Closing the CANDU nuclear reactor fuel cycle with a Modified PUREX Processing System: Reducing, Refining and Recycling CANDU Spent Fuel

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Abstract

Currently, CANDU nuclear reactors in Canada utilize less than 1% of the potential energy in fresh fuel bundles before storing the remainder as nuclear 'waste' at on-site nuclear waste management facilities. With the modified PUREX processing system, it is economically possible to close the fuel cycle by extracting and converting the remaining 99% to usable energy. It is also feasible to separate out rare earths and other fission products, which decay much faster than the longer-lived actinides in the spent fuel and may be sold as precious metals. Overall, the process is designed to mitigate proliferation efforts, as well as produce a mixed-oxide fuel for reuse in CANDU reactors and metallic fuel for use in fast-neutron reactors. The multiple products generated create a profit centre.

1. Introduction

More than 50% of Ontario's electrical energy usage is provided by the nuclear energy sector [1]. A by-product associated with the production of this energy is the accumulation of spent CANDU nuclear fuel bundles, which are currently considered unusable nuclear waste and require approximately 400,000 years of storage before decaying to background levels of natural uranium [2]. There are approximately 45,000 tonnes of spent nuclear fuel in Canada and in Ontario alone this amount is projected to increase at a rate of 1,500 tonnes per year [2]. However, only 0.74% of the original incoming fuel in a spent fuel bundle is utilized to create energy, leaving approximately 99.26% of the original fuel as untapped potential energy [1]. This is equivalent to approximately 4,000 years of energy at current levels of nuclear energy production in Canada or $50 trillion of electricity at an average cost of 9.9 cents/kWh [2]. The spent fuel also contains many elements that hinder the fission reactions, known as fission products (FPs), but decay relatively rapidly (within 300 years) to stable elements including cadmium, molybdenum, rhodium, ruthenium and silver. The values of rhodium and ruthenium alone are approximately 1,175.00 USD/oz. and 85.00 USD/oz. respectively on today's markets [3], while the present stock pile of spent fuel has been estimated to contain over $740 million of valuable fission products [2]. In order to capitalize on this large energy and economic resource, as well as reduce unnecessary long term storage of radioactive material, a processing system is needed to close the fuel cycle by producing recycled fuel for nuclear reactors. The processing system proposed is a modification of the traditional plutonium-uranium extraction (PUREX) process to better serve the fuel processing needs of Canada's fleet of CANDU nuclear reactors.

2. Modified PUREX Process

2.1 Overview of Conventional PUREX

The conventional PUREX process is a liquid-liquid extraction technique implemented to separate uranium (U) and plutonium (Pu) from a complex mixture of radioactive fuel elements dissolved in nitric acid. Separation is achieved by using an extracting agent known as tributyl phosphate (TBP) in a normal paraffinic hydrocarbon diluent.

The PUREX process engenders concerns about proliferation due to the isolation of Pu and concerns about damage to the environment from the disposal of high-level liquid waste containing long-lived radionuclides. Furthermore, it does not separate long-lived minor actinides (also known as transuranics or TRUs) from the short-lived FPs. The former may be used as fuel in fast-neutron reactors (FNRs) with a fraction reused as CANDU fuel, while the latter decay to valuable metals. The original PUREX process needs to be modified and optimized to achieve these end products and overcome the aforementioned concerns.

2.2 Process Flow of the Modified PUREX Processing System

The scope of the modified PUREX processing system includes the processing of spent nuclear fuel, from delivery of CANDU spent fuel to the formation of fresh metallic FNR fuel and a mixed metal oxide (MOX) as potential fuel in CANDU reactors. The intermediate stages are shown in Figure 1.

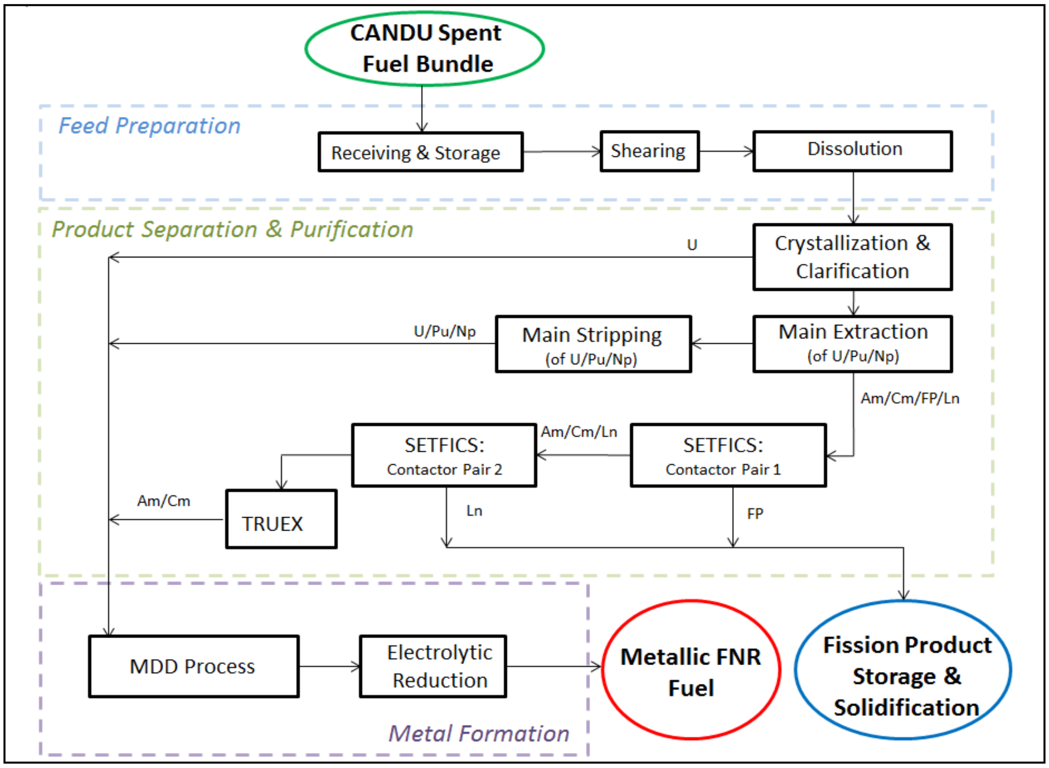

Figure 1: Overall Process Block Diagram for the Modified PUREX Process

2.2.1 Fuel Bundle Shearing

CANDU spent fuel processing via the modified PUREX process begins by removing the fuel rods, containing mainly uranium dioxide, from CANDU fuel bundles. The rods are mechanically sheared into small segments to achieve consistent pellets and high surface areas for dissolution [4].

2.2.2 Dissolution

After shearing, the uranium dioxide fuel pellets are dissolved in 4M nitric acid at 95°C in a multistage, counter-current flow, rotary dissolver. This dissolver operates continuously and provides effective mixing for dissolution at lower concentrations of acid [4]. Higher concentrations of nitric acid would yield higher rates of dissolution, but would also increase hazards due to higher acidity and corrosion rates. The main dissolution reaction in nitric acid of 10M or less is shown in Rx. 1.

$$3UO_2 + 8HNO_3 = 3UO_2(NO_3)_2 + 2NO + 4H_2O \quad \Delta H_r° = -367.6 \text{ kJ/mol} \qquad \text{(Rx.1)}$$

During dissolution, NO_x and radioactive iodine off-gases are emitted. NO_x is processed to reform nitric acid; while iodine is adsorbed using a silver nitrate impregnated silica gel [5]. The concentrated dissolver solution (approximately 300 g U/L) is sent for crystallization; while the undissolved zircaloy cladding is rinsed clear of fissile material. This zircaloy is the first value-added product extracted, which is reused as an alloying element in fast-neutron reactor (FNR) fuel.

2.2.3 Crystallization and Clarification

The uranyl nitrate in the dissolver solution becomes less soluble at lower temperatures, such that at 10-20°C approximately 70-80% of the highly concentrated uranyl nitrate crystallizes out as uranyl nitrate hexahydrate (UNH) crystals [6]. If desired, it is possible to achieve greater U crystallization with lower temperatures (e.g. up to 95% at -10°C and 99% at -30°C) [6]. The crystallization reaction is shown in Rx. 2.

$$UO_2^{2+} + 2NO_3^- + 6H_2O = UO_2(NO_3)_2{\bullet}6H_2O \quad \Delta H_r° = -20 \text{ kJ/mol} \qquad \text{(Rx.2)}$$

Crystallization and washing to remove any FPs (3 wash cycles for a decontamination factor of about 100 [6]) is done in a rotary, inclined, temperature-controlled, continuous crystallization device.

Crystallization of 75% U reduces the amount of aqueous and organic solution being processed by a factor of four, as well as the contaminated liquid waste requiring treatment. Crystallization also facilitates the production of pure uranium oxide by calcination of UNH crystals. Pure uranium oxide can be stored indefinitely using conventional means, sold or used to create MOX or metallic fuel. Concurrently, crystallization of the depleted U out of solution concentrates the amount of fissile material in solution enabling the production of fuel suitable for use in CANDU reactors (see Section 2.2.7).

2.2.4 Main Liquid-Liquid Extraction and Stripping – Extraction of U, Pu and Np as a Group

After crystallization the remaining solution is fed into a multistage centrifugal contactor to separate the residual actinides from FPs via counter-current, liquid-liquid extraction. Liquid-liquid extraction via multistage centrifugal contactors works similar to gravity mixer-settlers, where mass transfer of a solute from one phase to a second, immiscible phase is promoted by successive mixing and settling. In any given stage of the centrifugal contactor, mixing is achieved by means of a stationary agitation disc mounted on a central drum. After the chosen solute is transferred into the desired phase, the liquids are separated by centrifugal forces generated by a rotating bowl. This forces the heavier phase to the outer portion, while the lighter phase occupies the inner portion. The phases are directed out of the stage in separate streams by heavy and light phase weirs [7]. A schematic of this equipment is depicted in Figure 2.

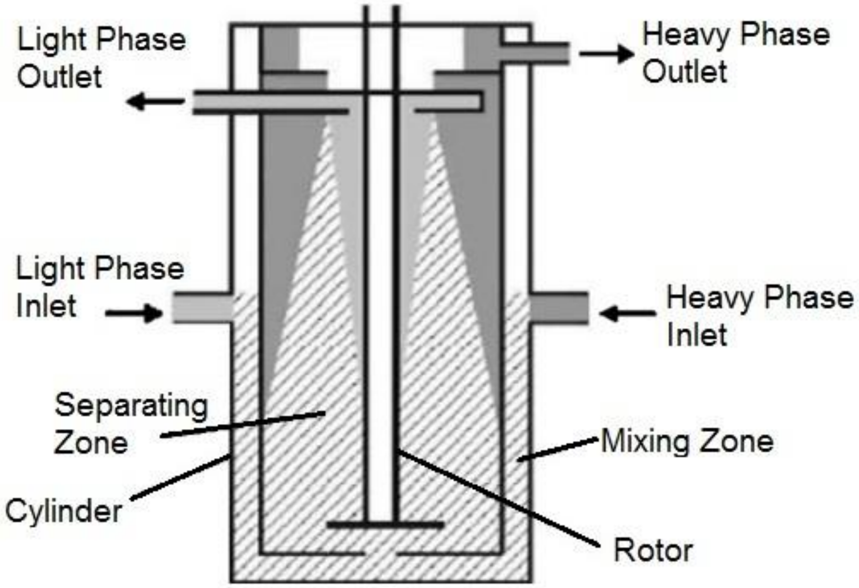

Figure 2 Schematic of a Centrifugal Contactor [8]

The extracting agent used is an organic solution of 30 vol. % TBP in n-dodecane (i.e. light phase). This organic is fed into one end of the contactor, while 2M fresh nitric acid and the dissolver solution (i.e. heavy phase) are fed counter-currently [9]. Provided that a high acid concentration is maintained, tetravalent and hexavalent actinides (i.e. U, Pu and Np solutes) are preferentially extracted from the dissolver solution into the immiscible organic TBP solution. The main reactions are shown in Rx. 3, 4, and 5.

$$UO_2^{2+} + 2NO_3^- + 2TBP = [UO_2(NO_3)_2*2TBP] \quad \Delta H_r° = -23 \text{ kJ/mol} \qquad \text{(Rx. 3)}$$

$$Pu^{4+} + 4NO_3^- + 2TBP = [Pu(NO_3)_4*2TBP] \qquad \text{(Rx. 4)}$$

$$NpO_2^{2+} + 2NO_3^- + 2TBP = [NpO_2(NO_3)_2*2TBP] \qquad \text{(Rx. 5)}$$

The remaining FPs and minor actinides exit in an aqueous phase. The loaded organic phase is then fed into another multistage contactor containing 0.2M of nitric acid [9]. This strips the U, Pu and Np into

an aqueous phase for further processing. Overall, the decontamination factor of FPs is greater than 10,000 for U, Pu and Np [6], more than sufficient to qualify these actinides as FNR fuel.
Centrifugal contactors used in this process have a lower volume hold up, achieve steady state faster, have shorter residence times and have improved instrumentation and control over other alternatives (e.g. mixer-settlers, pulse columns). They can also be rapidly restarted if necessary. These traits mitigate concerns about criticality and lessen the degradation of the TBP solvent due to reduced exposure to radiation [10]. They also have a high throughput, utilize minimal floor space and have a high efficiency (each mechanical stage is nearly one theoretical stage). These factors lead to a more compact and easily maintained system [10].

2.2.5 Solvent Extraction for Trivalent f-elements Intra-group Separation in a Carbamoylmethyl phosphine oxide-complexant System (SETFICS)

The minor actinides, mainly americium (Am) and curium (Cm), plus the lanthanide and non-lanthanide FPs leaving in the aqueous stream of the main extraction are separated using SETFICS. The process is composed of two multistage centrifugal contactor pairs.

The first pair is designed to extract minor actinides and lanthanides into an organic phase and to remove any nitric acid impurities. The CMPO-TBP-n-dodecane solvent is fed at one end of the first contactor as the organic stripping agent, running counter-current to a 10M nitric acid solution. The organic phase extracts all trivalent actinides and lanthanides from the aqueous phase via Rx. 6 (using M^{3+} for a generic actinide or lanthanide metal) [11].

$$M^{3+} + 3NO_3^- + 3CMPO = M(NO_3)_3CMPO_3 \quad \Delta H_r° = -21 \text{ to } -27 \text{ kJ/mol} \qquad \text{(Rx. 6)}$$

The remaining (non-lanthanide) FPs are sent for product treatment and then storage (see Section 3.2). The organic stream is reacted with hydroxylamine nitrate (HAN) in the second contactor to remove acid impurities.

The second multistage centrifugal contactor pair is designed to separate the lanthanide FPs from the minor actinides. In this case the lanthanides and actinides in the organic are fed counter-current to an aqueous solution containing diethylene triamine pentaacetic acid (DTPA) as a chelating agent, as well as HAN. DTPA preferentially forms complexes with minor actinides and extracts them into an aqueous phase according to the equilibrium shown in Rx. 7 with a generic trivalent actinide Ac^{3+} [12].

$$Ac(NO_3)_3CMPO_3 + DTPA^{5-} = AcDTPA^{2-} + 3NO_3^- + 3CMPO \quad \Delta H_r° = -14 \text{ to } -16 \text{ kJ/mol} \qquad \text{(Rx. 7)}$$

This reaction has been shown to achieve trivalent actinide-lanthanide group separation with a decontamination factor greater than 10 [6]. Another advantage of DTPA is that it is composed of only C, H, O and N, and thus may be incinerated leaving no large volume for disposal [13]. The remaining lanthanide FPs leaving with the organic are stripped into an aqueous phase using dilute nitric acid in the final contractor and are sent for product treatment with the other FPs.

2.2.6 Transuranic Extraction (TRUEX)

The trivalent minor actinide product in DTPA and HAN, which leaves the SETFICS process, is treated further in a multistage centrifugal contactor process known as TRUEX. Before being processed into an oxide or FNR metal fuel, the minor actinides must be extracted out of the DTPA and HAN solution into an aqueous dilute nitric acid phase. This works like the first contactor pair of SETFICS, where CMPO-TBP-n-dodecane extracts trivalent actinides in one contactor and in the second contactor dilute nitric acid is used to strip them into an aqueous phase [9]. This is shown in Rx. 8 and 9.

$$\mathrm{AcDTPA^{2-} + 3NO_3^{-} + 3CMPO = Ac(NO_3)_3CMPO_3 + DTPA^{5-}} \qquad \Delta H_r° = 14 \text{ to } 16 \text{ kJ/mol} \qquad \text{(Rx. 8)}$$

$$\mathrm{Ac(NO_3)_3{\bullet}3CMPO = Ac^{3+} + 3NO^{3-} + 3CMPO} \qquad \Delta H_r° = 21 \text{ to } 27 \text{ kJ/mol} \qquad \text{(Rx. 9)}$$

2.2.7 Actinide Oxide Formation

The main non-FP outputs of the product separation and purification section of the modified PUREX process are in the form of a U/Pu/Np stream, a stream of minor actinides in nitric acid and purified UNH crystals. For fuel production, the aqueous streams containing actinides must be converted into an oxide (for CANDU fuel) or a metal form (for most efficient FNR fuel use). The oxide form is unavoidable as direct reduction strategies are impractical.

The U/Pu/Np stream is oxidized separately to re-form MOX fuel suitable for CANDU reactors. As 75% of the initial U, as well as fission products, have been removed from the U/Pu/Np stream, the fissile Pu-239, Pu-241 and U-235 content is concentrated to around 4 times its original value. The Pu-239/Pu-241 composition increases from 0.27% in spent CANDU fuel to about 1.08%, while the remaining U-235 after crystallization increases to about 0.23% from 0.06% before crystallization [1]. This gives a total of 1.31% fissile material, which is even greater than the 0.72% necessary for typical CANDU fuel.

Two operational orientations exist for handling the minor actinide stream, as they can be oxidized along with the U/Pu/Np stream for CANDU fuel or oxidized separately, reduced to a metal (see Section 2.2.8) and used as FNR fuel. As mentioned, the UNH is oxidized to form a relatively benign oxide that can be stored, sold or used to produce more FNR fuel.

A process called Modified Direct Denitration (MDD) is preferable for precipitation of actinides as oxides. To make a mixed actinide oxide, the designated actinide stream (i.e. U/Pu/Np, minor actinides or UNH) is treated with ammonium nitrate to form a ratio of ammonium ions to actinide of 2:2.6 [14]. This re-dissolution and ammonium treatment is required, as direct conversion of actinide nitrates to an oxide results in a glassy product unfit for further reduction treatments or for use as fuel [14].

The MDD solution is metered into an electric, inclined rotary kiln for continuous operation, fine powdered products, smooth operation and better control, with less temperature fluctuations, uniform off-gas and constant product accumulation [15]. The general chemical reaction for this process is shown in Rx. 10 for Pu. It occurs at temperatures of up to 500°C [14].

$$\mathrm{2NH_4NO_3 + Pu(NO_3)_4 \rightarrow PuO_2 + 2N_2O + 4H_2O + 4NO_2 + O_2} \qquad \Delta H_r° = 197.9 \text{ kJ/mol} \qquad \text{(Rx.10)}$$

To facilitate this treatment, air is blown counter-current to the solid flow to purge the evaporation and decomposition products, including water vapour, nitric acid vapour, NO_x and N_2O.

2.2.8 Reduction

After the formation of actinide oxides the compounds may be removed from the process for storage or be reduced to a metallic form for FNR fuel. Based on process simplicity and efficiency electrolytic reduction is the preferable method.

After MDD, the mixed actinide oxides are transferred into fuel baskets (a type of porous ceramic magnesia filter) and loaded into a large electrolytic cell composed of a cathode, anode and electrolyte under an argon atmosphere. The metal oxides within the fuel basket function as the cathode. An inert, conductive ceramic serves as the anode, and a $LiCl/Li_2O$ (3 wt. %) molten salt at 650°C is the electrolyte. When a voltage of about 2.40V is applied, the metal ions of the oxides are reduced to their base metals [16]. Concurrently, oxide ions formed are oxidized to oxygen gas and continuously vented from the cell by inert argon gas to maintain a constant oxygen partial pressure necessary to maintain the anode potential [17]. Typical efficiencies are 99.7% for U, >97.8% for Pu, 98.8% for Np and >90% for Am [16]. The half-reactions and overall reaction for some metal actinide, M, are as follows:

Cathode process: $M_xO_y\,(s) + 2y\,e^- = x\,M\,(s) + y\,O^{2-}$ (Rx.11)
Anode process: $O^{2-} = \frac{1}{2}\,O_2\,(g) + 2e^-$ (Rx.12)
Overall process for UO_2: $UO_2\,(s) = U\,(s) + O_2\,(g) \quad \Delta H_r^\circ = 1084\ \text{kJ/mol}$ (Rx.13)

After reduction, the cathode material is in the form of a metallic lump of U, Pu, Np and other minor actinides. This metal represents the final product of the modified PUREX processing system and may be transferred to a metallurgical treatment facility to be smelted, casted into FNR fuel pins and sealed into the FNR fuel bundle.

2.2.9 Process Efficiency

The following table summarizes overall efficiencies estimated for the modified PUREX process, including both the overall recovery of FPs and actinides in pure form (i.e. the amount extracted compared to the amount fed), as well as their final distribution between the main output streams. Efficiencies have been estimated based on a collection of lab, pilot and industrial scale process data.

Table 1: Overall Process Extraction Efficiencies

	Overall Recovery in Pure Form (wt. % of original)	**Composition (wt. %) of Output Streams**	
		Actinide Product Stream	**Fission Product Stream**
Fission Products	98.02	0.87	99.95
Actinide Product	99.70	99.13	0.05
Sum	*N/A*	*100*	*100*

3. Waste Mitigation

3.1 NO_x Recovery

During normal operation approximately 7kg/h of NO_x gas is produced. The gas is recycled to make nitric acid that is reused as process solvent. To enable the conversion, NO_x gas is fed counter-currently to oxygen and water in an absorption tower, where it is oxidized to NO_2 and forms nitric acid. Any NO present is recycled in the process.

3.2 Liquid Waste Treatment

Non-recyclable liquid waste streams containing FPs need to be pre-treated before being transported and stored at a storage facility. Storing liquid waste for extended periods of time can be disadvantageous when compared with solid waste, as leaks or spills can occur. Furthermore, storage of liquid is more difficult, costly and is less appropriate for subsequent transport. To select an appropriate solidification technique for the FPs a number of factors were considered. Due to the radioactivity and associated decay heat of FPs the solid product must display thermal stability and resistance to radiation. The solid must be chemically stable to prevent the FPs from leaching or becoming volatile [18]. Lastly, the volume for storage should be reduced significantly.

The most developed process for managing PUREX high-level liquid waste is vitrification. The approach has been adopted by a number of French facilities, including La Hague, as well as some U.S. Department of Energy facilities, including the Hanford Waste Vitrification Plant [18]. The vitrification process is used to incorporate the liquid into a solid form of highly stable borosilicate glass blocks. Ammonium nitrate is added to promote thermal stability before the mixture is calcined in an inclined, rotating kiln with a temperature of about 600°C [18]. The calcined material flows by gravity into a glass-making furnace where it is mixed with glass frit. After heating at about 1150°C, the molten product is poured into stainless steel storage containers, which are sealed and loaded into flasks for transportation to a storage facility [18]. This yields a significant reduction is waste. For example, one tonne of LWR spent fuel results in only 70 to 80 liters of solid waste [18].

4 Economics

An economic analysis was performed on the preliminary design of the modified PUREX process. The analysis included looking at the capital cost, operating cost and revenue generated by the plant. A cash flow analysis was done for an assumed 40-year plant life [19], and economic metrics such as net present value (NPV), internal rate of return (IRR) and payback period were calculated to determine if the processing plant design was economically feasible. The analysis was done based on processing approximately 16,000 CANDU fuel bundles per year, which equates to 304 tonnes of heavy metal (HM) per year.

The capital cost included the cost of each major piece of equipment (including the associated installation, piping, electrical and instrumental costs), the cost of building the plant (including land, material, and shielding), as well as indirect costs (such as contractor fees), and a contingency. The operating cost included the cost of raw materials, utilities, and both direct and indirect labour costs, which includes an allowance for equipment replacement. The revenue was calculated by assuming that all of the metal actinide products would be sold at a price based on the price of U. Thus, calculating the yearly cash flow with depreciation, taxes, and inflation, a cash flow analysis was generated. From this cash flow a NPV calculation showed the project was profitable at an IRR of 12%, which was above the minimum acceptable rate or return of 10% often used by the nuclear industry [20]. From the cost analysis, the project was economically feasible, and could be made more profitable if the throughput or the price at which the product is sold at is increased.

A cost analysis of an existing PUREX plant showed a capital cost of approximately $1 billion [21]. This varied greatly with the economic analysis performed here where the calculated capital cost was approximately $60 million. A similar discrepancy is observed with the operating cost where a report states that the general cost of PUREX processing and MOX fuel fabrication is approximately $2250/kgHM [22]. The calculated cost of the current analysis was $115/kgHM.

5 Design Benefits and Recommendations

Spent nuclear fuel contains the fissile isotopes U-235, Pu-239 and Pu-241. Their existence at higher concentrations raises concerns about the potential proliferation of nuclear weapons [23]. The conventional PUREX process is designed to separate the Pu, which if used in the processing of civilian spent fuel would inadvertently increase the chances of proliferation. The modified PUREX process has been designed to ensure that Pu is consistently in a mixture solution with other actinides or fission products at concentrations that preclude their use as explosives. At no point during the process is Pu separated from the other fuel elements, which is in line with proliferation resistance efforts put in place by the CNSC and the IAEA.

The initial research and design was performed for recycling CANDU fuel approximately 10 years old. The spent fuel available is greater than 10 years old and thus it is expected that the radioactivity of some of the isotopes would have decreased. Additionally, the process is adaptable to recycling various types of fuel. The current industrial use of the PUREX process is for light water reactors (LWR) and pressurized water reactors (PWR). With a few considerations and modifications, it is expected that the process will be able to recycle FNR fuel as well [24]. The U/Pu/Np stream obtained at the end of the process is highly fissile due to high Pu239, Pu241 and U235 content (approximately 1.31%). Since this is higher than the fissile content in fresh CANDU fuel (approximately 0.72%), the MOX product of this stream can potentially be used to refuel CANDU reactors. The pure U stream extracted at the crystallization stage could also be directly used to refuel an FNR. The ability of the PUREX facility to process a wide variety of fuels and have the output fuel be used in various reactors closes the fuel cycle, while extracting the maximum energy with minimal losses.

Areas recommended for further investigation include the recycle of waste streams, where a more detailed recycling percentage would be established for the inclusion of recycling tanks. Numerous assumptions were made when performing the economic analysis with regards to the various rates and the MARR, typically 10% for the nuclear industry [20]. Furthermore, as the process has not yet been implemented on the scale and in the configuration described, large uncertainties exist for the cost estimates. A more detailed analysis would have to be performed to determine the economic feasibility of the modified process more accurately. Another recommendation would be to consider extracting stable fission products after processing the spent fuel. If you consider the value of all of the individual amounts of the multitude of fission products, it can be calculated that one tonne of this material would be worth about $2.5 million [2]. Since approximately 85% of these FPs are already stable, it becomes practical to consider extracting the most valuable stable components, such as rhodium and ruthenium, without the 300 year wait required before the total radiotoxicity has decayed to background levels of natural uranium [25]. This possibility is being investigated. Lastly, while industrial complexes for the PUREX process exist, this particular modified version has not been tried industrially. Therefore, the process would have to be tested on a pilot plant scale to verify the conclusions and efficiencies provided within this preliminary design.

6 Conclusion

The proposed modified PUREX process is designed to extract a spectrum of valuable products. These include zirconium, purified FPs, major and minor actinides, as well as metal and metal oxide nuclear fuel. Reprocessing these spent nuclear fuel bundles has multiple purposes. Firstly, it will decrease the amount of nuclear waste requiring storage in Canada by recovering the actinide components of the spent fuel to be reused as fuel for nuclear reactors. A further motivation arises due to the economic benefits from the potential revenue stream of about $50 trillion in non-carbon electrical energy production from the recovered actinides and $740 million through the isolation of the FPs in the spent fuel. The process proposed to accomplish these benefits offers simplicity and reliability through the use of liquid-liquid extraction techniques, most of which have a long track record of implementation on an industrial scale. It also offers high separation efficiency, producing an actinide product and fission product stream with purities of **99.13%** and **99.95%** respectively. Overall, this option should provide the processing system with multiple products to allow for economic operation regardless of market price fluctuation or the construction of an FNR within Canada, as well as aid in the public perception of the nuclear industry as a more sustainable energy sector.

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8 References

[1] NWMO, "Moving Forward Together. NWMO Triennial Report 2008 to 2010," 2011. [Online]. Available: http://www.nwmo.ca/uploads_managed/MediaFiles/1721_triennialreport2008to2010.pdf.

[2] P. Ottensmeyer, "Candu Fuel Waste Re-used, Recycled, Eliminated; $45 trillion of Carbon-Free Electrivity via Fast Neutron Reactors," July 2012. [Online]. Available: http://www.peo.on.ca/DIMENSIONS/julyaug2012/PolicyEngagement.pdf.

[3] "Precious Metals Investing - Prices, Mining Stocks and News," 04 February 2013. [Online]. Available: http://www.infomine.com/investment/precious-metals/. [Accessed 04 February 2013].

[4] R. Jubin, "Spent Fuel Reprocessing," in *Introduction to Nuclear Chemistry and Fuel Cycle Separations Course, Consortium for Risk Evaluation With Stakeholder Participation*, Nashville, 2008.

[5] T. J. Daryl R.Haefner, Methods of Gas Phase Capture of Iodine from Fuel Reprocessing Off-Gas: A Literature Survey, Idaho National Laboratory , 2007, p. i.

[6] T. Takata, Y. Koma, Sato, Koji, Kamiya, Masayoshi, A. Shibata, K. Nomura, H. Ogino, Koyama, Tomozo and S.-i. Aose, "Conceptual Design Study on Advanced Aqueous Reprocessing System for Fast Reactor Fuel Cycle," *J Nucl Sci Technol,* vol. 41, no. 3, pp. 307-314, 2004.

[7] Rousselet-Robatel, "Rousselet Robatel Model LX Multistage Centrifugal Extractor," [Online]. Available: http://www.rousselet-robatel.com/products/pdfs/multistage-centrifugal-extractor-operatingprinciple.. [Accessed October 2012].

[8] J. McFarlane, J. Birdwell Jr., C. Tsouris and H. Jennings, "Process intensification in continuous base-catalyzed biodiesel production," in *Proceedings of the AIChE Annual Meeting*, 2008.

[9] M. Nakahara, Y. Sani, Y. Koma, M. Kamiya, A. Shibata, T. Koizumi and T. Koyama, "Separation of Actinide Elements by Solvent Extraction Using Centrifugal Contactors in the NEXT Process," *J Nucl Sci Technol,* vol. 44, no. 3, pp. 373-381, 2007.

[10] R. Herbst and M. Nilsson, "Standard and advanced separation: PUREX processes for nuclear fuel reprocessing," in *dvanced Separation Techniques for Nuclear Fuel Reprocessing and Radioactive Waste Treatment*, 2011, pp. 141-173.

[11] T. J. Stockmann and Z. Ding, "Uranyl Ion Extraction with Conventional PUREX/TRUEX Ligands Assessed by Electroanalytical Chemistry at Micro Liquid/Liquid Interfaces," *Anal. Chem.,* vol. 83, no. 19, pp. 7542-7549, 2011.

[12] S. Leguay, T. Vercouter, S. Topin, J. Aupiais, D. Guillaumont, M. Miguirditchian, P. Moisy and C. Le Naour, "New Insights into Formation of Trivalent Actinides Complexes with DTPA," *Inorg. Chem.,* 2012.

[13] T. Todd and R. Wigeland, "Advanced Separation Technologies for Procesing Spent Nuclear Fuel and the Potential Benefits to a Geologic Repository," in *Separations for the Nuclear Fuel Cycle in the 21st Century,* Washington D.C., Oxfrod University Press, 2006, pp. 41-55.

[14] Kitts and F.G., "Pilot-Scale Demonstration of the Modified Direct Denitration Process to Prepare Uranium Oxide for Fuel Fabrication," Oak Ridge Nationoal Laboratory, Oak Ridge, 1994.

[15] L. Felker, R. J. Vedder, E. Walker and E. Collins, "Product Conversion: The Link between Separations and Fuel Fabrications," in *Atalante 2008*, Montpellier, 2008.

[16] C. E. Till and Y. I. Chang, "Electrolytic Reduction Step," in *Plentiful Energy: The Story of the Integral Fast Reactor,* CreateSpace, pp. 211-217.

[17] S. Herrmann, R. King, K. Durstine and C. Eberie, "Pilot-Scale Equipment Development for Lithium-Based Reduction of Spent Oxide Fuel," in *American Nuclear Society 3rd Topical Meeting DOE Spent Nuclear Fuel and Fissile Materials Management*, Charleston, 1998.

[18] R. Kneif, Reprocessing and Waste Management, 2008, pp. 559-593.

[19] "Major International Conference on Nuclear Plant Life Managements Opens in Shanghai," 18 February 2011. [Online]. Available:

http://www.iaea.org/newscenter/news/2007/npp_extension.html. [Accessed 04 February 2013].

[20] W. N. Association, "The New Economics of Nuclear Power," Carlton House, London, UK, 2006.

[21] J. Haire, "Nuclear Fuel Reprocessing Costs," Oak Ridge National Labratory, Hilton Head Island, South Carolina, 2003.

[22] "Nuclear Fuel Cycle Cost Comparison Between Once Through and Plutonium-Recycling in Pressurized Water Reactors," EPRI, Palo Alto, CA, 2009.

[23] D. Levy, "MYTH: Recycling spent nuclear fuel increases the risk of weapons proliferation," [Online]. Available: http://www.ornl.gov/info/ornlreview/v41_3_08/article11.shtml. [Accessed 25 01 2013].

[24] R. Jubin, "Spent Fuel Reprocessing," [Online]. Available: http://www.cresp.org/NuclearChemCourse/monographs/07_Jubin_Introduction%20to%20Nuclear%20Fuel%20Cycle%20Separations%20-%20Final%20rev%202_3_2_09.pdf. [Accessed 25 01 2013].

[25] U.S. Department of Energy, "Blue Ribbon Commission on America’s Nuclear Future," Washington, D.C., 2012.