), the Molten Salt Reactor (MSR), the Sodium-Cooled Fast Reactor (SFR), and the Supercritical-Water-Cooled Reactor (SCWR).¹⁴

Both the VHTR and SCWR are thermal reactors with open fuel cycles, meaning the fuel is burned once in the reactor and then stored on site until it is shipped to a repository. Thus they will not be discussed further in this paper, as the open fuel cycle does little to minimized nuclear waste. The remaining reactor concepts utilize a closed fuel cycle in which fuel is burned in the reactor and is then reprocessed to remove detrimental isotopes. This reprocessed fuel can then be reformed into fresh fuel rods. The GFR is a helium-cooled fast reactor with the potential for the generation of hydrogen and full recycle of actinides, thus reducing radioactive waste. However, it runs at a very high temperature, 850°C, which creates the need for new materials capable of resisting damage in extreme environments. For this reason many of the fuel options are ceramics.³ The LFR utilizes liquid lead or lead bismuth alloy as a coolant. It is a fast reactor with a closed fuel cycle. Fuels being considered are nitrides or metallic. Molten lead has many benefits compared to other coolants; it is relatively inert and abundant.³ The MSR is fueled by a circulating liquid of sodium, zirconium and uranium fluorides through graphite channels. The MSR has an integrated fuel cycle, meaning that it can be employed as a fuel burner or breeder

depending on reactor characteristics.³ Additionally, molten salt has a high thermal conductivity, which allows for the MSR theoretically obtain a much higher thermal output than other reactor designs.³ Perhaps, the most developed of the fast reactor concepts is the SFR. This reactor concept has two potential designs, a large plant run on MOX fuel with an output of 500-1500 MWe and a central reprocessing station utilizing an aqueous reprocessing method.³ The second option is a small to medium plant using metallic fuel with an output of 150-500MWe and a pyrometallurgical reprocessing facility located at the reactor site.³ Both are designed with the management of plutonium and other actinides in mind and possesses passive safety features such as a long thermal response time, large margin to coolant boiling and coolant that operates at near atmospheric pressure.³

Fast reactors could be a solution to many of the problems associated with nuclear power and the waste it produces. Fast reactors utilize unmoderated fast neutrons to maintain the nuclear reaction. Hence, water cannot be used as a coolant. The high energy neutrons in a fast reactor have a much higher ratio between splitting and the capture of neutrons than the thermal neutrons of a LWR. Thus, in a fast reactor the U-235, Pu-239 and a greater fraction of the transuranic elements are fissioned. This gives fast reactors an advantage in fuel efficiency and reduces the time in which the spent fuel must be isolated.

In the early days of nuclear power, nuclear engineers assumed that spent fuel from a LWR would be burned in fast reactors, which would in turn be used to create more reactor fuel. This would be done through neutron irradiation of fertile materials by placing a blanket of U-238 or Th-230 around the reactor core. This blanket would be set a distance away from the core and catch some of the neutron leakage from the core to generate Pu-239 and U-233 from the U-238 or Th-230, respectively.¹⁵ However, due to concerns over proliferation of the plutonium for use

in nuclear weapons the United States breeder reactor program was cancelled when President Carter banned the reprocessing of nuclear fuel in 1977. This banned any reprocessing such as PUREX and the creation of plutonium in fast reactors.

Of these design concepts the sodium cooled fast reactor and its associated fuel cycle are the topic of this paper. Additionally, this design concept is analyzed to ascertain how well it satisfies the Generation IV criteria of sustainability, enhanced safety, proliferation resistance and economic competitiveness. This selection was made due to the greater amount of technological experience with sodium cooled reactors.³ The Argonne National Laboratory have been working with sodium cooled reactors with metallic fuels as well as the associated pyrometallurgical process since the 1980s.¹⁶ The Russian's have had some success with mixed oxide fueled, sodium cooled reactors with their BN-600 plant. France has also developed two commercial sodium cooled fast breeder reactors utilizing mixed oxides fuels, the Phenix and SuperPhenix. The United States developed the Southwest Experimental Fast Oxide Reactor, in northwest Arkansas to test the safety features of the sodium cooled, mixed oxide configuration. In conjunction with using fast reactors, a new approach to reprocessing spent waste known as pyroprocessing is addressed.

1.3 Fuel Cycle

Pyroprocessing is a dry process that has been under development at ANL for reprocessing metallic fuel from the EBR-I and EBR-II reactors. It is a dry process utilizing salts and high temperatures to separate uranium and other actinides from the fission products. This process possesses advantages over PUREX in that it is more compact, less complex, has less waste streams, and creates a highly radioactive fuel that makes diversion to weapons difficult.¹⁷

Present day nuclear reactors have what is known as the once through nuclear fuel cycle. Uranium ore is mined, then processed into yellowcake(U_3O_8). The uranium is then reduced to metal and combined with fluorine to form uranium hexafluoride gas. Isotope separation then takes place via gas diffusion or gas centrifuges. The goal of this procedure is to enrich the content of U-235 to 4% for nuclear reactors or upwards of 90% for weapons or small nuclear reactors such as those found in nuclear submarines. The enriched uranium is then converted to uranium dioxide and cast into pellets, which are then placed in fuel rods. After the fuel rod is spent, it is transferred to a Spent Fuel Pool filled with water to allow the fuel rods to cool and shield them from releasing radiation. The rods are typically stored for ten to twenty years before being moved to dry cask storage. These dry casks are constructed of steel and filled with an inert gas. The casks are stored on site because approval for the Yucca Mountain repository has not occurred.¹⁸

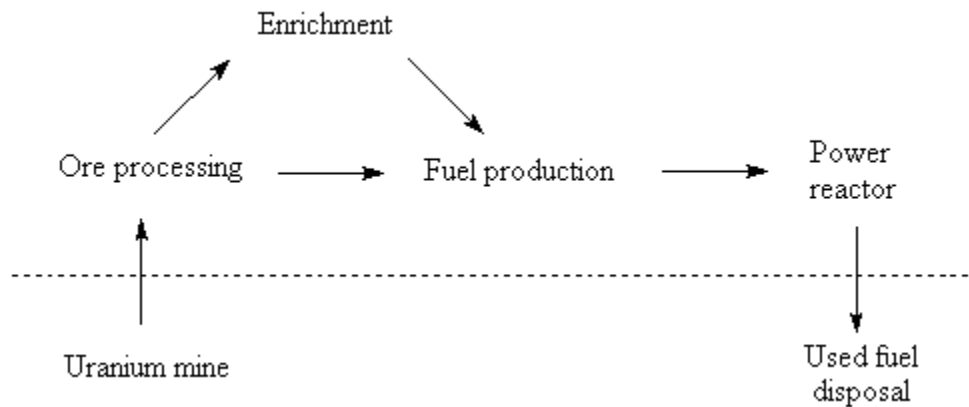


Figure 1: Flow sheet of Open-Fuel cycle in which uranium ore is mined, refined via centrifuge or gas diffusion and then converted to uranium dioxide pellets to be place in fuel rods. Following burning in a reactor, fuel is placed in a cooling pond awaiting disposal at Yucca Mountain.

Fast reactors possess advantages over LWR when it comes to choice of fuel. This advantage arises from the use of the fast neutron spectrum, in which most of the transuranic isotopes will undergo fission. Transuranics or transuranium elements are those with an atomic

number greater than 92, the atomic number of uranium. All transuranium elements are unstable and decay radioactively into other elements. The fuel cycle of the fast reactor is known as a closed fuel cycle because the spent fuel is reprocessed and reused as opposed to the once-through fuel cycle. The closed fuel cycle is far more efficient than the once-through because most of the energy in the mined uranium is burned, and the waste would mostly be composed of the fission products.¹²

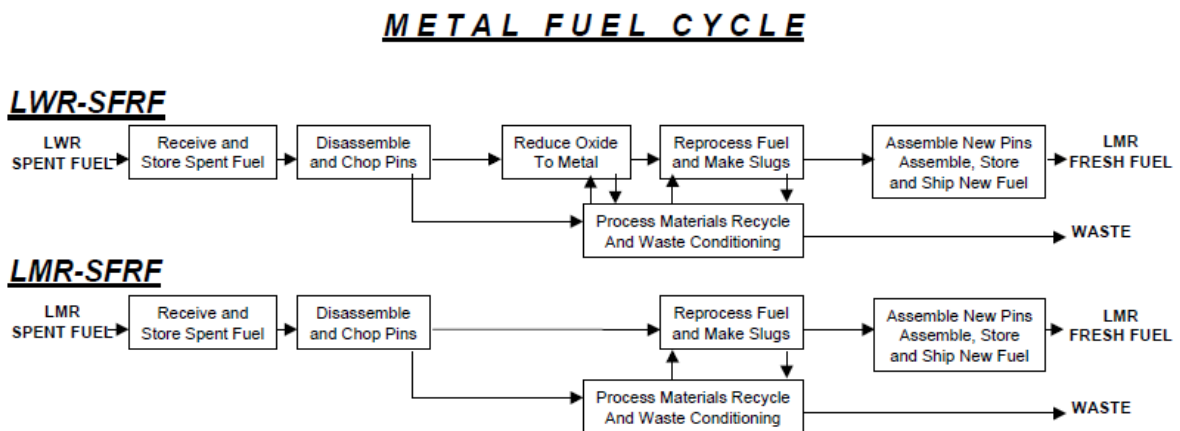


Figure 2: Flow sheet for Closed fuel cycle utilizing sodium cooled fast reactors. Spent oxide fuel from Light Water Reactors(LWR) is chopped up, and reduced to a metal for reprocessing. New metallic fuel rods are formed and burned in a fast reactor. Once burned, fuel is again chopped and reprocessed with waste being collected during the initial oxide reduction step and all reprocessing steps.¹⁹

The fast reactor fuel cycle would begin at the back end of the open fuel cycle with oxide reduction of the spent fuel to convert it to metal. Metallic fuels are the ideal for fast reactors because metal fuel benefits from simple fabrication, high thermal conductivity, and small Doppler reactivity feedback.^{20, 21}

There are several different approaches to reprocessing of spent nuclear fuel. Current methods are aqueous and derived from the PUREX process. This paper will focus on the fuel cycle of the Integral Fast Reactor(IFR) which is a prototype design based on the work of the Advanced Liquid Metal Reactor. The IFR is a sodium cooled fast reactor that burns metallic fuel and performs reprocessing on site thus limiting the shipment of highly radioactive materials to startup.¹⁶ Additionally, the sodium cooled system with pyroprocessing was scored first overall in a Department of Energy study of the Generation IV reactor designs.²²

1.3.1 Voloxidation

Nuclear fuel rods from a LWR are composed of either uranium dioxide or a mix of uranium and plutonium oxides, MOX. For use in an IFR concept the spent fuel must undergo two processes: voloxidation and electrolytic oxide reduction to reduce the fuel to metal.¹⁷

Voloxidation, volumetric oxidation, is a process under research at the Idaho National Laboratory(INL) in collaboration with the Korea Atomic Energy Research Institute(KAERI). It is an procedure with the purpose of decladding spent oxide fuel and removing some of the volatile fission products such as Cs, Tc, Mo, I, Kr, and Xe.^{23,24} The entire procedure must be done remotely due to the high radioactivity of the spent rods. The fuel rods are heated in an oxygen rich environment at temperatures between 500-700° C for 1-2 hours, which results in oxidation of UO_2 to U_3O_8 via the reaction $3\text{UO}_2 + \text{O}_2(\text{g}) \rightarrow \text{U}_3\text{O}_8$.²⁵ The volume increase associated with this reaction causes the fuel claddings to rupture, as seen in Figure 3 below.



Figure 3: Spent Fuel after voloxidation. It can be seen that chopped fuel rods have been reduced to powder due to voloxidation process and that fuel rods have been ruptured.²⁵

After the oxidation of uranium dioxide, the temperature is further increased to excess of 1000°C to promote the oxidation of fission products which escape as gasses. The gasses can then be collected and stored. The removal of fission products via voloxidation simplifies later reprocessing be it aqueous or pyroprocessing.²⁴ In the experiments conducted by INL up to 98% of Tc and 30-40% of both Cs and Mo were successfully removed.²⁴

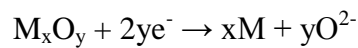
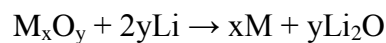
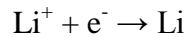
1.3.2 Electrolytic Reduction

Following voloxidation the oxidized fuel needs to be reduced to a metallic state. This can be accomplished by a process under development at INL and KAERI, known as electrolytic reduction. The basics of the process are as follows;

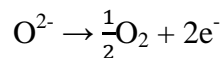
“... spent oxide fuel is crushed and loaded into a permeable steel basket, which is submerged in a molten salt electrolyte of LiCl-1 wt% Li₂O at 650°C. A power supply is connected to the fuel basket (the cathode) and to a submerged platinum wire (the anode). A controlled electric current is passed between the cathode and anode to effect the reduction of oxide fuel to metal and the formation of oxygen gas on the platinum wire surface. The reduced metal remains in the submerged cathode basket and the oxygen gas is allowed to surface and dissipate from the electrochemical cell.”²⁶

In addition, fission products such as cesium, barium, strontium and iodine which are salt soluble diffuse into the molten salt.²⁶

The chemical reactions that take place at the cathode are:



And at the anode:



where M is uranium or a transuranium element.²³

The benefit of this procedure is that it can be carried regardless of the extent of oxidation of the metal. Idaho National Laboratories performed electrolytic reduction experiments with spent LWR fuel, UO_2 , from Belgium Reactor-3 and the Korea Atomic Energy Research Institute performed electrolytic reduction of U_3O_8 , after voloxidation of spent fuel.²⁶ Both experiments performed were bench scale and aimed at adapting the pyrochemical process for metallic fuels, developed by Argonne National Laboratory, to oxide fuels. Results of INL tests showed reduction efficiencies of “99.7% for uranium, ... 97.8% for plutonium, >98.8% for neptunium and 90.2% for americium.”²⁶ Following the electrolytic reduction, the Li-Cl salt containing the Cs, Sr, and Ba fission products can be converted into ceramic waste by heating a mixture of

zeolite and salt and heating to approximately 950°C.²⁷ The reduced metallic fuel is now ready to be sent to the electrorefiner.

1.3.3 Electrorefining

From this point on the process of refining the fuel is identical to that of refining spent metallic fuel from a fast reactor, as can be seen from Figure 2. Electrorefining is accomplished by placing spent fuel in a steel basket and submerging it in a molten LiCl-KCl electrolyte at 450°C. A solid iron cathode for recovery of uranium and a liquid cadmium cathode (LCC) for plutonium and minor actinides are lowered into the molten salt, and the steel basket is configured as the anode.²⁸ A simple diagram of this configuration is shown in Figure 4, below.

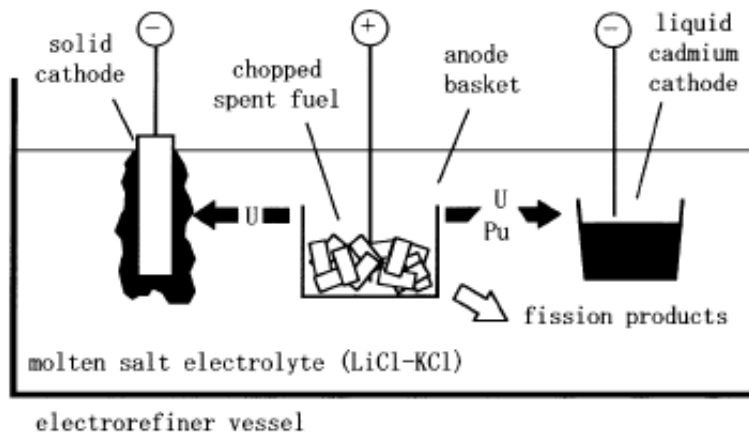


Figure 4: Simple schematic of an electrorefiner vessel. Shown is the anode basket containing chopped spent fuel, the solid cathode for collecting uranium, the liquid cadmium cathode for collecting U, Pu and other transuranics. Not shown is the stirring bar that promotes convection.²⁹

The actinides in the fuel baskets are anodically dissolved into the electrolyte while fission products accumulate as chlorides in the molten salt. Uranium is deposited on the solid cathode “because the free energy change of chloride formation for uranium is negatively less than those of the other actinide elements.”²⁹ The “transuranium elements are stabilized in the liquid

cadmium cathode due to their very low activity coefficients in liquid cadmium.”²⁹ Research on electrorefining has been conducted by Argonne National Labs, Central Research Institute of Electric Power Industry(CRIEPI), and Japan Atomic Energy Research Institute. These tests have all been laboratory scale, with the exception of those performed at ANL; which has conducted engineering scale tests with the Mk-IV and Mk-V electrorefiners shown in Figure 5. The Mk-V electrorefiner performed without a liquid cadmium cathode due to the shutdown of the Experimental Breeder Reactor-II and the lack of need for minor actinide retrieval for fuel fabrication.¹⁷

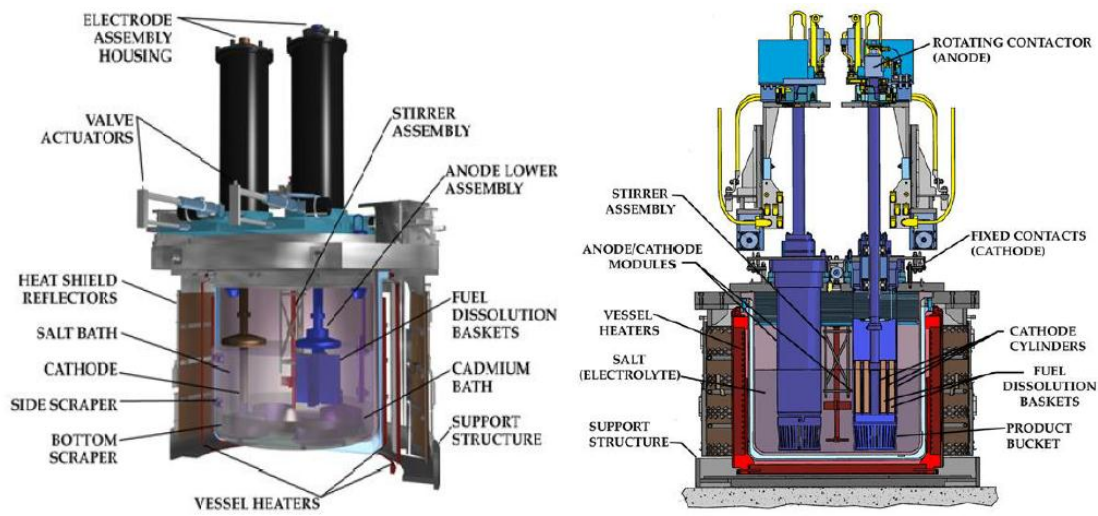


Figure 5 of Mk-VI(left) and Mk-V(right) electrorefiners, respectively. Both refiners performed in the same manner, however, the Mk-V lacked a LCC and has a higher throughput.³⁰

The Mk-VI electrorefiner was built to process spent EBR-II driver fuel and the Mk-V to process the blanket fuel, which exists in much larger quantities.³⁰ Other than differences in purpose and lack of a liquid cadmium cathode for collection of transuranics, both electrorefiners function in the same manner. $CdCl_2$ is added at the beginning of each run to cause oxidation of some uranium metal to U^{3+} to promote deposition on the cathode. The U^{3+} travels through the

salt by forced convection from a stirring bar and is reduced to metal at the cathode.³⁰ A scraper is employed in both models to prevent excessive buildup of uranium dendrites. The collection of transuranics is accomplished in a two step process involving anodic dissolution and cathodic deposition. In anodic dissolution the basket is configured as the anode and a pool of cadmium as the cathode. Uranium and minor actinides travel through the salt and are reduced at the cadmium and dissolve.³⁰ In cathodic deposition the cadmium pool containing dissolved transuranics is configured as the anode and the steel mandrel as the cathode.³⁰ Mk-VI tests have achieved U dissolution of 98% and upwards of 99% Pu dissolution.¹⁷ In addition to Pu and U recovery; Np, Am, and Cm are deposited on the liquid cadmium cathode.²⁸ This results in no pure Pu, which adds proliferation resistance to the procedure. In using a cadmium cathode, actinides accumulate in cadmium which requires distillation for future use in fuel recycle. Tests performed at INL have recovered greater than 99wt% of the cadmium via cadmium distillation.¹⁷

There are two waste forms generated in electrorefining, metal and ceramic. As more electrorefining runs are done, chlorides of fission products begin to accumulate in the molten salt. These chlorides must then be disposed of in a ceramic form and replaced, or purified of fission products and reused.³¹ The creation of the ceramic waste for the LiCl-KCl salt is identical to that of the LiCl-LiO₂ process discussed in the electrolytic reduction section. A flow sheet of the steps involved in the process of recycling salt, from both electrolytic oxidation and electrorefining, is shown in Figure 6.

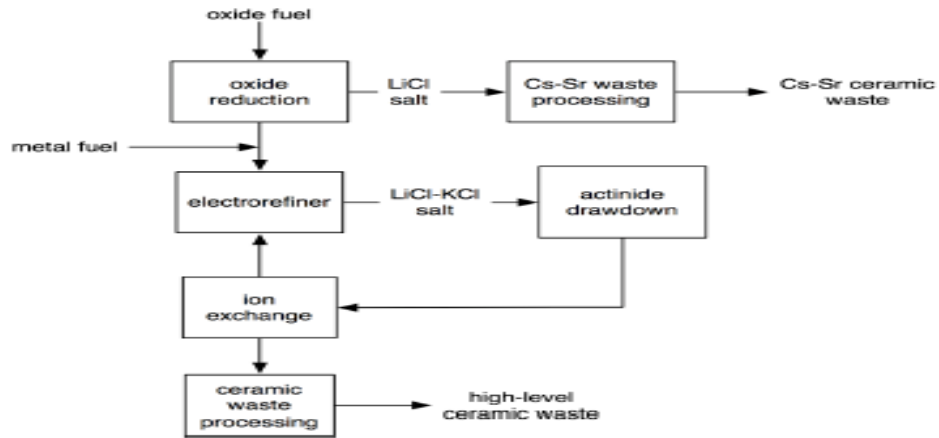


Figure 6: Flow sheet for ceramic waste in pyroprocess. Li-Cl salt is processed to removed Cesium and Strontium fission products and contain them in ceramic.²⁷

The second waste form is composed of “the stainless steel cladding, the noble metal fission products(Zr, Mo, Ru, Tc, Te, Rh, Pd, Nb, others) and some residual actinides that do not react in the electrorefining process.”³¹ The metallic waste can contain up to 4% noble metals and 11% uranium.³⁰ Stainless steel and zirconium-wt 15% from the EBR-II fuel make up the remainder of the metallic waste.³² Homogenous metallic ingots have been produced in experiments at ANL by placing the cladding hulls leftover from electrorefining in a distillation furnace and heating to 1100°C at 1 torr for many hours to remove salts from the hulls.³² The hulls and additional zirconium are then placed in a crucible within a vacuum induction furnace and heated at 1600°C for three hours.³² The result is a disc shaped ingot ready for storage in a repository.

1.3.4 Fuel Fabrication

There are many methods of casting fuel rods under development including injection casting, centrifugal casting and continuous casting.²⁰ All have challenges and advantages but the

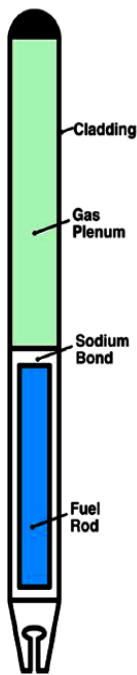


Figure 7:
Schematic of a
basic metallic
fuel rod.³³

most accepted method is that of injection casting.^{20,33} In this process the components of the fuel rod (U, Pu, Zr & MAs) are loaded into a crucible. Those with a low melting temperature are placed on the bottom, to reduce the ability of elements with high vapor pressures from escaping. The furnace is in a vacuum at the beginning of each run. Once the mold has been lowered into the melted alloy, the furnace is pressurized with argon gas, forcing the alloy into the mold.²⁰ The entire fuel rod is composed of four parts; the fuel element containing U, U-Pu or a mix of U-Pu and minor actinides, a sodium bond between the fuel element and cladding, a fission gas collection plenum, and a stainless steel cladding of austenitic composition or nickel-based alloy.³³ The sodium allows for heat to be easily transferred to the cladding and reactor coolant. The plenum collects fission gasses released during burn-up. A simple schematic can be seen in Figure 7.

1.3.5 Reactor Startup

The metallic or MOX fuel can now be inserted into a fast reactor. The initial startup fuel is obtained from the reprocessed spent light water reactor fuels. All further refueling can be accomplished with natural uranium, depleted uranium, decommissioned nuclear weapons or spent LWR fuel.^{15,33} Fast reactors can be run in three different modes; breeder, burner and breakeven, depending on the materials used in the blanket. To run as a fuel breeder uranium-238 is used as the radial blanket around the core, to increase breeding ratios axial blankets are also utilized within the core assembly.¹⁹ As the nuclear reaction progresses, some neutrons are slowed due to inelastic scattering. These slow neutrons can be absorbed by U-238 which, following an absorption of a neutron, decays to Pu-239 after two beta particle emissions. This plutonium can

be used as startup fuel for other fast reactors. If run in a fuel burning mode, the radial blankets are replaced by steel reflectors and the axial blankets are removed. This operational mode burns both fissile and fertile elements, and with pyroprocessing will only output activation products and fission products.¹⁹ Activation products are materials that have been irradiated, such as fuel cladding, and fission products are elements created as a result of the fission reaction. The breakeven mode is a mix between the two prior modes with the axial blankets removed while maintaining the radial blanket. The core design of the S-PRISM reactor by General Electric can be seen below in Figure 8. The S-PRISM reactor is a modular sodium cooled fast reactor with metallic fueled based off research from the Advanced Liquid Metal Reactor(ALMR) and the Integral Fast Reactor(IFR).¹⁹

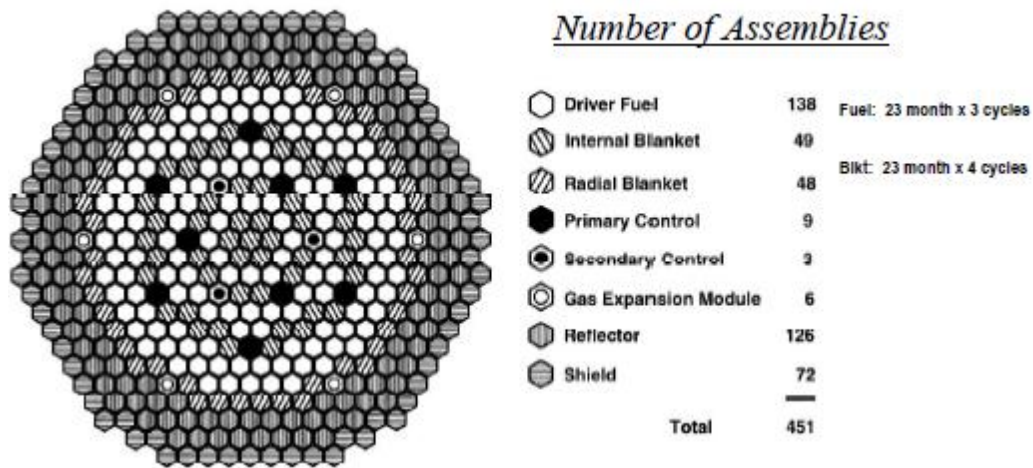


Figure 8: S-PRISM Core Layout-Internal blankets are referred to as Axial blankets. This reactor could function as a burner or breeder reactor.¹⁹

1.3.6 Plant Design

The two common designs for sodium cooled fast reactors are the “Pool” design and the “Loop” design seen in Figure 9. The loop design pumps liquid sodium from the reactor vessel to an intermediate heat exchanger containing non-radioactive sodium, which passes through a

secondary heat exchanger containing water. The water then passes through turbines to generate electricity. The purpose of the dual heat exchangers is to minimize the chance of radioactive sodium-24 coming into contact with water or air in the event of a pipe rupture. The pool design is similar in its intermediate and secondary heat exchangers; however, rather than pump radioactive sodium out of the reactor core, the secondary coolant loop enters the core. This limits the possibility of radioactive release. The pool type design was part of the IFR concept as well as the S-PRISM design by General Electric. Electromagnetic pumps are used but natural convection can cool the reactor core sufficiently in loss of power scenarios.²¹ Figure 9 shows the different schematics of both pool and loop plant designs.

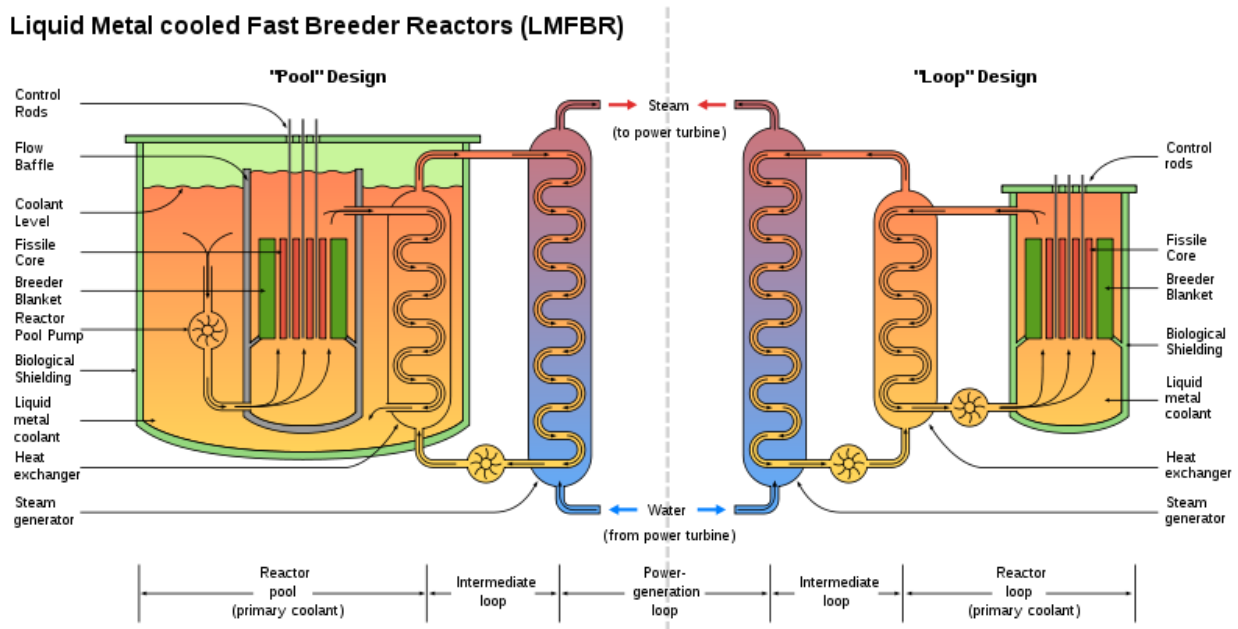


Figure 9: Basic Pool and Loop plant designs. Pool designs benefit from the intermediate loop being contained within the reactor coolant pool, reducing the risk of sodium-14 exposure to the environment.

2. Analysis

2.1 Sustainability

Sustainability as defined by the Generation IV initiative is split into two categories; waste minimization and resource utilization.²² Fast reactors excel at efficient use of resources due to their higher neutron spectrum, allowing for a wider range of actinides to be used as fuel sources. Waste minimization is could be achieved with the development of the pyro-process to recycle most of the transuranic elements, leaving only fission products and activation products to be sent to a repository. The closed fast reactor cycle coupled with an advanced recycling process could reduce the radiotoxicity of the waste to require about 1000 years of isolation rather than 100,000 years as planned for the Yucca Mountain Repository.²²

2.1.1 Resource Utilization

Studies have found that given the expected growth of nuclear energy in the future, current uranium resources will only last until mid-century with the current fuel cycle.¹² If there is no growth in nuclear power uranium resources will last 80 years. However, technological advances in mining and extraction of uranium from the oceans could expand potential reserves.³⁵ Figure 10 taken from the DOE roadmap for the Generation IV initiative shows three scenarios: one in which fast reactors are not introduced, another if fast reactors are introduced by 2030 and if introduced by 2050. According to the graph, only by introducing fast reactors by 2030, the date established by the Gen IV initiative, will it be certain that the uranium reserves will not be exhausted.

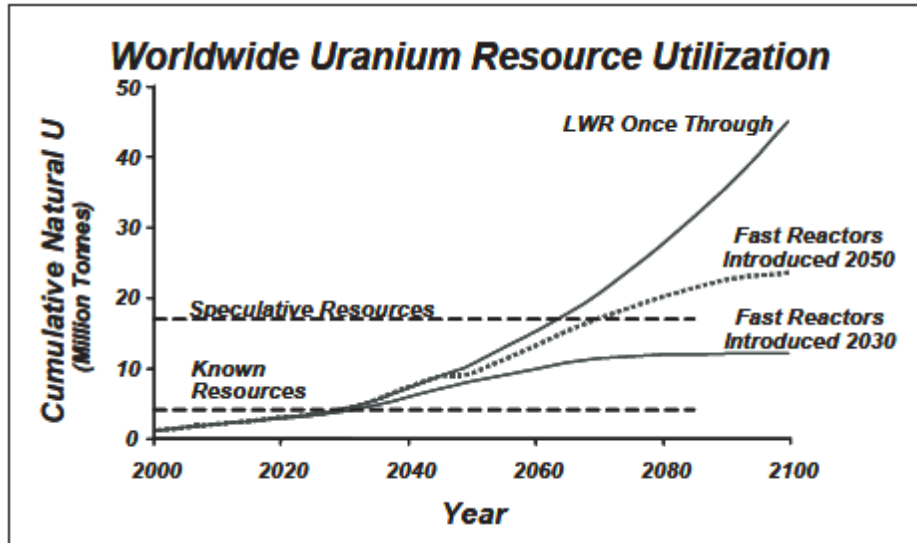


Figure 10: Uranium consumption over time for various fast reactor introduction scenarios.

However, speculative resources could potentially be higher due to improvements in mining technology.³

Fast reactors can burn reprocessed spent fuel from LWR or weapons grade plutonium, thus eliminating the need for additional uranium mining. Due to the higher energies of neutrons in a fast reactor nearly 99% of the energy can be extracted from the fuel.¹² Additionally, once the reactor has reached critical it, can create more fuel for the start-up of other fast reactors via breeding and sustain its self with occasional shipments of natural or depleted uranium. The purpose of adding U-238 is to replace heavy metal atoms that have fissioned. In the United States there are currently 480,000 tons of depleted uranium and 1.2 million tons worldwide. If breeding is considered, uranium resources can potentially last many thousands of years.¹¹

2.1.2 Waste Minimization

The closed fuel cycle of fast reactors possesses advantages over the open cycle in terms of waste minimization, this is due to the reduced volume of radioactive waste at the end of the cycle. This is true for fast reactors utilizing MOX fuel via PUREX and metallic fuel via

pyroprocessing. However, the latter has the advantage of only two waste streams, metallic and ceramic, and produces no High Level Liquid Waste(HLLW) in the form of organic compounds. The pyro-process also has less gaseous discharge. Only tritium and Kr-85 are released into the argon cover gas, compared to the PUREX process, which off-gasses H-3, Kr-85, in addition to C-14, and I-129.³⁶ Additionally, it has been shown in laboratory scale tests that the pyroprocess can be adapted to recycle the HLLW created in the PUREX process.²⁸

Whether run on MOX fuel or metallic fuel, fast reactors will have the advantage of a harder neutron spectrum, causing a higher burnup of actinides. Differences between burning metallic and MOX fuels are relatively insignificant in regards to the transmutation of minor actinides. Metallic fuels have a small advantage of a slightly higher breeding ratio, require less Pu enrichment, and have a 10% larger minor actinide transmutation amount.³⁷ Thus the fuel choice does not have a major impact in the transmutation and reduction of minor actinides.

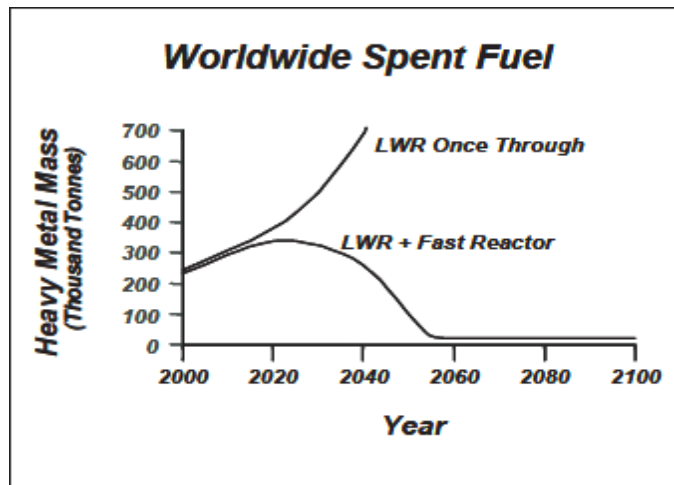


Figure 11: Spent Fuel accumulation vs. time under the assumption that nuclear power expands to compensate for fossil fuel. The LWR + FR scenario envisions a symbiotic relationship where fuel is first burned in a LWR and passed on for refinement and burning in FRs.³

If a pyro-process or advanced aqueous process were utilized, most of the uranium and transuranics would be removed. As a result this reduces volume limitations placed on a repository, as uranium and transuranics account for 97% of the mass of current spent fuel rods. Taking into account the maturity of the PUREX process, it is likely that the first fast reactors will utilize MOX fuel. However, this would require a change in U.S. policy or further development of PUREX based processes which extract plutonium and the transuranics as a whole; UREX+, NUEX, and COEX. From Figure 11 above, it can be seen that if the open cycle were to continue unimpeded, worldwide spent fuel would more than double by 2040. If fast reactors are steadily introduced to burn spent LWR fuel, stockpiles can be reduced and altogether eliminated through a symbiotic relationship, in which LWR irradiate fuel which is then passed to fast reactors for further irradiation and transmutation. Therefore, fast reactors are a potentially viable means to reduce the impact of nuclear power and maximize the potential of uranium reserves. However, most reprocessing techniques that comply with U.S. law are still in their infancy and require more research. In addition, it has yet to be proven that pyroprocessing would be as efficient on a commercial scale.

2.2 Proliferation Resistance

The goal of proliferation resistance is to have intrinsic features of fuel materials and processes such that³⁸:

- The fuel has a high content of actinides
- Group recovery of actinides
- Final products are highly radioactive leading to fatal dose in large quantities
- Alteration of plant for weapons purposes must be more difficult than a covert reprocessing facility

- Integrated concepts limiting the transport of nuclear materials.

These criteria eliminate the PUREX process due to the extraction of pure plutonium as an intermediate step, which can be diverted towards nuclear weapons. The pyroprocess avoids the separation of pure plutonium by extracting all transuranic elements in one group. This mixture of plutonium and minor actinides is highly radioactive, with a neutron dose 140 times higher than pure plutonium from LWR spent fuel.³⁹ This high radioactivity requires the fuel to be handled remotely in a shielded environment thereby giving it a degree of self-protection.

The high neutron dose and decay heat of this actinide mixture make it unsuitable for manufacture of nuclear warheads. The entire manufacture process would need to be done remotely and if constructed, the heat radiated would melt the chemical detonator and alter the shape of the fissile sphere.⁴⁰ If the obstacle of decay heat were to be overcome, the warhead would detonate prematurely, resulting in a small release of energy and the dispersal of radioactive waste.³⁸ The same effect or greater could be accomplished with a conventional explosive and spent fuel at a greatly reduced cost.

The implementation of an integral system is beneficial because it limits the shipment of potential divertible nuclear materials. In the case of the IFR, potentially divertible materials are only shipped to a reactor during start-up and all other shipments can consist of fertile materials unsuitable for diversion towards weapons.^{15, 34} The risk from shipping potentially divertible material for reactor start-up is no greater than shipping the same material to Yucca Mountain. Surveillance can also be employed for all aspects of the fuel cycle, from reprocessing to burn-up, due to all facilities being housed in a single secure complex.

In order to achieve proliferation resistance a hypothetical fast reactor would likely operate as a fuel burner or in break-even mode. This requires the elimination of radial blankets of fertile material surrounding the core with which plutonium is bred via neutron leakage. However, research has been conducted to study the ability of breeding proliferation resistant plutonium through the incorporation of minor actinides, specifically Np-237 and Am-241 which decay into Pu-238.^{40, 41} By breeding plutonium that is 12-20% Pu-238, weapons construction can be discouraged due to Pu-238 high spontaneous fission rate and high decay heat.⁴⁰

By making the diversion of reprocessing facilities and nuclear plants more difficult than a covert PUREX plant, a high degree of proliferation resistance can be achieved. However, this requires reprocessing techniques that reprocess the plutonium and the minor actinides as a whole. In this respect, the sodium fast reactor employing metal alloy fuel is favored over a MOX fueled reactor due to the use of the pyro-process.

2.3 Safety

In a post-Chernobyl age, safety is a major consideration in the design of nuclear facilities and inherent safety features, those lacking human intervention, are emphasized. Limiting possibilities for human error are crucial to building public confidence, as the Three Mile Island and Chernobyl accidents were largely the result of operator error. The event at the Fukushima plant resulted from the tsunami damaging the coolant pumps. With the pumps offline, the water around the reactor and in cooling ponds boiled off, leaving the fuel rods exposed and without coolant. As a consequence, safety features must be robust enough to contain and limit reactor damage in the event of multiple system failures.

Safety features are divided into two categories by the DOE: intrinsic, those having to do with plant design, and extrinsic, such as offsite emergency response and containment.³ Being that

extrinsic safety features are based on policy and protocol rather than design, therefore I will not be addressing these. There are three major transient events that must be accounted for within a reactor design: loss of flow transient(LOF), transient overpower(TOP) and loss of heat sink(LOHS). Typical examples of these transients are; electromagnetic pump malfunction within the core, accidental removal of control rod and loss of flow within the secondary sodium loop, respectively. Despite differing nuclear and thermal characteristics between MOX and metallic fuel, both reactor types share many safety features to mitigate or prevent transient events.

In considering operational safety, a few key terms must be defined. Two terms of importance are the fuel temperature coefficient of reactivity, also known as the Doppler coefficient and the void coefficient of reactivity. The Doppler coefficient is defined as the “change in reactivity per degree of change in the temperature of the nuclear fuel.”⁴² Particular elements such as U-238 absorb more neutrons from the fission process as temperature increases, resulting in the fission reaction stabilizing. The void coefficient is defined as the “rate of change in the reactivity of a water reactor system resulting from a formation of steam bubbles as the power level and temperature increase.”⁴² This definition applies to any liquid cooled reactor system. Other coefficients of importance are thermal and power coefficients of reactivity. The thermal coefficient of reactivity corresponds to the change in reactivity with respect to the temperature of the coolant, while the power coefficient of reactivity corresponds to the change in reactivity as the power level of the reactor increases.

Most current sodium cooled reactor designs possess positive void coefficients of reactivity as well as negative Doppler coefficients. The creation of voids within the sodium coolant leads to less scattering with the sodium atom resulting in a harder neutron spectrum. The

hardening of the spectrum causes more neutrons released per fission and an increase in the fission of fertile materials leading to an increase in reactivity.⁴³ The positive void coefficient of reactivity creates obstacles for the deployment of fast reactors as all reactors with this property are illegal to construct in the United States following the Chernobyl meltdown. Thus efforts are directed towards designing a reactor with a negative void coefficient or neutralizing its effect with negative power and thermal coefficients.

Sodium cooled fast reactors(SFR) possess features that make them well suited to exploit the concept of passive safety. The key factor being their high boiling point that allows liquid metal reactors, such as the SFR, to operate at approximately atmospheric pressure and with coolant temperatures that are several hundred degrees below its boiling point.⁴⁴ This large thermal margin allows significant time for negative reactivity feedback mechanisms to counteract the transient. Tests at RAPSODIE reactor in France, EBR-II and the FFTF in the United States, were conducted to assess the robustness of passive safety features for sodium cooled fast reactors.

The RAPSODIE reactor in France is an experimental nuclear reactor designed to test passive safety features for use in the Phenix reactor. RAPSODIE was a loop-type reactor with a thermal output of 40MW. In 1983 the plant's protective system was intentionally disabled and the reactor was subjected to a unprotected loss of flow transient at 50% power.⁴⁴ The reactor's outlet temperature rose initially but automatically reversed due to the negative temperature coefficient of reactivity of sodium.⁴⁴

The Experimental Breeder Reactor-II(EBR-II) is a fast reactor at Idaho National Laboratory. It is a pool-type reactor with a thermal output of 62.5 MW and metallic fuel. Two

tests were conducted at EBR-II in 1986. At full power and with protective systems disabled, the cooling pumps were shut off resulting in an unprotected loss of flow transient(ULOF). Reactor power spiked and then dropped off to anticipated event temperatures within 300s. This was due to negative temperature and power coefficients, as well as natural convection of the sodium.⁴⁵ Peak temperature during the test was 1150 F granting a large thermal margin to the sodium boiling temperature of 1650F.⁴⁵ A second test was performed at full power, in which flow in the secondary sodium loop was stopped, thereby preventing heat to leave the primary pool causing a loss of heat sink transient(LOHS). The temperature of the sodium within the reactor was quickly reduced to levels lower than normal full power.⁴⁵

Both the RAPSODIE and EBR-II tests demonstrated the feasibility of utilizing fast reactors negative coefficients of reactivity to prevent catastrophe in unanticipated events. However, both reactors were much smaller than a typical commercial plant. The French Phenix reactor produced 563MW of thermal energy for example. The Fast Flux Test Facility(FFTF) was constructed to gain further insight into fast reactor design and operation. The FFTF produced 400 MW of thermal energy utilizing MOX driver fuel. The FFTF adopted a Passive safety test program in 1986. However, the reactor possessed a power density level much higher than commercial designs, due to the nature of the irradiation experiments it performed, thereby limiting its ability to test passive safety features.⁴⁶ Tests performed verified that natural circulation of sodium is a viable means of avoiding a LOF transient. Gas expansion modules(GEMS) were also studied as a passive means of rapidly inducing negative reactivity into the core.⁴⁶ Results from LOF tests at the three reactors can be seen in Figure 12 below.

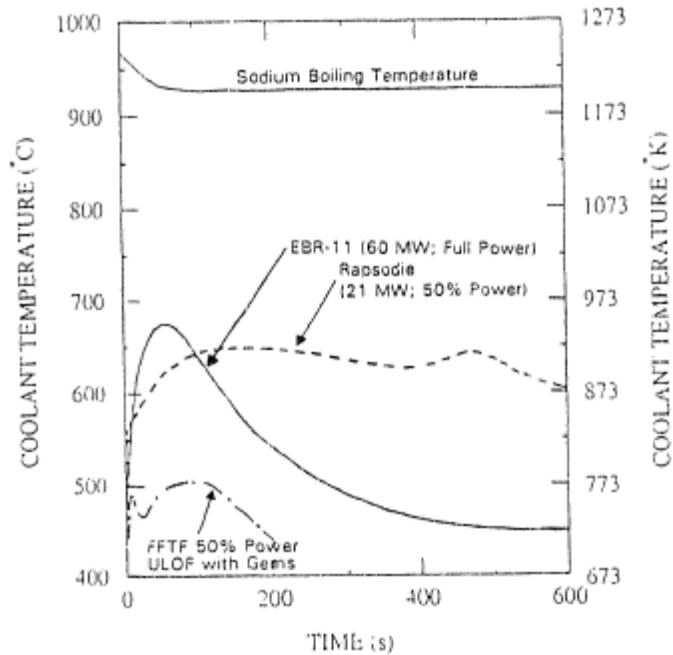


Figure 12: Unprotected loss of flow transients in the EBR-II, Rapsodie and FFTF reactors. All reactors performed well and had sizable margins before coolant boiling.⁴⁴

In addition to transient reactor events, the risk of the sodium coolant reacting with water and air is of serious concern. The most severe fire occurred in Japan's Monju reactor in 1995, when a pipe carrying sodium ruptured, exposing it to the air resulting in a large fire. This accident resulted in no radioactive release or injuries. The severity of the accident was minimized because the rupture occurred in the secondary sodium loop carrying non-radioactive sodium.

While fast reactors have shown appealing characteristics for passive safety, most tests have been performed on a small scale and further research would be required before such safety features could be considered acceptable for a commercial scale. Sodium leaks and fires are also a large problem that has plagued most fast reactor programs. Taking into account the recent

events at the Fukushima plants, nuclear plants will have to be held to higher standards of safety due to public concern. If the SFR program is to be implemented, safety features must be thoroughly vetted and sodium leaks must be contained due to the reactive nature of sodium when it comes into contact with water or air.

2.4 Economic Competitiveness

The largest hurdle facing the incorporation of fast reactors into the United States energy plan is achieving economic competitiveness with thermal reactors. Various studies have been conducted assessing the potential of fast reactors as a source of power. Given that such studies must rely on various assumptions, vastly different conclusions have been reached. Assumptions on the future expansion of nuclear power, uranium ore prices, cost of construction, reprocessing and maintenance are some of the many factors that must be considered. Factors such as cost of construction and reprocessing are difficult to quantify, which has caused a discrepancy in predictions, largely between scientists and economists.

Researchers at Argonne National Laboratory reached the conclusion that fast reactors could be competitive with LWRs if certain events take place in the future. The most critical being a sharp increase in the price of uranium ore following a large worldwide expansion of nuclear power.⁴⁶ It is also noted that while fast reactors may not be competitive presently, further research and development will lead to increases in efficiency and reliability. Scientists working on the European Fast Reactor program also concluded that their conceptual design could be competitive with thermal reactors.⁴⁷

Opposite conclusions were reached by a group of public policy experts and scientists at Harvard. In a 2003 report they concluded that direct disposal of spent nuclear fuel would be far cheaper than reprocessing and burning in fast reactors. In addition, they state that uranium ore

prices will remain low throughout the century.⁴⁹ In addition they found that for fast reactors to become viable, uranium ore prices would have reach to prices of \$360 per kilogram, far about the current price of ~\$90 per kilogram.⁴⁹ A more recent study by the International Panel on Fissile Materials reached a similar conclusion; that fast reactors reliability issues lead to increased cost and an uranium ore price of \$340 per kilogram would be necessary for fast reactors to be competitive.

Given the vastly different conclusions about the competitiveness of fast reactors and the number of factors that must be taken into consideration, it is difficult to discern whose model is correct. However, it appears the most important factor in the competitiveness of fast reactors is the price of uranium ore. While it has been rising in recent years, it would seem unlikely that it would reach prices proposed in the Harvard and IPFM studies.⁴⁸ However, an increase in nuclear power development worldwide could create such a situation. In the current global climate, such a situation is hard to predict, public opinion of nuclear power has dropped in United States due to the Fukushima event, although it is still higher than much of the globe.⁵⁰ If public opinion is unfavorable, then it is unlikely that funding will be allocated to research and design of experimental fast reactors.

3. Conclusion

Utilizing sodium cooled fast reactors to dispose of long lived transuranics while producing energy is a new way of utilizing the breeder reactor program of the 60's and 70's. While this program possesses many strong points, it is not without major obstacles that would need to be overcome if it were ever to be implemented on a commercial scale in the United States. If the fuel cycle includes the pyro-process, sodium cooled fast reactors have the potential to recycle spent fuel, reduce the load on Yucca Mountain, and maintain a degree of proliferation

resistance equal to what is currently employed. For the pyroprocess to be successfully implemented, it will need to demonstrate cost effectiveness on a commercial scale as well as maintaining strict safety protocols. Additionally, safety considerations are likely to be a major issue with the development of such a program due to the highly radioactive nature of the fuel, and the reactivity of sodium.

Sodium leaks will need to be contained far better than what was demonstrated at the Phenix and SuperPhenix reactors in France which were shut down for extended periods of time due to sodium leakage in the coolant loop. The Monju fire in Japan is likely to generate concern, as well as the Fukushima disaster. However, the sodium leaks never resulted in radiation leakage or deaths and the Fukushima reactor was an old design that could not employ natural convection to release decay heat, which all modern reactors can do. With research and development, sodium cooled reactors could minimize the risk of sodium leaks or other designs employing more benign coolant could become more developed.

Due to recent events, all nuclear plants, current and proposed will be held to a higher standard. Development and implementation of active and passive safety features leads to increased costs. In addition to any additional costs from increased safety, fast reactors are currently not viable economically unless uranium prices quadruple. This situation could arise from a large worldwide expansion of nuclear power but in the current climate that seems unlikely. While the United States still holds a favorable view of nuclear power, other parts of the world are retreating on nuclear development. If current trends continue, then uranium supplies could potentially last past mid-century. This gives ample time to develop fast reactor designs aimed at reducing the nuclear waste stockpile. Fast reactor programs despite their benefits are

simply unviable economically, in the near future. However, their potential benefit for the future should not be overlooked.

Ultimately, the success of fast reactors hinges upon their ability to ensure safe, reliable operation at a competitive price. It has been demonstrated that fast reactors are safe and could reduce our stockpile of nuclear waste while supplying energy for many hundreds of years. However, more research must be done to increase their reliability and make the process cheaper to implement. If these obstacles are overcome, fast reactors have the potential to solve the energy crisis as we know it today

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