HYBRID SYSTEMS USING SMRS: A PATH TOWARDS SUSTAINABILITY IN NUCLEAR AND CANADIAN ENERGY PRODUCTION

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A Thesis Submitted to the School of Graduate Studies in Partial Fulfilment of the requirements for the Degree Master of Applied Science

Abstract

Nuclear technology development in Canada has been relatively stagnant since the 1980s, when CANDU reactors were first implemented into the power grid. Reprocessing technologies such as pyroprocessing and the fluoride volatility method would introduce new opportunities for numerous industries throughout Canada. Transmutation of minor actinides and fission products have been proven to ease requirements of fuel repositories due to the reduction in radioactivity. Economic advantages from implementing SMRs in various industrial systems, including Canadian oil sands, would increase efficiency while decreasing CO_2 emissions.

Acknowledgements:

I would like to thank my supervisor Prof. John Luxat for providing oversight, leadership, and encouragement.

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1 Introduction

Global electricity production is mainly dominated by high carbon emitting sources, which is causing the interest in other electricity avenues to be explored and executed [1]. The production of electricity using renewable sources has increased; however, large-scale industrial operations and regions of large population densities also require electrical support from a baseload [2]. The baseload is a constant supply of power by an energy producer to the public to meet fundamental energy demands. Due to the energy grid's requirement of stability and robustness of supply, intermittent renewable sources cannot be the only contributor. Therefore, in order to not only replace coal power, but to maintain a baseload, nuclear and hydro (where available) must provide the baseload until more technology is available. Canada has set a goal to have 90% of the country's electricity produced by non-emitting sources by 2030 [3]. An international forum was established to develop the next generation of nuclear reactors, Generation IV (GEN IV) reactors, which includes the Small Modular Reactor (SMR). In order for Canada to reach its electricity production goals, the SMR should be considered.

The SMRs are described as new technologies varying in size, design features, and cooling types [4]. Most SMR designs employ novel approaches that will affect the certainty of how the plant will perform under normal and accident conditions [4]. Many SMRs are being marketed as flexible and unique in both operation and application. However, one cannot simply insert a new technology into a community or society and expect acceptance and success. One design aspect of some SMRs is the possibility of being operated with little or limited supervision. The idea behind this design choice is to allow the reactor to enter remote communities, including indigenous communities, and theoretically require little maintenance and operation staff, and minimal disruption to their way of life; although, this may not be entirely beneficial to the communities being impacted in both direct and indirect manners. One benefit of conventional nuclear operations is the guaranteed jobs that are supplied to the communities that surround the development. Remote operation would not provide many jobs, and therefore, communities would receive minimal training and understanding of the entire nuclear industry. The area would be subjected to a potential threat of radioactive exposure, and would have very few improvements to their way of life or local economy. Also, transportation of the radioactive materials would require the routed communities to approve of the risk, without necessarily receiving any benefit. Another risk with remote operation, which must be considered, is the limited amount of trained staff and support in case of an event. Although the SMR may be designed to protect itself, operations staff are typically present to oversee the reactor to ensure ideal operation. If the remote operation has any delay or interference (e.g. cyber security issues), the reactor and surrounding communities may be at risk. One apparent benefit of SMRs within the northern communities, is a clean power source instead of the diesel generators that are currently being used. Due to the changing environmental conditions, transportation of diesel fuel is already becoming difficult, such as the ice roads melting in the summer. The infrequent delivery of fuel would allow the risk of power loss to be reduced.

Several SMR designs can also be utilized on an industrial scale, with larger electricity production and higher process heat. Resource extraction such as oil refining or mining is being considered as a benefit of SMR designs; however, the current policies and licensing process may require modification in order to allow hybrid systems and SMRs to be implemented.

The infrastructure required to support many of the suggested SMRs would require policy revisions as well as societal acceptance. The only power producing reactor currently in operation in Canada is the Canadian Deuterium Uranium (CANDU) reactor, which uses natural uranium fuel. One of the appealing qualities of the CANDU reactor is it does not require any enrichment, and therefore increases the proliferation resistance. Since all of the proposed SMRs require enriched fuel, an enrichment facility would be required, or enriched fuel would need to be shipped in from elsewhere.

Currently, all of the spent fuel from the CANDU reactors are first cooled in wet storage for at least 6 years, then are shipped to dry storage [5]. The fuel bundles are kept intact to prevent any contamination, exposure, and again, would increase proliferation resistance. Currently, the Nuclear Waste Management Organization (NWMO) plans to have all the spent fuel placed underground in a Deep Geological Repository (DGR) indefinitely.

2 Literature Review

2.1 Reprocessing

A major challenge of the nuclear industry is the looming presence and management of nuclear waste [6]. An alternative to merely disposing of the spent nuclear fuel directly into a repository, is reprocessing and reusing elements of the spent fuel. Various methods for reprocessing are being studied to reduce waste generation and increase fuel efficiencies. As of 2011, the global demand for uranium is 67000t/year, which is expected to double by 2030 [6]. The increasing demand for uranium will put a strain on Canada's resources and potentially affect the CANDU fuel cycle that is already established.

The reprocessing of nuclear fuel involves the separation and purification of uranium and plutonium products from the fuel removed from the reactor [7]. Once recovered, the uranium and plutonium can be repurposed and converted into a recycled oxide fuel, and reinserted into a reactor. The remaining material includes fission products (FPs) and minor actinides (MAs), which are typically vitrified and disposed of. New designs of reactors involve technologies that help reduce waste generation and radioactivity of the nuclear waste by transmutation, which is discussed in Section 2.2.

2.1.1 DUPIC

The Pressurized Water Reactor (PWR) spent fuel contains fissile material, 0.9 wt% of U-235 and 0.5 wt% Pu-239, which could be used for CANDU reactors after some fission products are removed. The direct use of PWR spent fuel in CANDU (DUPIC) recycling is a PWR-CANDU symbiotic fuel cycle, and is popular in countries that have both reactor types, such as the Republic of Korea [91].

2.1.2 PUREX

The United States produced the Plutonium Uranium Redox Extraction (PUREX) process in 1940, and has continued to be the dominating reprocessing technology since the 1960s. Figure 1 shows a schematic representation of a typical PUREX process. The PUREX process consists of: a) head-end plant, b) chemical separation, c) conversion, and d) disposal. The head-end portion of the plant is to remove the fuel from the cladding. The fuel is then placed in HNO_3 solution to prepare for the chemical separation using solvent extraction to produce different aqueous nitrate products. The separate nitrate products are then converted to solid oxide products. The disposal portion of the PUREX process is a significant flaw, as there are many solid, liquid, and gaseous wastes and effluents that arise [7].

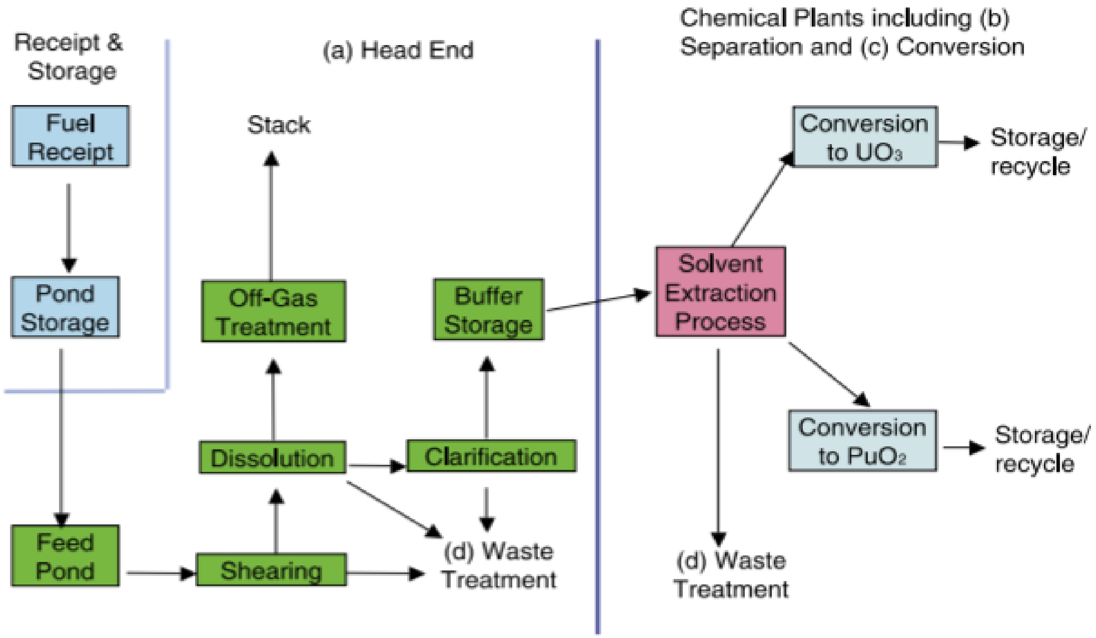


Figure 1: PUREX representation of head-end, chemical separation, conversion, and waste management [7].

Chemical separation flow sheets are typically similar, and numerous cycles are used to separate and purify uranium and plutonium from all other elements. Further effort is being put into reducing the solvent extraction steps, separating plutonium and uranium earlier in the process, minimizing waste by using salt-free reagents, and upgrading the contacting equipment [7].

Although the PUREX process is heavily established and widely used, the technology is unlikely to be used on Canadian soil. Canadian nuclear oversight highly protests the separation of plutonium from other elements, as it decreases the proliferation resistance [59].

2.1.3 Fluoride Volatility Method

The fluoride volatility method (FVM) can be used to reprocess oxide spent fuels from a variety of different reactors, including fast reactors. The technology would allow the reprocessing of fuels with inert matrices, high burnup, high plutonium content, and a short cooling time. The FVM was initially promising for those who were interested in the deployment and reprocessing of fast reactor technologies, as PUREX is not suited for fast reactor spent fuel. The FVM is based on a separation process, and the properties of various heavy metals' abilities to form volatile hexafluorides, whereas others are non-volatile trifluorides [8].

2.1.3.1 FVM Description

The advanced pyrochemical reprocessing method is considered a promising technology for oxide spent fuels from GEN IV fast reactors, especially fast breeders with a closed uranium-plutonium cycle [18]. The inspiration for this reprocessing method is from the need for GEN IV reprocessing methods, and is one of the only non-metallurgical processes. Advanced fuel cycles including Mixed Oxide (MOX) fuels with high amounts of plutonium, inert matrices, and very high burn-up, can turn to this method; whereas hydrometallurgical methods would not perform adequately due to the high radioactivity or solubility in nitric acid [18].

The FVM is based on a separation process, which relies on the properties of uranium, neptunium, and some plutonium to form volatile hexafluorides. Other fission products and higher trans-plutonium elements would form non-volatile trifluorides [18]. The range in properties of elements within spent fuel allows for several separation processes which could be brought by several fluorinating agents (e.g. BrF_3, BrF_5, ClF_5), or pure fluorine gas [18]. The reaction time in which fluorination would occur is dependent on the choice in gas; pure fluorine gas would provide a fast and efficient reaction.

The initial operations involved in the FVM can be considered separate and preparatory. The removal of the cladding material, and the transformation of fuel into powder are required before the true FVM can begin. The removal of the oxide fuel cladding can be done in a furnace; whether it be zircaloy or stainless steel, both can be fully removed. The pellets transforming into powder can be done mechanically or by voloxidation, which is the partial oxidation of UO_2 to U_3O_8, which would be difficult to achieve for the fluorination process [8].

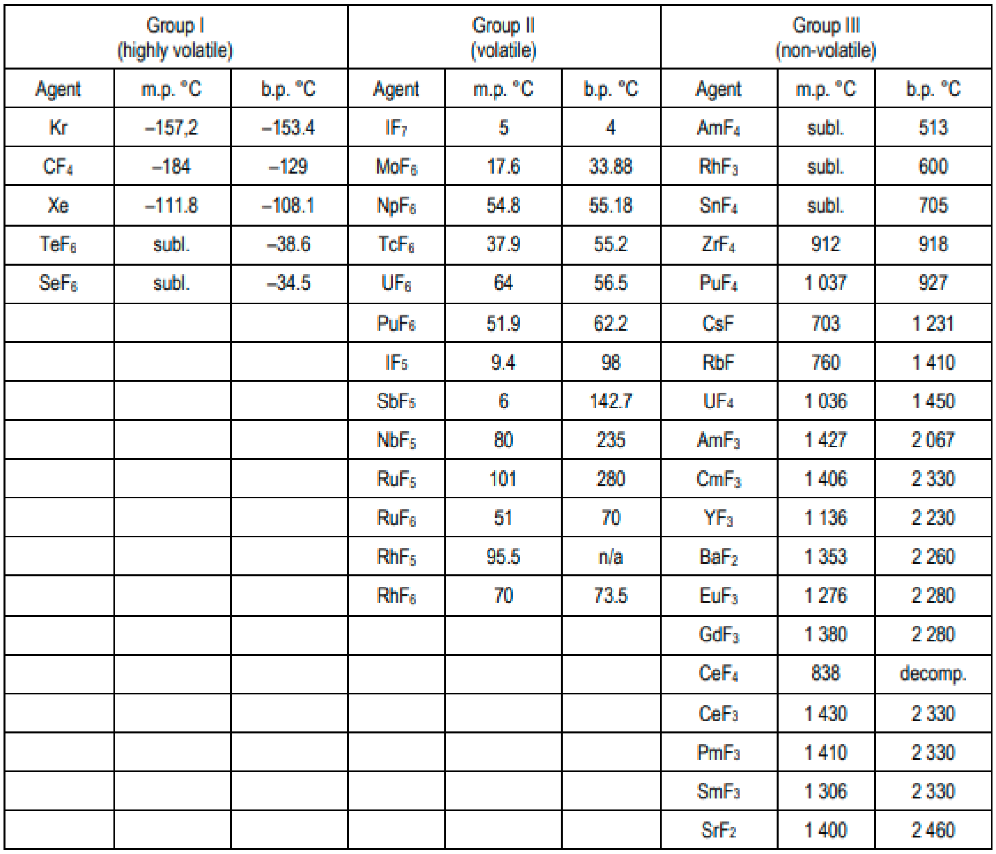
The reaction between the oxide fuel powder and fluorine gas is highly exothermic. The ignition temperature is around 250C, and the flame can reach as high as 1700°C, requiring a robust reactor chamber, and maintaining an intensive cooling system. Principle fluorination reactions of major fuel components can be found in Table 1.

Table 1: Chemical equations involved in major fluorination reactions [8].

Group Name	Chemical Equation	Standard Enthalpy $\Delta_r H^o_{298.15}$
Uranium	$UO_2(s) + 3F_2(g) \rightarrow UF_6(g) + O_2(g)$	-1062.4 kJ/mol U
	$U_3O_8(s) + 3F_2(g) \rightarrow 3UF_6(g) + 4O_2(g)$	-955.8 kJ/mol U
Plutonium	$PuO_2(s) + 2F_2(g) \rightarrow PuF_4(s) + O_2(g)$	-722.4 kJ/mol Pu
	$PuO_2(s) + 3F_2(g) \rightarrow PuF_6(g) + O_2(g)$	-693.1kJ/mol Pu
	$PuF_4(s) + F_2(g) \leftrightarrow PuF_6(g)$	---
Lanthides	$2Ln_2O_3(s) + 6F_2(g) \rightarrow 4LnF_3(s) + 3O_2(g)$	---
Minor Actinides	$NpO_2(s) + 3F_2(g) \rightarrow NpF_6(g) + O_2(g)$ $NpO_2(s) + 2F_2(g) \rightarrow NpF_4(s)+O_2(g)$ $NpF_4(s) + F_2(g) \leftrightarrow NpF_6(g)$	-907.9kJ/mol Np -844.7kJ/mol Np ---
	$2Am_2O_3(s) +6F_2(g) \rightarrow 4AmF_3(s) + 3O_2(g)$	---
	$2Cm_2O_3(s) +6F_2(g) \rightarrow 4CmF_3(s) + 3O_2(g)$	---

As described in Table 1, one can see that the majority of the partitioning is done within the fluorination reactor. The various elements within spent fuel range from highly volatile to non-volatile (Table 2). The volatile products leave the apparatus, and the non-volatile remains in the bottom in the form of ash. If more separation is desired, the volatile fluorides can be sent through sorption, condensation, or distillation processes [8].

Table 2: Distribution of the fluorinated spent fuel according to volatility [33].



Group I (highly volatile)			Group II (volatile)			Group III (non-volatile)		
Agent	m.p. °C	b.p. °C	Agent	m.p. °C	b.p. °C	Agent	m.p. °C	b.p. °C
Kr	−157,2	−153.4	IF_7	5	4	AmF_4	subl.	513
CF_4	−184	−129	MoF_6	17.6	33.88	RhF_3	subl.	600
Xe	−111.8	−108.1	NpF_6	54.8	55.18	SnF_4	subl.	705
TeF_6	subl.	−38.6	TcF_6	37.9	55.2	ZrF_4	912	918
SeF_6	subl.	−34.5	UF_6	64	56.5	PuF_4	1 037	927
			PuF_6	51.9	62.2	CsF	703	1 231
			IF_5	9.4	98	RbF	760	1 410
			SbF_5	6	142.7	UF_4	1 036	1 450
			NbF_5	80	235	AmF_3	1 427	2 067
			RuF_5	101	280	CmF_3	1 406	2 330
			RuF_6	51	70	YF_3	1 136	2 230
			RhF_5	95.5	n/a	BaF_2	1 353	2 260
			RhF_6	70	73.5	EuF_3	1 276	2 280
						GdF_3	1 380	2 280
						CeF_4	838	decomp.
						CeF_3	1 430	2 330
						PmF_3	1 410	2 330
						SmF_3	1 306	2 330
						SrF_2	1 400	2 460

The fluorination of uranium to UF_6 is spontaneous, due to its volatility (Table 2); however, plutonium hexafluoride is thermally unstable and can only be confidently obtained with excess fluorine gas. Neptunium hexafluoride (NpF_6) behaves similarly to plutonium, but more thermally stable, making it easier to acquire [18]. Although, if one desires to secure an independent stream of uranium and plutonium, the chemical properties of neptunium adds challenges. The reactions of NpF_6 with NaF, and the physical-chemical data of neptunium hexafluoride contaminates both aforementioned elemental streams [8]. Sorption and desorption with sodium and magnesium fluorides are methods to remove neptunium. Sorption and desorption are merely the attaching and detaching of molecules to each other, respectively. The flow chart of the sorption-desorption cycle is shown in Figure 2.

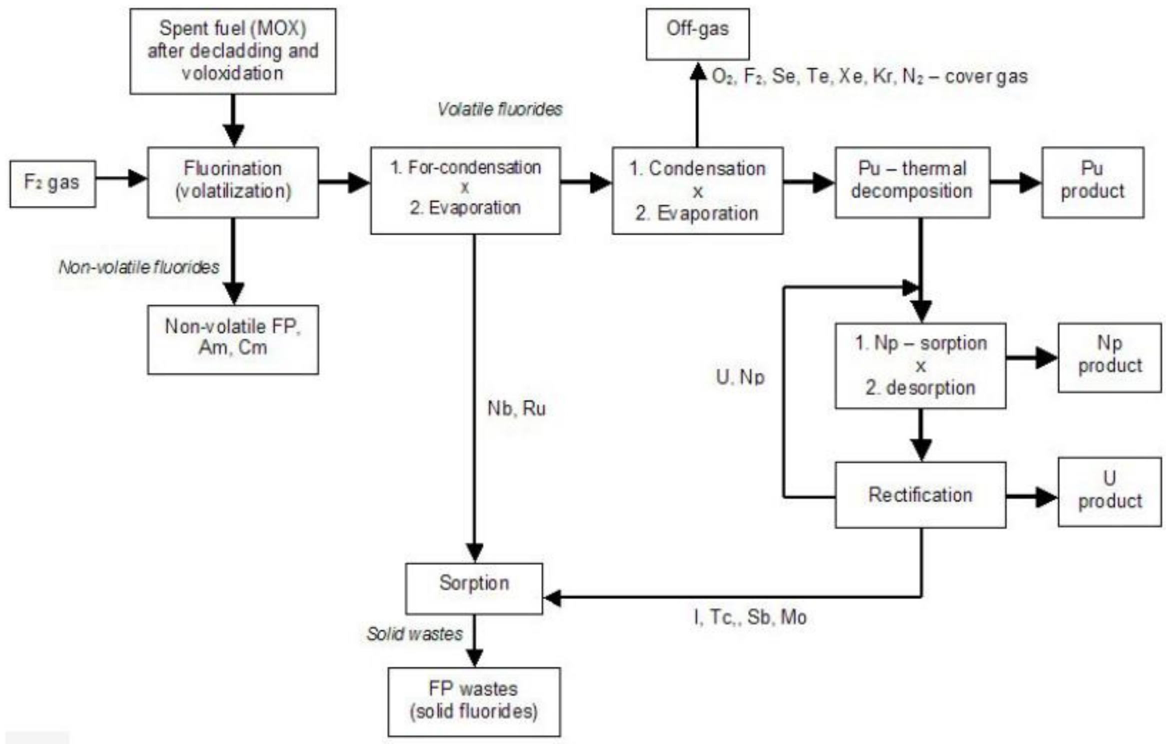


Figure 2: Process flow of fluoride volatility method [44].

All three heavy metal fluorides are completely sorbed on sodium fluoride at 100°C. Uranium and neptunium have successful desorption while passing fluorine gas through the bed; the desorption of plutonium hexafluoride is impossible and formed complex $PuF_4 \cdot 3NaF$, which is thermally stable in the fluorine gas flow. Separation of uranium and plutonium is possible through irreversible sorption of NpF_6 on MgF_2, as uranium is not sorbed on MgF_2 but neptunium hexafluoride has a sorption efficiency of 60-70% [8].

Certain compounds tend to accompany uranium hexafluoride throughout the cycle, and therefore requires a purification process to remove MoF_6, TcF_6, IF_5, and SbF_5 from the stream. Distillation has been proven to be an adequate process in which uranium hexafluoride is separated, where UF_6 is in liquid form [8].

2.1.3.2 FLUOREX

A reprocessing system was designed for the intention of having specific decontamination factors (DFs) for re-enrichment and storage. Reprocessed uranium and plutonium should be pure to reprocess MOX fuels in Light Water Reactors (LWRs), but a high DF may not be required for Fast Breeder Reactors (FBRs) [19]. The design of the fluoride volatility and solvent extraction (FLUOREX) allows for a flexible DF, which is a hybrid of fluoride volatility and solvent extraction (Figure 3).

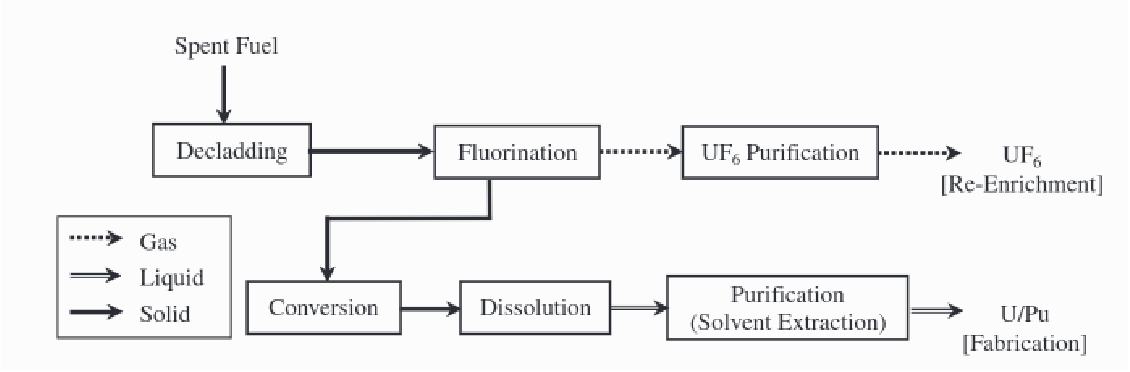


Figure 3: FLUOREX flow chart [8].

As shown in Figure 3, a pure uranium hexafluoride product is achievable through adsorbents like NaF, which is then able to be transferred for re-enrichment or simple storage. The other stream includes plutonium, uranium, and fission products that are dissolved in nitric acid and prepared for the PUREX method (purification). The interface between the FVM and PUREX needs to be perfected in order for the FLUOREX method to be successful. The non-volatile fluoride residues are converted to oxides soluble in nitric acid. Removing fluorine from the fluorination residues will prevent the production of insoluble plutonium tetrafluoride [19].

2.1.4 Pyroprocessing

The pyroprocessing method is a high-temperature electrochemical process in which spent nuclear fuel is converted into a metal fuel for a fast reactor [9]. Similar to the previous methods, the spent fuel must endure the head-end process before the actual reprocessing technology is performed. The flowchart of the pyroprocessing method is shown in Figure 4.

Figure 4: Korean flow diagram of pyroprocessing [9].

The head-end process involves voloxidation, where the dense oxide fuel pellet is oxidized and transformed into a powder of U_3O_8 at a high temperature. The powder is then prepared by voloxidation to the correct porosity for the following electrochemical process. Within electroreduction, the oxide fuel is reduced to metal feed within molten Li_2O-LiC salt, and the high heat fission products are dissolved in the salt. The metal feed is sent to electrorefining, where pure U is recovered within the LiCl-KCl salt, and transuranic elements and rare-earths are dissolved in the salt. Electrowinning recovers the uranium and transuranic elements from the salt [9].

2.1.4.1 Pyroprocessing Description

Although initially designed for the processing of metallic fuels, oxide fuels can be processed with an additional step. Using Li_2O-LiCl molten salt, the submerged metallic fuel would endure oxide reduction to prepare for the electrorefining process [32]. The inticing element of pyroprocessing is the electrorefining stage, in which uranium and transuranic (TRU) elements are separated from noble metals and fission products. Transuranic elements are elements which have atomic numbers $Z>92$. Plutonium could also be separated with uranium and minor actinides, which could be done by controlling the current or cathode potential. The potential to separate uranium and plutonium is cause for concert, as the International Atomic Energy Agency (IAEA) established a safeguard with which to detect the diversion of nuclear materials (U and Pu) in significant quantities [32]. In order to protect the facilities integrity and material flow, new technologies must be implemented to prevent material diversion.

Electrochemical Reduction

The following is a description of the FCC-Cambridge electroreduction process [9]. Electrochemical reduction has advantages for fuel processing: no troublesome handling of lithium metal; concentration of oxide ions in the electrolyte medium can be kept low, which is thermodynamically favourable for the reduction of actinide and lanthanide elements; simpler process than metallothermic reduction [9]. Although many molten salts were tested, the superior system included molten LiCl due to: the lower operating temperature, as LiCl has a melting point (605°C); a higher current efficiency; O^{2-} has higher solubility; and the higher compatibility with the following electrochemical process with the electrolyte being LiCl-KCl [9,40].The electrochemical process, shown in Figure 5, consists of a cathode basket and a platinum anode. The basket is loaded with oxide fuel, which could be in various physical forms (rod-cut, powder, porous pellets, etc.), and both the anode and the cathode are submerged in the molten LiCl. L_2O is added to the medium to increase the reaction rate and prevent anodic dissolution.

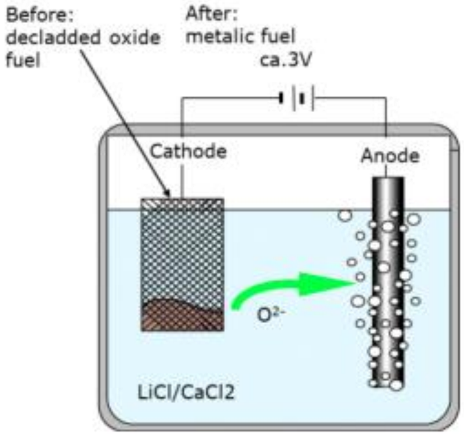


Figure 5: Schematic layout of electroreduction process [33].

As uranium is the prominent actinide in spent fuel, understanding the electrochemical conduct of uranium oxide is essential as a representative of all others. Beginning with U_3O_8 in the cathode, direct ionization results in the three different oxidation states, as follows: $LiUO_3$, U_4O_9, and UO_2. Further oxidation of UO_2 occurs in two paths, directly and with the aid of the lithium metal, as shown below.

$$UO_2 + 4e^- \rightarrow U + 2O^{2-} \quad \text{(Eq. 1)}$$

$$Li^+ + e^- \rightarrow Li \quad \text{(Eq. 2)}$$

$$UO_2 + 2yLi \rightarrow xU + yLi_2O \text{ or } 2yLi + yO^{2-} \quad \text{(Eq. 3)}$$

Carbon materials are typically used as anodes, particularly graphite due to the high electrical conductivity and low cost. When carbon is used for the FFC-Cambridge electroreduction process, the electrode becomes consumable as the metal oxide's O^{2-} ions are released and produces CO and CO_2. The use of carbon anodes for nuclear fuel reprocessing has been explored by many and shows poor reduction and carbon contamination of the metal oxide; therefore, inert anodes have been investigated as well, such as platinum and tungsten [9]. The reduction rate of the metal oxide is determined by the diffusion of O^{2-} ions to the bulk salt.

Fission products are soluble in the molten salt, in which they accumulate. The presence of the alkali fission products in the salt significantly prevents the reduction of UO_2, such as KCL or CsCl. If Cs is removed prior to the electrorefining, the lifetime of the salt bath would be longer, and more fuel would be refined in each batch. One way to remove Cs is in the head-end process is high temperature voloxidation, in which the oxide fuel is heated with oxygen [34]. Other fission products and actinides include Ce, Sb, Ru, Nb, and Kr.

The diffusion of the spent fuel's O^{2-} ions within the cathode basket is slower than that of the salt. Therefore, the fuel's characteristics, density and dimension, heavily affects the reduction rate as well. For example, low density and small sized spent fuels have higher reaction rates. However, powder fuel types are not pertinent for pyroprocessing because losses can occur, and powdered fuels would make the nuclear accounting more difficult [9].

The material that surrounds the cathode must be porous to allow the O^{2-} ions to flow from the salt, but smaller than the fuel's particles. Materials such as magnesia and stainless steel were investigated, stainless steel being more effective. Wire mesh baskets were made of stainless steel with multiple layers with 30-34% of the area being open for O^{2-} diffusion [9].

Another material that is considered for the cathode is solid aluminum. Actinides and fission products that remain in the salt can be selectively removed using electrodeposition (Figure 6). Throughout the electrorefining process, the fission product concentration within the salt increases, as they are dissolved from the fuel. Once a 'critical' fission product concentration is reached, when further selectivity would be problematic, the process is stopped. The actinides are to be removed, and the salt can be either recycled or disposed of. Inert chlorine gas is a proposed solution to remove the actinides from the anode; specifically, vacuum distillation for residual salt removal, chlorination of alloys yielding actinides and aluminum chloride, and performing sublimation for aluminum recovery from the chloride [36]. One significant problem of the chlorination process would be the complete chlorination of An-Al alloys without forming volatile An chlorides. Chlorination was studied using UAl_3 alloy and U-Pu-Al alloys, the conversion ratios (CRs) to chlorides were up to 99.8% and 97.0%, respectively. No losses of actinides were detected at 423K [36].

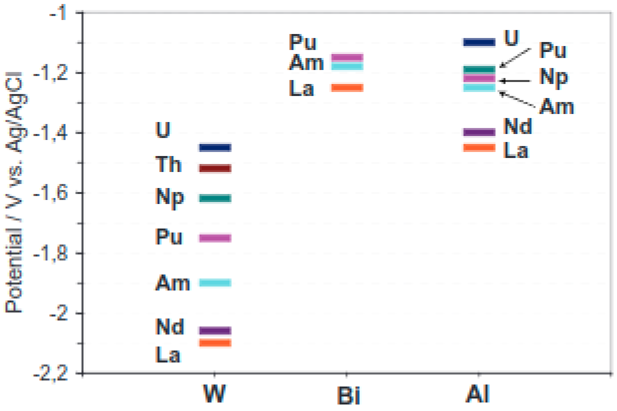


Figure 6: Experimentally determined reduction potentials of some actinides and lanthanides on different cathode materials [36].

A group-selective recovery process for actinides from nuclear fuel was developed for aluminum cathodes. Electro-separation of actinides and fission products in the molten salt occurs at 723 K, by applying a constant current between the metallic fuel within the cathode basket and the aluminum anode. The actinide cations move to the anode where they form An-Al alloys [36]. Alkali, alkaline, earth metal, and rare earth fission products are left within the salt due to controlled deposition potentials.

As O_2 gas is generated in the salt, the anode shroud provides an avenue for release and capture. Specifically, a stainless steel mesh around the anode was also proven effective, and did not endure damage or corrosion [9]. Ceramic materials were also investigated for anode materials, specifically $La_{0.33}Sr_{0.67}MnO_3$, $SrRuO_3$, $LaNi_{0.6}Fe_{0.4}O_{0.3}$. The dissolution of the cations (e.g. Sr, Ni, Fe), resulted in a chemically unstable compound [35].

The distance between the anode and the cathode basket, and the size of the electrodes would need to be optimized for the costs and volume of the salt. A large surface area would provide a high current and a faster reaction; however, this is not necessarily costly.

In a small scale-experiment [9], 60g of spent oxide fuel was reduced to a metal, most of which being uranium, plutonium, and neptunium (97%). Cs, Sr, and Ba were dissolved within the molten salt; however, 29% of the fission products remained within the spent fuel pellet.

Electrorefining

After the oxide fuel endures electrolytic reduction, the resulting metal fuel goes through the electrorefining process. As this is a pyro-process, there is no aqueous solution. The reduced metal acts as the anode, and contains uranium, transuranic elements, and rare earth elements, and is dissolved in LiCL-KCl eutectic salts. As the current is applied from the anode to the cathode, the uranium is dissolved, producing uranium ions soluble in the salt [42]. The metallic uranium is recovered as a solid dendrite on the cathode and the other elements remain in the salt to be transferred to the electrowinning process [41]. TRU elements that began within the feed material are also dissolved and form chlorides within the salt. The presence of two cathodes allows for lone distribution of uranium on one, and a U-TRU distribution on the other. Lanthanides present in the feed are also dissolved in the salt but do not deposit on the cathode along with the TRU elements [42].

The commercial process involves graphite cathodes, and the anode basket can load up to 50 kg of metal. The throughput of the electrorefiner is determined by the surface area of the anode load and the UCL_3 concentration within the salt. To achieve a high throughput, a high current should be applied, but is limited by the cut-off potential of the anode, with the noble metal retention of the anode basket. For example, when 32 kg of U metal was loaded in the anode basket and the salt's UCl_3 concentration reached 5.8%, the current limit is 325 A, and achieved a throughput of 17.9 kg U in one day. If the initial metal load is increased, and the allowed concentration of UCl_3 in the salt is increased, the upper limit of the current is increased and results in an increased throughput. A vacuum evaporation is required to remove the salt from the uranium deposits prior to the ingot casting process. In the ingot process, the uranium dendrite is fortified into a cylinder which can be used for fast reactor fuel or stored for future use [41].

For the U-TRU anode, phase separation occurs at a lower melting point, and the salt and metal can be separated using a bottom-pour crucible. Once separated, the U-TRU can be fabricated and used for fast reactor fuel [42].

Electrowinning System

A key step for proliferation resistance is the electrowinning step in which a liquid cadmium cathode is used to seize TRU along with the uranium from the salt [41]. At Korea's Atomic Energy Research Institute (KAERI) (Figure 7), the electrowinning process involves three steps: liquid cadmium cathode electrowinning for simultaneous recovery of U and TRU; residual actinide recovery process for recovery after the electrowinning process; and the cadmium distillation of the cathode product, removing U, TRU, and residual actinides [43].

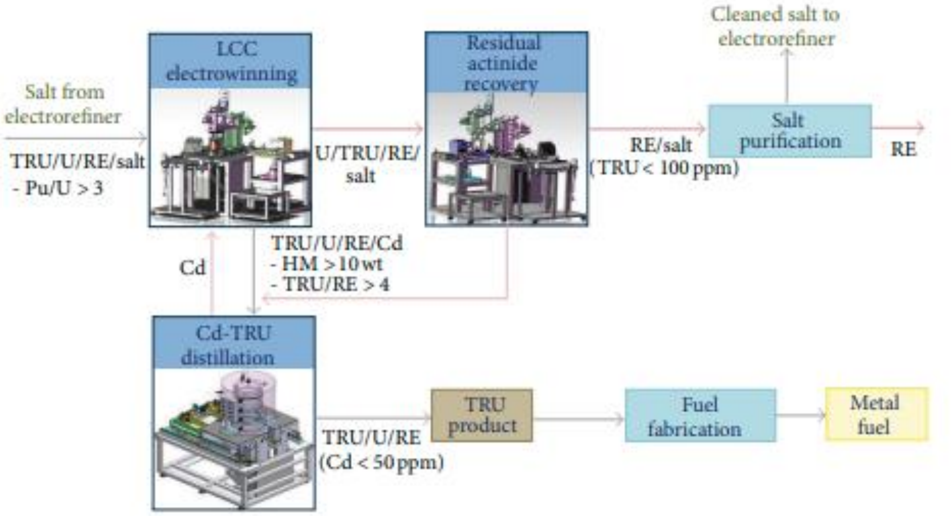


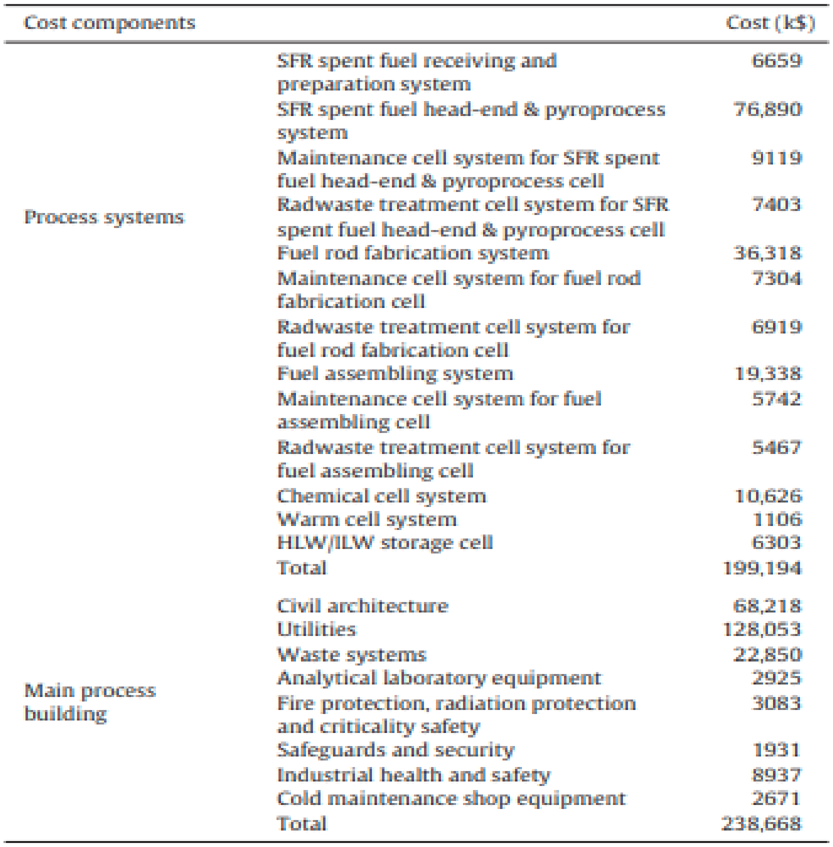
Figure 7:Schematic diagram of electrowinning of KAERI [43].

The dendrite deposition of the uranium ion on the cadmium cathode prevents co-deposition of U and TRU elements. A mesh type assembly for the liquid cadmium cathode prevents the uranium from growing into dendrites, increasing the uranium recovery and non-proliferation. The mesh assembly encourages the uranium to be pushed towards the liquid cadmium to allow for further deposition. A similar process was developed for the recovery of actinides in the salt, in which the ions are deposited onto a liquid cadmium cathode within a $CdCl_2$ oxidant [43].

2.1.4.2 Pyroprocessing Costing

An economic analysis performed by Choi et al. [53], determined the cost of spent fuel pyroprocessing, metal fuel fabrication, and decay storage. In 2007, the INL determined the nominal cost for pyroprocessing PWR spent fuel to be $1500/kgHM within a 40-year lifetime. In 2009, the values were adjusted and resulted in the nominal value being $2300/kgHM. The costs include head-end operations and separation costs. However, in 2010 KAERI estimated the cost of pyroprocessing PWR spent fuel to be $781/kgHM with a capacity of 400tHM/year. The cost includes construction, operation and maintenance, and decommissioning. The reprocessing of Sodium Fast Reactors were deemed to be more expensive in both the INL's analysis ($6000/kgHM) and KAERI's ($5511/kgHM) in 2009 USD. The KAERI also performed an estimation for a Sodium-cooled Fast Reactor pyroprocessing facility, and the costs associated with different aspects can be seen in Table 3.

Table 3: Direct cost details for process systems and main process building [54] in 2009 USD.



Cost components		Cost (k$)
Process systems	SFR spent fuel receiving and preparation system	6659
	SFR spent fuel head-end & pyroprocess system	76,890
	Maintenance cell system for SFR spent fuel head-end & pyroprocess cell	9119
	Radwaste treatment cell system for SFR spent fuel head-end & pyroprocess cell	7403
	Fuel rod fabrication system	36,318
	Maintenance cell system for fuel rod fabrication cell	7304
	Radwaste treatment cell system for fuel rod fabrication cell	6919
	Fuel assembling system	19,338
	Maintenance cell system for fuel assembling cell	5742
	Radwaste treatment cell system for fuel assembling cell	5467
	Chemical cell system	10,626
	Warm cell system	1106
	HLW/ILW storage cell	6303
	Total	199,194
Main process building	Civil architecture	68,218
	Utilities	128,053
	Waste systems	22,850
	Analytical laboratory equipment	2925
	Fire protection, radiation protection and criticality safety	3083
	Safeguards and security	1931
	Industrial health and safety	8937
	Cold maintenance shop equipment	2671
	Total	238,668

Table 4 shows the unit costs throughout the PWR fuel cycle. Although the PHWR values shown in Table 4 would be of use to Canada, as CANDU reactors would have similar costs, the reprocessing of the PHWR is not included in this analysis, so it can be assumed the PWR processing values would be similar, and therefore would be $2300/kgHM [53].

Table 4: PWR fuel cycle costs [53].

Step (Unit)	Idaho National Laboratory (Shropshire et al., 2009)			Electric Power Research Institute (Machiels and Sowder, 2010)			Harvard University (Bunn et al., 2003)			OECD/NEA (OECD/NEA, 2006a)			The Korean Government (Ministry of Knowledge and Economy, 2012a, 2012b), Korea Atomic Energy Research Institute (KAERI, 2010)		
	Low	Nominal	High	Low	Nominal	High	Low	Nominal	High	Low	Nominal	High	Low	Nominal	High
PWR capital ($/kWe)	2600	3900	6500							1200	1600	1900			
PWR O&M[a], D&D[b] ($/kWe)	55	66	80												
SFR capital ($/kWe)	3400	4600	8500							1200	1900	2300			
SFR O&M, D&D ($/kWe)	60	70	85												
Uranium ($/kgU)	30	75	260	104	169	520				20	50	80			
Conversion ($/kgU)	5	10	15	5	10	20	4	6	8	3	5	8			
Enrichment ($/SWU)	85	110	135	90	150	210	50	100	150	80	100	120			
PWR fuel fabrication ($/kgHM)	200	250	300	150	200	250	150	250	350	200	250	300			
PHWR fuel fabrication ($/kgHM)	115	135	155												
SFR metal fuel fabrication ($/kgHM)	Included in SFR fuel pyroprocessing							1400	2600	5000	Included in SFR fuel pyroprocessing				
PWR interim storage ($/kgHM)	100	120	300		150		100	200	300	40 (fixed), 5 (per year)	50, 5	60, 5		477[*]	
PHWR interim storage ($/kgHM)	100	120	300											209[*]	
Cs–Sr decay storage ($/kg Cs–Sr)	10,000	22,500	35,000												
PWR fuel pyroprocessing ($/kgHM)	1800	2300	2700											781	
SFR fuel pyroprocessing ($/kgHM)	3000	6000	9000							1000	2000	2500		5511	
PWR spent fuel packing, disposal ($/kgHM)	450	750	1130	$200/kgHM for packing plus $2500/m^3 for disposal										691[*]	
PHWR spent fuel packing, disposal ($/kgHM)														418[*]	
HLW conditioning, packing, disposal ($/kg FP)	3825	11,500	14,725							100,600 ($/m^3)	201,200 ($/m^3)	402,000 ($/m^3)			

[a] O&M (operation and maintenance).

[b] D&D (decommissioning and decontamination).

[*] From the Korean government.

2.1.5 Head-end Processes

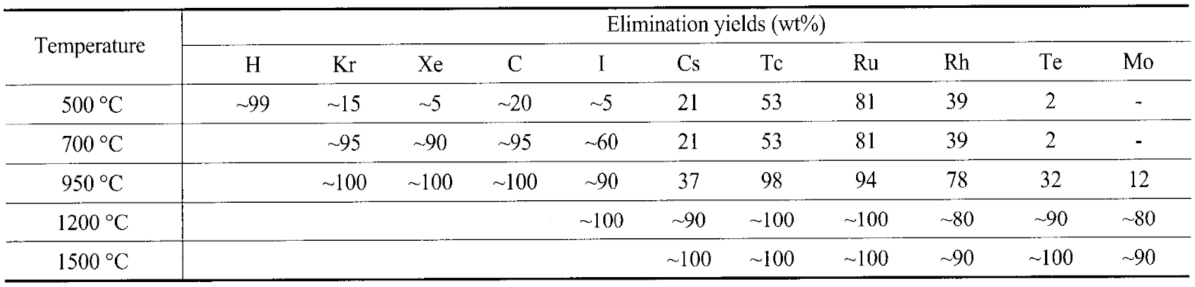
Before any reprocessing can begin, the fuel assemblies or bundles must be dismantled. PWR fuel assemblies are about 4m in length including instruments and nozzles. After the fuel assemblies are taken apart, the fuel rods are extracted and cut in the axial direction into shorter rods. The smaller fuel rods are decladded then transported for further processing. Common decladding practices involve either mechanical methods or voloxidation [33]. The stripped fuel cladding is sent to treatment and disposal, while the UO_2 is sent to the next process.

Mechanical decladding was developed for use with the DUPIC reprocessing method. The efficiency for fuels with a burnup less than 35 GWd/tU is 99%, but decreased to 74% for fuels with 60 GWd/tU [41]. Therefore, for fuels with higher burnup, a decladding method needs to be developed to achieve a higher efficiency. Oxidative decladding achieved higher efficiencies when paired with a rotational device [41].

2.1.5.1 Voloxidation Pretreatment

By exposing the uranium dioxide fuel to oxygen, the fuel oxidizes and becomes U_3O_8. In preparation for electroreduction, for pyroprocessing, the U_3O_8 is transformed to a powder to increase the surface area and thus improve the rate of the reaction. Oxidation reduces the density of the fuel from 11 g/cm^3 to 8.3 g/cm^3, therefore the volume is increased. While the uranium is being oxidized, some of the fission products and metal elements are transformed to volatile oxides and are sent to the off-gas treatment system. As shown in Table 5, as the temperature of the system increases, the yields are higher, but the sintering of the powder occurs at 1000^{o}C [38].

Table 5: Elimination Yields of the Fission Products in the Voloxidation Process [38].



Temperature	Elimination yields (wt%)										
	H	Kr	Xe	C	I	Cs	Tc	Ru	Rh	Te	Mo
500 °C	~99	~15	~5	~20	~5	21	53	81	39	2	-
700 °C		~95	~90	~95	~60	21	53	81	39	2	-
950 °C		~100	~100	~100	~90	37	98	94	78	32	12
1200 °C					~100	~90	~100	~100	~80	~90	~80
1500 °C						~100	~100	~100	~90	~100	~90

Implementing voloxidation as a pre-treatment for reprocessing methods can be considered to prevent contamination of products and machinery involved in reprocessing technologies. The off-gas treatment which follows the capture of the fission product from voloxidation includes a vacuum which effectively traps fission products such as: Cs using a fly ash filter, Tc by a calcium-based filter, and I by a silver-impregnated filter [41].

2.1.6 Economic Comparison

The Harvard Study looked at the comparison between direct disposal or reprocessing for LWR fuels, to determine which is more financially viable. The items included in the cost comparison are listed in Table 6.

Table 6: Cost items included in the Harvard Study cost comparison after on-site storage.

Direct Disposal	Reprocessing
Interim Storage	Transport to Reprocessing
Transport to Repository	Reprocessing Method
Waste Conditioning	Disposal of Reprocessing Wastes
Disposal	Recycle of Recovered U and Pu*

*recycling includes a credit for the reduction in fuel requirements.

The amount of savings from the recycled fuel is determined by the price of fresh uranium. Therefore, an increase in the price of uranium will decrease the difference in cost between Direct Disposal and Reprocessing [20]. Since the price of uranium is increasing still, there is a significant chance that the breakeven price of uranium will be surpassed before the NWMOs waste management plan, Adaptive Phase Management (APM), reaches Phase 3.

Before a formal recommendation is made, the NWMO needs to explore the FVM in Canada extensively; for example, appropriate uranium extraction efficiency, burnup of recycled fuel within a CANDU lattice (using WIMS), and further steps required for the use of plutonium ash [20].

Although certain countries, such as the US, have been reprocessing to reclaim fissile materials (e.g. Pu^{239}) for nuclear weapons, reprocessing could have positive global impacts as well [55]. The spent fuel from all of the nuclear power plants in the US are expected to have an inventory of $7.71*10^4$ metric tons of heavy metal (tHM), with a total activity of $3.46*10^4$ MCi. Although the spent fuel volume is a small amount of High Level Waste (HLW), it results in most (>95%) of the activity. Since global safeguarding is essential, all grades of plutonium would require protection [55].

2.2 Transmutation

Nuclear transmutation is the deliberate conversion of one chemical element or isotope into another. Transmutation can be achieved through nuclear reactions. Transuranic elements are the elements that are greater than 92, which is the atomic number for uranium; these elements are unstable and decay to other elements so their transmutation is of interest. Also, the transmutation of minor actinides, fission products, and fissile material within spent fuel can change the properties of the fuel to something more desired. Minor actinides can be transmuted to fission products, which although are more radioactive, decay more quickly. Long-lived fission products can be transmuted to isotopes that are more short-lived, and may even release energy to generate power [10].

One of the advantages of nuclear transmutation is the reduction of radiotoxicity; which reduces the engineering requirements for the repositories, simplifying designs, which could help increase public acceptance. Radiotoxicity is defined as:

$$Toxicity(i,k) = \frac{A(i)}{DAC(i,k)} \qquad \text{(Eq. 4)}$$

where A(i) is the activity of isotope i (Bequerel), and DAC(i,k) is the derived air concentration of isotope i in air or water, index k (Bequerel). If more than one isotope is involved, a summation is performed over all isotopes present in the mixture [92]. The heat production is reduced, which will alter the design of the packaging and therefore change the total capacity of the facility. Lastly, the volume of high-level waste is reduced [6]. Transmutation of nuclear isotopes is only perceivable if total radiotoxicity of nuclides being sent to the geological repository is considerably reduced. Therefore, the reinsertion of nuclides is only justified if the radiotoxicity production rate does not exceed the elimination rate [26].

2.2.1 Fission Products: Actinides

Fission Products are the result of the fission of a heavy atom and are generally unstable. Depending on the fission product, the isotope begins to decay and transforms into daughter isotopes. Fission products may have high activities (Cs^{137} and Sr^{90}) and long half-lives (Tc^{99}, $2.0*10^5$ years; I^{129},$1.6*10^7$ years), but actinides and their daughter products are liable for most of the radiotoxicity after 500 years of disposal (Figure 8). Pu^{239} dominates the radiotoxicity after several hundred years, followed by Np^{237} which have half-lives of $2.41*10^4$ and $2.14*10^6$ years, respectively [55].

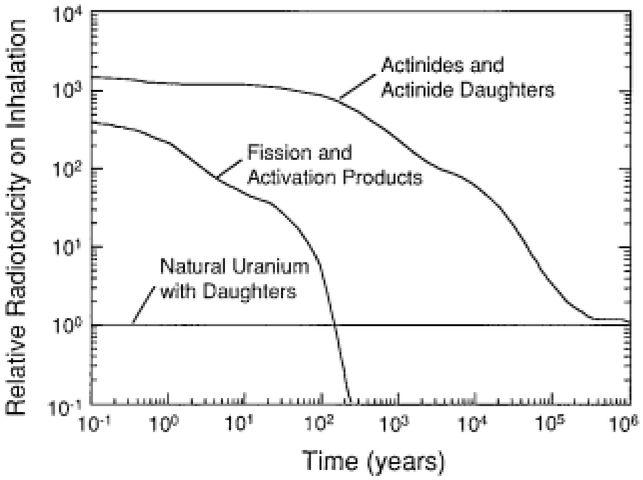


Figure 8: Relative radiotoxicity on inhalation of spent nuclear fuel with a burnup of 38 MWd/kgU [55].

2.2.2 CANDU Experience

There are multiple factors to consider when selecting the source of CANDU spent fuel contents. There are two sources that have been selected to ensure consistency throughout the calculations performed in future sections. One source, Waysywich 1993 [28], provides the isotopic content of

CANDU spent fuel after one year of cooling (Table 7). The sum of the actinides shown within Table 7 includes Cm, Am, Pu, and U, therefore the remaining actinides have a total weight of 1803.45g/Mg HM. Similarly, the fission products Cs and Sr are included in the shown mass of the fission products, therefore the remaining fission products have a total mass of 6844g/Mg HM.

Table 7: Principal isotopic content of one Metric Ton of CANDU fuel after cooling for one year [28, 29].

Name of Isotope	weight (g/Mg HM)	Half-Life (y)	Specific Activity (TBq/g)
Cm-242	0.145	$4.38*10^{-1}$	$1.2*10^{2}$
Cm-244	0.145	$1.80*10^{1}$	3
Am-241	11.06	$1.32*10^{2}$	$1.3*10^{-1}$
Pu-239	2660	$2.41*10^{4}$	$2.3*10^{-3}$
Pu-240	1040	$6.50*10^{3}$	$8.4*10^{-3}$
Pu-241	175.8	$1.40*10^{1}$	3.8
Pu-242	51.4	$3.73*10^{5}$	$1.5*10^{-4}$
U-235	2258	$7.04*10^{8}$	$8.0*10^{-8}$
U-238	9.85E5	$4.47*10^{9}$	$1.2*10^{-8}$
Actinides (total)	9.93E5	--	--
Cs-137	262	$3.02*10^{1}$	3.2
Sr-90	112	$2.89*10^{1}$	5.1
Fission Products (total)	7218	--	--

The other source of CANDU spent fuel contents comes from the NWMO [47], specifically a CANDU used fuel bundle from either Bruce or Darlington stations which has an initial mass U/bundle of 19.25kg, aged for a total of 30 years (e.g. 10 in wet, 20 in dry). Although it was initially assumed the burnup was 220 MWh/kgU, it was later realized that this is an overestimation, and therefore it is assumed that the burnup was the known average of 175 MWh/kgU, or 7.5MWd/kgU. The weight, half-life, and specific activity of the radionuclides of interest are listed in Table 8.

Table 8: Isotopic inventory of spent UO2 CANDU fuel after 30 years of cooling [29, 47].

Name of Isotope	Weight (g/kg U initial)	Half-Life (Years)	Specific Activity (TBq/g)
Ac-225	$4.24*10^{-12}$	$2.74*10^{-2}$	$2.1*10^{3}$
Ac-227	$3.57*10^{-9}$	$2.18*10^{1}$	2.7
Am-241	$2.78*10^{-1}$	$4.33*10^{2}$	$1.3*10^{-1}$
Bi-210	$1.11*10^{-15}$	$1.37*10^{-2}$	$4.6*10^{3}$
C-14	$1.01*10^{-4}$	$5.70*10^{3}$	$1.6*10^{-1}$
Cl-36	$2.00*10^{-4}$	$3.01*10^{5}$	$1.2*10^{-3}$
Cs-135*	$3.61*10^{-2}$	$2.30*10^{6}$	$4.3*10^{-5}$
Cs-137*	$1.76*10^{-1}$	$3.01*10^{1}$	3.2
I-129*	$5.45*10^{-2}$	$1.57*10^{7}$	$6.5*10^{-6}$
Np-237	$4.05*10^{-2}$	$2.14*10^{6}$	$2.6*10^{-5}$
Pa-231	$8.82*10^{-6}$	$3.28*10^{4}$	$1.7*10^{-3}$
Pa-233	$1.37*10^{-9}$	$7.39*10^{-2}$	$7.7*10^{2}$
Pb-210	$1.81*10^{-12}$	$2.22*10^{1}$	2.8
Pd-107*	$7.38*10^{-2}$	$6.50*10^{6}$	$1.9*10^{-5}$
Po-210	$3.07*10^{-14}$	$3.79*10^{-1}$	$1.7*10^{2}$
Pu-239	2.68	$2.41*10^{4}$	$2.3*10^{-3}$
Pu-240	1.28	$6.56*10^{3}$	$8.4*10^{-3}$
Pu-242	$1.03*10^{-1}$	$3.74*10^{5}$	$1.5*10^{-4}$
Ra-223	$5.00*10^{-12}$	$3.13*10^{-2}$	$1.9*10^{3}$
Ra-224	$2.46*10^{-10}$	$1.00*10^{-2}$	$5.9*10^{3}$
Ra-225	$5.54*10^{-12}$	$4.08*10^{-2}$	$1.5*10^{3}$
Ra-226	$5.32*10^{-10}$	$1.60*10^{3}$	$3.7*10^{-2}$
Ra-228	$1.91*10^{-10}$	5.75	$1.0*10^{1}$
Rn-222	$3.42*10^{-15}$	$1.05*10^{-2}$	$5.7*10^{3}$
Sb-126	$3.10*10^{-10}$	$3.38*10^{-2}$	$3.1*10^{3}$

Se-79*	$1.39*10^{-3}$	$2.95*10^{5}$	$2.6*10^{-3}$
Sm-147	$9.63*10^{-2}$	$1.06*10^{11}$	$8.5*10^{-1}$
Sm-151*	$2.20*10^{-3}$	$9.00*10^{1}$	$9.7*10^{-1}$
Sn-126*	$6.53*10^{-3}$	$2.30*10^{5}$	$1.0*10^{-3}$
Sr-90*	$6.80*10^{-2}$	$2.88*10^{1}$	5.1
Tc-99*	$2.38*10^{-1}$	$2.11*10^{5}$	$6.3*10^{-4}$
Th-227	$8.22*10^{-12}$	$5.11*10^{-2}$	$1.1*10^{3}$
Th-228	$4.78*10^{-8}$	1.91	$3.0*10^{1}$
Th-229	$1.10*10^{-6}$	$7.34*10^{3}$	$7.9*10^{-3}$
Th-230	$3.76*10^{-6}$	$7.54*10^{4}$	$7.6*10^{-4}$
Th-231	$6.80*10^{-12}$	$2.91*10^{-3}$	$2.0*10^{4}$
Th-232	$4.86*10^{-1}$	$1.41*10^{10}$	$4.0*10^{-9}$
Th-234	$1.43*10^{-8}$	$6.60*10^{-2}$	$8.6*10^{2}$
U-233	$8.41*10^{-3}$	$1.59*10^{5}$	$3.6*10^{-4}$
U-234	$4.89*10^{-2}$	$2.46*10^{5}$	$2.3*10^{-4}$
U-235	1.70	$7.04*10^{8}$	$8.0*10^{-8}$
U-236	$8.26*10^{-1}$	$2.34*10^{7}$	$2.4*10^{-6}$
U-238	$9.82*10^{2}$	$4.47*10^{9}$	$1.2*10^{-8}$
Y-90	$1.77*10^{-5}$	$7.30*10^{-3}$	$2.0*10^{4}$

*Isotopes of interest [26]

2.2.2.1 Transmutation of Actinides in CANDU

The CANDU reactor already provides a used nuclear fuel solution for the LWR with the DUPIC fuel cycle. The CANDU design is adaptable to actinide transmutation as well, with minimal reprocessing. Since CANDU uses heavy water as the moderator, there is a high neutron economy; therefore, a high TRU destruction rate can be achieved as there are fewer neutrons being absorbed in the moderator. Also, the CANDU on-line refueling feature allows for the actinides to be placed in desired locations with the exposure time to be controlled and manipulated separately from the regular fuel bundles [60].

Am and Cm were chosen for reinsertion into the CANDU using multiple methods due to their long presence in spent fuel. One has Am and Cm ranging from 5-60% volume in the centre pin of a fuel bundle. One method uses recycled uranium fuel throughout the CANDU core with an enrichment of 0.9%, while 14-25wt% of Am/Cm is placed in 30 periphery channels. Figure 9 shows the pathways for Am to transmute.

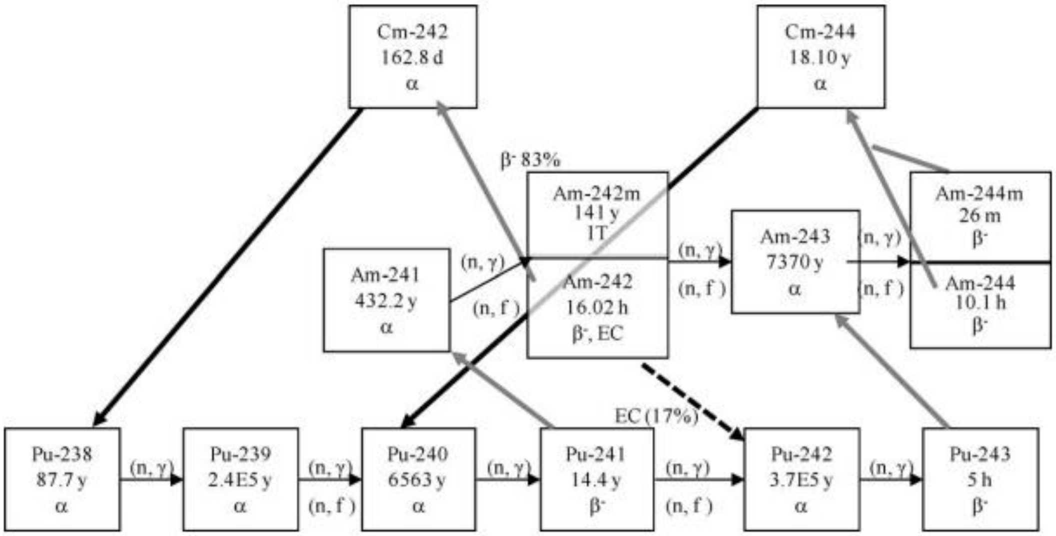


Figure 9: Two main transmutation pathways of Am-241 [60].

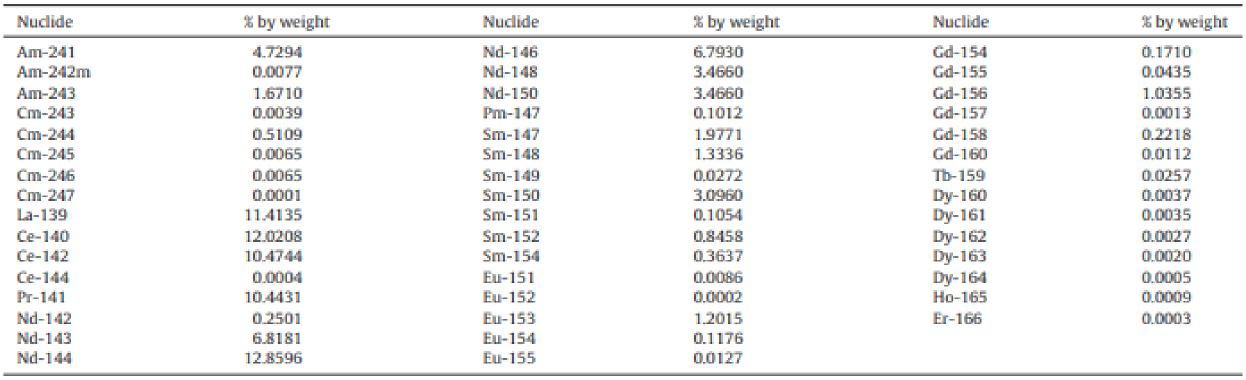
As one could determine from Figure 9, there are a few different ways to achieve the transmutation of Am-241. The pathways affect the decay heat of the spent fuel, and result in the Cm and Pu production. Initially, Am-241 captures a neutron and creates either Am-242 or Am-242m. From there, more transmutation options are available. Am-241m has a high fission-cross section, so transmutation would easily be achieved through this path. Am-242 beta decays to Cm-242 which then alpha decays to Pu-238, which will result in the eventual fission of Pu-239. Am-242m could also capture a neutron to Am-243, which would capture another neutron to become either Am-244m or Am-244, both of which have short half-lives and beta decay to Cm-244. Cm-244 has a relatively short half-life, and decays to Pu-240. The contributors to decay heat of spent fuel are Cm-244, Cm-244, and Pu-238; therefore, the transmutation of these isotopes would be beneficial for post-reactor fuel handling.

The fissile isotopes shown in Figure 9 are Am-242m, Cm-244, Pu-239, and Pu-241. The other isotopes would act as a poison and capture neutrons, resulting in the reduction in the coolant void reactivity of the bundle in which these isotopes are located. Although, even through the capture of neutrons from non-fissile isotopes, plutonium could still be produced, resulting in future fission. Any creation of Cm within the process is relatively short lived, and thus could be placed into short-term storage instead of long-term, or be transmuted further [60].

Heterogeneous Target Pin

In order to reduce fuel handling before the reinsertion of Am and Cm, the isotopes were not separated from the lanthanides during reprocessing (Table 2.9). Therefore, the material of interest (Am/Cm/Ln) was combined with an inert matrix (ZrO_2). The new zirconia material was placed in the centre pin of a CANDU flexible (CANFLEX) fuel bundle, the remaining pins contained Low Enriched Uranium (LEU) (1.0%), then were irradiated in a CANDU 6 reactor simulation (Figure 10). The fuel initially was enriched to 4% with an exit burnup of 50MWd/kgIHE and was cooled for 10 years prior to reprocessing. As aforementioned, the amount of Am/Cm/Ln varied from 5-60% by volume. In order to achieve greater destruction of Am and Cm, a demountable fuel bundle was used, so after the first irradiation, the centre pin would be removed and reinserted with fresh fuel in the remaining pins [60].

Table 9: Isotopic composition of centre pin, containing Am, Cm, and Ln [60].



Nuclide	% by weight	Nuclide	% by weight	Nuclide	% by weight
Am-241	4.7294	Nd-146	6.7930	Gd-154	0.1710
Am-242m	0.0077	Nd-148	3.4660	Gd-155	0.0435
Am-243	1.6710	Nd-150	3.4660	Gd-156	1.0355
Cm-243	0.0039	Pm-147	0.1012	Gd-157	0.0013
Cm-244	0.5109	Sm-147	1.9771	Gd-158	0.2218
Cm-245	0.0065	Sm-148	1.3336	Gd-160	0.0112
Cm-246	0.0065	Sm-149	0.0272	Tb-159	0.0257
Cm-247	0.0001	Sm-150	3.0960	Dy-160	0.0037
La-139	11.4135	Sm-151	0.1054	Dy-161	0.0035
Ce-140	12.0208	Sm-152	0.8458	Dy-162	0.0027
Ce-142	10.4744	Sm-154	0.3637	Dy-163	0.0020
Ce-144	0.0004	Eu-151	0.0086	Dy-164	0.0005
Pr-141	10.4431	Eu-152	0.0002	Ho-165	0.0009
Nd-142	0.2501	Eu-153	1.2015	Er-166	0.0003
Nd-143	6.8181	Eu-154	0.1176		
Nd-144	12.8596	Eu-155	0.0127		

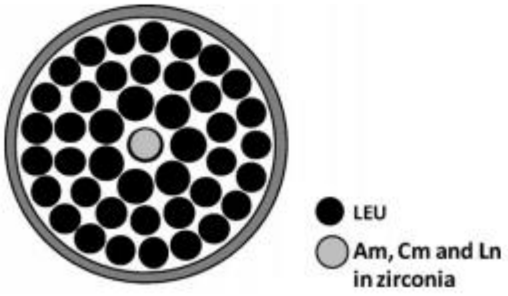
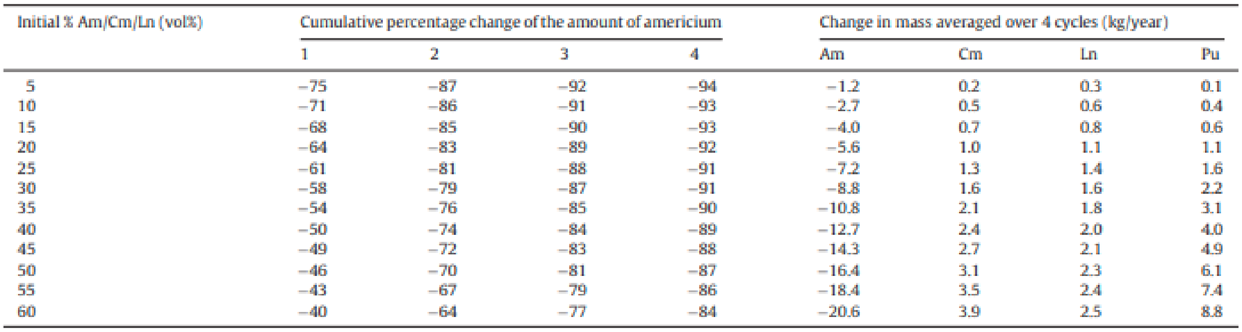


Figure 10: CANFLEX fuel bundle design with actinide target [60].

After each irradiation, the exit burnup would increase, and the Coolant Void Reactivity (CVR) reduction would decrease. Throughout the reactor, different fuel bundles would be in different irradiation stages in order to average the effects of the CVR reduction reactor-wide [60]. The cumulative percentage change of Am and the mass change of Am, Cm, Ln, and Pu per year is shown in Table 10.

Table 10: Transmutation percent of Am, average mass of Am, Cm, and Ln per year averaged over the four cycles [60].



Initial % Am/Cm/Ln (vol%)	Cumulative percentage change of the amount of americium				Change in mass averaged over 4 cycles (kg/year)			
	1	2	3	4	Am	Cm	Ln	Pu
5	−75	−87	−92	−94	−1.2	0.2	0.3	0.1
10	−71	−86	−91	−93	−2.7	0.5	0.6	0.4
15	−68	−85	−90	−93	−4.0	0.7	0.8	0.6
20	−64	−83	−89	−92	−5.6	1.0	1.1	1.1
25	−61	−81	−88	−91	−7.2	1.3	1.4	1.6
30	−58	−79	−87	−91	−8.8	1.6	1.6	2.2
35	−54	−76	−85	−90	−10.8	2.1	1.8	3.1
40	−50	−74	−84	−89	−12.7	2.4	2.0	4.0
45	−49	−72	−83	−88	−14.3	2.7	2.1	4.9
50	−46	−70	−81	−87	−16.4	3.1	2.3	6.1
55	−43	−67	−79	−86	−18.4	3.5	2.4	7.4
60	−40	−64	−77	−84	−20.6	3.9	2.5	8.8

The negative values shown in Table 10 represent a reduction in mass; although there is a large decrease in mass of Am, the mass of Cm increases, as Am transmutes to Cm. As expected, the inventory of lanthanides increases, as they do not fission, but are fission products. Specifically, as the actinides fission, more lanthanides are being produced. More Pu is being produced as the transmutations occur, as expected, which occurs mostly in the first cycle. When there is a low inventory of actinides and lanthanides (5-10%), there is a large destruction of Pu in cycles 2-4; for 15-45% volume there is destruction of Pu in cycles 3 and 4; for 50-60% initial Am/Cm/Ln/Pu destruction occurs in cycle 4 [60]. As shown in Table 10, comparing the initial mass of Am/Cm/Ln at 5% and 60% has significant differences in the change in mass for Am, at 1.2kg/year and 20.6kg/year, respectively.

Heterogeneous Target Channel

The following scenario has a CANDU 6 reactor being fuelled with 0.9%RU, with 30 peripheral channels around the outside of the reactor filled with an Am/Cm inert matrix (Figure 11). The wt% of Am/Cm in the centre pin of the target channels are provided from a Takahama-3 reactor with an exit burnup of 47.03GWd/MTIHE with a 30-year decay period, the Am and Cm were removed from the rest of the isotopes.

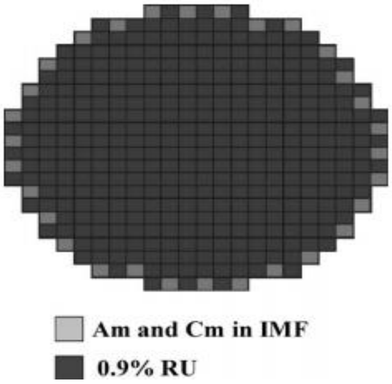


Figure 11: Schematic diagram of Am/Cm target model of a CANDU 6 core [60].

The channels were selected for transmutation due to their minimal effect on reactor operation. The wt% of Am/Cm inert matrix fuel (IMF) are 14%, 19%, 26%, and 35%, with SiC used as the IMF material. The selection of an IMF is due to the transparency to neutrons from a physics perspective [60].

Full Core Time-Average calculations were used to monitor maximum channel and bundle powers, with maximums of 6660kW and 790kW, respectively. The refuelling ripple, or power increase upon refueling, for the channels and bundles was monitored using an instantaneous model, with maximums of 7080kW and 845kW, respectively [60].

Four different fuel bundles were modelled for the carrier of the actinides, with each bundle having a different mass of heavy elements. The destruction of americium using the 30-peripheral channel scheme does not depend on the fuel bundle, but the support ratio (Figure 12). For example, a support ratio of 4 means an LWR that provides 4 GWe produces enough americium for 1GWe of CANDU 6. A lighter bundle would have a shorter residence time to achieve the same percentage of transmutation than with a heavier bundle, as there is less Am/Cm in the former. For any bundle with a given Am/Cm initial loading, more transmutation of that bundle would occur if the bundle were to sit in the reactor for longer. However, if the bundle is in the reactor for longer, the throughput of Am would be lower, thus lowering the ratio.

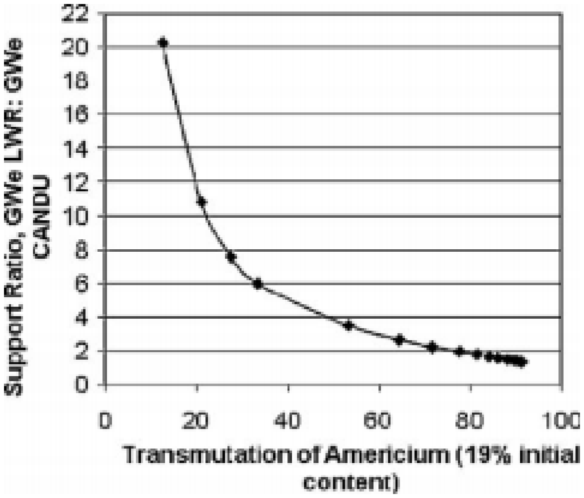


Figure 12: Relationship between the support ratio and the transmutation for the CANFLEX bundle with 19% initial concentration of Am/Cm by volume [60].

There is an inverse relationship between the support ratio and the destruction of Am; if a high support ratio is desired, a low destruction rate is obtained. If a higher destruction of Am is desired, more GWe of CANDU is required. Once-through applications can use support ratios to indicate how much of minor actinides is required in various scenarios. For a chosen destruction rate and support ratio, the residence time can be chosen for either a lighter or heavier fuel bundle design [60].

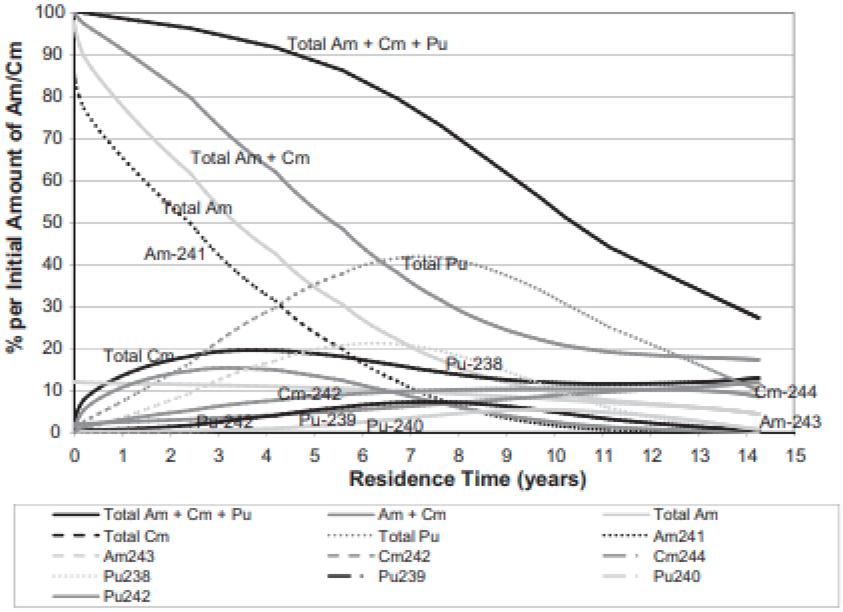


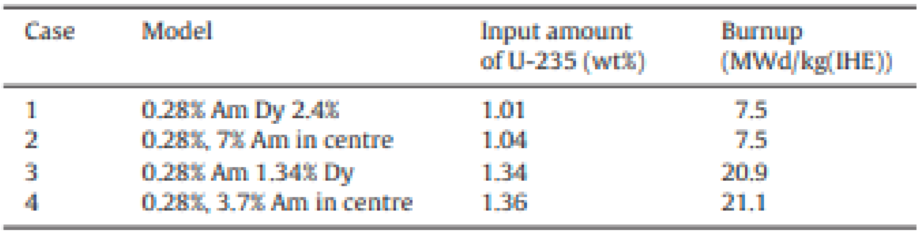
Figure 13: Isotopic evolution of the significant transuranic nuclides in the Am/Cm fuel for the 21-element fuel bundle with an initial composition of 19% Am/Cm volume [60].

There is an initial increase in plutonium from the decay of Cm-242 and from the electron capture of Am-242. The isotopic composition is shown in Figure 13 for the 21-element fuel bundle with a 19% Am/Cm bundle; other combinations would have the same trends, with lengthening axes. If minimal operational change is desired, the 30-peripheral channel method would be optimal. The power contribution of the outer channels is small to begin with, so altering their compositions would not affect the entire core's operation. Also, instead of the neutrons being lost from the reactor, these channels would optimize their interactions with transmutation. However, in this scenario, the transmutation is quite low at less than 10%. The transmutation rate per year was achieved for the 21-element bundle, shown in Figure 13. Higher transmutation would be achieved if the support ratio were reduced for all of the bundles [60].

Homogeneous Full Core

The following method only has americium separated from the rest of the actinides. The weights of the americium isotopes Am-241, Am-242m, and Am-243, are 73.8%, 0.12%, and 26.1%, respectively. The Am isotopes are mixed with recycled uranium (RU). RU is used because Am acts as a neutron poison, so additional fissile material was required to compensate for reactivity loss. Two burnups were used (7.5MWd/kgIHE, 21MWd/kIHE), along with the CANFLEX fuel bundle design. The four cases that were performed are shown in Table 11.

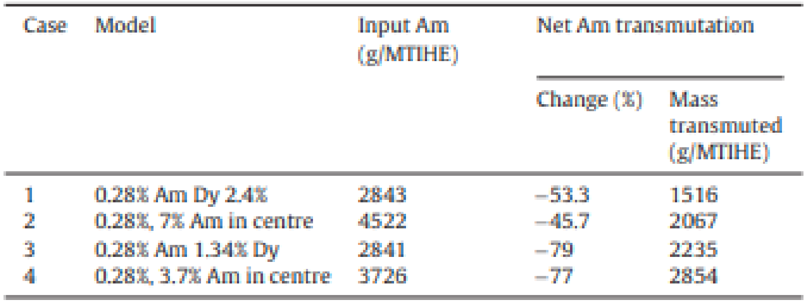
Table 11: Parameters for the Am/LEU fuel [60].



Case	Model	Input amount of U-235 (wt%)	Burnup (MWd/kg(IHE))
1	0.28% Am Dy 2.4%	1.01	7.5
2	0.28%, 7% Am in centre	1.04	7.5
3	0.28% Am 1.34% Dy	1.34	20.9
4	0.28%, 3.7% Am in centre	1.36	21.1

Each case has the neutron poison (Am) present in the centre pin of the fuel bundle to lower the CVR. The results for each of the cases in Table 11 are presented in Table 12 below. The cases with the most Am in the centre pin, 2 and 4, have the most Am transmutation of 2067g/MTIHE and 2854MTIHE, respectively. Although the transmutation of the Am is the most reduced, the transmutation fractions are only 45.7% for 7% Am and 77% for 3.7% Am. Therefore, the reduction in fraction of Am transmutation is less significant at higher burnups (79% and 77%).

Table 12: Results for Am/LEU [60].



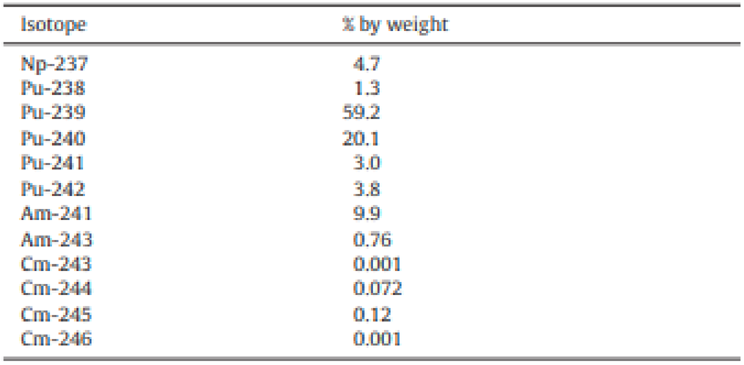
Case	Model	Input Am (g/MTIHE)	Net Am transmutation	
			Change (%)	Mass transmuted (g/MTIHE)
1	0.28% Am Dy 2.4%	2843	−53.3	1516
2	0.28%, 7% Am in centre	4522	−45.7	2067
3	0.28% Am 1.34% Dy	2841	−79	2235
4	0.28%, 3.7% Am in centre	3726	−77	2854

The largest support ratio comes from case 2, with 20.7; however, the Am transmutation change is the lowest at 45.7%. The higher transmutation fraction cases require up to 6.1GWe from a PWR to support [60].

Group-Extracted TRU-MOX in CANDU

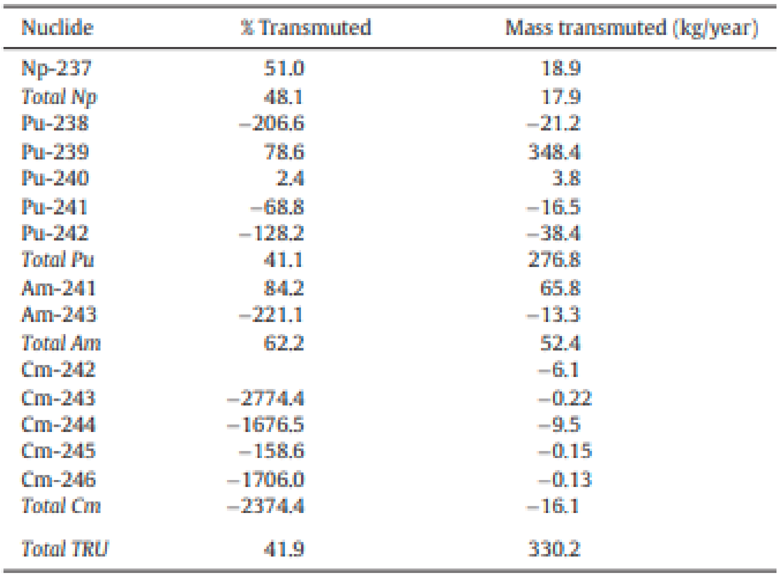
All transuranic elements, Pu, Np, Am, and Cm, were extracted from LWR spent fuel with their respective amounts. LWR fuel that had been cooled for 30 years, was then reprocessed to recover the TRUs. The TRUs were mixed with natural uranium to make a MOX fuel to be irradiated in a CANDU 6. Table 13 and Table 14 show the isotopic input of the TRU and the resulting transmuted isotopes.

Table 13: Isotopic composition of the transuranic nuclides used for the input [60].



Isotope	% by weight
Np-237	4.7
Pu-238	1.3
Pu-239	59.2
Pu-240	20.1
Pu-241	3.0
Pu-242	3.8
Am-241	9.9
Am-243	0.76
Cm-243	0.001
Cm-244	0.072
Cm-245	0.12
Cm-246	0.001

Table 14: Transuranic nuclides that are transmuted in the TRU-MOX method [60].



Nuclide	% Transmuted	Mass transmuted (kg/year)
Np-237	51.0	18.9
Total Np	48.1	17.9
Pu-238	−206.6	−21.2
Pu-239	78.6	348.4
Pu-240	2.4	3.8
Pu-241	−68.8	−16.5
Pu-242	−128.2	−38.4
Total Pu	41.1	276.8
Am-241	84.2	65.8
Am-243	−221.1	−13.3
Total Am	62.2	52.4
Cm-242		−6.1
Cm-243	−2774.4	−0.22
Cm-244	−1676.5	−9.5
Cm-245	−158.6	−0.15
Cm-246	−1706.0	−0.13
Total Cm	−2374.4	−16.1
Total TRU	41.9	330.2

The fuel used in this scenario was the 43-element CANFLEX design, with 4% TRU by weight; however, in the centre pin Dy in a zirconia matrix was used to reduce the CVR. The exit burnup was 43.4MWd/kgIHE, a support ratio of 11.2, with a total transmutation of 41.9% (Table 14). The high neutron economy in the CANDU reactor provides an effective environment for the destruction of the fissile nuclides. The initial fissile content of the TRU was 63%, and it dropped to 30%. There is a 40% reduction in decay heat of the used fuel at 1000 years when compared to LWR spent fuel, which would significantly impact the capacity of a geological repository [60].

The time average model revealed maximum channel and bundle powers to be 6300 and 820kW, respectively. The instantaneous model revealed maximum channel and bundle powers to be 7200kW and 910kW, respectively. For the instantaneous model, the values are higher, as expected, which accounts for the immediate power increase as the fresh fuel bundles are added to the core. All values are within normal CANDU operating conditions [60].

Conclusion of Methods

The methods were chosen to impose a minimal effect on the existing CANDU reactor's operation. The centre pin to carry the actinides was chosen as a method to mimic the "low void reactivity fuel" that has a centre neutron absorber and slightly enriched uranium in the remaining fuel pins. The heterogeneous model chose the peripheral channels to prevent any major impact on the reactor operation, while utilizing the neutrons in the designated areas. The online refueling of CANDU allows for a residence time to be easily manipulated, as desired. High neutron fluence allows a high destruction of the actinides, as only a small portion of the reactor is being dedicated to transmutation; however, the total throughput of actinides is then limited. The heterogeneous methods would also be able to work in unity with the LWR fleet [60].

The homogeneous methods achieved significant transmutation of actinide fuels, and achieved the order of several kilograms per year for one reactor. However, the fuel and the actinides are subject to the same neutron fluence, which reduces the destruction of actinides. Future reprocessing and transmutation would need to occur to achieve high destruction to compare to the heterogeneous methods [60].

2.2.3 PWR Experience

As most of the reactors that are currently in operation are PWRs, a lot of research has gone towards the PWR fuel cycle, and increasing its efficiency. Within PWRs, reactivity control relies on the insertion of control rods, and the injection of boron into the coolant. Boron absorbs neutrons, and by adjusting the concentration within the coolant affects the neutron activity.

About 0.4 wt% of the PWR spent fuel mass is composed of the long-lived fission products, cesium, strontium, technetium, and iodine; 1 wt% is composed of plutonium and minor actinides [65]. Three prominent minor actinides in the spent fuel, Np-237, Am-241, and Am-243, have large thermal neutron capture cross sections. Upon the absorption of a neutron, these isotopes become Pu-239, Am-242, Am-244, Cm-243 and Cm-245, all of which have large fission cross sections. Therefore, the insertion of the original minor actinides will not only transmute efficiently, but will also produce power. However, the initial presence of the minor actinides in the reactor would cause a loss of neutrons due to their large thermal capture cross sections, causing the initial reactivity to decrease; therefore, the minor actinides act as burnable poisons in PWRs [64].

Different core designs in which to incorporate minor actinides for transmutations would impact the reactors in many ways. Liu et al. (2014) investigated different core designs for the PWR for transmutation while monitoring the k_{eff} after minor actinide insertion. The k_{eff} is the reactivity coefficient, which identifies the criticality and whether the reactor can sustain chain reactions. The k_{eff} and the neutron flux was monitored after different minor actinides were added, as well as the transmutation rates of the minor actinides, and the inventory of the transuranic nuclides after a 300-day exposure [64]. A uniform distribution and a heterogeneous distribution are described in Section 2.2.3.1 and Section 2.2.3.2, respectively.

The loading of MOX fuel and minor actinides into a PWR has similar behaviours. Due to the large resonance integral of U-238 and Pu-240, a large negative doppler coefficient is applied when the PWR is in operation. Relative contents of U-238 and Pu-240 are smaller when minor actinides are added to the PWR core. It was decided that a 1% weight would not significantly alter the physics of the reactor core [64].

2.2.3.1 Homogeneous Distribution

The minor actinides that were inserted into the reactor uniformly are presented in Table 15, with their respective ratios from spent fuel. As shown in Table 15, the transmutation of Np-237 would greatly reduce the volume of the spent fuel globally. Table 16 shows the effect of the k_{eff} after the insertion of a homogeneous minor actinide distribution.

Table 15: The ratio of each MA nuclide in the depleted fuel of a PWR [64].

MA Nuclides	N-237	Am-241	Am-243	Cm-244	Cm-245
Ratio (%)	56.2	26.4	12	5.12	0.28

Table 16: Effects of minor actinide presence on keff [64].

Content of MA nuclides in nuclear fuel (%)	0	1	2	3
k_{eff}	1.00296	0.85349	0.75504	0.68492

Even a presence of 1% can impact the k_{eff} significantly. The consumption of neutrons is due to the presence of the isotopes with large thermal capture cross sections, such as Np-237, Am-241, and Am-243. Not only is the presence of the minor actinides in the fuel affecting the initial k_{eff} of the reactor, but it would also affect the lifetime of the fuel loading. The initial core fuel loading is often increased by inserting burnable absorption materials to compensate. The burnable poisons increase core lifetime without compromising control safety, control rod requirements are more relaxed, and core power distributions could also be improved [64].

The inclusion of minor actinides in the fuel within the early stages of the fuel cycle could potentially compensate for the early excess reactivity during the PWR operation. Theoretically, minor actinides can replace some burnable poisons during the beginning of the PWRs fuel cycle. However, due to the uniform distribution of the minor actinides, the initial change in reactivity would be very high as the minor actinides deplete, causing a reactivity mismatch. It was determined that the homogeneous distribution of a minor actinide - uranium dioxide mixture is not feasible.

2.2.3.2 Heterogeneous Distribution

A heterogeneous distribution of minor actinides would avoid the initial reactivity drop due to spatial self-shielding. The affective absorption cross section of the minor actinide rods that are heterogeneously distributed would be reduced, and thereby minimize the reactivity mismatch [64]. A heterogeneous distribution was achieved by simply replacing fuel rods within a fuel assembly with minor actinide transmutation rods (Figure 14). In this scenario, the transmutation rods were completely separate from the uranium fuel.

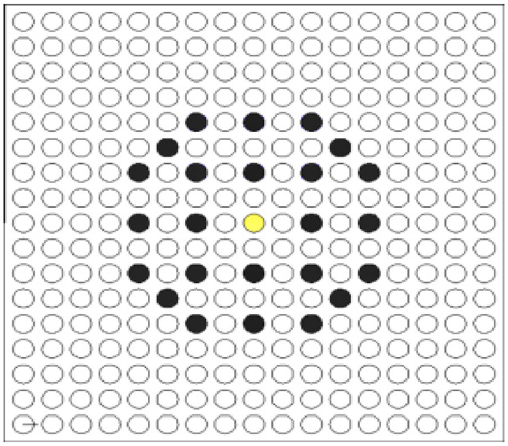


Figure 14: Transmutation rod distribution pattern in fuel assembly [64].

Upon the addition of the minor actinide rods, the k_{eff} was monitored, as shown in Table 17. As one could see, each minor actinide affects the k_{eff} differently when separated. If all of the minor actinides were mixed, there is a reduction in the k_{eff} with a total of 288 transmutation rods throughout the core. The total weight of minor actinides in the core is only 1% that of the fuel. When compared to the effects of a homogeneous minor actinide presence, there is much less (Table 16).

Table 17: keff after adding different MA nuclides to the PWR core [64].

MA Nuclides	Without MA	Np-237	Am-241	Am-243	Cm-244	Cm-245	Mixed MA
k_{eff}	1.00296	0.998636	0.98728	0.98713	1.00060	1.20007	0.98634

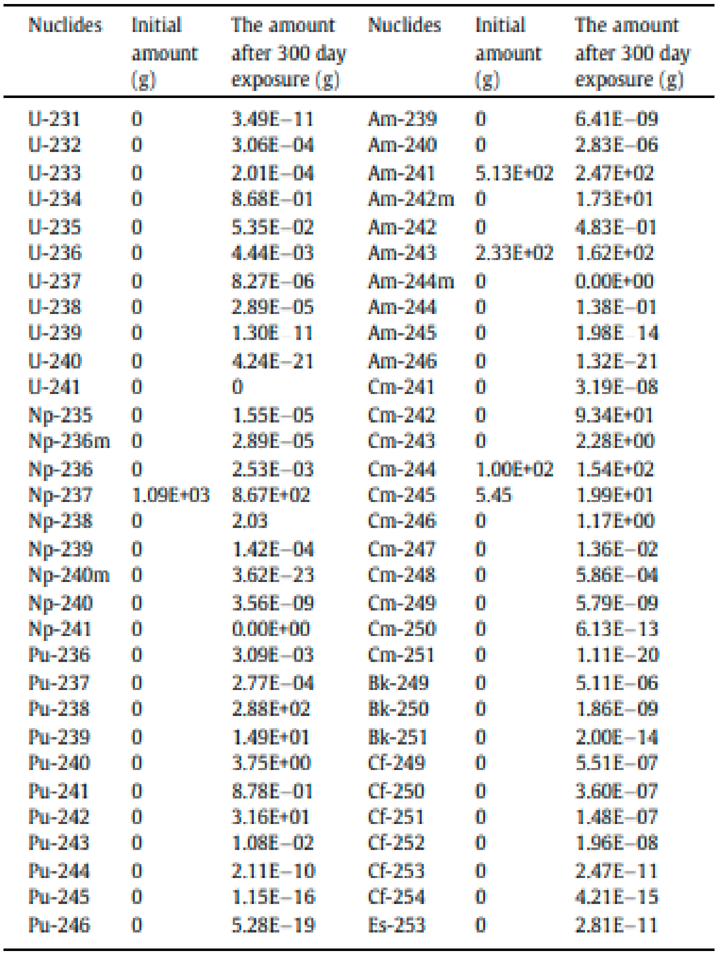
The reduction in impact indicates that the spatial self-shielding of the heterogeneous distributions plays an important role in the prevention of the reactivity drop. Also, the reactivity mismatch and control difficulties could be overcome by heterogeneous distribution of minor actinides [64]. One could conclude that the initial presence of minor actinides behave similarly to burnable poisons. Therefore, minor actinide materials could be used to partially substitute burnable poisons in PWRs [64].

The transmutation rate that was experienced by the minor actinides in the PWR is shown in Table 18 and all the residual actinides that are produced are shown in Table 19. The direct fission rate indicates that most of the minor actinides present were not transmuted by fission. After the 300-day exposure, the initial amount of 1941.45g of minor actinides is reduced to 1908g.

Table 18: Disappearance rate of MA after 300-day-exposure in PWR [64].

MA	Initial Amount (g)	Residual Amount (g)	Disappearance Rate	Direct Fission Rate
Np-237	1.09E+03	8.67E+02	20.5%	0.358%
Am-241	5.13E+02	2.47E+02	51.9%	0.609%
Am-243	2.33E+02	1.62E+02	30.5%	0.299%
Cm-244	1.00E+02	1.54E+02	-54%	/
Cm-245	5.45E+00	1.99E+01	-265%	/

Table 19: Residual nuclides after MA nuclide 300-day-exposure in PWR [64].



Nuclides	Initial amount (g)	The amount after 300 day exposure (g)	Nuclides	Initial amount (g)	The amount after 300 day exposure (g)
U-231	0	3.49E−11	Am-239	0	6.41E−09
U-232	0	3.06E−04	Am-240	0	2.83E−06
U-233	0	2.01E−04	Am-241	5.13E+02	2.47E+02
U-234	0	8.68E−01	Am-242m	0	1.73E+01
U-235	0	5.35E−02	Am-242	0	4.83E−01
U-236	0	4.44E−03	Am-243	2.33E+02	1.62E+02
U-237	0	8.27E−06	Am-244m	0	0.00E+00
U-238	0	2.89E−05	Am-244	0	1.38E−01
U-239	0	1.30E 11	Am-245	0	1.98E 14
U-240	0	4.24E−21	Am-246	0	1.32E−21
U-241	0	0	Cm-241	0	3.19E−08
Np-235	0	1.55E−05	Cm-242	0	9.34E+01
Np-236m	0	2.89E−05	Cm-243	0	2.28E+00
Np-236	0	2.53E−03	Cm-244	1.00E+02	1.54E+02
Np-237	1.09E+03	8.67E+02	Cm-245	5.45	1.99E+01
Np-238	0	2.03	Cm-246	0	1.17E+00
Np-239	0	1.42E−04	Cm-247	0	1.36E−02
Np-240m	0	3.62E−23	Cm-248	0	5.86E−04
Np-240	0	3.56E−09	Cm-249	0	5.79E−09
Np-241	0	0.00E+00	Cm-250	0	6.13E−13
Pu-236	0	3.09E−03	Cm-251	0	1.11E−20
Pu-237	0	2.77E−04	Bk-249	0	5.11E−06
Pu-238	0	2.88E+02	Bk-250	0	1.86E−09
Pu-239	0	1.49E+01	Bk-251	0	2.00E−14
Pu-240	0	3.75E+00	Cf-249	0	5.51E−07
Pu-241	0	8.78E−01	Cf-250	0	3.60E−07
Pu-242	0	3.16E+01	Cf-251	0	1.48E−07
Pu-243	0	1.08E−02	Cf-252	0	1.96E−08
Pu-244	0	2.11E−10	Cf-253	0	2.47E−11
Pu-245	0	1.15E−16	Cf-254	0	4.21E−15
Pu-246	0	5.28E−19	Es-253	0	2.81E−11

The transmutation of each individual rod was reduced due to the spatial self-shielding; only the actinides located on the surface are transmuted. Therefore, using entire rods with the sole purpose of transmutation can be deemed unfeasible [65].

2.2.3.3 Minor Actinide Burnable Poison Rods

Studies show that the cross sections for minor actinides are larger in thermal regions than in fast regions; therefore, minor actinide transmutation in thermal reactors may be very effective. All previous transmutation distributions require either a change in the fuel composition, the configuration of the reactor core, or the structure of the fuel rods. However, due to the well-established nature of the PWR, this is unlikely to be implemented.

Hu et al. (2015) examined the transmutation characteristics when loading the transmutable minor actinides in the burnable poison rods. In particular, the k_{eff} was monitored to try to find an optimal minor actinide mixture for effective transmutation and PWR operation [65].

The PWR core was simulated in a specific arrangement to observe the transmutation. The fuel was set to an enrichment of 2.1% and the boron concentration was kept low (1074 ppm) to ensure the burnable poison rods would experience most of the neutron interactions. The full core's distribution of burnable poison assemblies and burnable poison rods are shown in Figure 15 and Figure 16, respectively.

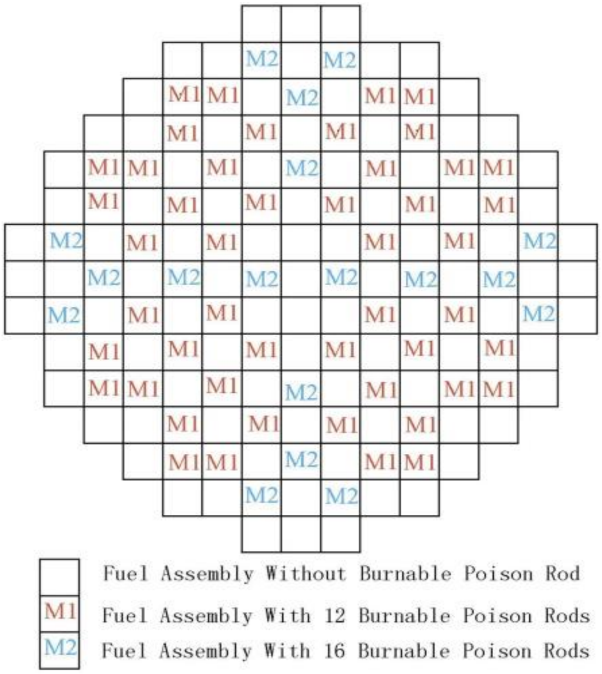


Figure 15: The arrangement of burnable poison rod assemblies in a PWR core [65].

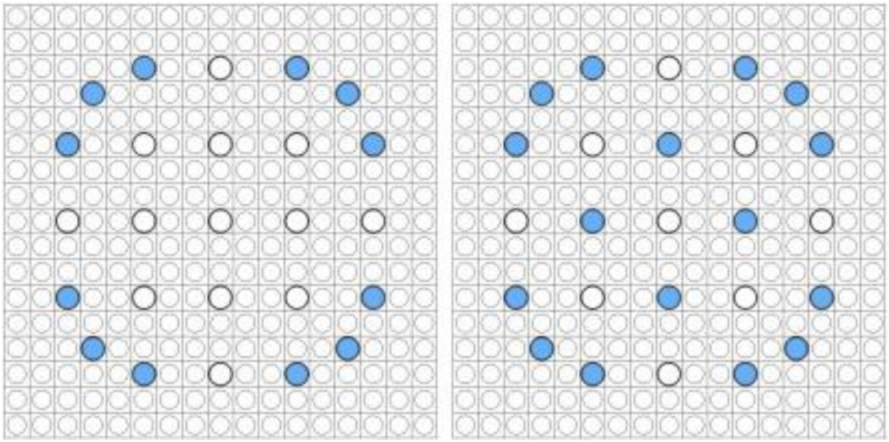


Figure 16: The distribution of 12 and 16 burnable poison rods in a fuel assembly [65].

Layered Burnable Poison Rods

The minor actinides to be inserted back into the reactor were set to have the same ratio that the nuclides from 3GWt, 33,000MWd/t spent fuel after 10 years of cooling, as shown in Table 20. To reduce the difficulty of implementation, only the burnable poison rods were redesigned to accommodate for the minor actinide presence. Figure 17 shows the burnable poison rod with and without the minor actinide sheath.

Table 20: Ratio of MA nuclides in MCNP simulation calculation [65].

Nuclides	Np-237	Am-241	Am-243	Cm-244	Cm-245
Ratio (%)	41.8	47.86	8.62	1.63	0.09

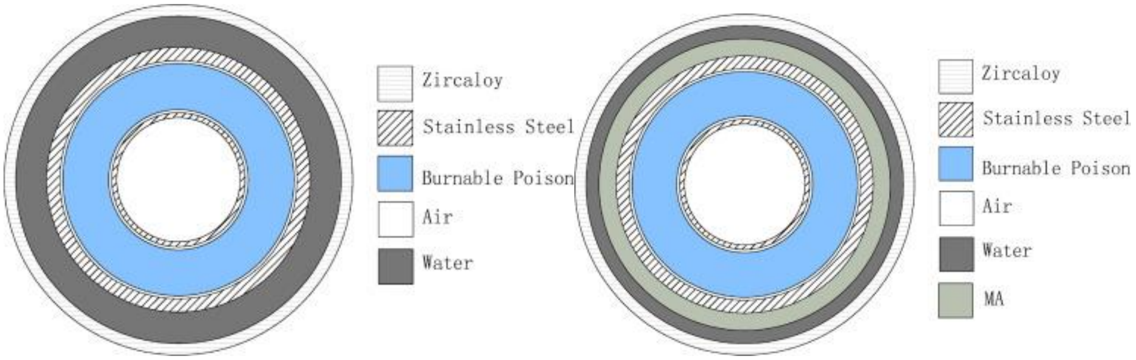
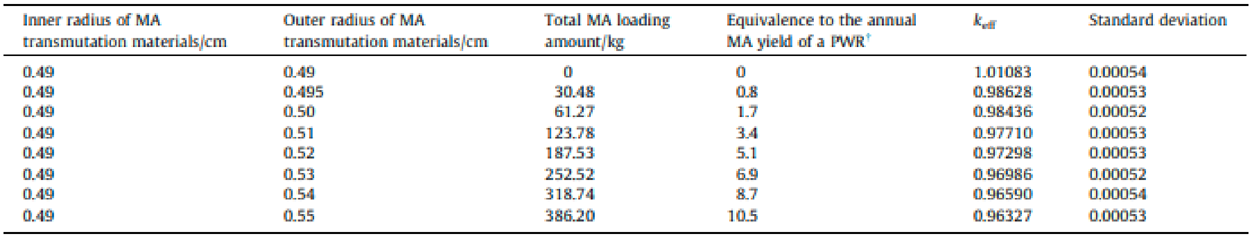


Figure 17: Burnable poison rod and transmutation rod cutaway [65].

The inner radius of the transmutation rod was set, and the outer radius was adjusted to alter the amount of minor actinides to be exposed. The resulting k_{eff} can be seen in Table 21. As the coating thickness increased, the PWR initial reactivity dropped correspondingly, as expected. The positive reactivity needs to be adjusted to compensate for the neutron absorption. Therefore, the enrichment of the fuel needs to be increased, the boron concentration needs to be reduced further, or some burnable poison rods need to be removed, in order to have the PWR return to criticality [65].

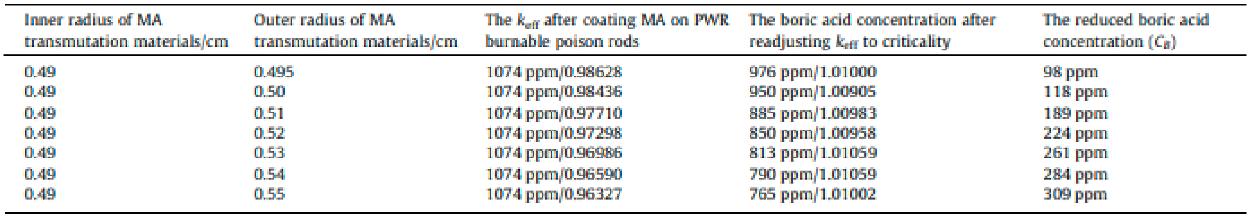
Table 21: Calculation results after coating MA transmutation materials in burnable poison rods [65].



Inner radius of MA transmutation materials/cm	Outer radius of MA transmutation materials/cm	Total MA loading amount/kg	Equivalence to the annual MA yield of a PWR†	k_{eff}	Standard deviation
0.49	0.49	0	0	1.01083	0.00054
0.49	0.495	30.48	0.8	0.98628	0.00053
0.49	0.50	61.27	1.7	0.98436	0.00052
0.49	0.51	123.78	3.4	0.97710	0.00053
0.49	0.52	187.53	5.1	0.97298	0.00053
0.49	0.53	252.52	6.9	0.96986	0.00052
0.49	0.54	318.74	8.7	0.96590	0.00054
0.49	0.55	386.20	10.5	0.96327	0.00053

The boric acid was adjusted to account for the large reduction in criticality [65]. The actual amount of boric acid was calculated by readjusting the concentration, allowing the PWR to return to criticality, and calculating the concentration again. As shown in Table 22, after the initial concentration of boric acid, 1074ppm, as the minor actinide thickness increases, the k_{eff} decreases, and the boric acid concentration is reduced. Therefore, adding minor actinides to a PWR would act as adding additional poisons to the reactor. The poisons that are currently present in the coolant would need to be reduced to keep the criticality, and to increase the negative temperature coefficient. The reduction of boric acid in the primary coolant can reduce the possibility of the critical accident in a fuel cycle, which is a distinct advantage [65].

Table 22: Partial substitution of minor actinides for boric acid [65].

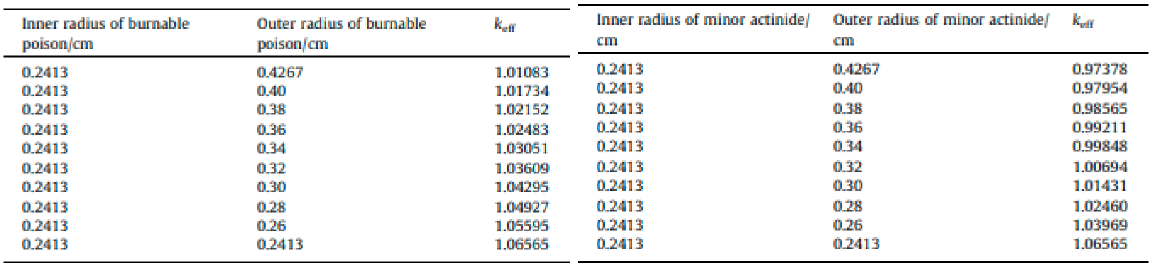


Inner radius of MA transmutation materials/cm	Outer radius of MA transmutation materials/cm	The k_{eff} after coating MA on PWR burnable poison rods	The boric acid concentration after readjusting k_{eff} to criticality	The reduced boric acid concentration (C_B)
0.49	0.495	1074 ppm/0.98628	976 ppm/1.01000	98 ppm
0.49	0.50	1074 ppm/0.98436	950 ppm/1.00905	118 ppm
0.49	0.51	1074 ppm/0.97710	885 ppm/1.00983	189 ppm
0.49	0.52	1074 ppm/0.97298	850 ppm/1.00958	224 ppm
0.49	0.53	1074 ppm/0.96986	813 ppm/1.01059	261 ppm
0.49	0.54	1074 ppm/0.96590	790 ppm/1.01059	284 ppm
0.49	0.55	1074 ppm/0.96327	765 ppm/1.01002	309 ppm

Complete MA substitution in Burnable Poison Rods

After comparing the neutron absorption of the burnable poison and the minor actinide material, the reactivity control ability of the two materials can be determined. Table 23 shows the effects of the burnable poison and the minor actinides on the k_{eff}. Clearly, when the rods contain minor actinides, the effect on the k_{eff} is less than the burnable poisons. Therefore, minor actinides can be used as burnable poisons; however, their excess reactivity is stronger than conventional burnable poisons [65].

Table 23: Effect on PWR keff of burnable poison and minor actinide material [65].



Inner radius of burnable poison/cm	Outer radius of burnable poison/cm	k_{eff}
0.2413	0.4267	1.01083
0.2413	0.40	1.01734
0.2413	0.38	1.02152
0.2413	0.36	1.02483
0.2413	0.34	1.03051
0.2413	0.32	1.03609
0.2413	0.30	1.04295
0.2413	0.28	1.04927
0.2413	0.26	1.05595
0.2413	0.2413	1.06565

Inner radius of minor actinide/cm	Outer radius of minor actinide/cm	k_{eff}
0.2413	0.4267	0.97378
0.2413	0.40	0.97954
0.2413	0.38	0.98565
0.2413	0.36	0.99211
0.2413	0.34	0.99848
0.2413	0.32	1.00694
0.2413	0.30	1.01431
0.2413	0.28	1.02460
0.2413	0.26	1.03969
0.2413	0.2413	1.06565

Since the minor actinides are also able to fission, and transmute to fissile materials, the total replacement of burnable poison rods with minor actinides may change the physical characteristics of PWRs. Therefore, the time in which the transmutations occur is important; if the minor actinide transmutation materials could burn out faster than the fuel burnup, as the burnable poison does, then later in the PWR life the minor actinides would contribute negligible negative reactivity [65].

2.2.4 Fast Reactor Experience

As mentioned in the previous section, minor actinides act as poisons in thermal reactors, thus a higher percentage of U-235 is required to compensate. The effectiveness of the transmutation of minor actinides is dependent on the conversion ratio; a theoretical consumption rate of 1g/MWt-day is obtained for a conversion rate of 0, because the fuel does not contain any uranium [66]. Fast reactors such as sodium-cooled, lead-cooled, and gas-cooled, have a larger ratio of fission-capture cross sections and larger neutron consumptions in general [69]. For fast the reactors, the choice of coolant is negligible in the viewpoint of minor actinide transmutation [66].

Various studies have been put forward to examine the use of fast reactors for minor actinide transmutation. Takeda (2016) aims to harmonize the minor actinide transmutation and reduce the sodium void reactivity while using a homogeneous distribution. A new calculation method was used to understand the minor actinide transmutation of individual actinides. Conventional burnup calculations are performed, and the burnup dependent fluxes are determined for specific regions in the core. Only the relevant isotopes are considered in each region, and the fluxes are used to establish the parent and calculate daughter isotopes in each region. Essentially, the transmutation rate is dependent on the incineration rate by fission, and the net transmutation rate of the fuel (U and Pu). Two main variables that are established are: the amounts of the parent nuclide before and after exposure with the minor actinide amount of other minor actinides that resulted in transmutation of the parent; and overall fission and the net plutonium production [66].

After 1 year and 6 years of burnup, the minor actinides were calculated. After one year of exposure, there was a 5.5% decrease in the total inventory of minor actinides. The increase in Am-243 and Cm-244 causes the total minor actinide reduction to be reduced to 5.5%. The values for the 6-year exposure were found to be 6 times that of the 1 year burnup for the total reduction in minor actinides. However, the overall fission and the plutonium production are even larger. In fact, the overall fission increases linearly as the burnup period increases, but the plutonium production shows saturation behaviour at 9 years [66].

2.2.4.1 Molten Salt

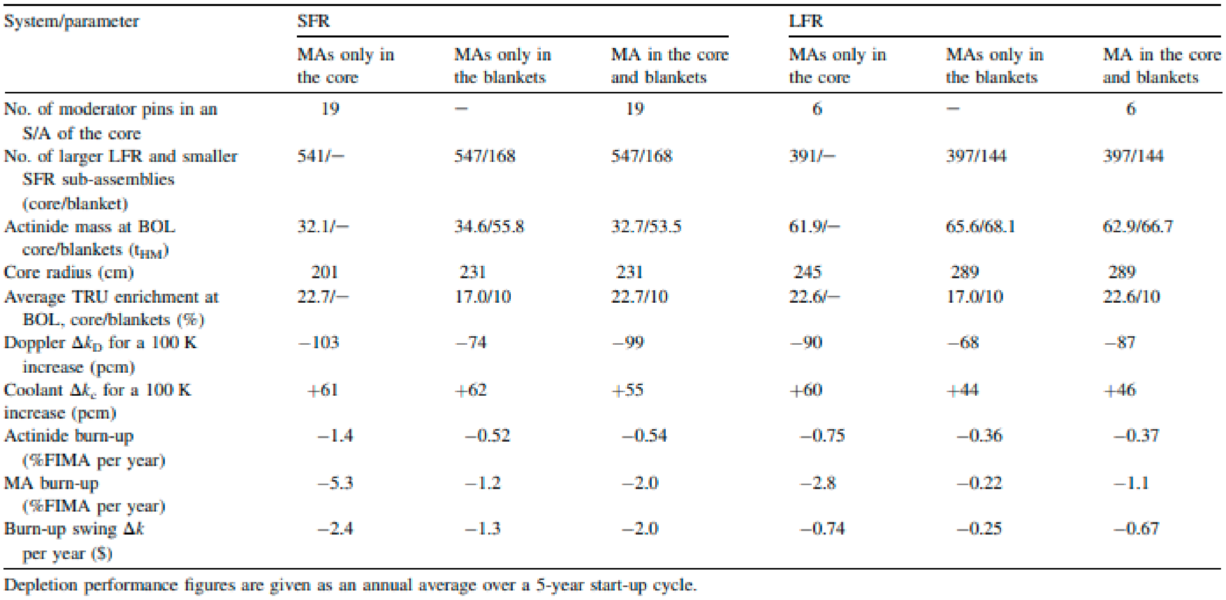
Tucek, et al. aimed to design reactor cores in which minor actinides would be consumed while plutonium would be produced, Sodium-cooled Fast Reactor (SFR) and Lead-cooled Fast Reactor (LFR) [70]. To compensate for the initial minor actinide amount, the starting fuel requires enrichment. In order to improve minor actinide transmutation, axial and radial blankets are investigated for both reactor types. Any plutonium bred in these regions is less-proliferation prone. Table 24 shows the characteristics for the LFR and SFR cores. Both cores have been adjusted to maximize the fuel economies and ensure safety is kept. Both cores use $(U,TRU)O_2$ mixed oxide fuel and in-core $UZrH_{1.6}$ moderators; the latter is employed to improve the temperature reactivity coefficients (Doppler and coolant). Moderators are placed in dedicated pins within the sub-assemblies [70].

Table 24: Design Parameters of SFR and LFR self-breeder cores [70].

Parameter	LFR	SFR
Power (MW_e)	600	600
Pellet outer radius (mm)	5.0	3.0
Clad inner radius (mm)	5.1	3.1
Clad outer radius (mm)	6.25	3.45
Pitch-to-diameter ratio (P/D)	1.5	1.2
S/A outer flat-to-flat (cm)	22.20	14.66
Pins per S/A	127	271
Length of upper plenum (cm)	100	100
Length of lower plenum (cm)	10	10
Active pin length (cm)	200	100
Length of upper/lower axial blankets (cm)	50	50

The TRU used in the fuel was from LWR spent fuel with a burnup of 41 HWd/t_{HM} after a decay of 30 years. Therefore, the minor actinide fraction within the TRU is 18%. Table 25 shows the results of three core/blanket configurations: homogeneous recycling of TRUs (Pu and MAs) with no radial and axial blankets, recycling Pu in the core and minor actinides in the blankets only, combined recycling of TRUs in the core and MAs in the blankets.

Table 25: Neutronic and burnup parameters of the 600MWe LFR and SFR self-breeders [70].



System/parameter	SFR			LFR		
	MAs only in the core	MAs only in the blankets	MA in the core and blankets	MAs only in the core	MAs only in the blankets	MA in the core and blankets
No. of moderator pins in an S/A of the core	19	–	19	6	–	6
No. of larger LFR and smaller SFR sub-assemblies (core/blanket)	541/–	547/168	547/168	391/–	397/144	397/144
Actinide mass at BOL core/blankets (t_{HM})	32.1/–	34.6/55.8	32.7/53.5	61.9/–	65.6/68.1	62.9/66.7
Core radius (cm)	201	231	231	245	289	289
Average TRU enrichment at BOL, core/blankets (%)	22.7/–	17.0/10	22.7/10	22.6/–	17.0/10	22.6/10
Doppler Δk_D for a 100 K increase (pcm)	−103	−74	−99	−90	−68	−87
Coolant Δk_c for a 100 K increase (pcm)	+61	+62	+55	+60	+44	+46
Actinide burn-up (%FIMA per year)	−1.4	−0.52	−0.54	−0.75	−0.36	−0.37
MA burn-up (%FIMA per year)	−5.3	−1.2	−2.0	−2.8	−0.22	−1.1
Burn-up swing Δk per year ($)	−2.4	−1.3	−2.0	−0.74	−0.25	−0.67

Depletion performance figures are given as an annual average over a 5-year start-up cycle.

It should be noted that the LFR required more actinide mass to achieve criticality than the SFR as the SFR has a superior neutron economy due to the tight pin lattice (P/D=1.2) compared to the LFR (P/D=1.5). Therefore, the SFR has a higher actinide burn-up rate; however, the lower initial actinide masses of the SFR causes a larger reactivity shift compared to the LFR. The unmoderated SFR core (MA only) experiences a larger negative Doppler effect as well as larger positive coolant temperature feedbacks than the LFR. The coolant temperature feedback is greater due to the spectral hardening, which is associated with the coolant heat-up/voiding [70].

The presence of $UZrH_{1.6}$ moderating pins improved the reactivity coefficients due to increased thermalization of the neutron spectra, improving the Doppler and coolant temperature feedback. Both reactors consumed minor actinides and bred plutonium (Table 26).

Table 26: Amount of annually generated/consumed transuranics in LFR and SFR self-breeders [70].

Parameter	SFR			LFR		
	MA in Core	MA in Blanket	MA in Core and Blanket	MA in Core	MA in Blanket	MA in Core and Blanket
Pu generated (kg/y)	-12	+198	+150	+14	+145	+110
MA consumed (kg/y)	-66	-65	-131	-67	-15	-104

Start-up cycle's annual average over a 5-year cycle length. Assuming all Cm-242 decayed to Pu-238.

As seen in Table 2.26, the highest consumption of MA is in the cores where MAs were placed in the core and in the blanket. When MAs were placed in the blankets only, there is a decrease of the U/Pu fraction in the core, which improves the breeding potential. Also, the SFR consumed more MAs (65kg/y) than the LFR (15kg/y). This is due to the larger self-production of minor actinides in the LFR core than the SFR, lower neutron flux, and lower absorption cross-section in radial blankets [70].

2.2.5 Breeding/Burning Reactors

The concepts of breeding reactors and burning reactors were introduced to maximize the utilization of the depleted uranium within nuclear waste. Breeding reactors would take depleted uranium fuel and produce fissile material which would be used as fuel in the future; Burner reactors rely on fissile material, and consumes various hazardous materials in the fuels during operation [11].

There is a potential to combine the two reactors, to a breed and burn (B&B) reactor. The B&B is unique by breeding plutonium in depleted uranium feed fuel and then burning the plutonium, without reprocessing the fuel. The reactor would begin with an enriched uranium fuel, to begin a chain reaction. TRU elements could be extracted from spent fuel and be used in the “fresh” B&B fuel [11]. The reactor relies on a once-through fuel cycle in which the plutonium is bred and burned without being removed from the core [12].

2.3 SMR Technology

Countries around the world are at various stages of nuclear infrastructure; many look towards nuclear energy for its minimal contribution to greenhouse gas emissions. Small Modular Reactors (SMRs) are more likely to be developed within the next few decades as larger GEN IV reactors will not be fully designed for another twenty years. SMRs have an electrical output of less than 350MWe, are deliberately small, and capitalize on their size to achieve specific performance characteristics [16]. SMRs are designed to be factory manufactured, transportable, and/or relocatable. Many designs allow for secondary benefits as well as electricity, including district heating, desalination, and aiding in the processes for other industrial by-products. Designers are targeting customers who would not benefit from a large economy of scale power plants: countries with smaller energy grids; villages, towns, and energy intensive sites in off-grid locations; rapidly growing cities or countries, hybrid plants for non-electric services [76].

The Canadian Nuclear Safety Commission (CNSC) is currently reviewing the SMR designs listed in Table 27. The pre-licensing vendor design review is an opportunity for the CNSC to view vendor technology and provide feedback without releasing confidential details of the design.

Table 27:Vendor design review agreements between Vendors and the CNSC.

Vendor	Name of Design	Cooling Type	Electrical Capacity (MWe)
*Terrestrial Energy	IMSR (Integral Molten Salt Reactor)	Molten Salt	200
Ultra SAfe Nuclear	MMR-5/10	High-Temp Gas	5/10
*LeadCold Nuclear	SEALER	Molten Lead	3
*ARC Nuclear Canada	ARC-100	Liquid Sodium	100
Moltex Energy	Stable Salt Reactor	Molten Salt	300
Holtec International	SMR-160	Pressurized LW	160
*NuScale Power	NuScale	Pressurized LW	60
U-Battery Canada	U-Battery	High-Temp Gas	4
GE-Hitachi Nuclear	BWRX-300	Boiling LW	300

*selected to be analyzed

2.4 Fuel Storage

Global disposal sites for low and intermediate level waste ranges from near-surface facilities to engineered geological repositories [13]. Site selection for waste storage is crucial, as the site in question needs to satisfy a list of requirements including: geology, hydrogeology, geochemistry, tectonics, seismicity, surface processes, meteorology, human-induced events, transportation, land use, population distribution, and environmental protection [13]. Public acceptance is a huge factor in site selection. Although many people in industrialized countries have a "not in my backyard" attitude towards nuclear developments [13], there are certain economic benefits to having the waste disposal site near. In Canada, there was initially community opposition to disposal sites, so the government halted the site selection to change the strategy. A co-operative five-phase programme taskforce was implemented to work closely with municipal communities [13].

The CANDU reactor fuels sit in wet storage for approximately 6 years before being transported to dry storage. Wet storage consists of an irradiated fuel bay, in which stockpiles of spent fuel bundles rely on the water to absorb decay heat. Canada currently does not have a permanent fuel storage solution, and therefore is simply relying on large casks to hold spent fuel until a long term solution is implemented.

2.5 Fuel Cycle Options

The development and disposal of nuclear waste will always be a requirement of the fuel cycle; however, elements of the nuclear fuel can be repurposed and recycled in order to “close” the once-through fuel cycle. In general, each fuel cycle begins with the front end, from mining to fuel fabrication. Uranium ore is found in different forms throughout the planet; however, where uranium is the least concentrated (<0.1%), in-situ leaching techniques can be used. Other cases use classical excavation, with an open pit for shallow surface deposits (<120m deep), and underground mines for deeper deposits [21].

After the acquisition of the uranium ore, it must be milled. Milling requires a mechanical treatment, specifically crushing and grinding. The uranium ore is thus converted to a finer powder, to ease uranium extraction. The separation of uranium is done using leaching techniques with either a strong acid or alkaline solution [21]. The result of the leaching is uranium oxide (U_3O_8) concentrate (yellowcake). The following steps are dependent on the type of fuel required. The uranium is either fabricated into fuel directly, or enriched, then fabricated into fuel.

The once-through cycle simply disposes of the fuel once it leaves the reactor. A closed-fuel cycle involves reprocessing spent fuel at least once. Spent fuel can be reprocessed multiple times to encourage using fuel elements to their maximum capacity.

2.5.1 Fuel Cycle Options Material Flow

There are many issues to consider when deciding whether or not to close the fuel cycle in a specific area or country. Firstly, there is a finite amount of uranium on earth; therefore, one should explore how the resource can be used more efficiently. There is already a large reservoir of nuclear waste, and more is being produced daily. The current waste management is to store the fuel underground until an unknown future date. Is this the most environmentally safe solution? There are countries that have large amounts of plutonium to use for weapons, and other countries (such as Canada) strongly oppose the use of any nuclear material being weaponized. Is there a way to ensure that nuclear materials could not be obtained for proliferation? Lastly, the economics of the aforementioned considerations needs to be competitive in order to ensure support from countries and energy vendors [14].

As each country has different energy needs, the chosen fuel cycle option should be chosen to have those needs met. The Republic of Korea (ROK) analyzed nuclear fuel cycle options for their country, which consists of 17 PWRs, 4 CANDU reactors, and 7 PWRs under construction. Due to the heavy presence of the PWR, the once-through (OT) cycle refers to the PWR-to-disposal option. Alternative cycle options are as follows: DUPIC recycling, PWR-MOX recycling, and Pyro-SFR recycling, depicted in Figure 18. The front-end (mining, milling, conversion, enrichment, and fuel fabrication) of these cycles would be the same as they all begin with the PWR operation [14].

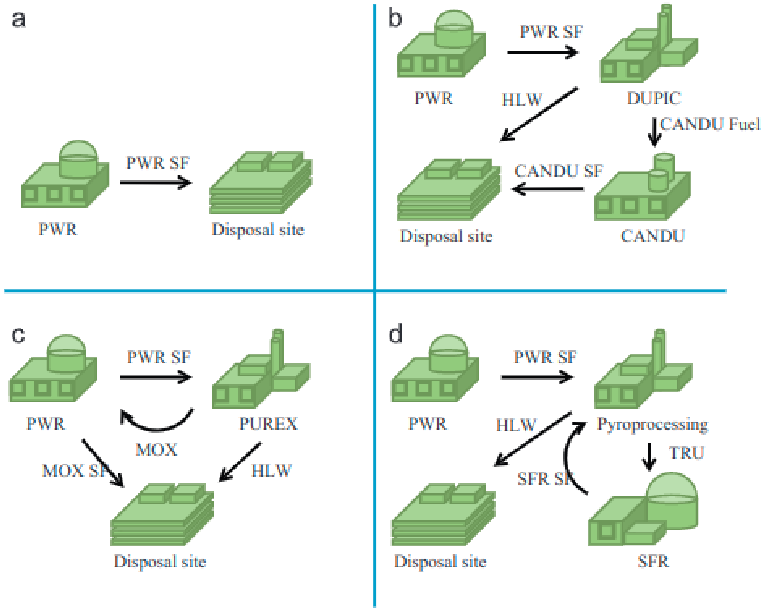


Figure 18: Schematic description of fuel cycle options [14].

The OT cycle (Figure 18a) spent fuel provides insight into the contents that the other cycles would be able to manipulate. Although, if there are large variances between initial enrichment and burnup between all of the PWRs, there will be differences in composition. Assuming the PWR initially has 3.5% enriched fuel, and a burnup of 35GWd/tHM, the spent fuel has 0.9wt% U-235 and 0.5wt% Pu-239, which would be sufficient for the use in the CANDU reactors (Figure 18b), providing that the fission products are removed. This cycle has already been heavily investigated for ROK. The reprocessing of the spent fuel involves a simple dry thermal/mechanical process, the Oxidation and Reduction of Oxide fuel. The DUPIC fuel would be disposed of rather than reprocessing further [14].

As discussed in Section 2.1.2, PUREX is the most well-known reprocessing technology for PWR fuels, and recovers both uranium and plutonium. Since there are thermally prevalent fissile actinides of plutonium, Pu-239 and Pu-241, it is also reprocessed to be fabricated into a mixed oxide fuel (MOX), which is made of UO_2 and PuO_2. Upon leaving the CANDU reactor, the MOX spent fuel will be disposed of (Figure 18c) [14].

Lastly, the sodium cooled fast reactor (SFR) is implemented in a fuel cycle option. The SFR uses fast neutrons of higher energy to burn fissile isotopes as well as transuranic elements, as discussed in Section 2.2.4 The cycle in question involves an SFR with a conversion ratio of 0.6067, which promotes both waste reduction and non-proliferation solutions. Pyroprocessing was developed for spent oxide fuels discharged from PWRs, and recycled metallic components containing TRUs from SFRs. A metal fuelled SFR using actinide-zirconium could be recycled using pyroprocessing as well. In this fuel cycle, the spent fuel from the SFR which contains TRU, would be repeatedly reprocessed using pyroprocessing. The recovered TRUs would then be reinserted in the SFR to close the fuel cycle, shown in Figure 18d [14]. All reactors involved in this study are described in Table 28.

Table 28: Characteristics of Reference Reactors, PWR, CANDU, and SFR for fuel cycle options [14].

Reactor Parameters	PWR	CANDU	SFR
Electric Power (MWe)	1000	713	600
Thermal Efficiency (%)	34.23	33	39.4
Thermal Power (MWt)	2921.4	2160.6	1522.8
Load Factor	0.85	0.9	0.85
Cycle Length (Full Power Day)	290	-	332
Total HM per core (tHM)	69	84.72	20.3
No. of Batches	3	-	6
Conversion Ratio	-	-	0.6067

In order to establish a quantitative comparison, each cycle was set to produce a total of 1TWh of electricity. A steady-state model was used to focus on the fuel cycle itself, so other economic, social, and environmental constraints do not need to be considered. However, in reality the readiness of certain fuel cycles (involving Pyroprocessing and SFRs) would require decades for deployment. Table 29 depicts the fuel content parameters that are relevant for reprocessing technologies.

Table 29: Parameters of the four fuel cycles [14].

Fuel Cycle	OT	DUPIC	PWR-MOX	Pyro-SFR
Enrichment	4.5% U-235	3.5% U-235	4.5% U-235	4.5% U-235
PWR Burnup (GWd/tHM)	55	35	55	55
Back-end PWR SF	-	OREOX loss: 0.1% (Kr, Xe, Cs)	PUREX loss: 0.1% (MA, FP)	Pyroprocess loss: 0.1% (FP)
CANDU Fuel	-	Burnup: 7.5MWd/kgHM Pu:0.92%, Fissile Pu:68%	-	-
MOX Fuel	-	-	Burnup: 55GWd/tHM Pu:8%	-
SFR Fuel	-	-	-	Burnup: 121GWd/tHM Pu: 33.8%, MA: 5.6%
Back-end for SFR SF	-	-	-	Pyroprocessing loss: 0.1%
SF Contents (wt%) U Pu MA	 98.51 1.27 0.22	 99.05 0.82 0.13	 93.44 5.71 0.86	 71.46 25.39 3.15

Resource utilization was the focus of this analysis, as well as waste generation [14]. Core ratios were calculated to find the optimal number of reactors in each scenario in order to maximize material usage. The core ratio for OT cycle is 100% as there is only one reactor within the cycle. Given the reactor load factor (Table 29), the reactor would need to be operating for 1176 hours to make 1TWh of electricity [14]. The ratio for the DUPIC cycle is 0.54, implying that it will take two PWRs to sustain the required material input for the CANDU. The electricity produced by the PWRs and CANDUs are 71% and 29% respectively, and a combined operating time of 1287 hours [14]. For the PWR-MOX fuel cycle, the enriched fuel PWRs contributed to 87.7% of the electricity, with the remaining PWRs using the MOX fuels. One MOX fuel PWR was supported by seven enriched fuel PWRs, and to achieve 1TWh was a combined time of 1176 h. Lastly, the Pyro-SFR relied on the assumption that all TRUs collected from the PWR were recycled and used in the SFR. One SFR and one PWR would be sufficient for equilibrium, operating at 396GWh and 604GWh, respectively, and with a combined time of 1487 hours [14].

Resource Utilization:

When the four cycle options are compared (Table 30), with respect to uranium requirements, the OT cycle requires the most, and the Pyro-SFR requires the least. Therefore, the three alternative fuel cycle options are considered competitive options, as they use uranium more efficiently, which is important because uranium prices are not consistent. The natural uranium requirement for DUPIC, PWR-MOX, and Pyro-SFR are 8.2%, 12.4%, and 39.6% less than the OT cycle [14].

Table 30: Mass flow summary through the four fuel cycle options based on 1 TWh electricity [14].

Cycle Stage	Once-Through PWR	DUPIC Recycling	PWR-MOX Recycling	Pyro-SFR Recycling
Milling/Mining (tU)	20.444	18.769	17.921	12.342
Conversion to Yellow Cake (tU)	20.424	18.75	17.903	12.33
Enrichment Waste (depleted tU)	18.209	16.09	15.961	10.992
Enrichment Product (tU)	2.215	2.66	1.942	1.337
Fresh Fuel put into PWR (tU)	2.213	2.657	1.94	1.336
Spent Fuel (tHM)	2.088	2.561	1.831	1.261
DUPIC Fuel (tHM)		2.558		
CANDU SF (tHM)		2.469		
PUREX Disposal (tU)			1.7	
PUREX Disposal (tHLW)			0.109	
PUREX for MOX Fuel (tPu)			0.022	
Pyro Disposal (tU)				1.088
Pyro Disposal (tHLW)				0.072
Pyro for SFR Fuel (tU)				0.041
Pyro for SFR Fuel (tTRU)				0.018
*Second Pyro of SFR SF (tTRU)				0.106
*Second Pyro of SFR SF (tU)				0.267
SFR SF (tHM)				0.737
Second Pyro Disposal HLW(tHM)				0.000373

Enrichment Waste (depleted tU) for MOX			0.252	
MOX Fuel (tHM)			0.273	
Disposal of SF (tHM)			0.259	
Total Disposal (tHM)	20.297	18.559	17.777	12.152373

Waste Generation:

Due to the inevitable generation and existence of waste, the nuclear industry has tried to decrease the volume, heat, and toxicity of waste. Compared to the OT cycle, the DUPIC and PWR-MOX fuel cycles are 118% and 12.4%, respectively, and no spent fuel is disposed of in the Pyro-SFR model. For the DUPIC cycle, the increase of spent fuels is due to the required increased enrichment for the specific cycle (Table 30). The volumes of the radioactive wastes produced, is mainly dependent on the reactor types within the cycles.

The CANDU reactor produces the largest amount of LILW-SL, and the Pyro-SFR produces the least. Due to the use of reprocessing technologies, especially PUREX, there is an increase in LILW-SL for both the PWR-MOX and DUPIC cycles. The geological disposal site requirements built for LILW-LL, and therefore the other cycles will be compared to the OT cycle. For the Pyro-SFR cycle, the capacity is 70%, and for DUPIC and PWR-MOX are both 180%. The HLW is mainly produced in the back-end of each cycle, resulting in the smallest contributor being the Pyro-SFR, because it allows for the high heat generating elements (Cs and Sr) to be separated as LILW-SL for decay, and TRUs are recovered and reused.

The activities of the waste streams are mainly from the fission products, which would establish the shielding requirements for transportation, storage, and other operations. As shown in Figure 19, the Pyro-SFR cycle has the least amount of activity, with a significant decrease after 100 years, due to the TRU utilization strategy [14]. As proliferation resistance is a major issue for the upcoming nuclear generations, the presence of plutonium in the spent fuel, if disposed, could be considered a plutonium reservoir. Also, plutonium is fissile and thus can be used with uranium to produce power; if disposed of, it could be considered wasteful. Therefore, maintaining plutonium and TRU within the fuel cycle would reduce proliferation and improve fuel economy [14].

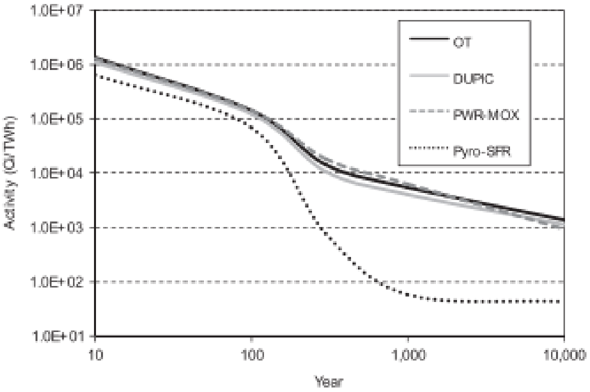


Figure 19: Activity of HLW per fuel cycle option over time [14].

2.5.2 Environmental Assessment of Fuel Cycle Options

With the expected growth in energy demands, while simultaneously trying to reduce greenhouse gas (GHG) emissions, countries should be looking to nuclear energy. With respect to GHG, there are fewer environmental concerns with nuclear energy, due to a much lower emission rate (6-10 $gCO2eq/kWh_e$)[21]. The social and political advantages involve the mining of uranium, as it is unlikely to cause friction and significant international tensions as is the case for oil and coal. Although there is a significant capital cost of a nuclear facility, the reduced dependence on uranium price is a benefit as well. Nuclear energy is being considered as an energy contributor in the future to meet electricity needs while maintaining the climate [21].

However, after the events of Fukushima-Daiichi, the public believes that the consequences of nuclear energy are severe for public health and the environment. Future developments in any industry, as well as nuclear, needs to prove the robustness and strength of the facilities going beyond the technical aspects. The nuclear industry must address technical, social, environmental, and economic implications. Sustainable development is defined as a "development that meets the needs of the present without compromising the ability of future generations to meet their own needs." Although it has been defined, a way to effectively measure sustainable development is up for debate, as not all aspects are quantifiable (societal development). Life cycle assessments have been widely used across industries to determine the most "sustainable" option; however, nuclear life cycle assessments result in a wide range of results due to the fuel cycle variances [21]. The following comparison involves a closed and open energy cycle that produces the same amount of energy produced [21]. The nuclear energy systems are based on those within France, with a twice-through cycle (TTC). France's fuel cycle front-end and back-end are briefly discussed in Section 4.4.1.1. However, for this analysis, certain assumptions were made for simplicity and due to the available data. Indicators were chosen based on dissemination and acceptance within the scientific community, and applicability for different energy sources. Table 31 shows the eight indicators used for the analysis of France's nuclear industry. The indicators were chosen as they are generally accepted within the scientific community and their applicability for a variety of energy sources. The calculations were performed by using the Nuclear Energy Life Cycle Assessment tool (NELCAS). The tool compiles and relates all energy and matter fluxes together throughout the fuel cycle. The NELCAS tool does not aim to replace a life cycle assessment, but merely shows the global footprint of the French nuclear energy system, as a leading nuclear country. Figure 20 shows the relative contributions depending on the fuel cycle step.

Table 31: NELCAS results for environmental and technological impact indicators for French TTC [21].

	GHG gCOzeq/kWhe	Atmospheric Pollution SOx mg/kWhe	Atmospheric Pollution NOx mg/kWhe	Water Pollution mg/kWhe	Land-use m2/GWhe	Water Consumption L/MWhe	Water Withdrawal L/MWhe	Tech Waste m/MWhe
Mining	1.704	14.242	19.73	263.072	144.1	12	12	1.5
Conversion	0.278	0.058	1.04	0.087	1.82	4.6	4.6	2.0
Enrichment	0.626	0.547	1.06	2.548	1.88	23	23	0.65
UOX Fabrication	0.035	0.013	0.05	0.021	0.93	0.3	0.3	0.23
Reactors	2.140	0.938	2.84	16.366	45.1	1460	72.318	20.15
Reprocessing	0.376	0.484	0.5	5.433	4.98	1.7	1.7	0.63
MOX Fabrication	0.027	0.004	0.035	-	0.13	0.1	0.1	0.18
Disposal	0.104	0.024	0.097	-	12.01	0.1	0.1	1.11
Total	**5.29**	**16.276**	**25.30**	**287.53**	**211.0**	**1507**	**72.364**	**26.4**

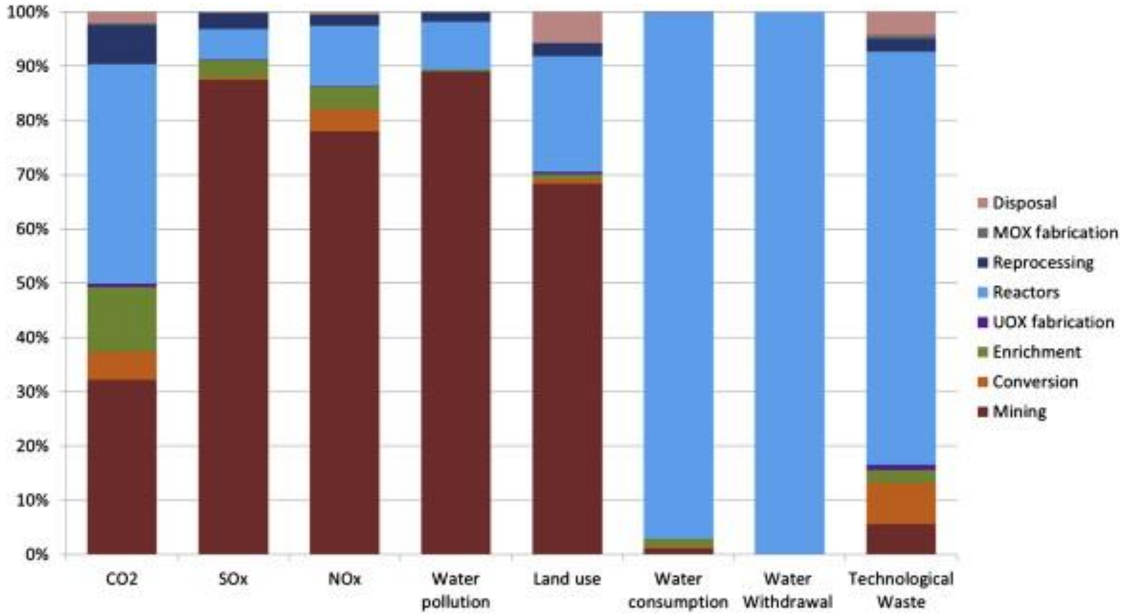


Figure 20: Relative contribution of each fuel cycle step to the indicators calculated in NELCAS for French TTC [21].

As shown in Table 31 and Figure 20, most of the SO_x/NO_x emissions, water pollution, and land usage, comes from mining. GHG emissions mainly come from mining and reactor operations. For the global fuel cycle, the total greenhouse gas emissions are estimated to be 5.29gCO$_2$eq/kWh$_e$, although the enrichment contribution is considered to be lower than other countries' assessments. The lower estimation is due to the French enrichment plant being stationed and fed by three PWRs. For example, the USEC enrichment facility, which is coal-powered, is 55gCO_2eq/kWh$_e$ [21].

Mining is the main contributor for the release of SO_x and NO_x at 87% and 78%, respectively, followed by the reactors and enrichment. Water pollution is dominated by mining and milling (89%) due to the releases of chemicals (non-radioelements) in water, especially sulphates.

Typically, land usage falls between 50 and 500 m^2/GWh$_e$ and therefore France falls in the middle with 211 m^2/GWh$_e$. Mining and milling contribute 68% of land use, with reactors at 21%. However, due to the majority of uranium ore primarily coming from underground mines, the land-use indicator is reduced by a magnitude of two [21]. Water consumption and withdrawal are dominated by reactor operations at 97% and 99.9%, respectively. Since more than half of the reactors in France require cooling towers, there is a net water consumption of 1500L/MWh$_e$, but only for those reactors. Due to the nature and technologies of the front-end operations, mining and milling also consume water. For example, mining techniques need to be considered, as in-situ leaching requires more water; and in the areas in which mines are typically located, water needs to be conserved as much as possible.

As uranium reservoirs deplete, the efficiency with which nuclear fuel is used will become more important. France's twice-through cycle, where 7647t of natural uranium is required to produce 408TWh$_e$, has an efficiency of 53.4GWh$_e$/kgU$_{nat}$. Many assumptions were made in order to simplify calculations to obtain the values above, such as the omission of second and third order contributors.

As previously mentioned, the indicators above are environmental and technological; the additional indicators are acidification, eutrophication, photochemical ozone creation, and eco and human toxicity. Acidification, the formation of acid rain, mainly depends on the SO_x and NO_x, and therefore is mainly controlled by the mining and milling operations. Eutrophication is linked to the increase of phosphorus and nitrogen compounds in water, which releases NO_x, and leads to the proliferation of seaweeds, asphyxiating ecosystems. Mining is the main contributor, followed by enrichment and reactor operations. Many simplifications were required to calculate the toxicity for ecosystems and humans. Therefore, an overestimation was made by assuming all effluents were released in fresh water. If salt water was included there would be a large dilution and therefore a significant decrease in the impact. Once again, mining is the major contributor with over 99% of the impact for both ecosystems and humans. Vanadium is the main contributor to eco-toxicity, followed by molybdenum. Molybdenum is the main contributor for human toxicity, followed by lenium, vanadium, and NO_x. Uranium only contributes to 2.5% of the eco-toxicity and 1.6% to human toxicity [21].

France examined the impact of their radioactive waste management. During the fuel cycle, radioactive wastes are released through aqueous media and into the atmosphere; noble gases, mainly radon, tritium, and C14 are main contributors. Since radon is only present in the uranium ore, all of its release is during the mining and milling phase, which is 53% of the noble gas emissions. Noble gases, tritium, and C14 are produced within the reactor through neutron interactions. Most gases are trapped within fuel pins, which are released during the reprocessing step. Most of the tritium (99.2%, 27.1kBq/kWh$_e$) is released as liquid wastes. A lot of waste is released during the reprocessing step, both gaseous and liquid (24.4kBq/kWh$_e$). In France, these releases are well below the regulation threshold set, and the impact is lower than 10μSv/year. One of the largest inhibitors of nuclear energy developments is the presence of nuclear waste, especially solid waste. Due to the absence of dismantling experience, data is restricted. Most of the very low-level waste is from mine tailings, leaching residues, and residual waste). Reactor operation is the main contributor of short-lived and long-lived intermediate level waste, which is assumed to be recovered during dismantling of the fuel cycle facilities. High level wastes are collected at reprocessing operation sites. France's management of the different waste is dependent on timescale and selected technical solutions. The long-lived intermediate level waste and the high-level waste are contained in different ways; although, in order for the geological repository to be efficiently used, storage of these materials will need to be altered.

France did perform a comparison of the twice-through cycle and the once-through cycle, the results of which can be summed in Figure 21. Depending on the indicator, the preferred fuel cycle is typically the twice-through cycle, where the most significant inclination towards the twice-through cycle is the amount of high-level waste, with over a 200% difference. However, the amount of HLW present in both scenarios is so small, that the 200% difference does not account for a significant volume.

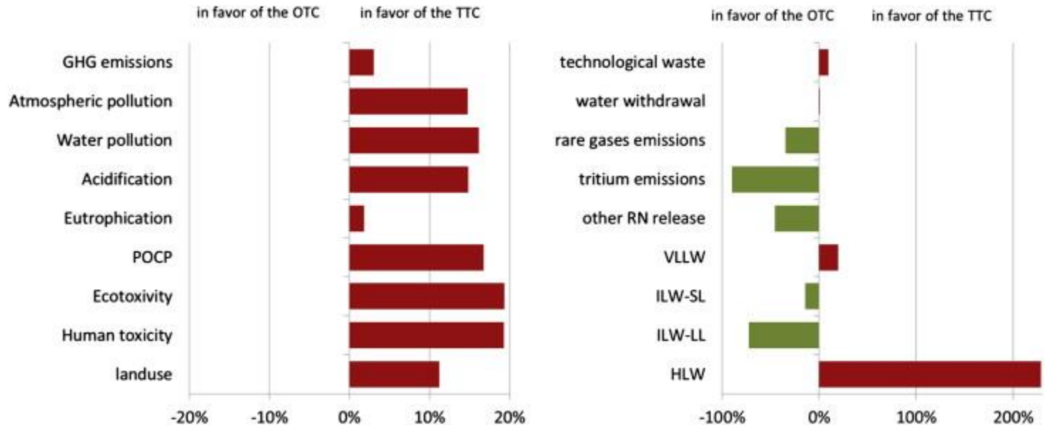
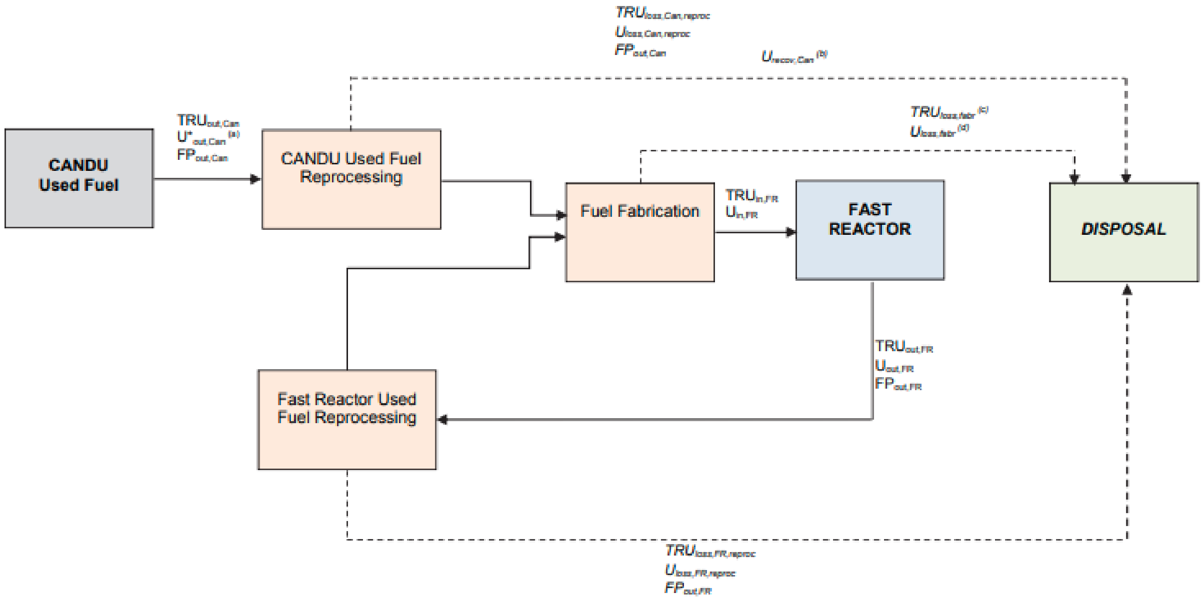


Figure 21: Impact indicators when comparing the French TTC to an OTC producing the same amount of electricity with the same PWR fleet [21].

2.5.3 NWMO CANDU Cycle Analysis

Ion [58] performed a high-level analysis of the advanced fuel cycle in which plutonium and minor actinides from CANDU spent fuel was burned in Fast Reactors (FRs). The FR spent fuel was also recycled continuously to produce energy. The entire cycle that was analyzed is shown in Figure 22. As mentioned in Section 2.2.5, FRs can operate as breeders or burners. A breeder reactor is attractive as it continuously creates its own fuel. A burner reactor requires a continuous source of fissile material, which could include TRUs, and could be used for waste management. The eventual wastes from the FR would accompany the waste from reprocessing CANDU spent fuel, and be deposited in the DGR. The waste from the metal cladding was not considered in this analysis. Also, the loss in TRU and U during reprocessing was deemed negligible and therefore ignored for mass flow purposes. The following analysis assumes there is enough CANDU spent fuel storage for the operation of two FRs to last 60 years.



Notes:

a) $U^{*}_{out,Can}$ is the total amount of uranium available in CANDU used fuel, out of which a smaller quantity ($U_{out,Can}$) is used for reprocessing and for fabrication of the FR fuel.

b) $U_{recov,Can}$ is the amount of uranium recovered from reprocessing of CANDU used fuel, and not used for fabrication of the FR fuel, which could be either stored for future re-use as make-up in the fast reactor or sent for disposal.

c) $TRU_{loss,fabr}$ represents the total TRU losses from FR fuel fabrication using reprocessed CANDU and FR used fuels.

d) $U_{loss,fabr}$ represents the total U losses from FR fuel fabrication using reprocessed CANDU and FR used fuels.

Figure 22: CANDU-FR nuclear fuel cycle used in analysis [58].

The reprocessing technologies are at various stages of development; however, the pyroprocessing technology was considered for this analysis due to its superior proliferation resistance compared to PUREX. It is assumed that the rate of pyroprocessing CANDU spent fuel is not limiting, and will be sufficient to supply the needs of the FRs. The efficiency of pyroprocessing is assumed to be 99.5 wt% recovery for TRU elements and 99% recovery for uranium, and the fuel fabrication has an efficiency of 99.9 wt% [58].

A pool-type, sodium-cooled fast reactor (PRISM) was chosen for the CANDU fuel cycle analysis with three conversion ratios (Table 32). The reference fuel for the FR is an alloy metal which contains uranium, plutonium, and zirconium. In order to achieve the different conversion ratios, the number and size of the fuel pins were adjusted. For example, the number of pins modified the linear power, and the fuel pin diameter modified the fuel volume fraction (requiring more TRU). A higher TRU enrichment increases the TRU fission relative to U-238 capture, resulting in a reduction of the TRU conversion ratio.

Table 32: Fast Reactor Characteristics [58].

Thermal Power (MW_{th})	1000		
Thermal Efficiency (%)	38		
Conversion Ratio (CR)	0.25	0.5	0.75
Discharge Burnup (HWd/t_{HM})	186.9	131.9	99.6

In terms of waste management, a low CR would be better for a burner reactor as it requires more external fuel and maximizes the burnup of the TRU. However, a low CR requires the enrichment of the TRU. Based on current technology, the CR between 0.5 and 0.6 would be optimal. The CANDU spent fuel used in this analysis has a burnup of 220MWh/kgU and contains uranium (0.9857), TRU (0.0044) and Fission Products (0.0099), with Pu-239 and Pu-241 making up 66wt% of the TRU [58]. The FR fuel composition is dependent on the CR, with the CR of 0.75 containing 54wt% Pu of the total TRU. Each conversion ratio has a different initial loading of heavy metals: 0.25 has $6.169t_{HM}$. 0.5 has $9.449t_{HM}$, and 0.75 has $13.436t_{HM}$. The TRU that is reprocessed comes from a LWR with a burnup of $50MWd/kg_{HM}$ with a cooling time of 5 years [58].

Mass Flow Modeling

The values used for the following calculations are found in Table 32. The mass of FR fuel required to generate $1GW_ey$ of electricity is calculated with:

$$m = \frac{365}{B_d \eta_{th}} \quad \text{(Eq. 5)}$$

where B_d is the discharge burnup (GWd/t_{HM}) and $\eta_{th}$is the thermal efficiency (%). The amount of TRU required per year for the operation of the FR from the spent FR fuel and the CANDU fuel is found by:

$$TRU_{in,FR} = \varepsilon_{TRU,r}\varepsilon_{TRU,f}(TRU_{out,CAN} + TRU_{out,FR}) \quad \text{(Eq. 6)}$$

where $TRU_{out,CAN}$ is the mass of the TRU reprocessed from CANDU fuel, $TRU_{out,FR}$ais the mass of TRU reprocessed from FR fuel, $\varepsilon_{TRU,r}$ is the TRU recovery efficiency from reprocessing, and $\varepsilon_{TRU,f}$ is the recovery efficiency from fuel fabrication. The mass of the start up FR fuel contents are:

$$TRU_{in,FRstart} = f_{TRU,start}m_{HMload} \quad \text{(Eq. 7)}$$

$$U_{in,FRstart} = f_{U,start}m_{HMload} \quad \text{(Eq. 8)}$$

where $TRU_{in,FRstart}$ and $U_{in,FRstart}$ are the masses of TRU and U in the fresh fuel (t), respectively; *f* is the fraction of the TRU and U in the fresh fuel at start-up. The same equations can be used for CANDU fuel calculations.

The TRU and U in FR frsh fuel at equilibrium are:

$$TRU_{in,FR} = f_{TRU,in} m\ P\ CF \quad \text{(Eq. 9)}$$

$$U_{in,FR} = f_{U,in} m\ P\ CF \quad \text{(Eq. 10)}$$

where $TRU_{in,FR}$ and $U_{in,FR}$ are the masses of TRU and U in the FR fresh fuel at equilibrium (t/yr), f is the fraction of U and TRU in fresh fuel at equilibrium, m is the mass of FR fresh fuel required to generate $1GW_e yr$, P is the electrical output per fast reactor (GW_e), CF is the capacity factor (%).

The FR fuel is assumed to be completely recycled. Although, additional TRU and U are required with each iteration of the FR cycle. For the second cycle, after one year of operation, the make-up amount of TRU required to be reprocessed for the FR fresh fuel is:

$$TRU_{mk,FR,n} = \frac{TRU_{in,FR,n}}{\varepsilon_{TRU,r}\varepsilon_{TRU,f}} - TRU_{out,FR,n-1} \quad \text{(Eq. 11)}$$

where $TRU_{mk,FR,n}$ is the mass of make-up TRU reprocessed for FR fresh fuel at equilibrium for years $2 \leq n \leq 60$; $TRU_{in,FR,n}$is the mass of TRU in fresh fuel at equilibrium for year n; $TRU_{out,FR,n-1}$ is the mass of TRU in FR used fuel at equilibrium from FR operation in year n-1; $\varepsilon_{TRU,r}$ is the TRU recovery efficiency of reprocessing and $\varepsilon_{TRU,f}$ is the recovery efficiency from fuel fabrication. The same equation is used to calculate the make-up amount of U required to be reprocessed for the FR fresh fuel, using the respective variables for U.

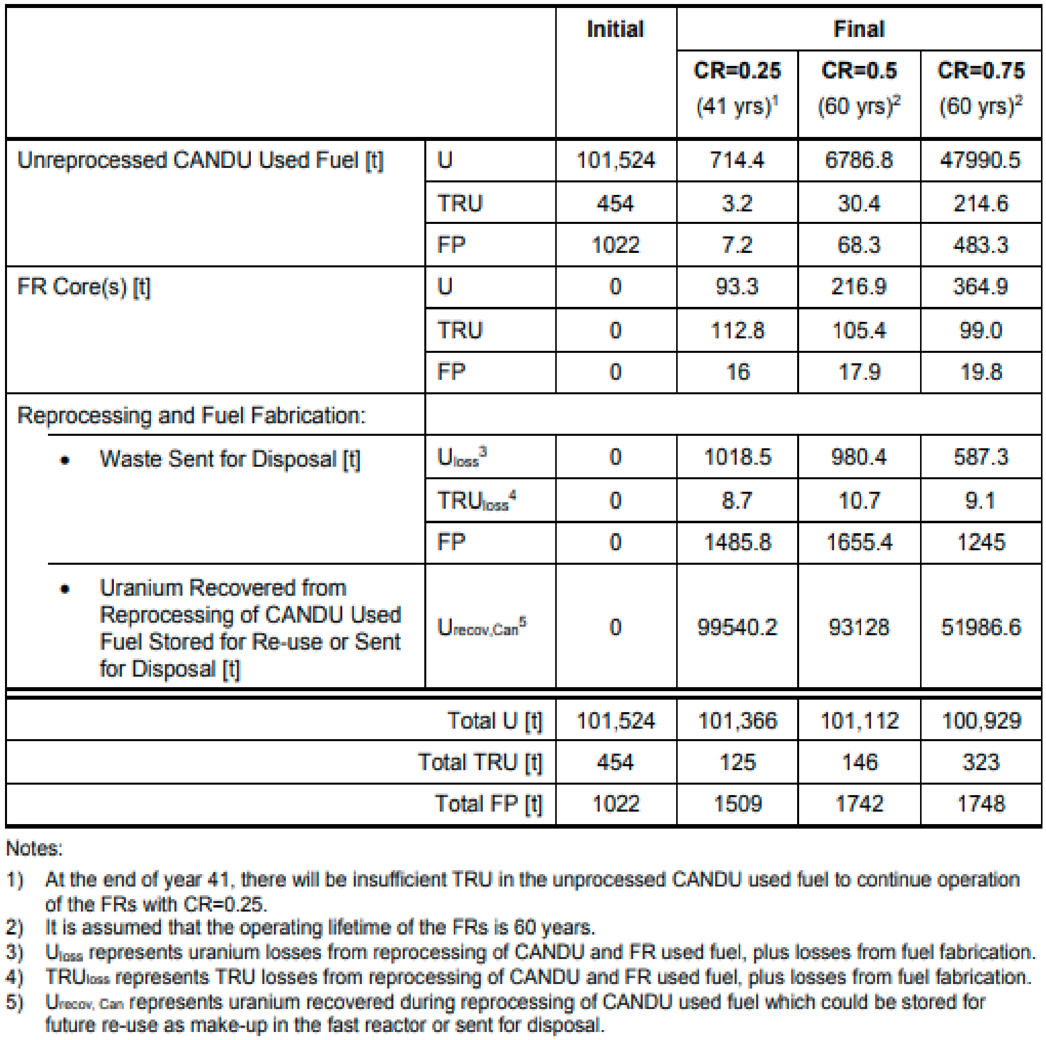
The TRU in the irradiated fuel in the FR core is estimated as:

$$TRU_{FR} = \frac{f_{TRU,in} + f_{TRU,out}}{2}(FR\ HM\ inventory\ -\ m\ P\ CF) \quad \text{(Eq. 12)}$$

where TRU_{FR} is the mass of TRU in the irradiated fuel at the start of the cycle; FR HM inventory is the mass of heavy metal in the FR core at the start of the cycle, which is estimated from the mass of heavy metal loading at equilibrium; m is the mass of FR fuel used to generate t_{HM}/GW_e; $f_{TRU,in}$ and $f_{TRU,out}$ are the fractions of TRU in the fresh fuel and used fuel at equilibrium, respectively.

Results

The NWMO performed two scenarios, one with two FRs with the sole intention of burning TRUs from CANDU reactors, the other was to replace the CANDU fleet with FRs and used the current inventory of CANDU spent fuel to fuel the FRs. Figure 23 shows the results of the second scenario.



		Initial	Final		
			CR=0.25 (41 yrs)[1]	CR=0.5 (60 yrs)[2]	CR=0.75 (60 yrs)[2]
Unreprocessed CANDU Used Fuel [t]	U	101,524	714.4	6786.8	47990.5
	TRU	454	3.2	30.4	214.6
	FP	1022	7.2	68.3	483.3
FR Core(s) [t]	U	0	93.3	216.9	364.9
	TRU	0	112.8	105.4	99.0
	FP	0	16	17.9	19.8
Reprocessing and Fuel Fabrication:					
• Waste Sent for Disposal [t]	U_{loss}[3]	0	1018.5	980.4	587.3
	TRU_{loss}[4]	0	8.7	10.7	9.1
	FP	0	1485.8	1655.4	1245
• Uranium Recovered from Reprocessing of CANDU Used Fuel Stored for Re-use or Sent for Disposal [t]	$U_{recov,Can}$[5]	0	99540.2	93128	51986.6
Total U [t]		101,524	101,366	101,112	100,929
Total TRU [t]		454	125	146	323
Total FP [t]		1022	1509	1742	1748

Notes:

1) At the end of year 41, there will be insufficient TRU in the unprocessed CANDU used fuel to continue operation of the FRs with CR=0.25.
2) It is assumed that the operating lifetime of the FRs is 60 years.
3) U_{loss} represents uranium losses from reprocessing of CANDU and FR used fuel, plus losses from fuel fabrication.
4) TRU_{loss} represents TRU losses from reprocessing of CANDU and FR used fuel, plus losses from fuel fabrication.
5) $U_{recov,\ Can}$ represents uranium recovered during reprocessing of CANDU used fuel which could be stored for future re-use as make-up in the fast reactor or sent for disposal.

Figure 23: System inventory for Scenario with 36 FRs and 103,000tHM of CANDU spent fuel [58].

The scenario with two FRs would not provide fast reduction of the CANDU TRU inventory, and it is estimated that it would take almost 1000 years of operation time for two FRs to burn through the 103,000 t_{HM} of CANDU spent fuel. Therefore, 15 generations of 2 FRs (or 30 FRs) would be required, during which more fission products would be produced by the FRs.

For the power generation scenario, there is insufficient TRU amounts remaining in unprocessed CANDU used fuel just after 40 years of operation for a CR of 0.25. After 40 years, the continued operation of 1 or two reactors would be able to sufficiently run using the remainder of the CANDU spent fuel. In both scenarios, the spent fuel would contain more fission products and fewer TRU; neither scenario is seen to have a net loss of the hazardous component of used fuel [58].

Gobien (2015) used the data above [58] to compare the current once through cycle with the proposed reprocessing and FR cycles [59]. The two fuel cycles are compared using radioactivity as a function of time and radiotoxicity as a function of time.

The waste inventory is shown in Figure 23, and upon reactor shut down, the remaining TRUs would either be disposed of or reprocessed for further use. It was estimated that 1486t of fission products, 1019t of uranium, and 8.7t of TRU were sent for disposal upon completion of the cycle with 36 FRs. The recovered uranium (99,540t) from the FRs was repurposed and not disposed of.

The total radioactivity is presented as Bq/kg$_{\text{wasteform}}$ and the radioactivity of the wasteform was divided by the mass of the fast reactor (400kg) and the CANDU (24kg), both shown in Figure 24. The FR radioactivity is dominated by the fission products, specifically Cs-137 and Sr-90, along with their daughters Ba137m and Y-90 for the first 300 years. From 300 to 1 million years, the radioactivity is dominated by Tc-99, and beyond is Zr-93 and Nb-93m. The CANDU spent fuel is initially dominated by the fission products with actinides dominating after a few hundred years. Due to a higher amount of fission products being loaded in the FR wasteform, the initial radioactivity is 1.5 times higher, but the FR radioactivity falls below the CANDU after the first few hundred years [59].

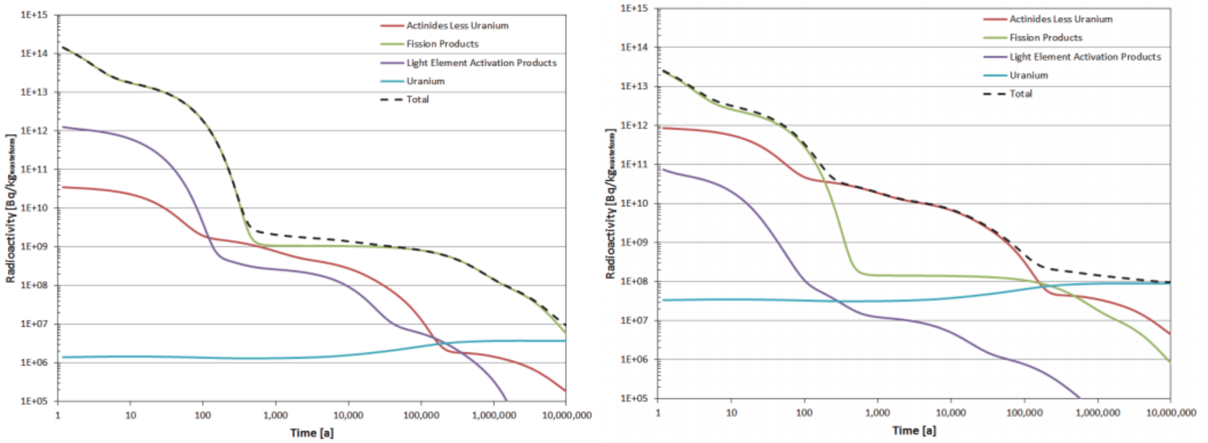


Figure 24: FR and CANDU Fuel Radioactivity [59].

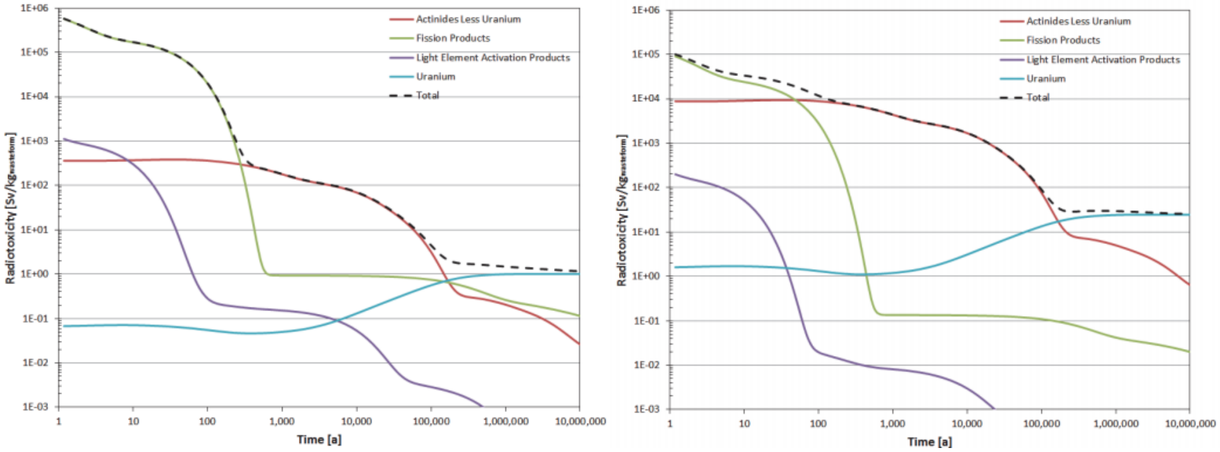


Figure 25: FR and CANDU Radiotoxicity [59].

Radiotoxicity from ingestion (Sv/kg$_{\text{wasteform}}$) was calculated by multiplying each radionuclide's radioactivity by that nuclide's respective ingestion dose for both the FR and CANDU fuel cycles (Figure

25). For both reactor cycles, the first few hundred ears are dominated by Sr-90 and Cs-137, while Am-241, Pu-239, and Pu-240 dominate between 1000 and 300,000 years.

2.5.4 SMRs and Canada's Fuel Cycle

Ottensmeyer (2019) performed an analysis on the potential relationship between CANDU reactors and SMRs in future developments [75]. Canada could implement SMRs throughout the country; however, many SMR designs require enriched fuel, which Canada does not make. At the current rate at which Canada is mining, exporting, and using the uranium, the 500,000 ton reserve will be exhausted by 2050 [75]. The current reservoir of CANDU spent fuel is approximately 60,000 tonnes; 240 tonnes of TRU, containing Np, Pu, and Am, which have been created during the exposure of natural uranium fuel within the CANDU reactors. For example, 240 tonnes of TRUs are able to start 24000 MWe fast SMRs almost doubling Canada's current nuclear power supply (13,500 MWe) [75]. If the chosen SMRs only consume the fissile components of the fuel (burners), the current uranium exploitation would be required by the CANDU fleet which would result in faster depletion of fissile material. Although, some SMR designs could supply CANDU with fissile material.

Reprocessing

The enrichment of U-235 is costly and proliferation-prone as it was initially developed for the creation of nuclear weapons [75]. The PRISM reactor developed by GE-Hitachi indicates that all CANDU spent fuel could be consumed while maintaining and transmuting fissile components. Therefore, enrichment facilities may not even be required as TRUs could be extracted economically, and transmuted in a proliferation-resistant manner [75].

Many SMRs require fissile concentrations of 15% to 20%, as small reactor cores naturally lose more neutrons before the opportunity to fission. Some SMRs require U-235, while other designs use concentrated TRUs from a used fuel source. TRU-based fuel contains all actinides and is a mixture of unpurified elements, which is essentially proliferation resistant as Pu-239 and other fissile isotopes are diluted with 30% fertile elements, which are nearly impossible to separate [75].

As of 2019, the estimate for the DGR lies between $18.3 and $28.4 billion, capacity depending. The current used fuel management cost of each CANDU fuel bundle is estimated to be $3,654. Comparison of the reprocessing methods analyzed are presented in Table 33. All reprocessing methods were limited to a CANDU reactor fuel load of 100 tonnes. The FVM met the through-put criterion in 4 months, permitting a larger yield. Each output was in metal form, ready for alterations depending on future fuel requirements. The modified PUREX was changed to leave excess plutonium, which is extremely costly, which explains the common assumption that the DGR is a more economical path.

Table 33: Cost breakdown of recycling used CANDU fuel by various methodologies [75].

	Mod. PUREX	Pyroprocessing	FVM
Capital- Overnight cost (land,constr,license, etc. ~30%)	\$186.9 M	\$80.2 M	\$11.5 M
Total annual cost (OM&A, taxes, 5% interest for 40 yrs)	\$37.2 M	\$11.2 M	\$5.6 M
Annual Thru-put (bundles)	5000	8050	5000
Cost per bundle	**\$7,432**	**\$1,368**	**\$1,114**

Neutron interaction in SMRs

Although SMR designs are proprietary, published information was used to estimate certain reactor characteristics to make “-like” reactors. The calculations performed took into account all the relevant fuel isotopes as well as structural components and heat transport fluids. Although Ottensmeyer examined multiple reactors, only the IMSR, NuScale, and ARC-100 reactors are recorded here (Table 34).

Table 34: Operational core characteristics for chosen SMRs and CANDU-like reactor [75].

	CANDU			IMSR			NuScale			ARC-100		
Reactor Spectrum	Thermal			Thermal			Thermal			Fast		
Neutron Energies	0.025 eV	100 keV	2 MeV	0.025 eV	100 keV	2 MeV	0.025 eV	100 keV	2 MeV	0.025 eV	100 keV	2 MeV
New-Neutron Ratio	1.34	0.136	2.40	1.95	-	-	1.95	-	-	1.12	1.24	2.46
#U_{235} Nuclei Used	1	1	1	1	-	-	1	-	-	1	1	1
#New Fissile Ratio: $U_{238} \rightarrow Pu_{239}/Pu_{240} \rightarrow Pu_{241}$	0.52	12.0	4.48	0.12	-	-	0.043	-	-	0.720	0.685	0.268

NuScale and ISMR are thermal small reactors and therefore only thermal neutron energies were calculated. The higher numbers for the new-neutron ratio indicates the higher initial fissile fuel content is compensated by controlling the neutron absorption [75]. Once a controlled neutron equilibrium of 1 is achieved, the interest in fuel consumption is the number of new fissile isotopes created for every fission, also known as the conversion ratio (last row). For thermal reactors, the fissile content at thermal energies cannot be replaced; however, for CANDU the ratio increases as more U-235 isotopes are transmuted to Pu-239, which yields 10% more neutrons per fission than U-235. The thermal SMRs have very low conversion ratios in comparison due to the high fissile content of the fresh fuel. The U-235 absorbs many neutrons that would be available to convert the fertile U-238 [75].

The neutronic behaviour in CANDU is dependent on the neutron's energy. A nascent neutron can have an energy around 2.1 MeV, which is moderated by the surrounding heavy water. The neutron has opportunities for many interactions throughout its path. If a neutron would re-enter the fuel with the energy of 100keV, a fission event would be considered a rare occurrence, and therefore the CANDU has a new-neutron ratio of 0.136 at this energy level. However, the cross-section of U-238 is much higher at that energy which results in a higher conversion ratio. But with few new neutrons being produced, a reactor would not be able to operate at this energy alone. For 2 MeV, there is a high fission cross-section for the fertile U-238 which adds to the total new-neutron yield. While U-235 is the lowest fissile cross-section, the new neutron ratio and the conversion ratio are 2.40 and 4.48, respectively. However, due to the heavily moderated CANDU design, the operation of a CANDU reactor in the high neutron energy range is not sustainable [75].

The energy loss of nascent neutrons of high energy can be avoided by replacing the moderator/coolant material. Currently, the most popular liquid coolant is sodium, which has been used for over 400 reactor-years in the fast spectrum [75]. The ARC-100-like reactor that was examined uses liquid sodium in the fast neutron spectrum. The SMR requires a high enrichment of U-235 and achieves a low conversion ratio at 2MeV. This is likely caused by the high cross-sections of the highly fissile material (including U-238 in the fast region), which leaves few neutrons to be captured and transmuted. As one could expect, at 100 keV, the reduction in energy causes the fission of U-238 to drop, which increases the production of Pu-239, increasing the conversion ratio. In fast reactors the 100keV range is where the neutron energy utilization peaks [75]. Although the ARC-100-like reactor does not achieve the fissile isotope equilibrium (CR=1), there is a shift in the neutron yield of 2.55 per fission at 2 MeV to 3.17 at 100 keV; this provides the reactor with a higher neutron ratio than the new fissile ratio, keeping the reactor operational at lower energies.

Most SMR designs require enriched fissile fuel, which is becoming more invaluable. However, although fast-spectrum reactors (ARC-100) require the enriched fuel, from Table 34, it is clear that the enriched fuel component is well maintained and can be augmented between fuel cycles. Once it is operational, the ARC-100 can contain fertile U-238 and non-fissile isotopes, which can fission at high energies or can transmute for future fissile consumption [75].

Fuel Costs

Since Canada currently refuses to develop uranium enrichment facilities, the acquisition of enriched fuel must be from a foreign source, and from local TRUs from CANDU spent fuel. Table 35 uses pyroprocessing and fluoride volatility to determine the cost for a hypothetical fuel requirement of 20 tonnes of uranium fuel. In all scenarios the end result has the same neutron yield; 20% enriched U-235 fuel and 20% TRUs.

Table 35: Comparison of enriched-fuel costs for SMRs [75].

	Total Tonnes	Fissile/TRU	CANDU used Fuel Bundles	Cost Per Bundle CAD	Total Cost CAD (million)
20% U_{235}	20	4	-	-	146.65
20% TRUs Pyro*	20	4	50,000	1,368	68.40/77.19*
20% TRUs FVM*	20	4	50,000	1,114	55.70/62.85*

*corrected for fissile content

For the GE-Hitachi PRISM core, the costs in Table 35 are nearly applicable; for other reactors, costs can be adjusted accordingly. The operation of a SMR is more economical and is between 1.9 or 2.5 times more economical than purchasing enriched-235 fissile fuel [75].

2.6 Hybrid Systems

Although there is a clear incentive to reduce the use of fossil fuels, the ability to easily adjust the electricity output is a feature that cannot be made by nuclear reactors alone. Variable electricity is currently dominated by the fossil fuel industry due to the affordable storage and the characteristics of the fuel itself. Fossil fuels can be adjusted based on demand, and nuclear reactors are not as easily manipulated; therefore, nuclear reactors generally provide a baseload. Due to the high capital cost of nuclear and renewable energy sources, it is difficult to consider replacing fossil fuels with cleaner options [15].

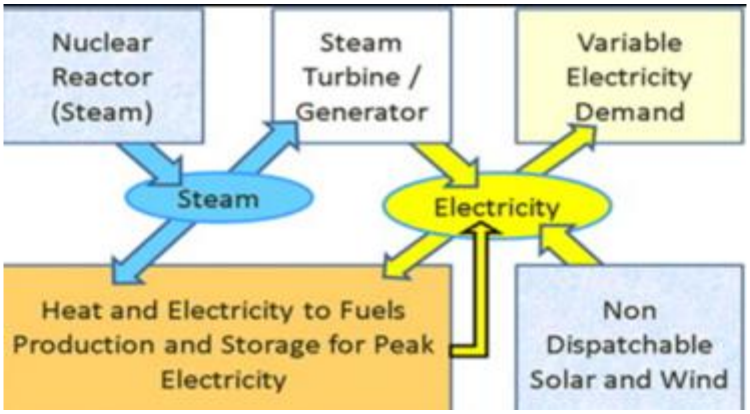


Figure 26: Hybrid nuclear-renewable system for variable electricity production [15].

Hybrid energy systems may be the approach to satisfy a variable energy demand while reducing greenhouse gas emissions (Figure 26). Hybrid systems behave in the following manner: nuclear and renewable energy sources operate at full load; renewable sources send electricity directly to the grid; steam from the nuclear source can be sent to turbines or other applications; varying demands in energy can be met with small economic penalties [15]. Due to the geographical dependency of geothermal and hydroelectric sources, many hybrid systems focus on renewable sources to pair with nuclear due to the versatility of the technologies. The hybrid technologies are able to address the variations in seasonal demand, but can also meet changes in hourly demand; the only question is the amount of storage required.

2.6.1 Electrical Storage

Applications combining the source for electricity and steam have the potential to economically absorb large quantities of variances for these resources. The hybrid technologies are designed to address variances in demand, whether they occur hourly, weekly, or seasonally. Short term variations in demand can be easily addressed with batteries, seasonal options are more scarce as fossil fuels prevent the need for such technologies. The non-electric markets could capitalize on the hybrid systems by using the process heat for other purposes: desalination, district heating, and high temperature heat for hydrogen manufacturing. The business would not necessarily fall under the energy production governance, and if the payback period is small, the financing requirements of fast growing populations would easily be met [76].

Since the US aims to reduce greenhouse gas (GHG) emissions by 80% by 2050, electricity would have to provide more energy than it is currently. In order to provide insight into future energy demand, California's electricity storage requirements were estimated for three resources, nuclear, wind, and solar. The hourly, daily, and weekly storage demands were compared for the three electricity sources. For daily and weekly time periods, storage technologies must be designed to hold the potential for at least the time period in question. Although there are technologies that exist to meet these requirements (smart grids, pumped hydro, etc.), seasonal storage is much more challenging to address [15]. If nuclear systems were strategically placed where production and energy demands were closely matched, nuclear energy would have a competitive edge compared to wind and solar, due to the reduced amount of storage systems required to meet variable demand.

For solar and wind systems, storage requirements are dependent on latitude and climate. Specifically, not only do the technologies themselves depend on the atmosphere in which they are stationed, but the electrical storage does as well. For example, sites near the equator would achieve great solar input, compared to sites with higher latitude and lower elevations. California is able to achieve a 21% storage rate if it combined a thermal daily storage solution with its solar power [15]. Due to inexpensive short-term storage options, a solar-thermal system would have a competitive edge compared to photovoltaics.

2.6.2 Heat Storage

Geothermal heat storage could take spent heat from a source, such as a nuclear reactor, at times of low electricity demand and store it in a subterranean cube of rock to create an artificial GWyr heat source. The heat from the rock could then be used during intermediate and peak electricity production by integrating a geothermal power system. The initial heat source could be a nuclear reactor (LWR, PWR, etc.), or a high-temperature solar-thermal system. If an LWR was implemented with the geothermal heat storage system, the heat transfer fluid would be pressurized water. Cold water would be heated in the heat exchanger from the primary reactor circuit's hot water. The heated water would then be injected into the top of permeable rock located underground, which would disperse the heat within the rock. Cold water would be removed from the rock and cycled back to the reactor's heat exchanger. When there is an increase in electricity demand, water would be injected in the bottom of the permeable rock to be heated as it passes through, which would then flow to the geothermal plant to produce more electricity than the nuclear facility could provide. Cold water would then be re-injected at the bottom of the rock [15]. This particular storage system allows for variations in rate of heat addition, storage capacity, and heat extraction, to meet the local demands.

The capital cost of the heat-storage system is lower due to rock being the storage material. Also, due to the lower cost of heat storage which is one third that of electricity, the cost of inefficiencies are less than those that store electricity [15]. A PWR-geothermal system was examined for potential issues(Table 36). Since it is not possible to insulate rock underground, the heat will transfer to cold rock; however, the heat losses will decrease as the system size increases. Meaning, as heat is added to the rock, the surface to volume ratio of the hot rock decreases, and the larger systems would have smaller relative losses. In order to keep losses within a few percent of total for seasonal storage, the total capacity should be at least 0.1GWyr. Pressurized water is ideal if the heat source is below 300°C. Since the secondary side of the CANDU heat transport cycle is less than 300°C, the water would need to be pressurized. At higher temperatures, water causes the dissolution of silica, so supercritical carbon dioxide can be used instead.

Table 36: Baseline assumptions for Heat Storage System [15].

Technology	Chosen Option	Constraint
Powerplant	PWR	T_{max}=273° C
Geofluid	Pressurized Water	P_{min} = 5.7MPa
Nuclear Coupling	Bypass Primary Side	Intermediate Heat Exchangers
Underground Reservoir Type	Enhanced Geothermal	Underground Stimulation Required to boost permeability
Power Cycle	Binary Power Cycle	Efficiency
Geology	Sandstone	Rock Properties
Depth	1-1.5km	Pressurized liquid water needed
Method to Create Permeability	Hydraulic Fracturing	Max permeability = 2D

To permeate the rock, three methods were identified, which would be chosen based on local geology and economics. Hydrofracture involves the drilling of wells into an already permeable rock. High pressure water is injected into the rock along with proppants, such as sand, which opens fractures within the rock. The proppants act as fracture stabilizers and maintain the width of the fracture when the injections stop. Cave-block mining creates underground rubble-zones with macroscopic fractures. Establishing the fractures would be performed similarly to standard mining operations. Levels would be separated by 50-500m, with explosives to be inserted at the bottom and top of each silo. Rubble chimneys would be created by these controlled explosions that would become primary heat storage columns, as the resulting crushed rock has high permeability. Selective dissolution is the removal of specific materials to create high permeability zones. For example, a geothermal heat system could be used to remove heavy oil with steam, which would lower the oil viscosity and the oil would drain to recovery wells and leave porous rock [15]. As there are existing fields with varying amounts of oil remaining within the rock, there are incentives to retrieve the heavy oil. One could even gradually move the geothermal reservoir over time to increase the oil recovery, which would be limited by the distances from the nuclear reactor.

2.6.3 District Heating, Desalination, and Off-Grid Communities

Population centres, especially in small villages and towns off-grid, require energy for both electricity and district heating. District heating requires the plant to be in close proximity to the demand. For off-grid communities waste heat can be used, which would increase the energy efficiency of the plant [79]. In 2016, plants which combined heat and power contributed 22% of the world's fossil fuel electricity, and 16% of the global electricity [77]. Although SMRs could produce either heat or electricity, the reactors that produce both would gain more profits, but capital costs are 5% higher than electricity-only units [77]. SMRs also have a potential to desalinate water, which would be beneficial for regions where the reservoir of natural fresh water is depleting; however, the Canadian market for desalination is small or non-existent [79].

Currently, off-grid communities are present in every province in the country, as shown in Figure 27. Due to their remote locations, many require the use of small diesel generators (Table 37).

Figure 27: Map of remote communities in Canada [79].

Table 37: Fossil fuel generation in off-grid communities in Canada by province/territory [79].

Province/ Territory	Total Number	Primary Diesel	Connected to Grid	Primary Renewable	Hybrid Diesel/ Renewable	Other FF
AB	2	1	1	0	0	0
BC	75	57	6	4	1	0
MB	7	7	0	0	0	0
NB	0	0	0	0	0	0
NL	29	26	1	0	1	0
NS	0	0	0	0	0	0
NT	37	21	8	4	0	2
NU	25	25	0	0	0	0
ON	38	31	0	0	1	0
PE	0	0	0	0	0	0
QC	45	24	20	1	0	0
SK	1	1	0	0	0	0
YT	21	4	16	0	0	1
TOTAL	280	197	52	9	3	3

Clearly, there is a demand for power in these communities. However, as each community varies in size, the amount of energy that is demanded varies. Figure 28 shows the power demands for each community. Many of the communities fall within 0.1 and 2MWe. The micro SMRs or battery-sized SMRs would be best suited for these reactors, such as the SEALER or the U-Battery. If the population were to suddenly increase and demand more power, the modular design of these reactors would allow for the ability for supplementary SMRs to be developed in the same plant.

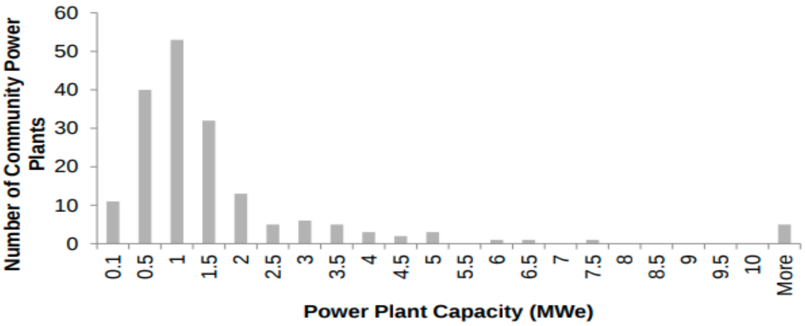


Figure 28: Histogram of current power plant capacity in Canadian off-grid communities [79].

2.6.4 Hydrogen Production and Storage

Hydrogen is an integral element used in many operations, including heavy oil conversion, desulfurization, and many more. For example, the US requires more than 10 million tons of hydrogen every year. The conversion of water to hydrogen has been a commercial technology for more than a century [15]. When there is a reduction in energy demand, and a resulting decrease in electricity price, the low-priced electricity can be converted to low cost hydrogen; however, the electrolyzer has a high capital cost and prevents this from occurring. If low cost electrolyzers can be produced: all electricity produced would be consumed, there would be no negative price for electricity, and large-scale renewables would be more viable. As shown in Table 38, only certain industries and markets could really benefit from hydrogen production [2].

Table 38: Characteristics and value (X) of nuclear hydrogen relative to other methods of hydrogen production for specific applications [2].

Application	O_2	CO_2-free	Large-Scale	Bulk Storage	Heat	Institutional
Small Local User		X				
Pipeline		X	X			
Chemical Industry		X	X		X	
Refinery		X	X		X	
Liquid Fuel Prod.	X	X	X		X	
Peak Electric Prod.	X	X	X	X	X	X

An alternative to the expensive electrolyzers are high-temperature electrolysis (HTE), which uses steam and electricity to convert water to hydrogen and oxygen, instead of electricity alone. The high temperature operations allow for the system to be reversed, and use hydrogen for electricity production during times of high demand. Industrial markets could prosper with the optimization of the electricity-to-hydrogen process; however, the reversible option could potentially reduce the cost of hydrolysis. Many industrial operations have implemented gas turbines to avoid blackout scenarios during peak hours, however, they are rarely required (>100h/year) [15].

The typical hydrogen production plant is between 25-50MWe. A Midwest electricity grid was examined where nuclear reactors provide heat (2857MWt) along with electricity (15438MWe), while producing a stream of hydrogen at 120.81 kg H_2/s. In times of high electricity prices, the reactors would produce electricity and the hydrogen reservoir would act as a fuel cell, and could produce 28500MWe with an efficiency of 40%. One significant limitation of hydrogen production is the location of each facility, which would need to be with or near a nuclear power station [15]. There are many potential reactors that can be implemented to the thermochemical cycle for hydrogen production, one just needs to determine the scale. The IAEA formed a collaborative project to determine the global readiness for the nuclear and thermochemical technologies to be combined on a larger scale [81]. Some countries have their own hydrogen production designs; however, the nuclear reactor is only providing heat for the thermochemical process. Therefore, depending on the hydrogen production technology, the only deciding factor is the heat required to achieve the splitting of water [81].

Similar to natural gas, hydrogen is stored underground at low cost, which allows for seasonal storage. As HTE is still in development, the primary sources of low-cost electricity can still be provided by wind and/or solar in the spring and summer months. Based on the experience of the natural gas industry, storing H_2 will be inexpensive [80].

The transportation of hydrogen may be required for processes that do not involve future electricity production. Ammonia production is required in many parts of the world for biomass, specifically fertilizer. The hydrogen-fuelled cars are also being developed, with liquid fuels of methanol, ammonia, and other hydrogen carriers to avoid the release of greenhouse gases. The transportation industry relies heavily on H_2, with the exception of the electric car [80].

2.6.5 Nuclear and the Oil Industry

Changes are being made to the way liquid fuels (gasoline, diesel, jet fuel) are being produced. The depletion of the high-quality crude oil is forcing the industry to turn to heavier feedstocks (heavy oil, tar sands, shale oil, and coal). The heavier feedstocks produce more greenhouse gas emissions per litre of fuel. The greenhouse gas emissions could be doubled per litre of fuel consumed as alternative feedstocks replace light crude oil [82].

2.6.5.1 Oil Shale and Kerogen

Oil shale contributes between 30% and 50% of the world’s fossil fuel resource, with the US having the largest reserves of oil shale. Oil shales have the highest concentration of fossil fuels, with the potential to produce more than a million barrels of oil per acre [15]. As there is great reliance in Middle Eastern oil, the ability to reap the elements from oil shale would ease the dependence on the Middle East.

Oil shale does not simply contain oil, but rather contains kerogen which must be converted. Kerogen is the fossilized plant residue, a precursor to oil and coal. In order to do so, the kerogen must be slowly heated (to 370°C) to a high-quality light oil, along with light gases. Large oil companies have been developing in-situ processes which use electricity to heat (resistance or microwave) the kerogen. The slow heating processes without oxygen exposure results in high quality light oils, leading to high yields of gasoline, diesel, and jet fuel.

Nuclear could provide high-temperature steam in pipes to provide heat to the kerogen in situ. Table 39 shows the comparison of nuclear and gas driving 50 thousand barrels of oil per day. Since the recovered natural gas is used to fuel the process, there is a large reduction in gas produced by the natural gas option. There is a factor of 10 reduction in carbon dioxide emissions from the nuclear-shale oil process instead of the natural gas. As of 2013, the US was importing approximately 10 million barrels of oil each day. If the US were to replace all oil imports completely, 330 modular reactors with 200GW$_t$ could produce sufficient shale oil.

Table 39: Heat balance for recovery of 50 thousand barrels of shale oil per day with heat from burning natural gas, or heat from a high temperature reactor.

	Input	Gas (Btu/D)	Electricity (MW$_e$)	CO_2 (tonne/D)	Oil (m^3/D)
Gas	91.9 Btu/D	$29.5\text{x}10^9$	105	5982	7950
Nuclear (HTGR)	1007 MW$_t$	$121.4\text{x}10^9$	74	483	7950

Due to the low thermal conductivity of rock, the industrial process of heating the rock would require months to years. However, due to the slow heat transfer, the rock does not require constant heat. The heat could only be applied during hours of low electricity demand (at night), and use the heat to produce electricity during the day. US shale oil primarily exists in the west where solar and wind are prominent. Therefore, a hybrid system could exist where variable electricity provided by nuclear-shale production would support the renewables in the low electricity cost market. In terms of a greenhouse footprint, the hybrid system is desirable, as other processes require fossil fuels to further produce fossil fuels. Also, the production of carbon dioxide is reduced, as the underground refining results in carbon as a residue which remains underground along with impurities, which are ultimately sequestered.

3 Objectives and Methodology

The objective of this study is to examine the technologies and strategies for Canada's nuclear industry: reprocessing spent fuel to close the fuel cycle; reducing the radioactive waste inventory through transmutation; implementation of hybrid systems to allow for accessible, clean, and sustainable energy sources throughout the country.

3.1 Reprocessing Calculations

PUREX is the most commonly used method throughout the globe, but due to the stream of plutonium that is produced, Canada is unlikely to invest in the technology; therefore, other methods will be examined.

Due to the number of different reprocessing techniques that are currently in development, and variations of those technologies, a simple calculation which determines the theoretical efficiency of each method has yet to be developed. There are multiple studies which show experimental data throughout each stage of the process to show losses within most methods. However, studies are currently focusing on PWR spent fuel, which would not be of much interest in Canada.

Since the majority of Canada's spent fuel reservoir contains oxide fuels, implementing pyroprocessing would involve an additional step, where the fuel is downgraded to a metal. Due to the extra cost, exposure, and potential losses, only one pyroprocessing method will be examined. The separation efficiency will be compared to that of the fluoride volatility method. The predicted efficiencies, the contents of the different streams, the cross section traits of each of the streams, and the variances in cost, will be discussed. The reprocessing calculations for the FVM and pyroprocessing will use CANDU fuel after it has been cooled for 1 year.

Using the methodologies discussed in Section 2.1, the same procedures were used to determine the efficiencies for CANDU spent fuel. The Harvard Study was also used to compare the cost of both Pyroprocessing and the FVM within Canada.

3.2 Transmutation Calculations

Every reactor is theoretically able to transmute fission products; however, the reinsertion of these isotopes may not be worthwhile. Each isotope has its own absorption cross-section which identifies the probability of neutron interaction depending on the neutron's energy. Many countries are exploring ways to transmute long-lived wastes: actinides, especially plutonium, minor actinides (Np, Am, etc.), and certain fission products. The incineration of plutonium draws a lot of attention due to the fissile properties, as well as the potential threat it poses outside of reactors [48].

The neutrons that are produced by fission are generated with an average energy of 2MeV, but can reach up to 10MeV. The presence of heavy and light nuclei causes the neutrons to lose energy as they scatter. Most of the transmutations that occur within fission reactors are caused by neutron absorption, resulting in fission (generating fission products), or emission of γ-rays. Various radioactive decays can occur (i.e. β^+, β^-, α), which result in a change in charge and number of neutrons.

A critical reactor at time *t* has a field of neutrons that can be described by flux density $\boldsymbol{\Phi}$(r,Ω,E,t) where *r* is the location, Ω is the direction of movement, and *E* is the neutron's energy, which is a solution to the homogeneous Boltzmann neutron transport equation with power normalization. Integrated in the scalar direction and by energy, results in the space-time dependent neutron flux, $\boldsymbol{\Phi}$(r,t). The neutron spectrum is defined by χ(r, E, t)= $\boldsymbol{\Phi}$(r,E,t)/ $\boldsymbol{\Phi}$(r,t) with χ(r, E, t)dE as the fraction of neutrons with energies between E and dE, at location r and time t.

In order to determine parameters for the creation and transmutation of nuclei, one should concentrate on one nuclei and its potential reactions. Pu^{241} will be used as an example: Pu^{241} (β^-) Am^{241}, $Pu^{241}(n,\gamma)Pu^{242}$, Pu^{241}(n, f) *FP*, Pu^{240} (n, γ)Pu^{241}[48].

The rate of change of the number of Pu^{241} (N_{Pu241}) is:

$$\frac{d}{dt}N_{Pu241}(t) = -\lambda N_{Pu241}(t)_{decay} - (\sigma_{c,Pu241} + \sigma_{f,Pu241})\,\phi N_{Pu241}(t)_{transmutation} + \sigma_{c,Pu240}\phi N_{Pu240}(t)_{creation} \qquad \text{(Eq. 13)}$$

where $\sigma_{x,n}$ is the microscopic cross-section for the reaction x at nuclide n, and λ is the decay constant. The effective reaction cross-sections $\sigma_{x,n}$ are given by:

$$\sigma_{x,n}(r,t) = \int_{0.001eV}^{10MeV} dE\ \sigma_{x,n}(E)\chi(r,E,t) = \int_{0}^{23.03} du\ \sigma_{x,n}(u)[E\chi(r,E,t)](u) \qquad \text{(Eq. 14)}$$

where lethargy $u=\ln(E_o/E)$, and E_o = 10MeV. The dimensionless lethargy distribution $\boldsymbol{\phi}$(u) = [Eχ(r, E, t)](u) is typically referred to as the neutron spectrum. The effective cross-section of each isotope is a function of the reaction type's cross-section, and the reactor's space and time neutron spectrum χ(r, E, t). Also, the rate of change of the cross-section is dependent on the neutron flux $\boldsymbol{\Phi}$(r,t), the local power at time *t*, and the number densities of the reacting nuclides. Using the equations above, a neutron spectra can be calculated and presented for thermal and fast reactors, followed by the effective cross-sections. The regions of resonances can be replaced with mean values to provide a simplified curve [48]. Using the method above, one could calculate the change of nuclide by hand; however, total accuracy in the calculation is discarded. Ideally, one would use a program such as "Serpent" to calculate the depletion equations, transmutation cross-sections, and nuclide inventory [49]; however, due to the rigidity of scope, these methods will only be discussed on the theoretical level, and simplified to the transmutation rate.

The experience discussed in Section 2.2 lead to a further understanding of the transmutation process; however, as aforementioned, transmutation calculations were not performed in this study.

3.2.1 Transmutation Rate

As mentioned in Section 2.2, the radiotoxicity of the actinides being sent to the repository should not exceed the sum of radiotoxicity being eliminated:

$$\mathrm{tox}_A < \mathrm{tox}_U \qquad \text{(Eq. 15)}$$

where tox_U is the sum of all uranium isotopes being burned within the nuclear reactor. Since the Long-Lived Fission Products (LLFPs) have a higher mobility in the repository, and thus have higher risk of exposure, it is ideal to have a reduced inventory of LLFPs. There is no quantitative criterion yet; however, a margin of one order of magnitude was established [26]. During transmutation within a reactor, fission products (unlike TRUs) do not contribute supplementary neutrons; therefore, the presence of fission products in a reactor would be a neutron consumption process. The neutron consumption is an important parameter to consider and assess when determining the sensibility of transmutation within a nuclear system [26]. The transmutation rate of a nucleus is characterized below:

$$T_J^{Trans} = \frac{ln2}{\sigma_{n,\gamma}^{J}\phi * 3.16 * 10^7}\mathrm{yr/s} \qquad \text{(Eq. 16)}$$

where T_J^{Trans} is the time required to incinerate *half* of the initial mass, $\sigma_{n,\gamma}^{J}$ is the cross section (barns), and ϕ is the neutron flux (n $cm^{-2}s^{-1}$). Therefore, transmutation of fission products would only be feasible if the transmutation rate is much less than the natural rate of decay. One can use the above equation to determine whether or not an isotope should be included in future transmutation considerations. The transmutation rate was calculated for CANDU spent fuel components in Section 4.2.1.

3.3 Policy Review

Current Canadian policies are adequate for the infrastructure in place, which mainly consists of thermal reactors, natural uranium fuel, and immovable reactors. The potential introduction of enriched fuel on a larger scale may require an adjustment of policies surrounding enrichment facilities in Canada. The introduction of movable reactors, especially those placed in the north, would require a new policy that would ensure not only the safety of Canadian citizens, but the safety of the fragile ecosystems.

The handling of enriched fuel, the reprocessing of spent fuel, and the transmutation of actinides would introduce new operations in Canada. Canadian policies surrounding enrichment and reprocessing are non-existent or opposed to the development of such technologies. Therefore, the grey areas in the policies may need to be filled in order for Canada to move into the twenty-first century, support GENIV technologies, and take advantage of more sustainable energy systems. However, in order to have new technologies approved, proof of strong proliferation resistance is required by all of the reviewed reactor designs and associated technologies.

Although a closed fuel cycle has been implemented in other countries, as discussed in Section 2.5 the concept would be new for Canadian soil. The NELCAS was used to determine the environmental impacts of closing the CANDU fuel cycle in Section 4.3.1.

Hybrid technologies are a relatively new concept; however, the opportunities for Canadian deployment and integration are numerous. All potential hybrid options within Canada will be explored and paired with potential SMRs, if applicable, in Section 4.4. The economic, societal, and environmental advantages will be investigated in order to thoroughly explore the sustainability of the systems.

Policies of countries that have well established strong nuclear presences will be discussed in Section 4.5. In order to keep alignment with the IAEA, reports of international requirements to which each county must adhere shall be discussed as well. The other countries' policies regarding reprocessing procedures, enrichment procedures, and Small Modular Reactor deployment could be used as potential guidelines for producing a Canada-specific policy.

4 Results

4.1 Engineering of Reprocessing

The nuclear fuel discharged from nuclear reactors is the main contribution to nuclear waste, especially where reprocessing does not take place. The main contributors to the hazardous waste are plutonium, neptunium, americium, curium, and some long-lived fission products (iodine, caesium, and technetium) [17]. Partitioning and Transmutation are considered ways to reduce the volumes and radiotoxicity of the waste. The long-term risk of a geological repository is dominated by the fission products, which tend to be more mobile than actinides [17]. The risk involves the release of radionuclides into the biosphere. However, the reprocessing of spent nuclear fuel would produce secondary waste, contaminated by long-lived radionuclides.

Most of the global uranium is sourced from Canada, Australia, and Kazakhstan. Since Canada not only uses natural uranium, but has easy access to uranium, it can be perceived that there is no need for reprocessing in Canada. However, reprocessing has many advantages that may benefit Canada's nuclear program in the long term.

4.1.1 Adapting Pyroprocessing Material Flow for CANDU

In the analysis performed by Ohta et al. [46], the electroreduction of the spent oxide fuels is simplified by dissolving an oxide ion from a cathode into the molten salt. The ion acts as a carrier of charge and therefore almost all salt-soluble fission products are dissolved and removed from the spent fuel basket. Spent LWR fuel was used for this analysis with the intention of theoretically providing Fast Breeder Reactor (FBR) fuel. Although LWR spent fuel has a smaller amount of plutonium and fission products compared to the FBR's spent fuel, attention is drawn to the handling of the uranium; the surplus of uranium will not be processed until required by LWRs and FBRs. Due to this assumption, the reduction and refining processes were simplified, as the collection of uranium occurred as an oxide [46].

A flow diagram of the adapted pyroprocessing of LWR spent fuel is shown in Figure 29. In the preprocess, or head-end process, any rare gases are extracted that may have been trapped in the fuel rods or non-fuel materials. Although the decladding treatment is not required for electrical conductivity, the pre-treatment resulting in fine grains or powder is easier to oxidize. The electrochemical reduction (shaded area) involves the recovery of UO_2 which is the main element of spent LWR fuel. Once the UO_2 is removed, the remaining elements that were not salt-soluble can be removed in additional processes. The resulting product is a low-decontaminated actinide alloy that contains a variable amount of metallic fission products. The metallic alloy can then be processed for FBR metallic fuels.

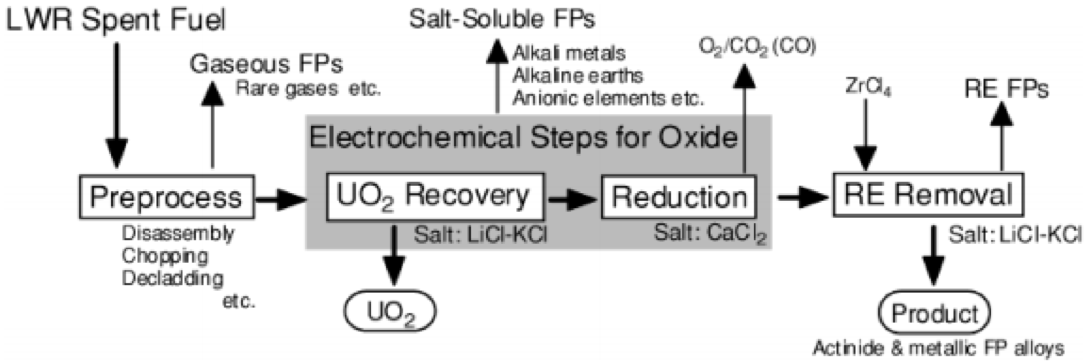


Figure 29: Flow diagram of pyroprocessing of LWR spent fuel by electrochemical reduction technology [46].

The decision to remove UO_2 from the spent fuel is to maximize the impact of plutonium within the FBR fuel; however, due to the small amount of plutonium present, it would require enrichment before insertion. Also, due to the reduction in volume of product, the capacity requirements for future processes is drastically reduced. The presence of noble metals (e.g. Ru, Rh, Pd) within either the recovered UO_2 or the product is dependent on the anodic dissolution of UO_2. If the anodic dissolution maintains below 70% of the total UO_2 then the recycling of noble metal FPs can be attained. The electrochemical reduction step and the rare earth material removal steps are performed in a similar fashion as described in Section 2.1.2.

The material balance was analyzed for both MOX and high-burnup UO_2, with discharge burnups of 40 and 48 GWd/t, respectively. Since CANDU fuel has a spent fuel burnup of 7.5 GWd/t, but does not contain PuO_2 within its fresh fuel, the high-burnup UO_2 will be considered a closer correlation to the CANDU's material flow; also, the removal of UO_2 will be ignored to align with Canadian proliferation resistance values. Table 40 shows the fuel discharged from a PWR which corresponds to the mass flow of Figure 30.

Table 40: Composition of high-burnup UO2 PWR spent fuel discharged with 5 years cooling time [46].

Contents	High-Burnup UO_2 wt%
Discharge Contents: U/ Pu/ MAs/ FPs	93.8/1.2/0.1/4.9
U^{234}/ U^{235}/ U^{236}/ U^{238}	0.0/1.3/0.7/98.0
$Pu^{238}/Pu^{239}/Pu^{240}/Pu^{241}/Pu^{242}$	2.3/56.9/22.9/12.1/5.6
$Np^{237}/Am^{241}/Am^{242m}/Am^{243}/Cm^{242}/Cm^{243}/Cm^{244}/Cm^{245}$	54.3/31.5/0.1/10.6/0.0/0.0/3.1/0.3

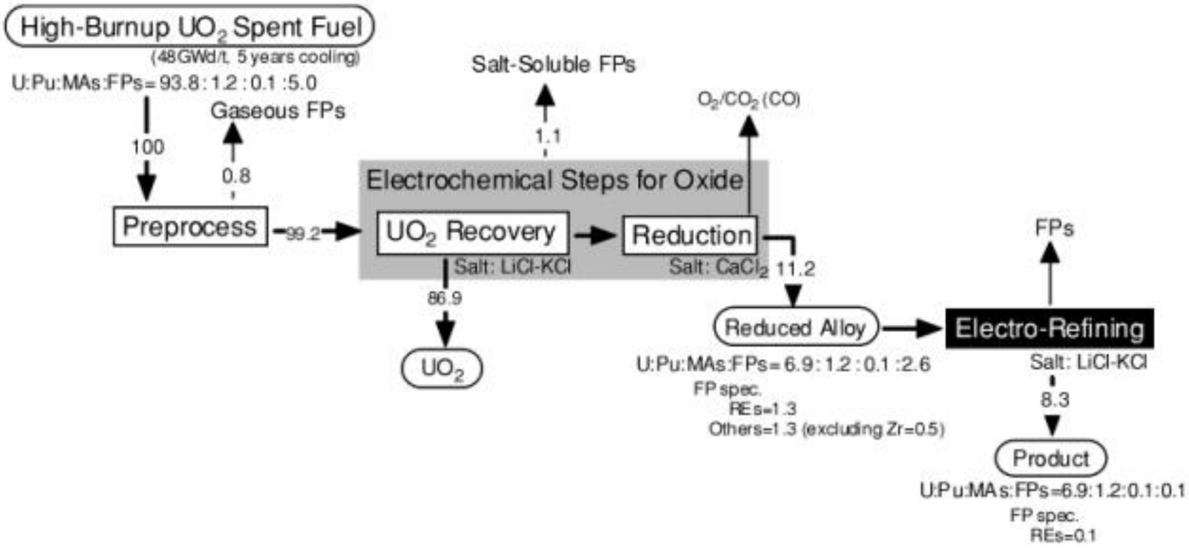


Figure 30: Material flow of Pyroprocessing of high-burnup UO2 spent fuel by electrochemical reduction technology [46].

As aforementioned, the separation or seclusion of plutonium in Canada is not an option, so if Pyroprocessing is to be put in operation, the option of removing UO_2 would not be implemented. Therefore, the methods used in [46] with the fuel contents specified in Section 2.2.2, the CANDU material flow using pyroprocessing was calculated.

Prior to processing, the CANDU spent fuel contents were U:Pu:MA:FP→99.43:0.41:0.08:0.08 and it is assumed that the same ratio of gaseous fission products exist in the CANDU fuel rods; therefore, there is a 16% reduction in the fission products during the head-end process, resulting in the spent fuel contents entering the electrochemical reduction step being 99.43:0.41:0.08:0.067. Since the removal of UO_2 is being ignored, the 99.43 wt% of uranium will remain unchanged, however, due to the salt-soluble fission products' ability to be removed, it is assumed that there is a 22% reduction in the additional fission product inventory during the oxidation stage. Therefore, the reduced alloy contents are now 98.57:0.29:0.15:0.049. Lastly, the electro-refining removes the remaining fission products except for the rare earths, which for CANDU reactors is less than 0.01wt%. The analysis performed by [46] is an idealistic scenario, as there are no assumed losses throughout each process, and there are no cross-contaminations of any product.

4.1.2 FVM and CANDU

Although there are many studies that prove the benefits and the effectiveness of the FVM with LWR and PWR fuels, CANDU fuels are of interest to the Canadian nuclear industry. The NWMO investigated the FVM with CANDU fuel (Figure 31), which essentially contains the same steps as described in Sections 2.1.3.

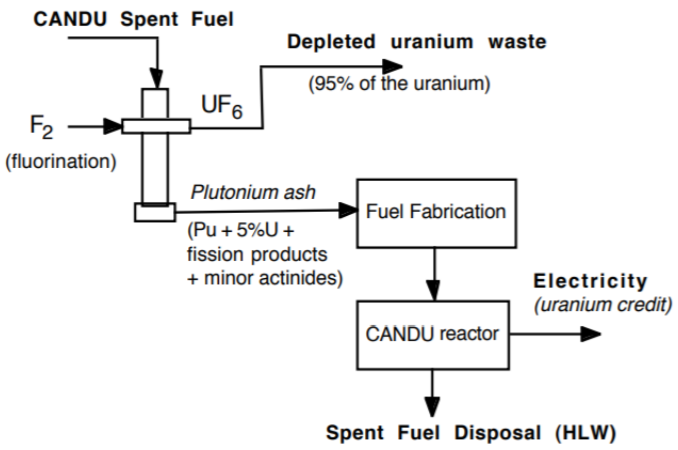


Figure 31: CANDU Spent Fuel and the Fluoride Volatility Method [20].

Decladding*:*

As with any reprocessing method, one must remove the fuel cladding. The NWMO looked at oxidative decladding, which causes the UO_2 to swell into U_3O_8 and turn into a powder. The cladding must initially be cut in order to expose the UO_2 to the oxygen gas, and also release the powder [20]. If the oxidative decladding method is used, the fuel is ready for the next step in the FVM. It should be noted that there is a release of fission product gases during the decladding process; however, these gases can be immobilized in clay filters and transported with the waste. The small amount of HLW produced during decladding due to exposure are all short lived [20].

Most of the recovered uranium would be obtained as uranium hexafluoride, with a volatile extraction efficiency of 95%. Although, since the stream contains mainly depleted uranium, it is considered low level waste, as long as few fission products are present. If one wishes to use the uranium, further processing and purification is required, which would increase the costs. Therefore, direct disposal of the depleted uranium (UF_6) stream would reduce disposal costs immensely, as 94% of the spent fuel volume (U) is now considered low level waste (LLW). The remaining 6% (Pu) is high level, and has high disposal cost.

Table 41: Composition of the fuel before/after 95% U extraction [20].

Fuel Element	Mass Before U Extraction (g)	Mass After U Extraction (g)	Weight % of Initial Spent Fuel
U-235	2.3	0.12	82%
U-236	0.7	0.04	
U-238	985.8	49.29	
Pu-239	2.5	2.5	6%
Pu-240	1.0	1.0	
Pu-241	0.2	0.2	
Pu-242	0.1	0.1	
FP + Actinides	7.4	7.4	12%
Total	1000	60.6	100%

According to Table 41, in order to obtain 1kg of recycled fuel material, 16.5kg of CANDU spent fuel is required. With a small amount of fissile material (Pu-239), this mixture could be used in a CANDU lattice and may still obtain a significant burnup. For instance, LWR ash fuel has 7% fissile material and can achieve 60MWd/kg [20].

The CANDU reactor is very versatile for fuel recycling from other reactors. The DUPIC fuel cycle uses PWR spent fuel in a CANDU. The recycled plutonium from the FVM above would benefit from cycling through the CANDU once again. However, the fabrication of fuel with highly radioactive materials poses a challenge. A new bundle design may be required to prevent exposure, and ensure the ideal power distribution.

Although the Pu ash saves disposal costs the first time, after it leaves the reactor a second time there is an even higher spent fuel cost. There is a significantly higher heat load from the higher accumulated burnup and a higher actinide concentration [20].

4.1.3 Canada Reprocessing CANDU Fuel

Although there is an economic incentive to reprocess LWR fuel, there is a small concentration of U-235 (approx. 0.23%) in spent CANDU fuel. The tail-ends of reprocessing facilities for enriched fuel are comparable to CANDU spent fuel (0.2-0.3%). Therefore, it would be deemed more financially viable to merely take the run-off from the already developed enrichment facilities [20]. Reprocessing CANDU fuel would be to recover plutonium, and other elements. The current technologies available (PUREX) are considered high cost, with significant proliferation issues. At this time, it is very unlikely that Canada will invest in the current technology for reprocessing; however, the continuing nuclear program in Canada may require alternative fuel cycle facilities.

4.1.3.1 Cost Analysis using Harvard Study

The Harvard Study [20] will be used to determine the \$/kg cost of the FVM and Pyroprocessing in 2004 dollars. Disposal of the spent recycled fuel (CHLW) could occur later than the time assumed for direct disposal’s unprocessed bundle; this would reduce the HLW disposal costs relative to the direct disposal [20]. In order to simplify the calculation, direct costs were used and the time frames of reprocessing and direct disposal were ignored. The goal of the following calculations is to achieve breakeven unit costs to compare to direct disposal:

$$C_D = C_R + f \cdot C_{LLW} + (1 - f) \cdot (C_{REC} + C_{HLW}) - (1 - f) \cdot (B_{REC} / B_{NAT}) \cdot (C_{FAB} + C_U) \qquad \text{(Eq. 17)}$$

Table 42: Chosen estimates of the breakeven price of uranium for FVM and Pyroprocessing [20].

Variable Name	Variable Description	Assigned Value
C_D	Direct disposal costs \$22B for 3.7 million bundles, each initially containing 19kg	300\$/kg
C_R	Cost estimate of reprocessing, which is significantly lower than PUREX	**See Table 43**
C_{LLW}	Disposal costs of low level waste, e.g. depleted uranium	$0.2C_D$
C_{REC}	Cost of remote fabrication of fuel bundles with varying plutonium contents and activities	**See Table 43**
C_{HLW}	Direct disposal cost of high level waste	$3C_D$
C_{FAB}	The credit associated with the fabrication of fresh fuel	60\$/kg
C_U	The price of uranium	156\$/kg
f	Fraction of spent fuel that ends up as depleted uranium	0.94
B_{REC}	Average discharge burnup that could be achieved with recycled fuel (depending on reactor design) SEALER(33MWd/kg), NuScale (30MWd/kg), IMSR (26-29MWd/kg)	40 MWd/kg
B_{NAT}	The burnup for the CANDU reactor with natural uranium fuel	7.5 MWd/kg

Table 43: Assumptions made for Pyroprocessing and FVM estimates [20,51,52].

Variable Name	FVM/Pyro Rationales and Estimates
C_R	Pyro: KAPF+s conceptual design, **699$/kg*** FVM: the dry process reduces cost, target price 10-20% lower than conventional process, aiming for: **200$/kg** and **400$/kg**
C_{REC}	Pyro: Assuming metal fuel fabrication, **1790$/kg*** FVM: Pu ash fuel, and similarities to OREOX fuel fabrication (high plutonium fuel fabrication): **600$/kg**

*adjusted to 2004 from 2010

As one could see from Table 43, using the Harvard method to calculate the cost per kg of fuel for both the fluoride volatility method and pyroprocessing, shows the significantly higher cost of pyroprocessing. However, there were many assumptions that were made, and therefore, the actual values are likely to be different.

4.2 Transmutation Experience

There is global experience in reprocessing spent fuel for the reuse of fissile material, particularly U^{235}. The attainment of high efficiency uranium usage requires multiple fuel recyclings, due to the material limitations. Typically, recycling involves the removal of cladding, removal of most fission products, and uranium implantation to increase the fissile content; however, recycling is often objected to due to the economic viability and proliferation concerns [11]. However, there are other benefits of reprocessing; which involves the transmutation of certain fission products to either reduce the volume of nuclear waste or the radiotoxicity. The danger in isotopic reinsertion is the absorption of neutrons that could otherwise fission. Therefore, strategic separation of isotopes could prevent unwanted neutron loss. In Section 4.2.1, the transmutation rate and neutron consumption of radioisotopes of interest is calculated.

4.2.1 Transmutation Rate and Neutron Consumption

The transmutation of LLFPs within an existing reactor would strain the control devices and eventually cause the chain reaction to stop. The spent fuel contents of CANDU fuel that was used to find the transmutation rates of each isotope present is shown in Table 44 and Table 45. Table 44 and Table 45 are associated with the spent fuels that were reinserted after one year, and 30 years, respectively. To determine if an isotope is worth reinsertion into a reactor for transmutation purposes, the abundance in fuel, the half-life, and the specific activity was considered.

Table 44: Transmutation Rates* in CANDU fuel reinsertion after 1 year cooling.

Isotopes	$\sigma_{n,\gamma}^{J}$(b)		Half Life (y)	T_J^{Transm}(yr)		Transmutation (Y/N)
	Fast	Thermal		Fast	Thermal	
Cm-242	0.055	2	$4.38*10^{-1}$	$3.99*10^{2}$	$1.10*10^{2}$	N
Cm-244	0.12	3	$1.80*10^{1}$	$1.83*10^{2}$	$7.31*10^{1}$	N
Am-241	1.22	98	$4.32*10^{2}$	$1.80*10^{1}$	2.24	Y
Pu-239	0.052	8.5	$2.41*10^{4}$	$4.22*10^{2}$	$2.58*10^{1}$	Y
Pu-240	0.12	50000	$6.50*10^{3}$	$1.83*10^{2}$	$4.39*10^{-3}$	Y
Pu-241	0.08	7	$1.4*10^{1}$	$2.74*10^{2}$	$3.13*10^{1}$	N
Pu-242	0.09	8	$3.73*10^{5}$	$2.44*10^{2}$	$2.74*10^{1}$	Y
U-235	0.46	11	$7.04*10^{8}$	$4.77*10^{1}$	$1.99*10^{1}$	Y
U-238	0.17	0.5	$4.47*10^{9}$	$1.29*10^{2}$	$4.39*10^{2}$	Y
Cs-137	0.003	0.04	$3.02*10^{1}$	$7.31*10^{3}$	$5.48*10^{3}$	N
Sr-90	0.00068	0.0054	$2.89*10^{1}$	$3.12*10^{4}$	$4.06*10^{4}$	N

*Neutron Energy: E_n= 0.2MeV (Fast), E_n = 1eV (Thermal) JEFF-3.3

Table 45: Transmutation Rates* in CANDU fuel reinsertion after 30 year cooling.

Isotopes	$\sigma_{n,\gamma}^{J}$(b)		Half-Life (y)	T_J^{Transm}(yr)		Transmutation (Y/N)
	Fast	Thermal		Fast	Thermal	
Ac-225	$5.50*10^{-1}$	$6.00*10^{2}$	$2.74*10^{-2}$	$3.99*10^{1}$	$3.66*10^{-1}$	N
Ac-227	$3.50*10^{-1}$	$6.00*10^{2}$	$2.18*10^{1}$	$6.27*10^{1}$	$3.66*10^{-1}$	N
Am-241	1.50	$1.00*10^{2}$	$4.33*10^{2}$	$1.46*10^{1}$	2.19	Y
Bi-210	$7.00*10^{-3}$	$9.00*10^{-2}$	$1.37*10^{-2}$	$3.13*10^{3}$	$2.44*10^{3}$	N
C-14	---	---	$5.70*10^{3}$	---	---	---
Cl-36	$1.96*10^{-3}$	1.58	$3.01*10^{5}$	$1.12*10^{4}$	$1.39*10^{2}$	Y
Cs-135	$5.39*10^{-2}$	1.29	$2.30*10^{6}$	$4.07*10^{2}$	$1.70*10^{2}$	Y
Cs-137	$3.16*10^{-3}$	$4.27*10^{-2}$	$3.01*10^{1}$	$6.94*10^{3}$	$5.14*10^{3}$	N
I-129	$1.09*10^{-1}$	5.13	$1.57*10^{7}$	$2.02*10^{2}$	$4.28*10^{1}$	Y
Np-237	$8.00*10^{-1}$	$7.20*10^{1}$	$2.14*10^{6}$	$2.74*10^{1}$	3.05	Y
Pa-231	$7.64*10^{-1}$	$1.62*10^{1}$	$3.28*10^{4}$	$2.87*10^{1}$	$1.35*10^{1}$	Y
Pa-233	1.00	$4.50*10^{1}$	$7.39*10^{-2}$	$2.19*10^{1}$	4.87	N
Pb-210	---	---	$2.22*10^{1}$	---	---	---
Pd-107	$5.22*10^{-1}$	$3.46*10^{-1}$	$6.50*10^{6}$	$4.20*10^{1}$	$6.34*10^{2}$	Y
Po-210	---	---	$3.79*10^{-1}$	---	---	---
Pu-239	$1.99*10^{-1}$	9.50	$2.41*10^{4}$	$1.10*10^{2}$	$2.31*10^{1}$	Y
Pu-240	$2.70*10^{-1}$	$2.00*10^{4}$	$6.56*10^{3}$	$8.12*10^{1}$	$1.10*10^{-2}$	Y
Pu-242	$1.65*10^{-1}$	7.50	$3.74*10^{5}$	$1.33*10^{2}$	$2.92*10^{1}$	Y
Ra-223	$1.15*10^{-1}$	$2.07*10^{1}$	$3.13*10^{-2}$	$1.91*10^{2}$	$1.06*10^{1}$	N
Ra-224	$6.41*10^{-2}$	1.91	$1.00*10^{-2}$	$3.42*10^{2}$	$1.15*10^{2}$	N
Ra-225	$1.33*10^{-1}$	$1.58*10^{1}$	$4.08*10^{-2}$	$1.65*10^{2}$	$1.38*10^{1}$	N
Ra-226	$1.00*10^{-1}$	2.30	$1.60*10^{3}$	$2.19*10^{2}$	$9.54*10^{1}$	Y
Ra-228	---	---	5.75	---	---	---
Rn-222	---	---	$1.05*10^{-2}$	---	---	---
Sb-126	$1.25*10^{-1}$	2.80	$3.38*10^{-2}$	$1.75*10^{2}$	$7.83*10^{1}$	N
Se-79	$7.09*10^{-2}$	1.80	$2.95*10^{5}$	$3.09*10^{2}$	$1.22*10^{2}$	Y
Sm-147	$3.70*10^{-1}$	9.70	$1.06*10^{11}$	$5.93*10^{1}$	$2.26*10^{1}$	Y

Sm-151	$4.90*10^{-1}$	$2.60*10^{3}$	$9.00*10^{1}$	$4.48*10^{1}$	$8.44*10^{-2}$	Y
Sn-126	$9.73*10^{-3}$	$2.85*10^{-1}$	$2.30*10^{5}$	$2.25*10^{3}$	$7.70*10^{2}$	Y
Sr-90	$1.46*10^{-3}$	$5.49*10^{-3}$	$2.88*10^{1}$	$1.50*10^{4}$	$4.00*10^{4}$	N
Tc-99	$2.71*10^{-1}$	4.44	$2.11*10^{5}$	$8.09*10^{1}$	$4.94*10^{1}$	Y
Th-227	$3.93*10^{-1}$	$2.55*10^{2}$	$5.11*10^{-2}$	$5.58*10^{1}$	$8.60*10^{-1}$	N
Th-228	$2.69*10^{-1}$	$2.70*10^{1}$	1.91	$8.16*10^{1}$	8.11	N
Th-229	$6.20*10^{-1}$	$5.60*10^{1}$	$7.34*10^{3}$	$3.54*10^{1}$	3.92	Y
Th-230	$4.10*10^{-2}$	$2.40*10^{1}$	$7.54*10^{4}$	$5.35*10^{2}$	9.14	Y
Th-231	---	---	$2.91*10^{-3}$	---	---	---
Th-232	$1.70*10^{-1}$	$7.50*10^{-1}$	$1.41*10^{10}$	$1.29*10^{2}$	$2.92*10^{2}$	Y
Th-234	$4.85*10^{-2}$	$2.87*10^{-1}$	$6.60*10^{-2}$	$4.52*10^{2}$	$7.65*10^{2}$	N
U-233	$2.16*10^{-1}$	$1.40*10^{1}$	$1.59*10^{5}$	$1.02*10^{2}$	$1.56*10^{1}$	Y
U-234	$2.08*10^{-1}$	8.86	$2.46*10^{5}$	$1.06*10^{2}$	$2.48*10^{1}$	Y
U-235	$3.10*10^{-1}$	$1.40*10^{1}$	$7.04*10^{8}$	$7.08*10^{1}$	$1.57*10^{1}$	Y
U-236	$8.00*10^{-2}$	1.10	$2.34*10^{7}$	$2.74*10^{2}$	$1.99*10^{2}$	Y
U-238	$1.30*10^{-1}$	$5.10*10^{-1}$	$4.47*10^{9}$	$1.69*10^{2}$	$4.30*10^{2}$	Y
Y-90	$4.29*10^{-2}$	1.02	$7.30*10^{-3}$	$5.11*10^{2}$	$2.16*10^{2}$	N

*Neutron Energy: E_n= 0.2MeV (Fast), E_n = 1eV (Thermal) JEFF-3.3

As shown in Table 44 and Table 45, there are multiple isotopes that have transmutation rates lower than their half-lives. Although these materials have low specific activities, and would not drastically reduce the reactivity of the spent fuel, uranium and plutonium are responsible for most of the spent fuel's volume. The removal of the plutonium isotopes and the uranium isotopes could be performed using a transmutation method, such as the fluoride volatility method.

If isotopes are transmutable, they could theoretically be selected to be inserted into a nuclear reactor; however, if their presence within the reactor would negatively affect the criticality, their transmutation ability may not be worthwhile. The transmutable isotopes shown in Table 46 and Table 47 have been selected to determine their respective neutron consumptions. The fission products in Table 47 all consume at least one neutron per transmutation, and therefore could be potential reactor poisons.

Table 46: Neutron consumption (DJ) of transmutable isotopes with their yields (YJ) of Actinides [57].

Transmutable Actinides (J)	D_J (neutrons consumed/fission)*	Y_J (Nuclei / Fission in Nuclear Power)*	Transmutation Rate (yr) (Fast)	Transmutation Rate (yr) (Thermal)
Am-241	0.95	3.239	$1.46*10^{1}$	2.19
Np-237	0.97	---	$2.74*10^{1}$	3.05
Pu-239	-0.73	2.884	$1.10*10^{2}$	$2.31*10^{1}$
Pu-240	0.38	3.086	$8.12*10^{1}$	$1.1*10^{-2}$
Pu-242	1.13	3.189	$1.33*10^{2}$	$2.92*10^{1}$
U-235	-0.55	2.435	$7.08*10^{1}$	$1.57*10^{1}$
U-238	-0.007	2.819	$1.69*10^{2}$	$4.03*10^{2}$

*MOX LWR with a moderator to fuel ratio of 4 and a neutron flux of $1x10^{14}$ n/cm^{2}s

Table 47: Neutron consumption (Dj*) of transmutable isotopes with their yields (YJ) of Fission Products*[26].

Transmutable Fission Product (J)	D_j^* (neutrons consumed/Transmutation)**	Y_J (Nuclei / Fission in Nuclear Power)**	Transmutation Rate (yr) (Fast)	Transmutation Rate (yr) (Thermal)
Tc-99	1.01	0.055	$8.09*10^{1}$	$4.49*10^{1}$
Pd-107	2.04	0.015	4.20*10	$6.34*10^{2}$
I-129	1.008	0.009	$2.02*10^{2}$	$4.28*10^{1}$
Cs-135	1.002	0.017	$4.07*10^{2}$	$1.70*10^{2}$
Sn-126	~2	0.0012	$2.25*10^{3}$	$7.7*10^{2}$
Se-79	~2	0.0004	$3.09*10^{2}$	$1.22*10^{2}$

*Transmutable Fission Products chosen based on availability of neutron consumption data
**LWR (UO_2) after 5 years of cooling

4.2.2 Reprocessing and Transmutation VS DGR

Before reprocessing is implemented in Canada, a cheaper process must be found [20]. Until then, a Deep Geological Repository is the safest way to isolate spent nuclear fuel from the environment with many passive barriers. Starting from the fuel bundle, it will be sealed in a steel and copper container, and surrounded by bentonite clay. The room in which the fuel bundles are located will be hundreds of meters below the surface beneath low permeable rock [59].

Ottensmeyer (2014) performed an analysis comparing the DGR to reprocessing CANDU spent fuels [74]. The fissile content of the CANDU fuel is low, and too low to be considered for further use in thermal reactors. However, the volume of CANDU fuel per GWh is 7 to 10 times larger than LWR fuel waste. The CANDU fleet in Ontario contributes 1400 tons annually to the current reservoir. The current solution involves the construction of a DGR, which can cost between $24 and $40 billion, depending on the capacity. Currently, 0.15 cents per kWh are being collected from the consumer with $2.9 billion has been accumulated from 2.5 million fuel bundles; however, this value is more than half of the smaller capacity for the DGR. Therefore, there would need to be an increase in charge of about 1.34 cents/kWh for the remaining 1.4 million bundles [74]. Table 48 shows the rates required to obtain the cost of the DGR.

Table 48: Future average rate required to meet DGR costs (in 2014) [74].

Planned DGR Capacity	Total Life Cycle Cost	Outstanding Balance	Future Avg. Rate (c/kWh)
3.6 million	$ 24.4 billion	$ 21.5 billion	1.34
4.6 million	$ 29.8 billion	$ 26.9 billion	0.90
7.2 million	$ 40.8 billion	$ 37.8 billion	0.57

However, 99% of heavy atoms (which have fissile potential), can be reprocessed into fast reactors. The FR could theoretically use one ton of spent CANDU fuel to produce electrical energy worth $1.2 billion [74]. The transmutations that are undergone in the FR would alleviate the requirements of a million year sequester in a DGR.

4.3 Closing the Fuel Cycle

4.3.1 CANDU Environmental Assessment

The assessment for Canada using the NELCAS method requires assumptions listed in Table 49. Many assumptions had to be made for the life cycle assessment of Canadian life cycle NELCAS. Wherever data was unable to be found, the value within the French NELCA was taken and adjusted with the Canadian nuclear electricity production. Canada produced 7 kT of uranium in 2018, however, 76% of the product is exported. Assuming the remaining uranium is solely used for CANDU reactors, 1.68 kT is used for fuelling 17 reactors [22]. The electricity produced by the CANDU reactors in 2008 was 598 GWh_e [23]. Assuming the average CANDU reactor is operational for 30 years, the total electricity produced in all of Canada by CANDU reactors is 17.9 TWh_e, and the total fuel used is 50.4kT. Considering that much of the data in this calculation was taken from France, there are a few values that are overestimated, due to the transportation costs. Since many contributors to the nuclear operations are within Canada, direct transportation is assumed to be rail or road. Table 50 lists the environmental and technical indicators. Using the values calculated in Table 49, Figure 32 was calculated to show the relative contributors for the chosen indicators.

Table 49: Fuel Cycle steps with assumptions and references for the life cycle NELCAS calculation.

Cycle	Canadian Site	Operation
Front-end of Fuel Cycle	Open-pit underground mine Fuel Fabrication	7kT required for one year in France, and 1.68kT is required in Canada, which is a 78% reduction in demand. Therefore, 22% of the contributions in France will be used for CANDU’s mining. Assuming fuel fabrication is performed at one site, it is assumed that the data is similar to France
Energy Production	17 Reactors	Data from Environmental Monitoring Programs provided by Ontario Power Generation will be used for on-site contributions for all reactors
Back-end	Bruce	Since Canada does not reprocess Nuclear Fuel, the environmental emissions will be limited to the dry storage contributions at one site.
Transportation	Between sites	Total distance was determined through rail or road.
Construction, Dismantling		Data derived from the provided reference in [21]

Table 50: NELCAS results for the environmental and technological impact indicators for the Canadian Once-Through cycle.

	GHG gCO_zeq/kWh_e	Atmospheric Pollution SO_x mg/kWh_e	Atmospheric Pollution NO_x mg/kWh_e	Water Pollution mg/kWh_e	Land-use m^2/GWh_e	Water Consumption L/MWh_e	Water Withdrawal L/MWh_e	Tech Waste m/MWh_e
Mining	257	2148	2976	39681	21.74	2.56	2.56	0.23
Conversion	191	39.8	713	59.65	1.25	3.15	3.15	1.37
UOX Fabrication	24	8.9	34.3	14.40	0	0.14	0.14	0.16
Reactors	0	7.9	168	14.64	0	0	40458	13.82
Disposal	71	16.5	66.5	0	0	0.07	0.07	0.76
Total	**543**	**2221**	**3958**	**39769**	**22.98**	**5.92**	**40464**	**16.33**

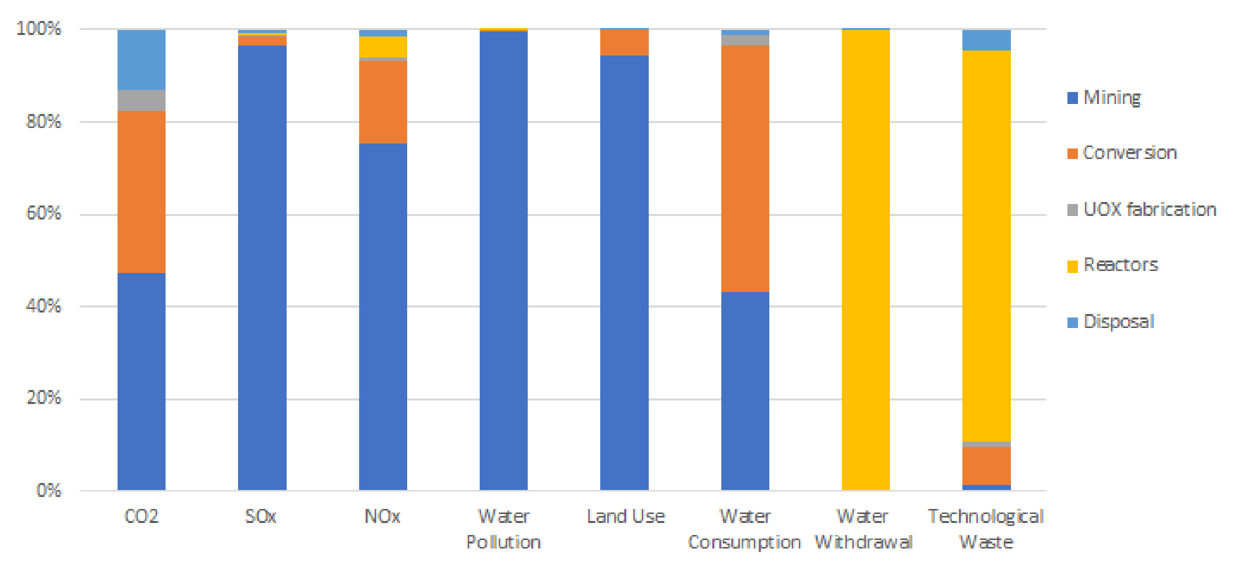


Figure 32: Relative contributions of each step in the CANDU fuel cycle to the environmental and technological impact indicators.

Greenhouse gas emissions, Atmospheric and Water Pollution

As shown in Figure 32, the largest contributors to GHG emissions are mining, fuel conversion, and disposal. Since Canada is heavily reliant on fresh fuel, the mining and conversion contributors are heavily weighted. Therefore, when compared to France, there is a significant difference between Canada and France, with 543 gCO_2eq/kWh_e and 5.29 gCO_2eq/kWh_e, respectively. Although, as one can see in Figure 32, reactors contribute more to the GHG emissions; this is likely a result of the chosen data for CANDU reactors. Ontario Power Generation releases Environmental Monitoring Program reports, where pollutants are listed [24]. Only the CO_2 emissions were considered, and the CO_2eq were omitted. For SO_x, NO_x, and water the main contributors are the mining operations.

Land Use

Due to the lack of processing facilities within Canada, for enrichment and reprocessing purposes, land usage is significantly reduced. Also, since most of the CANDU reactor sites have multiple reactors within the same facility, there is an efficiency in space usage. Therefore, once again, the main contributor to land use is the mining operations at $23m^2/GWh_e$. However, since the disposal values from France were used for CANDU calculations, the dry storage of CANDU spent fuel was not considered.

Water Consumption and Withdrawal

There was an assumption that the withdrawal of water for the CANDU reactor has a net value of zero, due to the presence of a water reservoir on each site, there are no cooling towers and therefore, no net water consumption. The D_2O usage was also assumed to be completely efficient, and all deuterium is recycled completely at the end of the reactor life. Therefore, the water consumption is completely dominated by mining operations. Water withdrawal includes the feedwater from the reservoirs to compress the steam within the reactor's secondary heat transport loop. The average intake of water from the reservoir (such as Lake Ontario) is 46000L/s [25]. Therefore, the resulting water withdrawal is significantly higher than that of the mining operations. France and Canada are both heavily dominated by water withdrawal.

<u>Resource efficiency</u>

In order to calculate Canada's current efficiency, a few assumptions had to be made. Therefore, the nuclear fuel efficiency in Canada is currently 0.355 GWh_e/tU_{nat} or $3.55*10^{-4}$ GWh_e/kgU_{nat}, which is significantly (150000 times) less than France's twice through cycle.

4.4 Hybrid Systems in Canada

4.4.1 Nuclear and Canada's Oil Sands

Currently, the bitumen is processed using natural gas-fired plants, which provides heat and steam. Since bitumen is too viscous to pump to the surface because of the high carbon-to-hydrogen ratio, the oil sands must find ways to recover the oil while upgrading the bitumen to higher quality oil. Almost 80% of the deposits in Canada are too deep for surface mining and require in-situ methods for recovery [83].

The current method for the Canadian oil sands is Steam-Assisted Gravity Drainage (SAGD), as shown in Figure 33. Two horizontal wells are drilled underground, the length of the wells range from 500 to 1000m, with depths as shallow as 40m. Slots are added to the wells to allow for the passage of the steam and extracted oil; steam is sent in at a high pressure on the bottom of the well and forms a steam chamber. The expansion of the steam causes the colder bitumen at the interface to lose viscosity and separate from the sand. The condensate and bitumen drain to the bottom of the well and will be pumped to the surface for further processing [83].

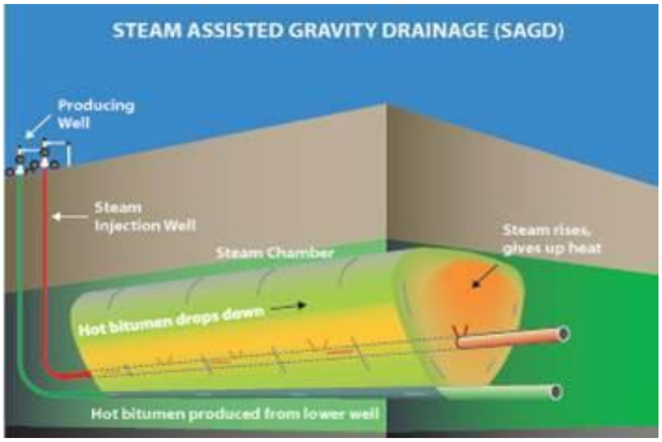


Figure 33: Schematic of SAGD [83].

The processing facility converts the emulsion into diluted bitumen, which is preferential for transportation. Further upgrades to the diluted bitumen can occur onsite or it can be transported elsewhere, to be converted into a synthetic crude oil (syncrude). Nuclear energy has been proven to be competitive to natural gas in providing the energy demands of the oil sands. In order to achieve a more sustainable, long-term energy source, a scenario in which a nuclear reactor provides the heat for the production of bitumen, electricity, and hydrogen for upgrading should be considered. The design has to have a 100,000 bbl/day bitumen processing capability [83].

Using high temperature steam electrolysis (HTSE), the facility produces hydrogen to upgrade the bitumen to syncrude. The electricity requirements for the 100,000 barrel per day syncrude plant is shown in Table 51, and the facility map options are shown in Figure 34.

Table 51: Onsite Electricity Requirements of nuclear powered syncrude plant [83].

Area	Connected Load / Heat (MW)	Average Demand / Power (MW)
Mine	27	20
Bitumen Extraction and Cleaning	200	150
Upgrader	115	86
Utilities and Offsites	44	33
Infrastructure	1	1
Total	387	290

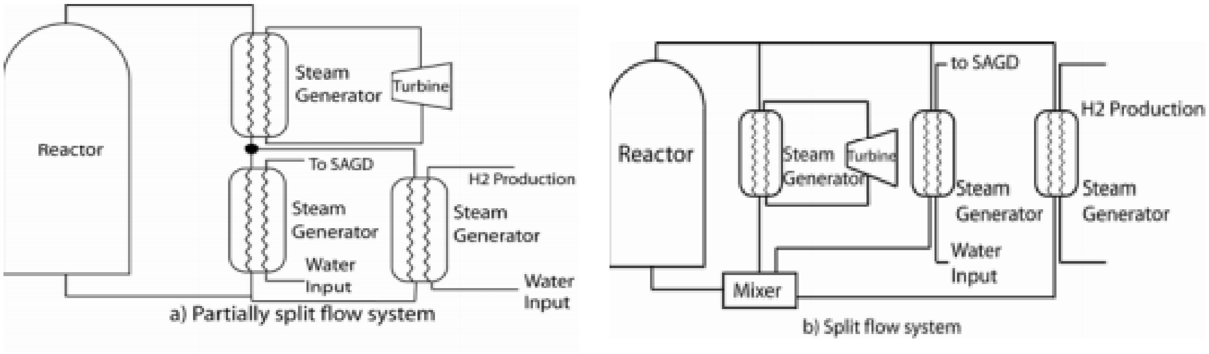


Figure 34: Options for hydrogen and electricity production systems [83].

Table 52 shows the reactors initially listed in Section 2.3 and the number of modules required to meet the needs of the scenario. The reactors that are (<50MWe) smaller were assumed to be uneconomical for this system. For example, the NuScale design would require a 12 module to provide enough energy; however, that is one of the benefits of its modular design.

Table 52: SMR units required to provide a minimum of 700 MWe.

Vendor	Name of Design	Electrical Capacity (MWe)	Reactors Required
*Terrestrial Energy	IMSR	200	4
*ARC Nuclear Canada	ARC-100	100	7
Moltex Energy	Stable Salt Reactor	300	3
Holtec International	SMR-160	160	5
*NuScale Power	NuScale	60	12
GE-Hitachi Nuclear	BWRX-300	300	3

All the reactors listed in Table 52 only address the capacity of the units; the potential to optimize the process, the efficiency, or the temperature of the steam was not considered. As you can see, the implementation of the larger SMRs would result in a surplus of electricity. The modular nature of the SMR designs with smaller power capacities would allow for more manipulation of reactor capacity to meet the system's specific needs.

4.4.2 Other Opportunities in Canada

As Canada has a wide range of demands, SMRs designs can be implemented to meet many requirements, while reducing or eliminating the environmental impact of the existing technologies. Table 53 shows the various applications throughout Canada, with the corresponding power requirements. Due to the privatized nature of many SMR designs, very little information was found regarding transport heat temperatures; therefore, the electrical power was only considered when determining SMR applications. However, chemical heat pumps can be incorporated in the hybrid systems where applicable [81].

Table 53: Canadian SMR roadmap applications, power requirements, and proposed SMR pairings.

Application	Total Power Demand	SMR
Oil Sands	210 MWe	IMSR, ARC-100, Moltex, SMR-160, BWRX-300
High Temperature Steam	25-50 MWe	MMR-5/10, NuScale
Remote Communities	<15 MWe	MMR-5/10, SEALER, U-Battery
Off-Grid Mines	30-150 MWe	ARC-100, NuScale,
Replace Coal Power	343 MWe	IMSR, ARC-100, Moltex, SMR-160, BWRX-300

From the list of vendors currently going through the review process by the CNSC, all vendors have an application for their design throughout Canada. As mentioned in Section 4.4.1, the oil sands require a minimum of 700 MW, including both heat and power demands. Large power reactors, such as the CANDU, would provide more power than what is required for the facilities.

4.5 Fuel Cycle Policies

National-building interests typically drive the nuclear power programs for each country, along with the desire to build nuclear weapons. Certain powerful nuclear states have limited certain aspects of global oversight to continue pursuing their own interests. For example, the European Atomic Energy Community (EURATOM) treaty wanted a global approach for common safety and waste regulations in the public interest, which has been reduced to a conglomerate of national initiatives. Therefore, the IEA and IAEA have implicitly limited their evaluations of nuclear power to the question of marketability [84]. However, the IAEA plays an important role in the control of nuclear proliferation; they carefully inspect nuclear operations in countries to ensure no material is diverted from peaceful operations for non-peaceful purposes. However, the extent of the IAEA's authority is uneven. For example, when North Korea was enriching uranium for nuclear weapons, and performed a nuclear weapons test in 2006, the IAEA notified North Korea for non-compliance with commitments to non-proliferation, and the technicians were expelled from the country in 2009 [6].

The implementation of SMRs into each country is being done using different methods for licensing. Table 54 shows the basic licensing concepts for selected countries. The regulatory frameworks approach can be prescriptive (US), or goal setting (UK). The licensing steps can be divided into one-step licensing: COL in the US, or site license in the UK. The Canadian Licensing process will be discussed in Section 4.5.2.

Table 54: Licensing Processes [85].

Country	Pre-Licensing	Political	New Facility	Regulatory Holds
Finland	Environmental Impact Assessment	Decision on Principle	Construction License	
USA	Early Site Permit Standard Design Certification		Combined Construction and Operating License Construction Permit, Operating License	Inspections, Tests, Analyses, and Acceptance criteria
Canada	Vendor Design Review		License to Prepare Site Environmental Assessment, License to Construct, License to Operate	
France	ASN Opinion on Safety	Multi Year investment plan	Authorization Decree for NPP Creation	
UK	Generic Design Assessment		Nuclear Site License	Established hold Points

4.5.1 International Influencers: France

After the first oil crisis in 1973, France decided to heavily invest in nuclear energy. Thus, France has grossly decreased its dependence on fossil fuels by having almost 80% of its electricity from nuclear sources. A benefit of which is the decrease in the country's CO2 emissions by a factor of 5, and a mean emission range between 70-100gCO2/kWh since 1993 [21]. In 2013, a total capacity of 63GWe was installed in France, with a production of 400-420 TWhe/year [21].

EDF and AREVA are France's two major industrial operators, who have helped France develop a complete fuel cycle, all within the country (omitting mining activities). France requires roughly 8000t of uranium ore each year to feed its nuclear fuel requirements [21]. Figure 35 refers to the main steps within the French fuel cycle, with values corresponding to 2010.

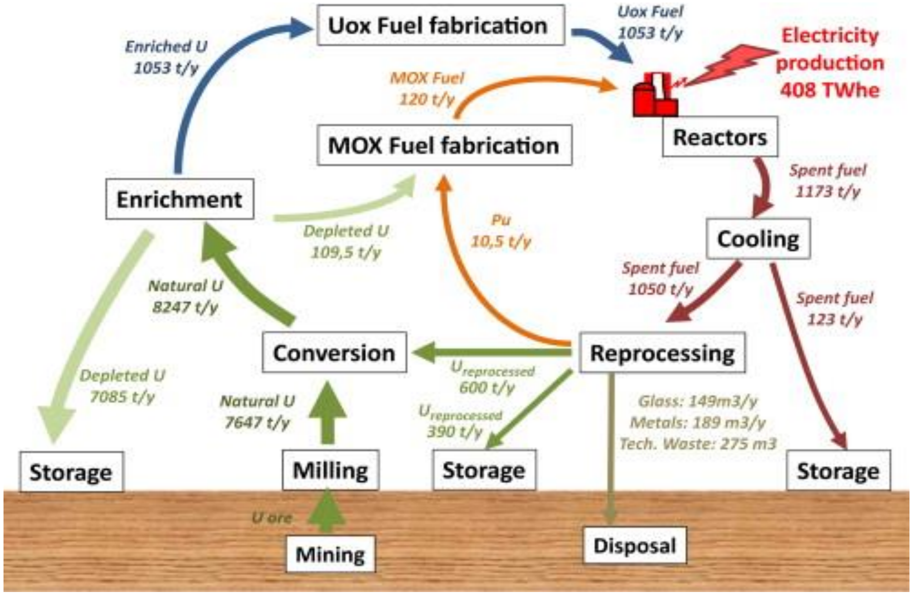


Figure 35: French reference fuel cycle and its representative streams (2010) [21].

After the milling of the natural uranium, the yellowcake is delivered to France, where the product is purified and converted into a gaseous UF_6 to prepare for enrichment. Before 2012, enrichment was mainly accomplished using gaseous diffusion; however, now it uses ultracentrifugation [21]. The depleted uranium cannot be used by the current institutions already available, and therefore is stored as a strategic stockpile. In order to maintain the energy demand 1200t of fresh fuel must be induced, and thus 1200t of spent fuel has to be disposed of yearly.

The back-end of France's fuel cycle is unique, as it contains a twice-through cycle, to recover uranium and plutonium. La Hague performs the separation process, PUREX, to obtain streams of the two heavy metals. MELOX is the process used to combine the plutonium and uranium to produce MOX fuel [21]. To prevent a stockpile of plutonium, France's policy aims to ensure all plutonium recovered by reprocessing is used in the MOX fuel. More than a third of the reactors in France use the MOX fuel within one third of the reactor. Since the PWR design lacks the ability to fission the even plutonium isotopes, the spent MOX fuel is not reprocessed. As there are possibilities for GEN IV reactors to burn any uranium isotopes, all spent fuels and depleted fuels are kept as strategic materials [21].

The recycling operations (PUREX) produce High Level Waste (HLW), which is encased in a borosilicate named R7T7 [21]. Lower levels of waste (ILW and LL) are compacted and cemented (technological waste). The French Act of 2006 states that radioactive waste is to be disposed of in a deep geological repository.

4.5.2 Canadian Policy Revision for Future Nuclear Development

If Canada would like to introduce innovative nuclear technologies, there may be some alterations that will need to be made to the current policies. However, one must first consider the non-proliferation and physical protection requirements as new technologies differ from the natural-uranium CANDU reactor. Considerations of a different fuel cycle include: manufacturing processes for different fuel types; transportation to a variety of locations; on-site storage and transportation of used fuel; reprocessing of spent fuel. Table 55 shows the different nuclear substances and their corresponding classes, which determines how the regulator wishes to handle the material.

Table 55: Definitions of Category I, II, and III Nuclear Material [87].

Item	Substance	Form	Quantity (I)	Quantity (II)	Quantity (III)
1	Plutonium	Unirradiated	2kg or more	2kg>m>500g	500g>m>15g
2	U-235	Unirradiated $\geqslant 20\%\ U^{235}$	5kg or more	5kg>m>1kg	1kg>m>15g
3	U-235	Unirradiated $20\% > U^{235} \geqslant 10\%$	N/A	10kg or more	10kg>m>1kg
4	U-235	Unirradiated $10\% > U^{235} > 0.711\%$	N/A	N/A	$m \geqslant 10$kg
5	U-233	Unirradiated	2kg or more	2kg>m >500g	500g>m>15g
6	Dep U, NU, Th, LEU (<10%)	Irradiated	N/A	m>500g Pu	500g Pu$\geqslant$m$\geqslant$15gPu
7	NU, Dep I, Th	Unirradiated	N/A	N/A	N/A

According to the Nuclear Safety and Control Act: Category I material must be used, processed, and stored in an “inner area;” Category II material must be processed, used, and stored in a “protected area;” Category III nuclear material must be processed, used, and stored in a “protected area” or “an area under direct surveillance by the licensee,” or “ an area to which access is controlled and that is designed and constructed to prevent unauthorized access to material using hand held tools” [87].

4.5.2.1 Enrichment, Reprocessing, and Transmutation Safeguards

A global concern regarding new nuclear technologies is the proliferation of nuclear weapons. Enrichment and fuel reprocessing are very sensitive, and are currently part of many peaceful nuclear programs, but can be used to produce highly enriched uranium and plutonium [6]. The general intent for safeguarding is to deter diversion of material for misusing nuclear material for military purposes. The measures that are typically in place include: nuclear material accountancy, containment and surveillance, design information verification, reports, and inspections [88]. The chosen nuclear material for a design does not affect the safeguard; however, it may affect the material attractiveness.

As nuclear power plants are currently viewed as “high-security sites,” facilities where Category I and II nuclear materials are stored, processed, or used are also to be considered “high-security sites.” Therefore, enrichment and reprocessing facilities shall also be considered “high-security sites” [88].

Pencer, J (2019) performed an analysis of three scenarios in which the current regulatory frameworks were kept with various fuel cycle options. In scenario one, the enrichment of the UO_2 fuel was kept under 10 wt%, and the fuel was sent once through the cycle. Compared to the CANDU fuel cycle, the conversion of the fuel would have UF_6 as the end product in order to prepare for future enrichment. The enrichment of the fuel would require Category III security measures. Once the fuel is in the reactor, the requirements would be similar to CANDU reactors, as well as the on-site and long-term storage requirements. If enrichment and/or fuel fabrication were to be done outside of Canada, the security measures would involve transportation across borders, with the associated precautions.

Scenario two involved a fuel with enriched fuel between 10 and 20 wt%, with a once through fuel cycle. Similar to scenario one, after the conversion of the fuel, the UF_6 would be the end product. The enrichment of the fuel would result in Category II measures due to the increased enrichment. As this scenario is once again a once through fuel cycle the power generation and fuel storage would have similar requirements to CANDU.

The last scenario, and the current proposal, involves recycled fuel. Due to many various fuel types for the SMRs, the scenario will remain generic. Transportation and storage of the used fuel is the first to be considered. The associated fuel requirements depend entirely on the irradiated fuel type. For example, CANDU spent fuel is considered a Category II which would have different requirements for transportation than LEU spent fuel, which is Category I. If the fuel coming from an SMR was already processed once, the contents would be assumed to contain Pu and/or U-233 and would require Category I security measures. Implementing reprocessing to reuse the enriched uranium and Pu would require Category I security measures, as well as the fuel fabrication. However, if absorber rods were to be made from minor actinides or other fission products, the required security precautions might change depending on the isotopes present. Reactor operation and storage would require Category I security; however, depending on the used materials, there may be exceptions [88].

4.5.2.2 Hybrid Systems and SMRs

One main concern for the deployment of SMRs throughout Canada is the remoteness of certain facilities; regulatory inspections would be difficult to uphold. Also, many SMR designs state that remote monitoring abilities make their reactor more attractive; however, the remote oversight may add challenges for material accountability. Especially for MSRs, as nuclear material circulates throughout the core, Category I materials may not adhere to the requirements in these designs [88].

With respect to the actual location of the nuclear materials, the local environment and society already takes the appropriate precautions. Canada has one of the most stringent nuclear licensing processes in the world. After the design review, the proposed locations must undergo numerous environmental assessments. The local communities must be supportive of the development if the vendors want to break ground. Oversight from the CNSC ensures a transparency in which the environment and surrounding people are protected [90].

4.5.2.3 Stakeholders

With respect to the present generations, the highest risk associated with nuclear fission power are the accidents that are caused by the facilities. Even if the probabilistic risk assessment reveals a miniscule probability, the hazards are very real. However, with each accident that occurs, more safety precautions, policy adjustments, and global safety standards are revised.

Banerjee et al. (2011) performed an analysis on a hypothetical company, with which they introduced new nuclear technologies to the public, with a combination of strategies. Table 56 and Figure 36 show the various stakeholders taken into consideration and the map of the strategy, respectively.

Table 56: Stakeholder attributes and management strategies [86].

Stakeholders	Power	Legitimacy	Urgency	Valence	Strategy
Shareholders	high	high	high	supportive	reinforcement
French Gov.	high	high	medium	supportive	reinforcement
Customers	high	high	medium/high	supportive	reinforcement
Employees	low/medium	high	medium	supportive	reinforcement
Suppliers	medium	high	medium/high	supportive	reinforcement
Regulators	high	high	high	obstructive	containment
NGOs	medium	high	high	obstructive	containment
Unions	low/medium	high	medium/high	supportive/obstructive	reinforcement/containment
Community	low/medium	high	high	supportive/passive	reinforcement/stabilization
Public	high	high	low	passive	stabilization
International Ags.	high	high	low/medium	supportive	reinforcement

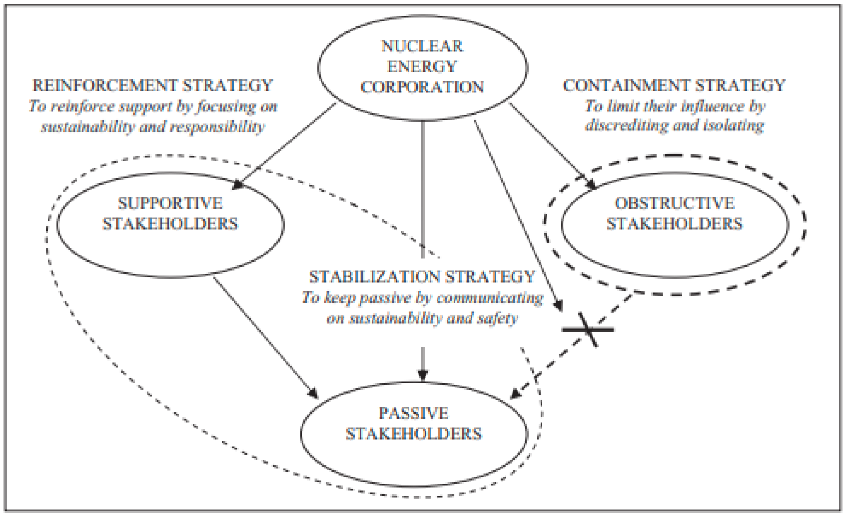


Figure 36: Stakeholder Management Strategies [86].

The implementation of SMRs and hybrid systems can be considered a step towards sustainable energy in the nuclear industry. The strategic engagement with the stakeholders that were deemed supportive involved business highlights, growth, and opportunities. For those interested in the sustainability strategies, the reduction of greenhouse gas emissions was also highlighted. The passive stakeholders were addressed with public relations campaigns to reassure the safety risks are being met, while “clean” aspects of nuclear energy are being pursued. During the stakeholder and community engagement forums, obstructive stakeholders were introduced to the sustainable development agenda, where the environmental initiatives were introduced; reprocessing was included in order to present methods to reduce radioactive waste [86].

5 Conclusion

Although many countries are investing in research in reprocessing technologies for future implementation, Canada's nuclear oversight does not encourage tampering with spent fuel. The CANDU reactor's natural uranium fuel is considered dependable and safe, which gains support from local communities. However, since uranium is a finite resource, with Canada exporting many of the recovered materials, the depletion of uranium will add strain on the Canadian fuel cycle within a few decades.

The most widely used reprocessing technology that is used on an industrial level is PUREX. Due to the stream of pure plutonium that is obtained during this process, Canada is extremely unlikely to implement this technology. Some countries use PUREX technology to develop nuclear weapons, and as Canada has no interest in nuclear warfare, a PUREX reprocessing plant would not be welcome on Canadian soil.

Two promising technologies are becoming more popular, pyroprocessing and the fluoride volatility method. Although the two methods have yet to be globally developed, the efficiencies and safety measures are promising. The proliferation-resistance for both methods is also superior to the PUREX method. Pyroprocessing has an adaptable process flow, in which the product could be altered depending on the needed future fuel. Considering the Canadian desire for minimal tampering, the pyroprocess was adjusted without separating the uranium from the other elements. However, the presence of FP and MAs within the spent fuel may not be desirable. After the head-end separation of materials, the pyroprocessing resulted in a 22% reduction in the fission product inventory after the oxidation stage. The NWMO has investigated the use of the FVM with CANDU spent fuel. The recovered uranium is not only most of the fuel's mass, but is depleted uranium and is considered low level waste; therefore, its separation and removal from other spent fuel material would have fewer requirements for disposal. One result of the FVM is plutonium ash, which can be inserted for transmutation. Although there is a savings in disposal costs after the first iteration, a second reactor exposure results in a higher heat load, burnup, and actinide concentration which adds requirements for handling of the ash, despite it being a smaller volume.

There are clear savings when choosing between newer reprocessing technologies and PUREX, but choosing between pyroprocessing ($1,368/bundle) and FVM ($1,114/bundle) can be left for the preferred technology and desired output. The economic advantage of reprocessing and closing the fuel cycle, versus direct disposal is not exactly clear-cut. There are environmental benefits to reprocessing, as there is a significant reduction in radioactivity and radiotoxicity. The DGR needs to be built regardless of the fuel cycle. However, the capacity requirement for high level waste can be decreased with reprocessing and transmutation of MAs and FPs.

The introduction of GEN IV reactors, specifically SMRs, would provide new opportunities for the nuclear fuel cycle, the economy, and energy production throughout the country. Deployment of SMRs would provide local communities with jobs and further insight to the nuclear industry, which would ultimately provide more support for the development of nuclear technologies. SMRs also have potential in industrial markets for resource extraction and hybrid energy systems. The production of hydrogen for times of peak-electricity demand could replace fossil fuel plants throughout Canada.

As the future of global electricity production moves towards GEN IV reactors, if Canada would like to continue to use nuclear energy beyond a few decades, alterations in fuel cycles, established technologies, fuel handling policies, and public perception will need to be adjusted.

6 References

[1] “Global Warming of 1.5C: Summary for Policy Makers” IPCC, 2018.

[2] Gorensek, M., Forsberg, M., “Relative Economic Incentives for M Hydrogen From Nuclear, Renewable, and Fossil Energy Sources,” International Journal of Hydrogen Energy 34, 2009.

[3] Canadian Small Modular Reactor Roadmap Steering Committee (2018). A Call to Action: A Canadian Roadmap for Small Modular Reactors. Ottawa, Ontario, Canada.

[4] Small Modular Reactors: Regulatory Strategy, Approaches and Challenges. CNSC, May 2016.

[5] Allan, C., Dormuth, K., “The Back End of the Fuel Cycle and CANDU” IAEA-SM-357/10.

[6] Silverio, LB., Lamas, W., “An Analysis of Development and Research on Spent Nuclear Fuel Reprocessing” Energy Policy 39, 2011.

[7] Birkett, J. E., et al., “Recent Developments in the PUREX process for Nuclear Fuel Reprocessing: Complexant Based Stripping for Uranium/Plutonium Separation,” CHIMA 59, 2005.

[8] Uhlir, J., Marecek, M., “Fluoride Volatility Method for Reprocessing of LWR and FR fuels,” Journal of Fluorine Chemistry 130, 2009.

[9] Choi, EY., Jeong, SM., “Electrochemical Processing of Spent Nuclear Fuels: An Overview of Oxide Reduction in Pyroprocessing Technology” Progress in Natural Science: Materials International 25, 2015.

[10] Liu, B., et al., “Transmutation of MA in the High Flux Thermal Reactor” Journal of Nuclear Materials 437, 2013.

[11] Greenspan, E., Heidet, F., “Energy Sustainability and Economic Sustainability with Breed and Burn Reactors,” Progress in Nuclear Energy 53, 2011.

[12] Greenspan, E. “Physics of Breed and Burn Nuclear Reactors,” University of California Berkeley, June 2010.

[13] Han, KW., Heinonen, J., Bonne, A., “Radioactive Waste Disposal: Global Experience and Challenges,” IAEA Bulletin, 1997.

[14] Park, BH., Gao, F., Kwon, E, Ko, W., “Comparative Study of Different Nuclear Fuel Cycle Options: Quantitative Analysis on Material Flow,” Energy Policy 39, 2011.

[15] Forsberg, C., “Hybrid Systems to Address Seasonal Mismatches Between Electricity Production and Demand in Nuclear Renewable Energy Grids,” Energy Policy 62, 2013.

[16] Locatelli, G., Bingham, C., Manchini, M., “Small Modular Reactors: A Comprehensive Overview of Their Economics and Strategic Aspects,” Progress in Nuclear Energy 73, 2014.

[17] Magill, J., et al., “Impact Limits of Partitioning and Transmutation Scenarios on the Radiotoxicity of Actinides in Radioactive Waste,” Nuclear Energy 42, 2003.

[18] Uhlid, J., Marecek, M., Skarohlid, J., “Current Progress in R&D of Fluoride Volatility Method,” Procedia Chemistry 7, 2012.

[19] Amano, O., et al., “Solubility of Lanthanide Fluorides in Nitric Acid Solution in the Dissolution Process of FLUOREX Reprocessing System,” Journal of Nuclear Science Technology, 41:1, 55-60, 2012.

[20] Rozon, D., Lister, D., “Reprocessing versus Direct Disposal of Spent CANDU Nuclear Fuel: A Possible Application of Fluoride Volatility,” NWMO Discussion Paper, January 2008.

[21] Poinssot, C., et al., "Assessment of the Environmental Footprint of Nuclear Energy Systems. Comparison Between Closed and Open Fuel Cycles," Energy 69, 2014.

[22] https://www.nrcan.gc.ca/science-data/data-analysis/energy-data-analysis/energy-facts/uranium-and-nuclear-power-facts/20070

[23] https://cns-snc.ca/media/nuclear_info/candu_performance.html

[24] n-rep-03443-10019

[25] IAEA-SM-353/50 XA0053581 CANDU 9 DESIGN SKW YU Atomic Energy of Canada Ltd, Mississauga, Ontario, Canada

[26] Salvatores, M., Slesarev, I., Tchistiakov, A., "The Transmutation of Long-Lived Fission Products by Neutron Irradiation," Nuclear Science and Engineering 130, 1998.

[27] https://canteach.candu.org/Content%20Library/20030101.pdf

[28] Wasywich, K. M., "Characteristics of Used CANDU Fuel Relevant to the Canadian Nuclear Fuel Waste Management Program," AECL-10463 COG-91-340, 1993.

[29] Code of Federal Regulations, "Energy: Containing a codification of documents of general applicability and future effect," p. 366-372, January 2010.

[30] Hocking, W., Cuclos, A. M., Johnson, L. H., "Study of Fission-Product in Used CANDU fuel by X-ray Photoelectron Spectroscopy (XPS) II," Journal of Nuclear Materials 209,1994.

[31] Grupen, C. "Introduction to Radio Protection: A Table of Frequently Used Radioisotopes," Graduate Texts in Physics, DOI 10.1007/978-3-642-02586-0, 2010.

[32] Zhou, W., Wang, Y., Zhang, J., "Integrated model development for safeguarding pyroprocessing facility: Part I-model development and validation," Annals of Nuclear Energy 112, 2018.

[33] "State-of-the-art Report on the Progress of Nuclear Fuel Cycle Chemistry," NEA No. 7267, OECD, 2018.

[34] Goode, J. H., et al., "Voloxidation - Removal of Volatile Fission Products from Spent LMFBR Fuels," Chemical Technology Division, Oakridge National Laboratory, January 1973.

[35] Kim, S. W., et al., "Chemical Stability of Ceramic Anodes in LiCl-Li_2O Molten Salt for Electrolytic Reduction in Pyroprocessing," Nuclear Engineering and Technology 48, 2016.

[36] Soucek, P., et al., "Pyrochemical Reprocessing of Spent Fuel by Electrochemical Techniques Using Solid Aluminum Cathodes," Energy Procedia 7, 2011.

[37] Ahn, B. G., et al., "Immobilization of Lanthanide Oxides Waste from Pyrochemical Process," Asian Energy Procedia 7, 2011.

[38] Yoo, J. H., et al., "A Conceptual Study of Pyroprocessing for Recovering Actinides from Spent Oxide Fuels," Nuclear Engineering and Technology, 2008.

[39] Koyama, T., et al., "Development of Pyro-processing Fuel Cycle Technology for Closing Actinide Cycle," Procedia Chemistry 7, 2012.

[40] Nagarajan, K., “Development of Pyrochemical Reprocessing for Spent Metal Fuels,” Energy Procedia 7, 2011.

[41] Lee, H., et al., “Pyroprocessing Technology Development at KAERI,” Nuclear Engineering and Technology 43, 2011.

[42] Williamson, M. A., Willit, J. L., “Pyroprocessing Flowsheets for Recycling Used Nuclear Fuel,” Nuclear Engineering and Technology, 43, 2011.

[43] Lee, H., et al., “Current Status of Pyroprocessing Development at KAERI,” Science and Technology of Nuclear Installations Article 343492, Hindawi, 2013.

[44] Uhlir, J., et al., “Current Progress in R&D of Fluoride Volatility Method,” Procedia Chemistry 7, 2012.

[45] Gao, F., et al., “Criticality Safety Evaluation of Materials Concerning Pyroprocessing,” Journal of Nuclear Science and Technology 48, 2011.

[46] Ohta, H., et al., “Pyroprocessing of Light Water Reactor Spent Fuels Based on an Electrochemical Reduction Technology,” Nuclear Technology, 150:2, 2005.

[47] “Postclosure Safety Assessment of a Used Fuel Repository in Sedimentary Rock.” NWMO TR-2013-07, p. 110, p.120-121, 2013.

[48] Wiese, H.W., “Actinide Transmutation Properties of Thermal and Fast Fission Reactors including Multiple Recycling,” Journal of Alloys and Compounds 271-273, 1998.

[49] Serpent Manual

[50] Cetnar, J., “General Solution of Bateman Equations for Nuclear Transmutations,” Annals of Nuclear Energy 33, 2006.

[51] Kim, S.K., et al., “Cost Analysis of a Commercial Pyroprocess Facility on the Basis of a Conceptual Design in KOREA,” Annals of Nuclear Energy 80, 2015.

[52] Kim, S.K., et al., “ Economic Assessment of Alternative Pyro-SFR Nuclear Fuel Cycle in Korea,” Progress in Nuclear Energy 6, 2013.

[53] Cho, S., et al., “Dynamic Analysis of once-through and closed fuel cycle economics using Monte Carlo simulation,” Nuclear Engineering and Design 277, 2014.

[54] Ko, W.I., et al., “Preliminary Conceptual Design and Cost Estimation for SFR Fuel Manufacturing Facility (SFMF),” Nuclear Engineering and Design 277, 2014.

[55] Ewing, R., “Nuclear Waste Forms for Actinides,” PNAS 96, p.3432-3439, 1999.

[56] Piet, S.J., et al., “Dynamic Simulations of Advanced Fuel Cycles,” Nuclear Technology 173, 2011.

[57] Salvatores, M., et al., “The Physics of TRU Transmutation - A Systematic Approach to the Intercomparison of Systems,” PHYSOR, 2004.

[58] Ion, M., “Some Implications of Recycling CANDU Used Fuel in Fast Reactors,” NWMO-TR-2015-11, 2015.

[59] Gobien, M., “Preliminary Hazard Assessment of Waste from an Advanced Fuel Cycle,” NWMO-TR-2015-22, 2015.

[60] Hyland, B., Gihm, B., “Scenarios for the transmutation of actinides in CANDU reactors,” Nuclear Engineering and Design 241, 2011.

[61] Takeda, T., et al., “Interpretation of Actinide Transmutation in Thermal and Fast Reactors,” Progress of Nuclear Energy 40 pp. 449-456, 2002.

[62] Ivanov, N.V., et al., “The results of the transmutation of fission fragments in the spectrum of neutrons of thermal and fast reactors,” Nuclear Energy and Technology 3, 2017

[63] Kazansky, Yu., Romanov, M.I., “Transmuting minor actinides with thermal reactor neutrons,” Nuclear Energy and Technology 1, 2015.

[64] Liu, B., et al., “Transmutation of minor actinides in the pressurized water reactors,” Annals of Nuclear Energy 64, 2014.

[65] Hu, W., et al., “Minor actinide transmutation on PWR burnable poison rods,” Annals of Nuclear Energy 77, 2015.

[66] Takeda, T., “Minor actinides transmutation performance in a fast reactor,” Annals of Nuclear Energy 95, 2016.

[67] Becker, B., et al., “Feasibility of a critical molten salt reactor for waste transmutation,” Nuclear Energy 50, 2008.

[68] Biss, K., Thomauske, B., “Shortening Transmutation time using the molten salt reactor,” Annals of Nuclear Energy 83, 2015.

[69] Yu, C., et al., “Analysis of minor actinides Transmutation for Molten Salt Fast Reactor,” Annals of Nuclear Energy 85, 2015.

[70] Tucek, K., et al., “Comparative Study of minor actinide transmutation in Sodium and Lead-cooled fast reactor cores,” Nuclear Energy 50, 2008.

[71] Ignatiev, V., et al., “Progress in Development of Li, Be, Na/F Molten Salt Actinide Recycler & Transmuter Concept,” Proceedings of ICAPP, Paper 7548, 2007.

[72] Ignatiev, V., et al., “Molten Salt actinide recycler and transforming system without and with Th-U support: Fuel cycle flexibility and key material properties,” Annals of Nuclear Energy 64, 2014.

[73] Vezzoni, B., et al., “Plutonium and Minor Actinides incineration options using innovative Na-cooled fast reactors: Impacting on phasing-out and on-going fuel cycles,” Progress in Nuclear Energy 82, 2015.

[74] Ottensmeyer, P., “Synergy Between CANDU and Fast-Neutron Reactor Technologies,” 10th International Conference on CANDU Maintenance, 2014.

[75] Ottensmeyer, P., “The SMR Challenge in Canada: Enriched Fuel; A Solution: Recycling of Used CANDU Fuel,” 4th Conference on Nuclear Waste Management, 2019.

[76] Kuznetsov, V., “Options for small and medium sized reactors (SMRs) to overcome loss of economies of scale and incorporate increased proliferation resistance and energy security,” Progress in Nuclear Energy 50, 2008.

[77] Lindroos, T., et al., “A techno-economic assessment of NuScale and DHR-400 reactors in a district heating and cooling grid,” Energy Sources, Part B: Economics, Planning, and Policy Vol 14., 2019.

[78] Varri, K., Syri, S., “The possible role of modular nuclear reactors in district heating: Case Helsinki Region,” Energies 12, 2019.

[79] Wojtaszek, D., “Potential Off-Grid Markets of SMRs in Canada,” Canadian Nuclear Review, CW-123700-CONF-048, 2017.

[80] Forsberg, C., “Futures for Hydrogen Produced Using Nuclear Energy,” Proceedings of INES-1, 2004.

[81] El-Emam, R., Khamis, I., “Advances in hydrogen production: results from an IAEA collaborative research project,” International Journal of Hydrogen Energy 44, 2019

[82] Forsberg, C., “Sustainability by combining nuclear, fossil, and renewable energy sources,” Progress in Nuclear Energy 51, 2009.

[83] Finan, F., et al., “Nuclear technology & CCanadian Oil Sands: Integration of Nuclear Power with In-Situ Oil Extraction,” Massachusetts Institute of Technology, 2005.

[84] Verbruggen, A., et al., “Assessment of Actual Sustainability of Nuclear Fission Power,” Renewable and Sustainable Energy Reviews 32, 2014.

[85] Soderholm, K., et al., “Licensing process characteristics of Small Modular Reactors and spent nuclear fuel repository,” Nuclear Engineering and Design 276, 2014.

[86] Banerjee, S., Bonnefous, AM., “Stakeholder Management and Sustainability Strategies in the French Nuclear Industry,” Business Strategy and the Environment 20, 2010.

[87] “Nuclear Security Regulations,” SOR/2000-209, Current July 2019.

[88] Pencer, J., “Security of the Fuel Cycle,” World Institute for Nuclear Security, Security for SMRs, November 2019.

[89] “Packaging and Transport of Nuclear Substances Regulations, 2015” SOR/2015-145, Current July 2019.

[90] “Small Modular Reactors: Regulatory Strategy, Approaches, and Challenges,” DIS-16-04, May 2016.

[91] Gao, F., Ko, W.I., “Modeling and System analysis of fuel cycles for nuclear power sustainability (I): Uranium consumption and waste generation,” Annals of Nuclear Energy 65, 2014.

[92] Tsoulfandis, N., “The Nuclear Fuel Cycle,” American Nuclear Society, 2013.