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Environmental Aspects of the Nuclear Fuel Cycle: Reactor and Reprocessing Technology

A Master's Thesis submitted for the degree of
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supervised by
Kaluba Chitumbo

Spencer Warren

0928193

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Affidavit

I, **SPENCER WARREN**, hereby declare

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2. that I have not prior to this date submitted this Master’s Thesis as an examination paper in any form in Austria or abroad.

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ABBREVIATIONS

ADS	Accelerator-Driven System
AFR	Away From Reactor
AGR	Advanced Gas Cooled Reactor
ALARA	As Low As Reasonably Achievable
AR	At Reactor
AVLIS	Atomic Vapor Laser Isotope Separation
CANDU	Canada Deuterium Uranium (Reactor)
EPR	European Pressurized (Water) Reactor
ESBWR	Economic Simplified Boiling Water Reactor
FHR	Fluoride-Cooled High Temperature Reactor
FP	Fission Products
HLW	High Level Waste
IAEA	International Atomic Energy Agency
ILW	Intermediate Level Waste
ISL	In Situ Leaching
LLW	Low Level Waste
LWR	Light Water Reactor
MA	Minor Actinides
MAGNOX	Magnesium Alloy No Oxidation (Fuel Cladding)
MHTGR	Modular High Temperature Gas Cooled Reactor
MOX	Mixed Oxide (Fuel)
MSR	Molten Salt Reactor
OTC	Once-Through Cooling
PM	Particulate Matter
PUREX	Plutonium-Uranium Extraction (Reprocessing)
RBMK	Reaktor Bolschoi Moschtschnosti Kanalny
SCWR	Super-Critical Water-Cooled Reactor
SIR	Safe Integral Reactor
SNF	Spent Nuclear Fuel
SFR	Sodium-Cooled Fast Reactor
TBP	Tri-n-Butyl Phosphate
TRISO	Tri-structural Isotropic (Fuel)
VHTR	Very High Temperature Reactor

ABSTRACT

Commercial nuclear generation is often associated with radioactive gaseous releases, although the impact of the nuclear fuel cycle extends well beyond this into areas such as tailings in the mining phase to high-level waste management in the reprocessing phase. These problems may be alleviated or compounded with changes in the reactor and reprocessing technology. The SFR and MSFR are projected to alleviate these products by altering the structure of the fuel cycle to require less natural uranium input and generate fewer amounts of waste to be ultimately stored in the form of non-recyclable actinides and fission products. The VHTR and SCWR on the other hand accomplish provide a form of environmental protection via passive safety features, and the VHTR may also be designed in the future to promote GHG-free energy generation by coupling to a hydrogen production facility. Reprocessing technology is coordinated in tandem with reactors and involves aqueous forms in the current closed fuel cycle and the possibility pyroprocessing in an advanced closed cycle, reducing liquid waste and providing proliferation-resistant group recovery in association with fast reactors.

1. INTRODUCTION

The idea of nuclear power can incite a great diversity of emotional responses among people. If considering your average man or woman, this may be a combination of such things as fear, awe, respect and above all uncertainty. In actuality such a mixture is healthy, especially in comparison to those channeled by the zealous fusion advocate or prophet of the nuclear apocalypse. However, considering news topics as the Fukushima reactor complex problems and the 25th anniversary of the Chernobyl disaster (April 26) there currently appears more potential for negative emotions in the spectrum to be unfairly preyed upon.

The Chernobyl accident which occurred in 1986 is widely considered as the gravest single event in chronicled nuclear and radiation accidents, having received a Level 7 designation on the IAEA INES scale. In this case, a miscalculated shut-down procedure brought together unmanageably hot fuel with coolant, leading to a violent steam reaction, and a subsequent (suspected) hydrogen explosion due to the mixing of steam and zirconium cladding. The containment building of the Chernobyl 4 RBMK was destroyed and facilitated the release of some 14×10^{18} Becquerels of radioactivity into the environment. (World Nuclear, 2011)

In Japan, an 8.9 magnitude earthquake activated the automatic shutdown function in Fukushima's BWRs on March 11. The tsunami which followed disabled the electrical supply needed to power the active cooling mechanisms, since decay heat is generated in the reactor vessel even after shutdown (equal to approximately 7% of operational power). (MIT NSE, 2011) Although emergency battery generators were able to provide cooling for a limited time, an improvised saltwater-boron combination was used thereafter, and efforts continue on this front. The Fukushima incident has ranged from 5 to 7 based on INES estimates, but assuming the secondary containment layer remains intact, the gravity should not supersede that of Chernobyl, which possessed no such precaution.

The recent event in Japan has served to highlight the predicament of the nuclear industry. Due to high up-front costs and public resistance in many countries, new construction of plants cannot proceed, and consequently many reactors are quite dated. Half of the reactors in the U.S., for example, are more than 30 years old (CNN, 2011), and the Fukushima reactors were emplaced nearly 40 years ago. This point is mentioned not to call skepticism to the extension plants lives to 60 or more years, as these have been approved, but to note that plant age conceals the fact that many newer technologies in commercial nuclear exist which have not been implemented due to cost barriers, lack of political capital, and a slew of other factors. Concepts such as underground reactor siting, enhanced cooling systems using different mediums and design approaches, and improved passive safety features (as seen with the SIR's large pressure vessel containing a minimal number of penetrations, for example) (Wilson, 1996) may have been designed and completed operational testing, but have not been implemented widely.

The lack of new designs in operation reflects the priority of safety on the nuclear power agenda, and noting some benefit of public skepticism and uncertainty, this element is important in ensuring protection of the environment as far as possible during all aspects of the nuclear fuel cycle. With improvements in safety, however, designs will typically become more uneconomical. The same can also be said of waste minimization and proliferation resistance as new technologies are developed, especially on the back end of the fuel cycle.

The aim of this paper is to detail these emerging technologies in selected areas of the fuel cycle, and assess any benefits they may bring to the environment, whether actively or preventatively. Despite its interconnectedness to all goals of the nuclear industry, economic aspects will only be considered secondarily, along with the subject of non-proliferation. Waste generation and management are more inherent in environmental protection.

In the following Section 2, the stages of the fuel cycle will be summarized, with the major environmental concerns in each stage addressed. In its entirety, the cycle progresses from the mining and milling of uranium ore, and then through purification, enrichments, and fuel manufacture. Thereafter, the fuel is burned in reactor, resulting in products which may be reprocessed, recycled, and/or disposed of.

Section 3 concerns what I consider to be the two most important as well as controversial aspects of the fuel cycle: reactor design and reprocessing. Ultimately it is the reactor plant which is responsible for safely housing sensitive fissile material, converting it into electricity, and isolating its hazardous byproducts from the greater environment. There is also an enormous potential for improvement in this department, as Jan Forsythe notes. Advanced 'Generation IV' reactors in general would consume 50% less nuclear fuel while producing 50% less waste and 100% less in thermal discharges to the environment. Meanwhile, reprocessing technology is central in the debate between the 'open' and 'closed' fuel. Whether the fission products from nuclear fuel are recovered and recycled is a matter of choice, but one which bears heavily on issues such as energy independence and nuclear security. Reprocessing is also a hazardous process which results in radioactive aerial and liquid discharges during normal operation. While mining, milling, and decommissioning (not covered) are the other principal processes creating waste, more precaution must be exercised during reprocessing and other back-end stages, since more radioactive emission from irradiated fuel will be more than million-fold in comparison to the original fuel assembly (10^5 TBq for every metric ton of fuel). (Ewing, 2008)

Reactor design and reprocessing were furthermore chosen for examination over other aspects of the fuel cycle for additional reasons: mining techniques are already well established and saltwater extraction has not shown great potential. Enrichment is contingent on reactor type and the well-established gas centrifuge technology appears to be the modus operandi for the foreseeable future given the energy intensiveness of gaseous diffusion and difficulties encountered with laser separation. Lastly, final disposal of HLW is one of the critical unsolved problems plaguing the nuclear industry, and as an operational program has not yet been implemented anywhere in the world, there is little experience from which to draw.

Section 4 will assess the information gathered on reactor and reprocessing technologies, and if possible envision any fuel cycle paths revolving around these two factors which might accrue to low environmental impact. Again, secondary aspects such cost and proliferation resistance may also be considered.

Comments in Section 5, the last segment, will recount the most significant lessons learned as well and take account of how the nuclear programs in various countries stand at the moment. Which fuel cycle options are preferred and for what possible reasons; and what are the prospects for the future of nuclear energy? Not only curiosity in the field of nuclear energy but also interest in other forms electrical generation is a primary motivating factor in my research, and the properties nuclear power in contrast to other sources will be useful to keep in mind during this section when it comes to such themes as centralized and decentralized power, base-load power versus intermittent (peaking power), and the coupling of power to other applications such as hydrogen production.

2. NUCLEAR FUEL CYCLE

This section introduces, in abbreviated fashion, the modern stages of the nuclear fuel cycle and their associated technologies. The core processes to be later examined are reactor and fuel combinations during the service period (middle of the fuel cycle) and reprocessing, which is exclusive to the closed cycle. This section will place these processes in the context of the 'entire' fuel cycle, including any intermediate steps, and familiarize the reader with the nature of fission generation and some of the environmental considerations related to this power.

Nuclear Fission, Radioactivity & the Environment

The basic premise of modern nuclear power is to spark nuclear fission in a Uranium atom using a thermal neutron ‘bullet.’ This first splitting propagates further fission reactions, with each reaction releasing approximately 200 electron volts (MeV) of energy, if we include the instantaneously released energy and the releases over time from the decay products. (National Physical Laboratory) This compares to only 7-8 MeV of input energy per neutron. (University of Wisconsin, 2007) As to how such an efficient ratio can be achieved in comparison to chemical reactions, the equation $E = mc^2$ provides some insight; extractable energy is derivable from the mass of atoms in question, multiplied by the square of the speed of light. If one takes ‘x’ grams of a substance, not all of the energy in this form of mass can be released. In order for this to happen according to the equation, the entire mass of inputs would have to be destroyed.

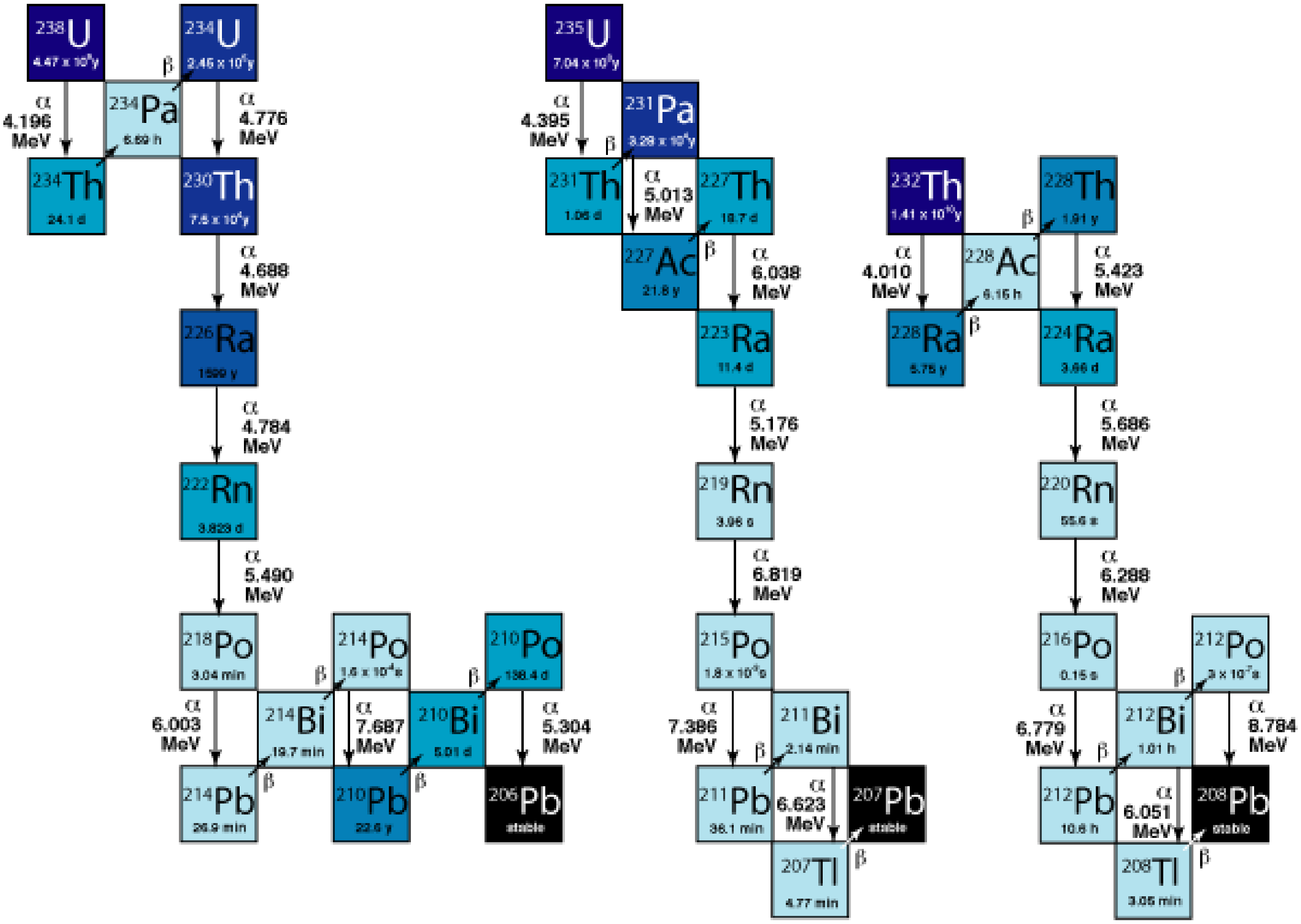
Annihilation as such is not possible. However, it is possible to acquire a fraction of this potential energy from fission, or ‘induced radioactive decay’. When a uranium atom is splits in typical fashion, a minute amount of matter is consumed in the process (transformed into heat), and it is in this small detail that the potency of nuclear fission lies.

U-235 + n > U-236* > Xe-140 + Sr-94 + 2n

The above equation shows the fission of Uranium 235 after absorbing a neutron. The susceptibility of uranium to this transformation into fission products and two neutrons lies in its high neutron to proton ratio. With 143 neutrons versus 92 protons, this value is nearly 2:1.
The same instability of Uranium is also responsible for its natural decay into smaller elements.

In order to relieve ‘tension’ generated between protons and neutrons, particle releases will occur which reduce the isotope in question to a lower state of energy. Alpha decay releases a Helium nucleus from the original core mass, while the most common form of beta decay (β–) converts a neutron into a proton and electron. The following table illustrates the decay of nuclear fuel sources over time; diagonal steps with no mass change represent beta decay, while vertical steps represent alpha decay.

Figure 1.1: Uranium Decay Series



Source: Macquarie University, Australia (2011)

In alpha and beta decay, a Helium nucleus and electron are released as radiation, respectively. Another form of radiation, gamma radiation (γ), also results in photon emissions, although these bear no consequence on the atomic masses of atoms and are merely symptomatic of the high energy levels of their nuclei. Altogether these discharges can be classified as ionizing radiation because they are capable of forcing electrons out of other materials that they run into, and in doing so creating (positively) charged ions in the environment. When the target environment is biological matter, these ions may participate in harmful reactions.

Fortunately, the different types of radiation are either limited in their effect or can be countered. Alpha particles have a small range of effect which can be blocked by dead skin tissue, beta radiation can be contained by a centimeter-thick layer of water, and gamma radiation requires more problematically a lead, steel, or concrete surrounding. (Wilson, 1996) Still, not all man-made radioactivities can be realistically shielded, and releases routinely occur into our atmo-, litho-, hydro-, and biospheres.

The effect of radioactivity in particular environments depends on the intensity and persistence of the radionuclides involved, and is often quite difficult to ascertain. For example, gaseous or suspended solid releases into the atmosphere from nuclear power facilities disperse within the low boundary layer (typically remaining within several tens of kilometers from the source) and become immeasurable after some time. Once on land or in a body of water, however, these unstable isotopes can become fixed to solid surfaces by chemical processes such as ion exchange or complex formation. This fixing has important implications in determining how radioactivity from fission and decay products will spread throughout an area, and is also known as absorption.

Radionuclides with stronger absorbing properties will attach to materials and move more slowly as a dissolved material in percolating water versus the actual water. (Wilson, 1996) Thus mobility of radionuclides, the opposite of absorption, is a keen environmental concern when it comes to issues such as groundwater contamination, although such mobility also means dilution from initial concentrations. Absorption and mobility in plants and animals is a different issue altogether; absorption entails incorporation into tissue and gradual elimination following ingestion, while mobile radionuclides will more likely be excreted and avoid accumulation. Elements such as Iodine and Cesium have a greater tendency to be absorbed in comparison Plutonium and Americium. The relevance of this mobility and absorptions is well exemplified is the issue of sheep in Wales having been contaminated with Cesium and remaining inedible for some time as a consequence of the Chernobyl fallout.

Radioactivity released by decaying nuclei, both in the form of photons or particles, can be measured indiscriminately in the SI unit of becquerels. One becqueral (Bq) translates to one particle release per second in a given source. If one wishes to determine how much radiation energy is absorbed in a kilogram of substance, or colloquially how “exposed” a material is to radiation, then the gray (Gy) SI unit can be applied. Depending on the mobility or absorbance of radionuclides in the environment, its exposure levels can change more quickly or slowly over time. While the “absorbed dose” of Gy does not take into account the effect of radiation on humans, this can be achieved by multiplying this value by a radiation weighting factor (WR) depending on the specific rays or particles involved. For example, alpha particles have a WR of 20 against the 1 WR of gamma and X-rays due to the ionizing havoc they can wreak on biological tissue. The number than results from the ‘Gy x WR’ calculation is measured in sievert (Sv) SI units. Additionally radiation doses can be targeted to specific parts of the body which may be more or less susceptible to uptake of radionuclides; this is also assessed in sieverts, with various organs having additional weighting factors. For the average person in the world, this “effective dose” accrued from natural sources of radiation amounts to several millisieverts per year. In comparison, the contribution of nuclear power generation to this effective dose equivalent measures in at approximately 0.1 microsieverts a year.* (Paschoa, 2004)

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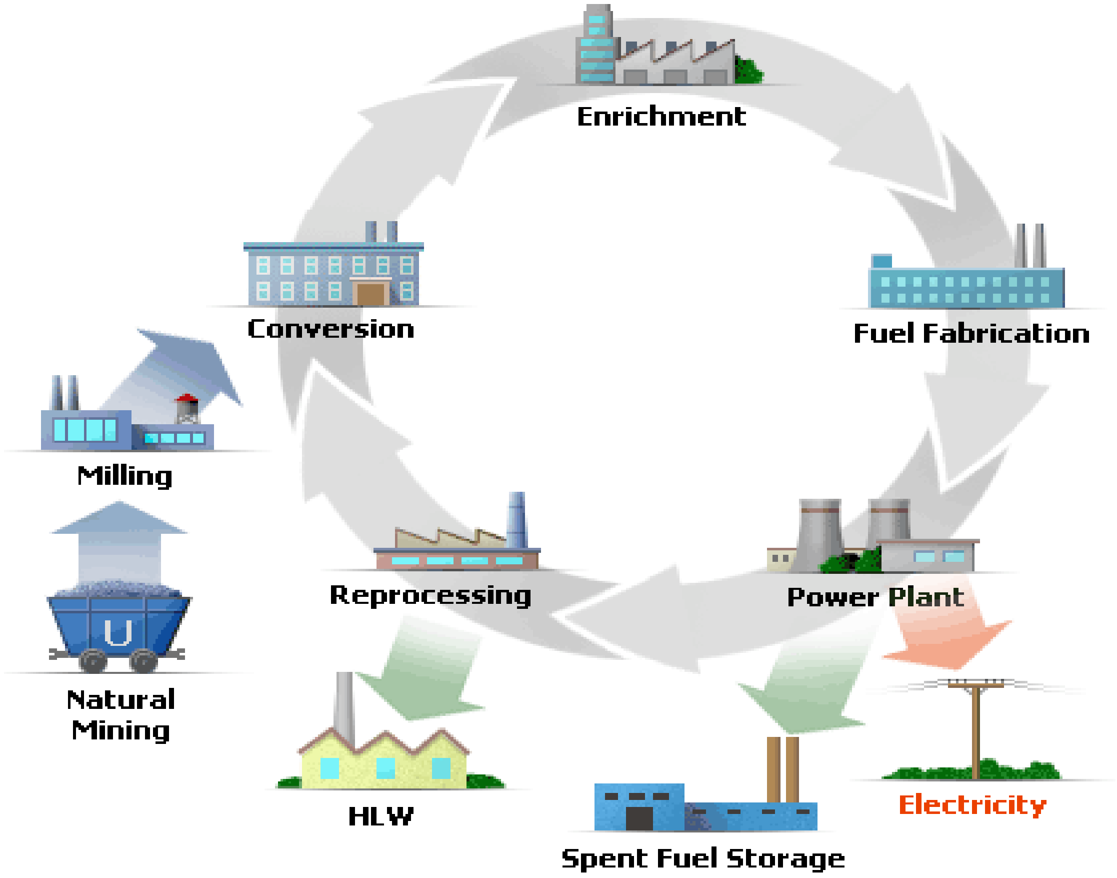
* The British National Radiological Protection Board recommends a 0.3 mSv annual human dose limit based on emissions from any one site (current or future).

Mining

A high amount of the proportion of natural radioactivity people receive comes from Radon, a decay product of Uranium (see Figure 1.1) Thus, where higher concentrations of Radon are found, there are also likely to be higher concentrations of Uranium present nearby. In some cases the Uranium content may be 1-4%, as with the high grade ore mined in Central Africa and Canada. (Haeberlin, 2004) Commercially obtainable from around a dozen different minerals, the uranium may be found in either oxide, mixed oxide, silicate, phosphate, vanadate, or carboniferous form. The initial extraction of these forms from their earthen repositories constitutes the first phase of the nuclear fuel cycle.

Figure 1.2 Nuclear Fuel Cycle



Source: University of Iowa, US (2010)

There are three general practices for mining uranium ore: conventional, in-situ leaching (ISL), and byproduct mining. Around 57% of uranium is produced through conventional techniques, either through open pits or underground operations. (Uranium Investing News, 2011)

Open-pit mining is used when ores bodies are found economically close to the surface, although of all processes involved this has the highest impact on the environment. Surface disruption is maximized, and due to the radioactivity in the ore, extensive watering is required to suppress the creation of airborne dust. After mining operations, the pit must also flooded or back-filled with waste, and it must be ensured that any tailings (unprocessed material around the ore) do not contaminate the surrounding environment as they may also contain radioactive-emitting substances. Underground mining offers less destruction to the surface, and is generally pursued when the ore is located more than 200 meters underground. (Wilson, 1996) Although destruction on the surface is limited to shaft penetrations, this method presents more hazards to workers since dust, rock falls, noise, vibrations, and radioactivity are more difficult to manage in a cramped work space.

In-situ leaching has come forward as a more environmentally considerate way of removing uranium from ore, as it foregoes a number of stages in the mining and milling processes such as physical extraction and size reduction (communition) by dissolving uranium in liquid and then pumping it to the surface. Additionally, no waste rock or tailings are generated, no dewatering of aquifers occurs since the installations do not break water tables, and lower volumes of hydrometallurgical contaminants are produced than in conventional mining. Both an acidic or alkaline compound may be used as leaching material, and acidic leaching solutions have been identified as having the highest potential for environmental contamination in the ISL process. The low pH (from sulphuric acid) in acid leaching results in the dissolution of heavy metals (as well as radionuclides in the ground) which can endanger the health of nearby soils and any proximal groundwater supplies. (IAEA, 2001) Today ISL account for 36% of the uranium recovered from the ground.

The remaining 7% production is classified as byproduct mining, where uranium is extracted secondarily from a material such as copper or phosphate. (Uranium Investing News, 2011)

Milling

Assuming the uranium does not undergo ISL, the solid is first crushed and ground in the milling phase. If there is also organic carbon mixed in with the uranium ore, this can be destroyed in a supplemental “roasting” phase which also provides greater efficiency in the subsequent leaching process. Ultimately, the uranium is precipitated from the leach solution into the uranium diuranate, also known as ‘yellowcake’, which is constituted of around 80% uranium oxide. (generally U_3O_8) (Hausen, 1998).

Pollutant sources from this phase include gaseous effluents, mill process residues, organic wastes such as solvents, humic acids, flotation agents, and barium-radium precipitates. Those sources which are collected into the tailings slurry collectively make up the most critical waste of the milling process, in volume nearly equivalent to the feed ore. The tailings will also contain heavy metals to some degree but more importantly around 70% of the radioactivity found in the mined uranium ore. (Long term radon emissions are provided chiefly by Radium-226 and Thorium-230) (IAEA, 1996) While tailings require ongoing monitoring and management, coupling mining and milling facilities together can allow better managements of such waste problems in while being economically advantageous. (Wilson, 1996)

Conversion

The yellowcake fashioned in milling still contains small quantities of undesirable elements such as cadmium or vanadium, which may be detrimental as a neutron poison or as the predecessor of a volatile fluoride compound unsuitable to the company of the uranium hexafluoride produced at the end of this conversion stage.

To remove these impurities the yellowcake is typically dissolved in nitric acid, yielding uranyl nitrate ($UO_2(NO_3)_2$), an oxidizing compound with very high toxicity. From there uranyl nitrate in solution undergoes evaporation to form UO_3, which is reduced with H_2 into UO_2. The uranium dioxide is reacted with hydrofluoric acid to produce UF_4, and this is finally combined with elemental Fluorine (F_2) under high temperatures to acquire uranium hexafluoride (UF_6). (Wilson, 1996) At this point in the fuel cycle, the fate UF_6 will be dependent on whether or not enrichment of uranium is required, and if not, into which sort of fuel it will be fabricated. In any case, it is very adaptable to whichever path selected, as its chemical and physical properties allow it to be stored as a solid below 57 degrees Celsius (assuming atmospheric pressure), manipulated as a gas above 57 degrees, or handled as a liquid under higher temperature and pressure. However, although UF_6 is nonreactive with oxygen, nitrogen, and other dry air ingredients, interaction with water or water vapor will create HF gas and uranyl fluoride, two corrosive compounds which can have severe health consequences if ingested or inhaled in sufficient amounts. (Argonne National Laboratory)

Enrichment

Since the Uranium-235 which is split to provide energy in reactors only makes up .711% of the element as it is found in nature (U-238 constituting 99.28%) and remains at this level in the UF_6 (Wilson, 1996), its concentrations must be increased to provide sufficient power in most reactors Magnox or CANDU. U-235 levels are elevated to 3-5% during the enrichment process, with the two major technologies of gaseous diffusion and high speed gas centrifuge each exploiting the mass differences between U-235 and U-238. In the UF_6 gas which is put through these two processes, only one isotope of Fluorine exists and thus cannot obscure this mass difference.

Gaseous diffusion was first used by the United States during World War II, and would become the first method of enrichment deployed on a large industrial scale. The UF_6 vapor is channeled through separation stages consisting of a diffusion chamber with a permeable membrane, a gas cooler, and a compressor assembly. The cooler is used to ensure low pressure conditions which facilitate more efficient diffusion through the membranes, although the separation factor remains very low at 1.0043. (Wilson, 1996) After diffusion in one separation stage, the slightly enhanced U-235 feed is forward to an additional stage while the depleted stream is cycled backwards, with the feeds being manipulated at each stage by a compressor, drive motor, and control valves. Collectively this system is known as a cascade, and a cascade may contain upwards of one thousand separation stages.

Gas centrifuge technology has replaced gaseous diffusion as the favored enrichment process due in part to higher separation efficiencies (based on absolute mass differences in uranium versus the square root of the relative difference with diffusion processes) which translates to a simpler cascade design. The uranium hexafluoride gas is fed into hollow cylindrical rotors which force the heavier U-238 to the walls of the casing through centrifugal force. The separation can also be augmented with countercurrent circulation provided by mechanical scoops, thermal convection, or a combination of the two. After the enriched product is siphoned from the axial flow and the depleted product from the radial flow, each can be solidified and stored or moved through the cascade to another rotor unit. Gas centrifuge cascades require only ten to twenty separation stages. (Chitumbo, 2011)

Although any environmental impacts of enrichment are dwarfed by other aspects of the fuel cycle*, specific interest for the purpose of this paper is directed to energy and resource consumption of enrichment plants. Conveniently, both of these variables can be accounted for in Separative Work Units (SWU), combining the kilogram amount of uranium feed to be processed and the energy required to enrich that amount to certain level. (Global Security, 2005) Depending on the intensity of the enrichment process, the tails or “waste” assay of depleted uranium will also vary, and given two trials with equal inputs and enrichment targets, the stream with the higher U-235 tails concentration will feature a lower SWU value. Likewise, if a certain amount of 3.5% enriched uranium is desired from two feeds of different quantity, comparatively lower amounts of energy would be necessary to process the greater quantity of un-enriched uranium at a higher tails assay. Thus there is some tradeoff between uranium stockpile depletion and energy consumption.

To give some idea of the advantage of centrifuge technology over diffusion, specific energy consumption in plants has been estimated at 100-300 kWh/SWU versus 2300-3000 kWh/SWU. (Global Security, 2005) While much of the details are classified, another enrichment technique utilizing lasers (AVLIS) has been touted with the potential to compete with centrifuge technology in the near- to mid term future (USEC, 1993), although a high degree of technical expertise is required and, as with other processes such as aerodynamic separation or chemical and ion exchange, the technology is difficult to apply on a commercial scale.

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* According to IAEA assessments, uranium content in effluents is very low at enrichment plants. Airborne PM consisting of fluorides and nitrogen- and sulfur oxides are also produced in minute amounts from various plant compartments. (TECDOC-1306)

Fuel Fabrication

To prepare uranium and possibly plutonium for burn-up in a common LWR, uranium fluoride must be converted into oxide form, although if uranium is to be used in a Magnox gas-cooled reactor, a metallic product must be produced. This path combines UF_4 (reduced from UF_6) with magnesium metal at exceptionally high temperatures, with the reaction beginning at 650 degrees Celsius and reaching a maximum of 1500 degrees. To handle such heat, the event takes place in graphite containment structure built within a stainless steel pressure, and surrounded by argon gas. Afterwards the metal is formed into large fuel rods (IAEA, 2002)

Metal Reaction: $\mathbf{UF_4 + 2Mg > U + 2MgF_2}$

The more common oxide path utilizes one of three chemical processes, one the “so-called Integrated Dry Route” and the other two aqueous. For the sake of simplicity only the “dry” process is detailed:

Oxide Reaction: $\mathbf{UF_4 + 2H_2O > UO_2 + 4HF}$

As with metallic fuel, UF_6 is first reduced to UF_4 with hydrogen combustion. Afterwards the UF_4 undergoes steam hydrolysis. These two steps happen in a single integrated kiln – hence the phrasing “Integrated Dry Route.” (IAEA, 2002) Whichever of the three processes is used, however, the end result is always a uranium dioxide powder, which is then formed into pellets, shaped, and loaded into zirconium-tin alloy cladding. The cladding is sealed at either end to complete the fuel rod, and in comparison to metal fuel rods these are thinner due to their lower conductivity.

Mixed oxide fuel can also be fabricated using recycled uranium and plutonium. This will either proceed along a dry process involving mechanic mixing of powders, or a wet process with plutonium and uranium nitrates combined in solution. Special shielding must be given to plutonium throughout the fabrication steps from pelletizing to assembly since it is a potent alpha particle, neutron, and gamma emitter. If the plutonium is particularly dated, increased gamma ray output may have to be accounted for due to plutonium-americium decay.

Overall the environmental impact from fuel fabrication can be expressed in the 1% emission fraction over the entire fuel cycle, counting both radioactive and non-radioactive emissions. (UNSCEAR, 1993) Of this, conversions of UF6 to UO2 along with scrap recovery are the most important activities releasing radioactive matter. (Shum, 1987) Accidents such a UF_6 cylinder rupture, hydrogen reduction furnace explosion, or a criticality event (more likely when handling metal fuel) could have additional emissions consequences; however, scrubber and ventilation systems, careful regulation of hydrogen and nitrogen reduction gases, and predominant handling of dry oxide powder act as safeguards for each of these scenarios. In terms of waste management, MOX fabrication involves the necessity for intermediate and final storage due to the involvement of plutonium. (IAEA, 1996)

Power Plant

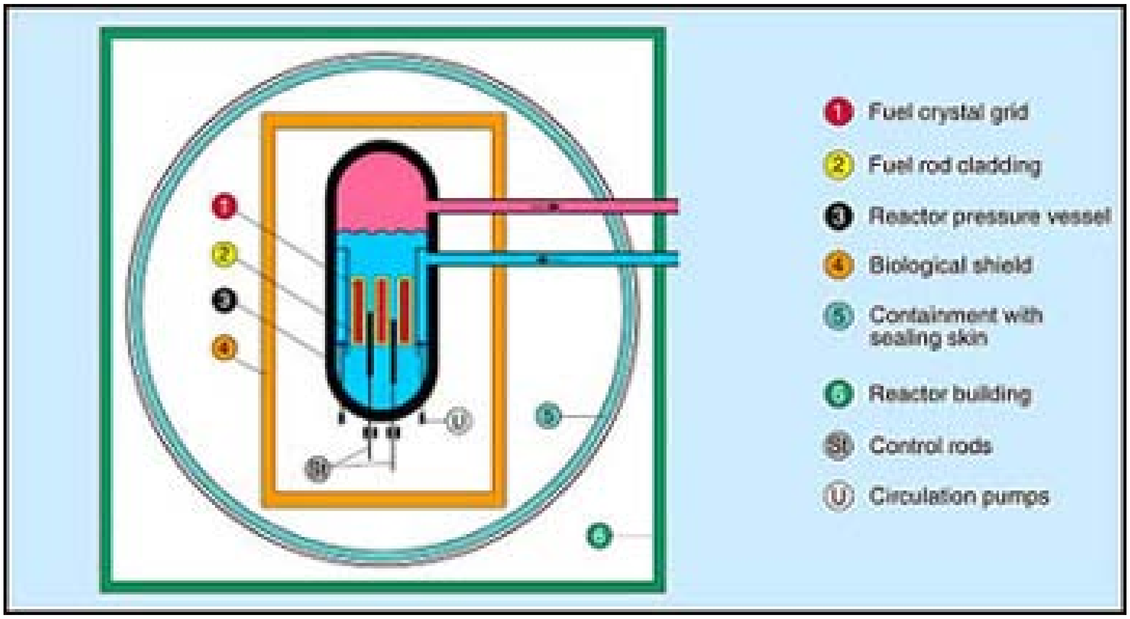
The middle of the nuclear fuel cycle, or service period, sees the fissile contents of the fuel partake in a regulated chain reaction, the energy from which is converted into steam, then used to drive turbines and create electricity. The chain reaction is initiated by a neutron source, which may come in the form of an external Cf-252 mass or from the uranium content in the fuel, since a number of these uranium atoms will already be undergoing spontaneous fission.* Starting the free flow of neutrons is often just a matter of removing neutron absorbers (poisons) from the reactor. This can be done chemically or mechanically via control rods. (Whitlock, 2011)

While the 'first' neutron catalyzes fission in a uranium atom, a number of neutrons are released in the aftermath. How many neutrons go on to participate in breaking up additional uranium nuclei is expressed with the term k_{eff}. If this value equals 1, then one neutron from the several released in each fission will go on to split another atom. This is known as criticality, and is the level maintained by a nuclear power plant during normal operation. With a k_{eff} less than one, neutrons will be absorbed by moderating material and the reaction cannot be sutained, and with a k_{eff} higher than one, the rate of fission will increase exponentially until fissile material becomes scarce. (Wilson, 1996) A high k_{eff} value is a feature of nuclear fission weapons, although this situation is inapplicable to commercial reactors because for one there is an insufficient concentration of U-235 involved. Nuclear weapons will typically feature uranium enriched to levels upwards of 85%, while the highest degree of enrichment in power reactors will approach 5%.

Without indulging too much in the variety of commercial power reactors operating around the world today, they are most commonly of the thermal class, and may range in electrical output from several hundred to around 1400 megawatts (MW). (Wilson, 1996) Notable models include the pressurized water reactors (PWRs) and boiling water reactors (BWRs) of the LWR class, Magnox and CANDU reactors employing un-enriched uranium as fuel, AGR reactors, and the RBMK design. PWRs dominate the commercial industry, although they are similar in principle to BWRs, using light water as both a neutron moderator and a coolant; the main difference is that the pressure in reactor core of a BWR is lower than that of a PWR so that water is directly boiled into steam, whereas the pressure in a PWR is dropped in a secondary heat loop outside of the core to produce steam. Each of these models use enriched uranium, unlike the CANDU and Magnox reactors developed separately in Canada and the UK. The CANDU is alternately defined as a pressurized heavy-water reactor because it uses deuterium water (^2H_2O) as a moderator; this absorbs fewer neutrons than would normal water so that more are made available to the lower concentration of U-235 in the fuel. Magnox reactors on the other hand achieve higher neutron capture in the fuel with via the magnesium alloy fuel cladding. The British AGRdesign is actually an updated version of the Magnox-based design, retaining both graphite moderation and carbon dioxide cooling while adding a nitrogen-injection system as an emergency shutdown neutron poison. Finally, the Russian RBMK class combines on some level an AGR and BWR; graphite serves as the moderator and water, as the coolant, is permitted to boil and be channeled from the core into the turbines. Unfortunately the initial Soviet-era setup made the coolant vulnerable to boiling and increasing the power level in the reactor, as very little neutron absorption was handled by the graphite. With absorption falling disproportionately on the coolant, positive feedback from coolant temperature to reactor power could have disastrous consequences, as demonstrated in the Chernobyl case. This particular flaw was corrected later, however, by increasing the concentration of U-235 to balance neutron absorption between the fuel and coolant. (Wilson, 1996).

The six aforementioned designs represent technical development up to the level designated as ‘Generation 2’, although construction of these models has for the most part been discontinued around the world. ‘Generation 3’ designs like the Enhanced CANDU, APWR and ABWR retain the basics of their Generation 2 ancestors while aiming to improve in areas such as fuel burn-up, thermal efficiency, and safety features, and Generation 4 technologies, to be seen in at least a decade if not longer, seek to address issues outside of electricity production such as waste management of SNF or hydrogen production. Several different concepts of the future will be covered in the following Section 3, including any documented means of providing safe operation. Measures for any new projects will likely fall under heavy scrutiny, especially given current media and public sensitivity to nuclear safety, and the basic containment provisions in Generation 2 LWR's cannot even be considered even a minimum if newer designs are to gain widespread approval. Nevertheless, Generation 2 containment mechanisms provide a basic model for understanding how fission products and other contaminants can be controlled.

Figure 1.3 Typical Reactor Safety Features



Source: European Nuclear Society (2011)

The crystal lattice of uranium dioxide fuel and Zircaloy cladding in LWRs act as the first two barriers against radioactivity leaks. The ceramic UO_2 features a melting point of around 2800 degrees Celsius, and any gaseous fission products (~1%) migrating from the fuel will generally be contained in a space at the top of the fuel rod (still within the cladding). (Wilson, 1996) The third barrier is the reactor pressure vessel, encapsulating former components and providing defense to pressure and temperature change, as well as radiation. From the vessel reaction heat is transferred out through the coolant and circulated back. The biological shield is found outside of the pressure vessel, and per name is responsible for protecting any personnel in the vicinity of the reactor. Radiation is absorbed here by lead, two to three meter thick reinforced concrete, or an equivalent material. The fifth layer is comprised of steel sealed off with air locks, and the sixth layer known as the secondary containment is made is reinforced of concrete, and responsible for neutralizing impacts occurring from the outside. (ex. explosives detonation or a plane crash. (Chitumbo, 2011) The potential of these barriers collectively to protect both workers and the public from radiation exposure (especially in the case of an aircraft impact) has seen the fire of controversy, although it was concluded in a 2002 study that even against weaker, dated containment buildings, multiple high speed artillery rounds or specially-designed bunker-penetrating ordnance would be required for a breach.**

In spite of these measures, however, environmental contaminants are still released outside of plants. Corrosion products are formed inside the reactor vessel and fission products can infiltrate the coolant as well if the fuel cladding is defective. (UNSCEAR 1977) (UNEP 1979) If cladding or "fuel failure" is assumed to be 0.25%, then worldwide gaseous releases of the fission products krypton and xenon are somewhere on the order of several terabecquerels every year, albeit distributed from a plume into trace amounts. Before they are released, however, they may also be stored for up to three days in delay tanks to reduce their radioactivity, given their short half-lives. Concerning liquid releases, tritium is the most important as it accounts for more than 99% of the radioactivity in such discharges. Once captured in floor drains and fed into retention tanks, both deuterium and the tritiated hydrogen are reduced to water to minimize the probability of a hydrogen explosion occurring. (Paschoa, 2004) Nevertheless, discharges of both kinds will be made based on the ALARA principle, considering dose reduction and the cost of such a reduction.***

Water demand and thermal pollution are also important concerns, since nuclear power plants use and consume more water than any other type of energy generator. This is in large part due to the lower temperatures and pressure at which steam operates in contrast to coal and natural gas plants, and translates to less efficient use of heat from the reactor. (POA, 2006) Impingement (and death) of aquatic species can occur during cooling water intake, and thermal shock incurred from discharges can redistribute the organism ecosystems. As temperature increases diminish the ability of a water body to carry oxygen while simultaneously accelerating the metabolism of species; some are unable to adapt, either in the short term from thermal energy intolerance or in the long term from decomposition of organic nutrients and oxygen insufficiency. (The longer term effects have been shown to lead to increased susceptibility to diseases and disruption to reproductive systems). (Pollution Issues, 2011)**** Problems of thermal pollution and water usage can be alleviated in several ways. For the first, reverse flow heat treatments in the intake pipes can follow a “fish-chase” procedure in which heat is gradually increased to drive off any fish species in the vicinity. Diffuser systems on the output end diluting the heat, or multiple, distanced outputs are other possible fixes. (Pollution Issues, 2011) But, minimizing the water required in the first place is perhaps a better solution. Pond and tower cooling systems recycling the water circulated through plants can reduce water withdrawal to approximately 2,000 – 4,000 liters per MWh in comparison to the 95,000 – 200,000 plus liters withdrawn by once-through cooling systems per MWh. (EPRI, 2002) In the future, of course, cooling systems for nuclear plants can be made even more effective, for example by using multiple cooling methods employing water and air, or by using different types of coolant such as molten salt which would furthermore allow the reactors to operate more efficiently at higher temperatures.

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* The uranium in fuel will also be undergoing decay, but spontaneous fission is a different concept in which neutrons from space split heavy nuclei. In the past, higher concentrations of fissile uranium are proven to have sustained a fission chain started by spontaneous or natural fission.

** From *Science* (2002): further studies indicated the public risk from such events as containment failure or meltdown to be limited to few, if any, direct deaths.

*** Based on International Commission on Radiological Protection principles, ALARA values must be justified by the benefit accrued by the public, marginal cost reduction, and an overarching dose limitation. From Health Physics Society (2009)

**** For survey of affected fish population in various nuclear and non-nuclear plants in California, see Steinbeck, J. (2008)

Reprocessing

When the fuel in a reactor has been irradiated near to the extent that fissile U-235 is no longer abundant enough to continue the chain reaction (and also considering the neutron poisoning properties of fission products in the fuel such as Xenon), the fuel assembly will be removed from the reactor. The spent fuel from a typical LWR will contain 0.8-0.9% Uranium-235, 95% Uranium-238, and 1% Plutonium, with the remaining 3% or so made up of fission products and transuranics. (IAEA, 1996) Whether the fuel is to be reprocessed or not, interim storage, generally on-site (AR), will be used in order to allow the heat and radioactivity of the SNF to decrease. Wet storage in water at least 2.5 meter depths, with concrete or stainless steel sealing is the preferred option, although dry storage can be used if the fuel cladding is prone to corrosion as with first-generation Magnox fuel. (Wilson, 1996) Following interim storage, the next course of action dictates whether the fuel cycle is ‘open’ or ‘closed’. The decision can also be postponed, in which case interim storage becomes indefinite storage, but in the case of the open cycle the waste contaminants will be conditioned for long-term storage and ultimately final disposal. With the selection of closed cycle reprocessing, the SNF is removed and treated in such a manner that some actinides and/or fission products are separated from the collective mass of high-level waste (HLW).

Reprocessing represents a closing or completion of the fuel cycle because the uranium and plutonium in SNF can be extracted, re-enriched if need be, and recycled as uranium or MOX fuel.* This capacity to produce additional energy in the future, the removal of volatile fission products, and the conversion of any unrecoverable or undesired radionuclides into stable waste form are the primary objectives of reprocessing.

Removing or penetrating the fuel cladding and dissolving the fuel in nitric acid are the first two steps in conventional aqueous (PUREX) reprocessing. The second of these in particular is of environmental concern as dissolution releases nitrogen oxides and Tritium, Iodine-129, Krypton-85, Carbon-14, and Ruthenium-106 fission products as off-gases. (Wilson, 1996) The off-gas stream enters a condenser and scrubber system where the recyclable nitric acid is collected and the bulk of NO_x liquified. Additional technologies such as caustic scrubbers (for Iodine-129 and Carbon-14), solid sorption devices, and high efficiency filters (for Ruthenium-106 and any PM) are applied to the remaining stream before it exits the stack. Krypton-85 presents perhaps the greatest challenge during off-gas treatment since it cannot be absorbed by aqueous scrubbing, although its releases are deemed to have a minor environmental impact and removal technology cost-ineffective for the potential benefits. (IAEA, 1996) (Wilson, 1996)

Nitric Acid Interaction with Uranium Dioxide:

$$\mathbf{3UO_2 + 8HNO_3 > 3UO_2(NO_3)_2 + 2NO + 4H_2O}$$

Treatment in nitric acid transforms Uranium into a hexavalent ion, which can be extracted by the molecule TBP in the next phase. TBP itself is a neutral ion, and requires nitrate anions to bind the metallic transuranics in solution. Some fuel elements, depending on their valence, however, are more extractable than others. Uranium and Plutonium (and also Neptunium) are highly extractable in the hexavalent and tetravalent forms, but species such as Np(V) and any trivalent forms from Plutonium and beyond are nearly impossible to isolate at the moment. Most fission products also present a similar dilemma within PUREX-style reprocessing, (Wilson, 1996) and capturing these elusive forms is one of the main focuses of current research and development.

TBP treatment results in co-extraction of Plutonium and Uranium, discriminating against other actinides initially. Thereafter the two major actinides are separated from another, a step which raises some concern about nuclear proliferation. Many flowsheets, though, will opt not to recover the Plutonium by reducing it to a trivalent form while maintaining Uranium with TBP solvent. (Wilson, 1996) In such an instance, the Plutonium could remain with the other actinides and fission products in aqueous form before being evaporated and stored in stainless steel tanks. Vitrification would occur thereafter; the metal nitrates transformed into oxides and combined with glass-forming materials at temperatures above 1000 degrees Celsius. After heating, the vitreous product is poured into stainless steel containers, (Smith, 1985) the ultimate destination of HLW and ‘end’ of the closed fuel cycle.

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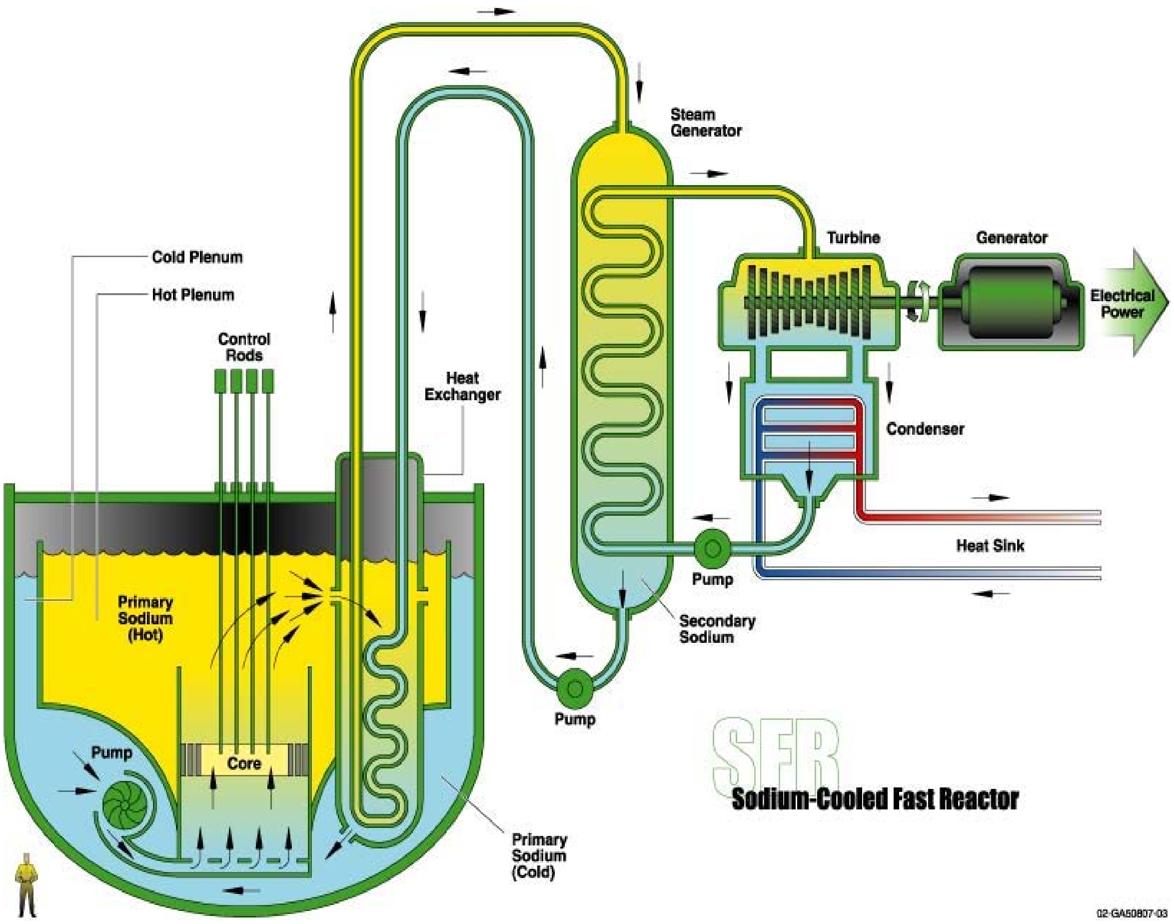
[*] Plutonium dioxide in MOX fuel amounts to 1-10% of content in thermal reactors and 25-35% in fast reactors. (Wilson, 1996)

3. SELECTED TECHNOLOGIES

A. Reactor Design

Sodium-Cooled Fast Reactor (GEN. 4)

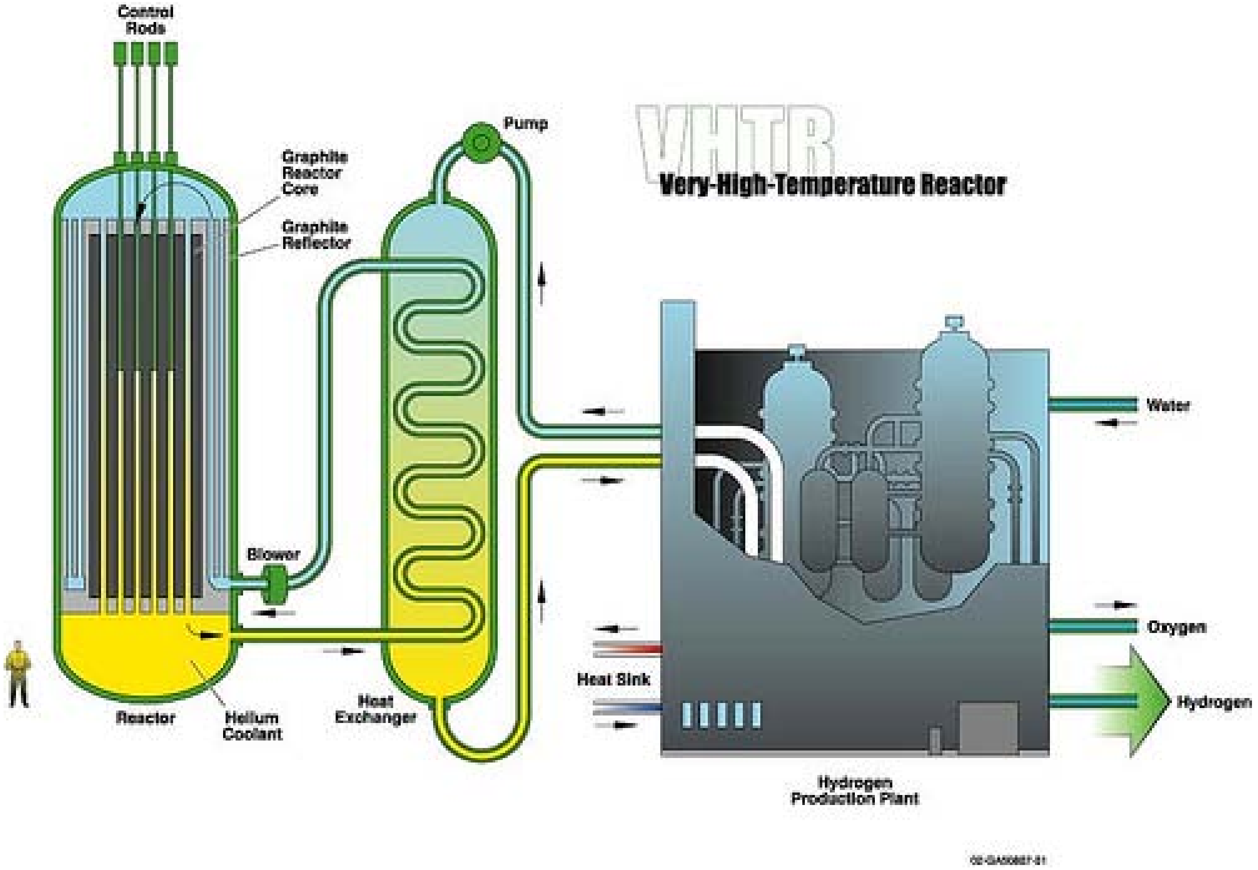


Source: Idaho National Laboratory (2011)

The SFR project aims to alleviate the waste nuisance of SNF by burning plutonium and other actinides, since fast neutrons are capable of splitting all isotopes of plutonium and uranium whereas thermal neutrons can only transmute those with odd numbers. (i.e. U-235, Pu-239) This also is means that the energy output from natural uranium fuel can be greatly improved over the 1% utilized in thermal systems. The graph below can be simplified into three systems: heat from the primary (radioactive) sodium bath is transferred to an intermediate, contained sodium loop, and finally to a power conversion system which will likely employ either water or carbon dioxide as the working medium. Build scenarios foresee either a medium-sized 150-500 MWe reactor using metallic fuel comprised of uranium, plutonium, and minor actinides, or a medium to large sized reactor burning MOX fuel. (Lineberry and Allen) Both models would use sodium as a coolant, but the first would require pyroprocessing prior to fuel preparation as this technique can only be applied to metal fuel; for the second, advanced aqueous processing would be used. Minor actinides are capable of undergoing transmutation in thermal reactors with this case, but they are considerably less efficient at absorbing neutrons.

While the SFR has advanced the farthest in terms of development of any Generation 4 reactors, one of its main problems is still cost competitiveness. Additionally, the inclusion of passive safety features must receive more attention if this design is to be realized in the next decade; for example, while the sodium coolant has a higher boiling point than the operating temperature and doesn't require pressurization, contract with water produces a violent reaction. If this were to occur in the secondary loop, release of radioactive content would not occur, but if contact occurred in the primary loop or coolant was somehow lost, the situation could become much more complicated and dangerous. (Lineberry and Allen) (Fanning, 2007)

Very High Temperature Reactor (GEN. 4)



Source: Idaho National Laboratory (2011)

As an offshoot of high temperature gas-cooled reactor design, the conceptual VHTR uses helium gas to cool the reactor and graphite as moderating material. As can be inferred from the name, the core is envisioned to operate at an unusually high temperature of around 1000 degrees Celsius, making it possible to produce hydrogen via a thermochemical process using sulfur and iodine. Additionally, cogeneration or direct gas propulsion through a turbine could provide electricity. (World Nuclear Association, 2010)

Safety features of the VHTR include passive heat radiation from the steel reactor casing away from the fuel, and an underground design which serves as containment and as a final sink for the reactor heat. (Ragheb, 2011) Other features are understood to be retained from similar reactor designs such as that of the MHTGR. This earlier design uses a separate shut-down cooling system with its own heat exchanger and cooling circulator as a back-up which, when combined with the primary heat transport system, can allow maintenance within 24 hours of shutdown. Separate from these two active systems, the MHTGR also includes a reactor cavity cooling system which functions passively and is installed in the concrete containment surrounding the reactor core. To elaborate on the meaning of a passive safety feature, it is one which works regardless of human interface and is therefore assured a higher degree of reliability. The RCCS is a prime example of this as it employs natural heat conduction, convection, and radiation and doesn't consist of any valves, controls, or circulating fans. (Silady and Millunzo, 1989)

Apart from the redundancy of the 3-system cooling, which is especially important given the nature of the reactor* and not-too-distant recollection of the failure of Japanese reactors to control decay heat, the MHTGR/VHTR design advertises further safety based on the single-phase (helium gas) coolant, low power density cores which can be designed in array (versus having one large core), and fuel properties. The coolant is significant because it is neutronically inert and is capable of resisting chemical attack by water should it be needed for emergency cooling. Likewise, if water should reactor with the graphite moderator, the reaction would be endothermic and thus act as another form of coolant. The ceramic fuel coating used, also known as TRISO, is one of the more interesting design aspects and also responsible for passive heat removal. Unlike conventional alloy cladding which would fail at temperatures lower than 1100 degrees Celsius, the silicon carbide coating the (usually) uranium dioxide pellets can withstand temperatures approaching 2000 degrees before decomposition ensues. (Silady and Millunzo, 1989)

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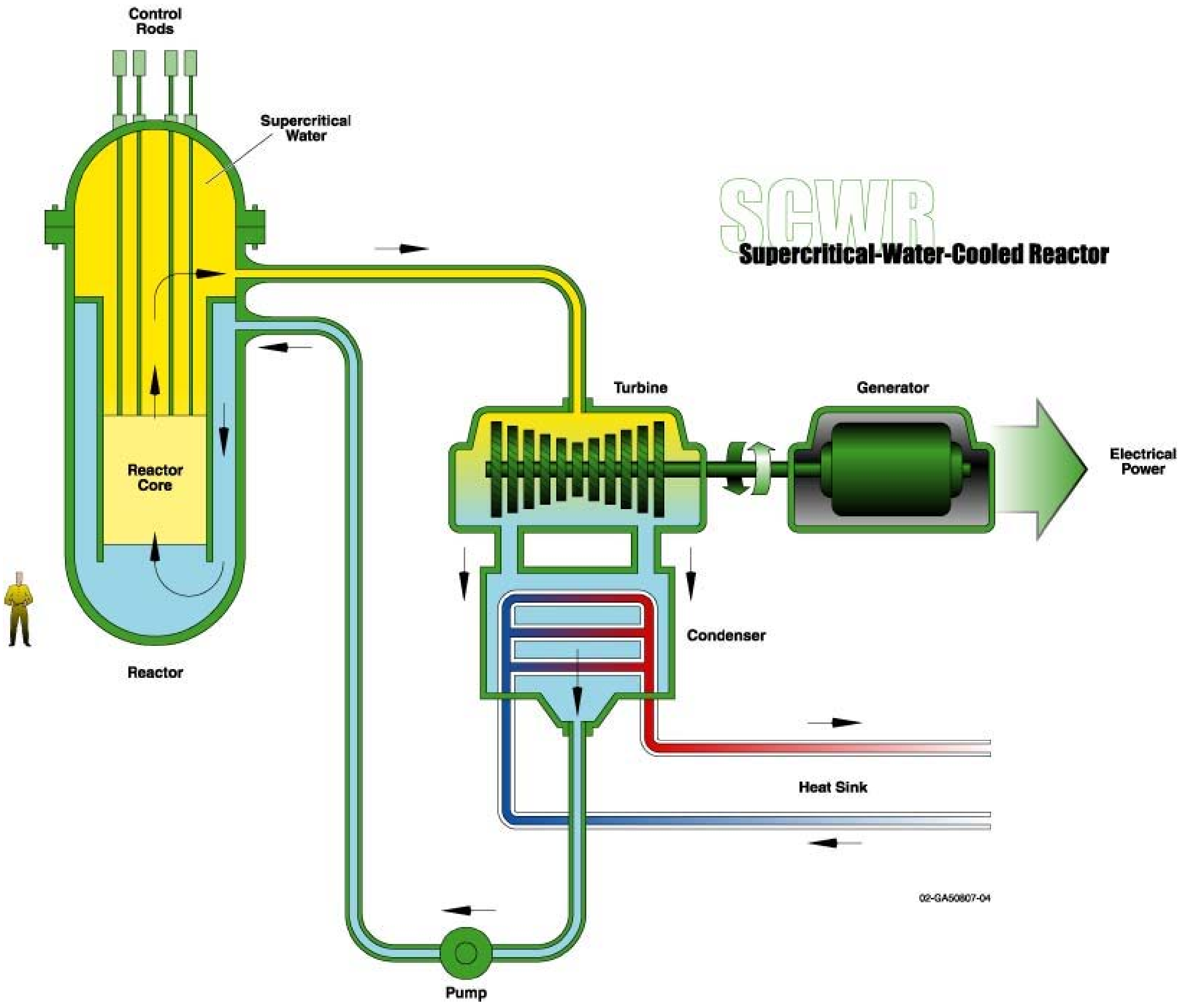
* MHTGT operating temperatures range from 500-600 degree Celsius versus the 1000 degree standard of VHTRs

Supercritical Water-Cooled Reactor (GEN. 4)



Source: Idaho National Laboratory (2011)

The SCWR takes its name from the high pressures and temperatures at which the reactor operates; specifically, at conditions above the thermodynamic critical point of water. (374 degrees Celsius, 22 MPa) Normal water coolant, which is forced through a single loop to drive the turbine as in a BWR, is able to achieve about 33% more thermal efficiency than the same coolant in most LWRs today. Unfortunately, this same aspect of water which makes it attractive for energy transfer also makes it difficult to design for. Water above the critical point is highly corrosive, and coolant loss and a positive void coefficient* further compound the problem. (GEN IV, 2010) (Ragheb, 2011) Uranium dioxide is designated as the fuel for a thermal plant, although the possibility of a fast-spectrum design with the recycled actinide fuel elements has not been excluded as an option.

Passive safety technology is assumed to be similar to transferrable from those found in current ESBWR reactors, a Generation III+ design evolved from the standard BWR. Isolation condensers located above the reactor core will gather heat from below and dump the heat into a water pool. The natural tendency of heat to rise in these pools will keep warmer water at the top while cooler water at the bottom of the pool is circulated to the reactor core. With several of these condenser-pool complexes working in such a gravity-driven cooling system, low water levels in the reactor, once detected, can be passively raised by releasing greater amounts from the pools. If the projected building costs (60-70% of typical LWRs) (ANSN, 2007) and enhanced safeguarding of radioactive emissions from the ESBWR are any indication for the SCWR, then this design could have promise as a reliable, cost-effective means of generating electricity in the future.

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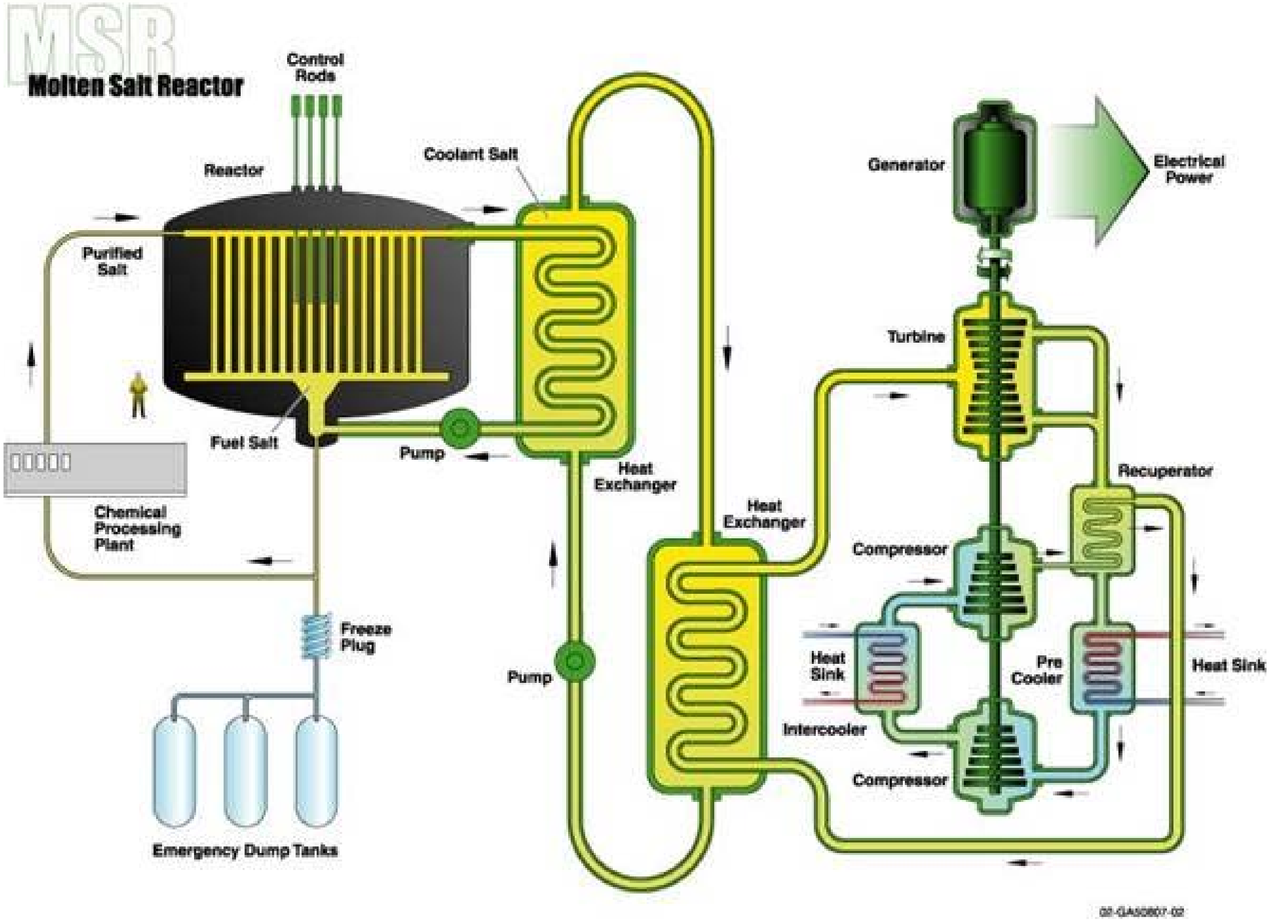
[*] Voids are formed as steam bubbles in the cooling water; with a positive void coefficient, neutron multiplication in the reactor will increase and the power level will increase. This positive feedback mechanism was detailed in Section 2

Molten Salt Reactor (GEN. 4)



Source: Idaho National Laboratory (2011)

Molten Salt reactor system are classed as one of two designs employing liquid salt as a primary coolant, the other being the Fluoride-Cooled High Temperature Reactor (FHR). Since the FHR incorporates elements from other designs such as the TRISO-style fuel from the MHTGR and a fast reactor plan identical to that accompanying the VHTR, this summary with focus on the more generic MSR. (GEN IV, 2010)

The thermal MSR system works by running uranium, sodium, and zirconium fluorides (salts) through a primary system going through the reactor. Heat is then transferred to a second, non-radioactive coolant loop also comprised of salts. The novel thing about this concept is that the fuel is also the coolant in the primary loop as a dissolved component. Compared to water, the salts have a similar heat capacity, but salts are able to sustain much higher temperatures in a reactor which translates to higher thermal efficiency and output. (Hoglund, 1997) They are also chemically inert and non-corrosive. (Dodds and Gat, 2008) While the fuel may not be critical passing outside of the reactor, once is passes through the graphite-laden core, neutron moderation there should produce criticality.

Safety features are abundant in an MSR. Criticality accidents are minimized because the negative temperature coefficient of the fuel does away with any need for control rods, and on-line fuel processing means that the reaction rate will not have to be increased, with the neutron-absorbing fission products being removed on a continual basis. Fuel processing is also related to a low source term value, source term being defined as the radiation which should be contained within a reactor, but which can escape in case of an accident. By removing these fission products with long lives, the source term is kept low, gaseous releases (see Section 2: Power Plant) are nearly eliminated, and only short-term radioactive risks will need to be managed in case of an accident. (Dodds and Gat, 2008) Lastly, accompanying the inherent passive safety of the fuel is the engineered feature of the Freeze Plug. This device works by freezing salt coolant in advance and storing it in locations such that, if the reactor heat somehow becomes unmanageable, the frozen salt plugs will melt, expanding and flowing into emergency dump tanks. (Ragheb, 2011)

Unique among the Generation 4 reactor concepts, the MSR has also been considered for fast reactor design, given that the negative temperature and void coefficients of the fuel would give it an advantage over solid-fuel fast reactors in terms of safety. Intrigue allowed, an MSFR would operate on a Thorium fuel cycle, generating fewer fission products during operation than the uranium fuel cycle while having the capacity to burn up fission products and actinides in shorter-term waste. (Merle-Lucotte et al., 2007) Since the Thorium fuel cycle has not already been mastered, however, this presents quite a challenge. Harnessing power from this mineral is a more difficult task because Thorium is not inherently fissile like U-235. It must first be transmuted into Th-233, and then allowed to undergo beta decay twice to reach the U-233 that can be fissioned.

B. Reprocessing Development

The type of extraction desired by reprocessing technologies can be classified into four groups: the major actinides including uranium and plutonium, the minor actinides, group extraction of all actinides, and fission products. While the major actinides represent the highest priority for the economic gain which can be realized from fuel recycle and the problematic heat and radiation generated in long-term storage by Plutonium, the other three divisions are essential to research if the problem of nuclear waste management is to be comprehensively tackled. The following technologies with be classified under the four aforementioned groups.

Major Actinide Separation

PUREX is the only mature reprocessing technology in use. As a hydrometallurgical (aqueous) process, it can be adapted to most forms of fuel, as well as condition any un-separated radionuclides for long-term storage. Many other aqueous processes are adapted from the PUREX flowsheet, although some methods such as UREX, DIAMEX-SANEX, and GANEX use extract molecules other than TBP. Non-aqueous routes still in development include pyrometallurgical reprocessing and experimental processes involving CO_2, Freon, or fluorides. The former is well-suited to metallic fuels which might be used in Generation 4 reactors (i.e. the Lead-Cooled Fast Reactor, which was not covered) or fuels with a high concentration of minor actinides. (IAEA, 2008)

Minor Actinide Separation

Minor Actinide recovery is accomplished generally by applying a PUREX-related process to the raffinate (liquid remains) from the initial PUREX actinide extraction. Key pursuits here include Americium and Curium extraction with DIAMEX-SANEX and Neptunium separation with COEX. No viable non-aqueous means currently exist for MA extraction, with vitrification to a solid a waste containment solution rather than recovery option. However, research on actinide extraction via liquid aluminum in molten fluorides is being researched in France. (IAEA, 2008)

Group Actinide Separation

Separation of all actinide remains the ideal path, like most ideals is quite far from attainment, however. This concept is closely linked with Generation 4 Fast Reactors, which in the course of today's uranium-based fuel cycle would burn fuel and transmute actinides from previous burn cycles. Following group reprocessing, the actinide collection would be returned to reactors while fission products are disposed of. The GANEX process details how this might proceed, beginning with spent fuel dissolution, initial recovery of uranium, co-extraction of all remaining actinides and lanthanides, separation of Plutonium from the minor actinides, and then collective back-extraction of Am, Cm, and Np. The pyrometallurgical alternative would see the SNF dissolved in liquid salts (as in on-line MSR processing) followed by extraction by way of selective precipitation, electrolysis, or other methods. (IAEA, 2008)

Fission Product Separation

Fission products present an extraction problem of their own because their utility is marginal in comparison to the actinide elements. Only I-129 and Tc-99 can be practically transmuted, while other important gamma and beta emitters such as Cs-135, Sn-126, and Se-79 cannot and must be vitrified with HLW. (IAEA, 2005) If any form of destruction were to be made possible in the future for Cs-137 and Sr-90, which produce almost as much heat in waste separately as the other fission products do combined, this could somewhat alleviate disposal problems. But as it is, these species only have around 300 years of radioactive life, which pales in contrast to the decay times of other radioisotopes. (IAEA, 2008)

Chart 3.1: Reprocessing Technology Information

Technology	**Fuel Type(s)**	**Separation**	**Maturity**	**Additional**
PUREX*	UO2, MOX (LWR/FR) +	Pu, U, MA, FP	Industrial	Economical, Efficient
COEX*	UO2, MOX (LWR/FR) BU+	Pu/U & Tc/I or Pu/U/Np & Tc /I	Near Industrial Scale	Gen. III, Prolif. Res.
NUEX	UO2 (LWR)	Spec. Unknown	Conceptual	No pure Pu extraction
THOREX	Irradiated ThO2	U(233)	Plant Scale	No MA separation
NEXT	MOX (FR)	U, Pu/Np, Am/Cm	Laboratory	All actinides recovered
REPA	UO2 (LWR)	U/Pu, Some FP	Laboratory	Nitrogen-based process
DIAMEX-SANEX*	UO2, MOX (LWR/FR) BU+	Am,Cm	Laboratory	PUREX/COEX-compatible
UREX Var.	UO2 (LWR)	Var. Actinides, FP	Laboratory	For Homo/Hetero Recycle
GANEX	Var. (LWR/FR)	U/Pu, MA, FP	Laboratory	For Homo Recycle in FR
DDP	UO2 (LWR), MOX (FR)	U/Pu/Np	Pilot Scale	For MOX Fabrication, Pyro.
Pyro-Process	FR/ADS Fuels	U/Pu/MA	Laboratory	Transmutation Prep.
FLUOREX	UO2 (LWR)	U or U/Pu, FP, MA	Laboratory	Hybrid Process

Source: Compiled from IAEA 2008 Data (TECDOC-1587)

4. DISCUSSION

Detailing how nuclear reactor and reprocessing technology may have an impact on the environment in the future, it is necessary to clarify several details. Environmental impact in the context of this paper refers to the effectiveness of design in preventing undue contact of radionuclides in areas outside of their containment as LLW, ILW, or HLW. It refers to any measures referred to throughout the discussion of the nuclear fuel cycle such as resource use.

In some respect both the reactors and reprocessing phases of the future will be naturally predisposed to environmental harm, regardless of which technologies are used. Higher electricity demand will likely necessitate the production of more reactors (assuming a nuclear renaissance), and more advanced designs will generate greater amounts of fission products and transuranic elements. The reprocessing industry will be tasked with then manipulating these byproducts and recovering nuclides such as Plutonium and Neptunium, which are long-term HLW problems. These materials will likely be used in fast reactors which break them into fission products *more* hazardous to humans and the environment in the short term than their predecessors. Thus, safety in the nuclear industry will have to progress as new technologies emerge, even as cost-competitiveness may become more difficult.

Advanced reactor design at least seems to address many problems incurred in the past, and overcoming such obstacles is perhaps one key to winning public acceptance. Avoiding volatile hydrogen reactions can be accomplished by converting Tritium at reactors into stable water to minimize this risk, and using fuels which do not involve Zircaloy cladding is another option, as this was a core ingredient in the Chernobyl disaster. The replacement of water coolant by other means may also be ultimately desirable, since the phase changes of water and its occasional tendency to form power-surging voids, and other mediums are able to cope with higher reactor temperatures for greater plant efficiency. Both cladding and coolant changes are possible with the TRISO fuel (in MHTGRs) using an inert ceramic cover with helium gas as the coolant, and molten salt which utilizes no express cladding and serves the dual purposes of fuel and coolant.

These two fuels and associated components also feature a high degree of passive safety, the most important concept for next generation reactors when abnormalities do occur during operation. Since passive safety does not require human interface, as is the case in Generation 2 reactors when personnel must occasionally manage reactor heat by pumping certain amounts of cooling water into the core, responsibility will reside with technology design and construction, and not with people and (to some extent) power supply. If passive safety combined with control room optimization are able to reduce the number of people working at such a sensitive location, and exposed to increased (if minimal) levels of radiation, then this can also be counted as a benefit.

Notable passive safety engineering included in Section 3 includes the isolation condensers designed for ESBWRs and SCWRs, and the Freeze Plug option in MSRs. The latter appears to be an extremely promising mechanism, not only in itself but because it is paired with other passive safety features in MSRs; it is no wonder many MSR design elements are incorporated into a project known as the 'Ultimate Safe Reactor', and the main concerned associated with this design is not necessarily the safety of the reactor per se but the complexity added with online fuel processing. Another safety mechanism which is not part of any Generation 4 reactor covered, but which has great relevance nonetheless, is the "Core Catcher"* system made for the EPR and other Generation 3 reactors. This works by gathering and cooling via gravity the fuel cladding and other materials should reactor meltdown transpire. By doing so, the device protects the containment building from being flooded with hot, radioactive material. When one considers the containment leaks in a number of the Fukushima reactors which are allowing radionuclides to escape into the environment, this is exactly the kind of technology which could stop such events from occurring.

One can ultimately only speculate how advanced reactor technologies might compare to one another since none have been deployed commercially and are still undergoing rigorous testing and experimentation. What can be better anticipated is the environmental benefit which may accrue due to reactor and reprocessing technologies (in tandem) altering aspects of the current fuel cycle. If uranium and plutonium are recycled with current PUREX-based processing, and refashioned into MOX fuel for use in present-day LWRs, then approximately 35%** less Uranium will be needed to be mined out of the earth, which is critical given the land degradation and tailings which are generated in the mining and milling processes, as well as the rising cost of this uranium. This step is already feasible, although waste management of actinides and fission products with PUREX is limited. If we look into the future of fast reactors with homogeneous actinide recycling and the UREX process also still in development, however, the potential for waste reduction may be able to reach 95%, with Cesium and Strontium remaining as the final impediments in HLW disposal. The fast reactors will also be able to tap into the energy content of fissile and transmutable actinides, which, in a once-through cycle, would be inaccessible. The energy attainable through recycling is estimated at upwards of one hundred times that recovered in a single cycle,*** so this has important implications in light of resource scarcity as well as use efficiency.

Of the additional reprocessing techniques yet to be mentioned, three should be mentioned due to their projected impact or lack thereof. COEX is a French aqueous design which represents a step up from PUREX reprocessing, is near industrial scale operation, and will be compatible with the emerging Generation 3 reactor designs. The change in COEX is that it will be able to process fuel having undergone more intense irradiation, and if coupled with the DIAMEX-SANEX process, it will be able to separate as great a variety of actinides and fission products as with multiple-stage PUREX processing. COEX also offers a higher degree of proliferation as found in PUREX since in each of its two paths Uranium and Plutonium are recovered together. Regarding the Thorium fuel cycle, the THOREX process raises some concern, since only recyclable fuel can be potentially recovered. And despite Thorium fission producing less hazardous products and Uranium fission, this is still an area which will need to be addressed if Thorium exploration is to continue and eventually linked to a Thorium fast reactor such as the MSR.

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* see UK-EPR Safety Document

** see IAEA(2005) Reprocessing Section

*** see Forsythe (2009) Recycling Section

5. CONCLUDING REMARKS

Environmental safety in the nuclear fuel cycle is a highly complex issue, and perhaps better analyzed in specific instance given the breadth of the supply chain and the variation in behavior of radionuclides from species to species. While the overall radioactivity dose transferred from nuclear plants to human (as mentioned in the Introduction) is on the order of .1 microsieverts per year compared to 3.1 millisieverts from natural sources (NRC, 2011), many people misconstrue any radiation exposure from an ‘unnecessary’ source into condemnation of an entire industry. This logic is flawed on several levels: one, nuclear power is not dispensable if energy demands around the world continue to escalate and action is to be taken against rising atmospheric carbon dioxide concentrations. Secondly, and I hope this paper provides verification, aspects of the nuclear fuel cycle such as reprocessing discharges, transport energy, mining tailings and water consumption and all other ‘hidden’ processes most certainly collectively outweigh the environmental impact of the dispersed Xenon and Krypton gases.

Still, even if there are many misconceptions about nuclear power, the development of certain reactors and reprocessing technologies in a fuel cycle scheme may not be justified in all cases. Applying district heating to nuclear plants may often be unpractical since plants are generally zoned away from other city sectors so that too much heat is lost in transfer. Additionally, nuclear reactors are a centralized form of electricity generation, and come with associated security, flexibility, and upfront-cost disadvantages; the smaller modular reactors designed for the future are an answer to this, but the question then becomes whether or not cost and the efforts needed to secure the fissile material are worth the diminished MW output. Desalination coupling has not proven successful, and hydrogen production depends on many factors in the future such as hydrogen economy safety and cost-competitiveness, not to mention the health of the nuclear industry. The most justifiable option in my opinion is still the use of fast reactors, simply based on it solving the waste issues of past, increasing fissile resource energy efficiency, and extending the life span of nuclear energy (should they still be deemed useful within this time frame) since nuclear fuel is a finite resource. The development of molten salt technology for the SFR is especially deserving of attention because advancement in the molten fuel department may also carry over to other industries such as solar power, in which centralized Concentrating Solar Power plants might use the liquid to retain heat directed and focused from mirrors onto a central receiver. Finally, I hold the idealistic notion that fast reactors might one day be used to consume military plutonium reserves, which are no doubt expensive and to maintain since plutonium requires constant shielding as in the fuel fabrication process. In the meantime, however, its destruction in thermal reactors as MOX fuel does some small consolation in security.

Glancing quickly at national policies in Table 5.1, it is interesting to consider why certain countries have selected fuel cycle paths as they have. Germany, which is currently in the midst of a political storm trending towards an 'Ausstieg' of nuclear power, clearly does not enjoy remaining indecisive on the fate of SNF, and has heavily researched the use of salt mines for final waste disposal as well as employed PUREX reprocessing. It may, however, be disadvantageous to continually shift position on reprocessing and final disposal, since interim storage is the least expensive option in the short term (40 years stages) and allows time to commit to a certain path without wasting resources in one of the other.* The United States on the other hand has committed to an open fuel cycle for supposed proliferation concerns, but this may change quickly if the it wishes to develop a more advanced commercial nuclear power fleet and remain a player in technology development (i.e. UREX, advanced reactors). This decision is also aggravated by the extreme dependence of the US on energy imports. For France, which has perfected the fuel cycle and is the leader in reprocessing development, an open fuel cycle would be unfathomable since about 80% of the country's power is provided from nuclear**; thus, more SNF waste is produced in comparison to other countries. It may even be that the momentum from having such a mature nuclear industry will necessitate the construction of fast reactors there. In any event the demonstration of France that nuclear capacity can be deployed safely on a large scale should be commended, even if it is not emulated in other nations.

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* This option could be economically disadvantageous if Germany opts to only dispose of or reprocess SNF rather than switching between interim storage (dissipating some of the radioactivity and heat) and reprocessing for higher HLW reduction/lesser storage requirements; reprocessing would also be discounted for the future in this case.

** Forsyth (2008)

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APPENDIX

A. FUEL CYCLE APPROACHES BY COUNTRY

SPENT FUEL MANAGEMENT APPROACHES IN DIFFERENT COUNTRIES

Country	Deferred Decision	Direct Disposal	Reprocessing
Argentina	◆		
Belgium	◆		●
Brazil	◆		
Bulgaria	◆		●
Canada		■	
China			●
Czech Republic	◆	■	●
Finland		■	
France			●
Germany		■	●
Hungary	◆		●
India			●
Italy	◆		●
Japan			●
Korea Rep. of	◆		
Lithuania		■	
Mexico	◆		
Netherlands			●
Pakistan	◆		
Romania		■	
Russian Fed.		■	●
Slovakia		■	●
Slovenia	◆		
South Africa		■	
Spain		■	
Sweden		■	
Switzerland	◆		●
UK			●
Ukraine	◆	■	●
USA		■	

***Note**: Some countries use different approaches for different types of fuel. Additionally, some countries follow one approach while evaluating different approaches that might be applied in the future.*

Source: IAEA (2011)

B. RADIONUCLIDE PROFILES

Plutonium

Plutonium (AN 94) is the heaviest element on Earth known to have existed prior to the formation of the planet. While its natural concentration is scarcely detectable, artificial concentrations and release due to its use in nuclear fuel and weapons is particularly problematic due to its biological toxicity as well as radioactivity. Plutonium makes produces about one third of the energy in nuclear reactors (although it is frequently a by-product rather than a primary fuel); one kg of Pu-239 will suffice to generate 10 kilowatt-hours worth of heat.

Tending to form insoluble compounds, plutonium possesses weak potential for mobilization within the food chain. The primary method of intake would occur from inhaling airborne particles, which would enter the blood stream through the lungs.

Pu-239, the most important isotope in terms of commercial and military application, is formed during the transmutation process converting U-238 to Np-239 and then to Pu-239. Both Pu-239 and 241 are fissile and make up the majority of plutonium in LWR reactor fuel, while Pu-238 and 240 are fertile, so that they may be converted into fissile plutonium via neutron capture. Pu-242 is a neutron poison which can lower the reactivity of fuel complexes. The high specific heat of plutonium which makes it (Pu-238 esp.) attractive for isolated thermoelectric generation also necessitates much consideration in waste storage and disposal efforts.

Radium

Four isotopes of Radium (AN 88) are capable of being formed during the decay chains of Uranium and Thorium. Highly chemically reactive and more than a million-fold radioactive than an equal mass of Uranium, Radium gained notoriety in the 1920's when a number of women employed in watch painting were poisoned by ingestion. Since radium gives off a blue luminescence as it decays, it was applied industrially for glow-in-the-dark products, and the workers made a practice of wetting their paintbrushes with their tongues in order to more accurately apply the paint. The "radium jaw" which developed in these women was characterized by profuse bleeding, anemia, and bone cancer. The last of these can be attributed to Radium's biological deposition in bone tissue, much like calcium. (Deposition here leads to deterioration of the marrow.) Tritium serves today as the functional replacement for Radium where such luminescence is required.

Radon

Descending from radium in nuclear decay chains, Radon accounts for most of the ionizing radiation received by the public. Its lethality first became apparent in mining operations, where as a gas it was readily inhaled by workers, contributing in many cases to lung cancer. Radon may also accumulate in buildings if they are not properly ventilated, resulting in more intense exposure. Possessing 36 isotopes, none of which are stable, Rn-222 and Rn-220 are the primary forms derived from uranium and thorium. Given their respective half-lives of 3.8 days and 56 seconds, alpha emission over time is extremely high compared to other radioisotopes, although Rn-220 is much less mobile.

Technetium

Technetium (AN 43) occurs as a fission product of uranium, and any amount managed by humans will have been artificially produced, as none of its isotopes are stable enough to persist very long in the natural environment. The most significant of its isotopes, Tc-99, is difficult to manage in spent fuel reprocessing as the pertechnetate compound which is formed will not be captured by most recovery techniques, which favor the cationic compounds formed by other fission products such as Cesium and Strontium. Transmutation of Tc-99, however, results in temporarily in Tc-100, which then through beta decay becomes Ruthenium. This neutron treatment in transmutation reduces Technetium's half life from 213,000 to 16 seconds.

Thorium

More naturally abundant than Uranium, Thorium (AN 90) can be mined out of the monazite found voluminously in Brazil, China, and Southern India. Th-232 is the only isotope to exist in such mineral deposits, and consequently requires no separation prior to being assaulted with slow neutrons in the Thorium fuel cycle (produces fissile U-233). Though not commercialized in modern reactors, the substance may be used in future Generation III reactors (Advanced LWR's) as well as Generation IV designs such as the Molten-salt reactor (MSR).
With a half-life of 14 billion years, Th-232's radioactivity is negligible. However, as with Neptunium and Uranium, Thorium's decay series includes the more potent Radium and Radon elements.

C. RADIONUCLIDE PROPERTIES

Fission Product	**Half-life (years)**	**TBq. (metric tons)-1**	**g.(metric tons)-1**
H-3	12	32	0.08
Kr-85	11	420	27
Sr-89	0.14	4,000	3.5
Sr-90	29	2,400	430
Y-91	0.16	7,600	7.8
Zr-93	950,000	0.08	490
Zr-95	0.18	16,000	19
Nb-95	0.1	520	21
Tc-99	210,000	0.6	880
Ru-103	0.11	7,200	5.7
Ru-106	1	33,000	240
Sb-125	2.7	520	12
Te-125	0.16	260	0.36
Te-127	0.3	1,000	2.7
Te-129	0.09	530	0.42
I-129	17,000,000	0.0016	250
I-131	0.02	0.08	<0.01
Cs-134	2	4,000	77
Cs-135	3,000,000	0.048	1,400
Cs-137	30	4,200	1,200
Ce-141	0.09	3,200	2.8
Ce-144	0.78	32,000	250
Pm-147	2.6	8,000	220
Eu-155	5	1,600	87

Actinide	**Half-life (years)**	**TBq. (metric tons)-1**	**g.(metric tons)-1**
U-235	710,000,000	<0.04	8,000
U-236	24,000,000	<0.04	4,000
U-238	450,000,000	<0.04	950,000
Np-237	2,000,000	<0.04	600
Pu-238	86	160	230
Pu-239	24,000	20	8,100
Pu-240	6,600	26	2,900
Pu-241	13	6,000	1,300
Pu-242	380,000	0.08	510
Am-241	458	30	230
Am-242	0.45	1,400	10
Am-243	7,800	0.8	100

Source: Compiled from EOLSS Data (2004). Figures based on 33 GWd(t).(metric ton)$^{-1}$ burn-up & 150 days cooling

D. RELEVANT TERMS

Actinides: a family of elements ranging from Actinium (89) to Lawrencium (103). Of these, the more abundant Thorium (90), Uranium (92), and Plutonium (94) serve as fuel material in nuclear reactors. The term ‘minor actinides’ is used to encompass elements in spent nuclear other than the fissile progenitors; these include Neptunium, Americium, and Curium.

Control rod: control rods absorb neutrons so that when fully immersed in the reactor core, the nuclear chain reaction may not proceed. Materials such as boron as used because as well as having a high neutron capture cross section, upon absorbing an electron, the resulting isotope is non-radioactive.

Criticality: a criticality event may occur when too much fissile material is concentrated in a location. While unintentional chain reactions can occur in test environment, they cannot generally be sustained very long, (nor result in an explosion) although enormous amounts of neutron radiation and heat will be generated. Normal criticality of k_{eff} =1 is required for stable plant operation.

LWR (light water reactor): a thermal reactor which uses ordinary waters as both coolant and neutron moderator. This is the most common design of today’s nuclear power plant. The more specific types of LWRs are pressurized water reactors (PWRs), boiling water reactors (BWRs), and supercritical water reactors (SWRs).

Moderator: in the reactor core, moderators are responsible for slowing down fast neutrons into thermal neutrons which can be captured by the fissile fuel, although as with control rods they will absorb neutrons to some extent. Moderating substances include most commonly water, heavy water (deuterium), and graphite, the latter two absorbing fewer neutrons per slowed neutron than the former.

MOX fuel (mixed oxide fuel): reactor fuel which contains more than one oxide of fissile material (typically uranium and plutonium oxide) as an alternative to natural or low-enriched uranium

Transmutation: the process of changing a chemical element into another element, which occurs artificially through neutron bombardment and naturally by radioactive decay. Transmutation using neutrons is an accelerated way of reducing the radioactive half-lives of actinides, (when applied to nuclear waste management) and in doing so favoring short-term radioactivity and heat management over long-term management. Actinides are essentially transformed into lower-order fission products.

Transuranic elements: those elements featuring atomic numbers great than that of Uranium (92). Each of these elements has been generated synthetically, either through neutron capture or nuclear fusion, as they do not possess the stability to occur in nature.