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A Step Towards Closing The *CANDU* Fuel Cycle: An Innovative Scheme for Reprocessing Used CANDU Fuel

F. Collins[1] and D. Lister[1]

[1] UNB Nuclear, Department of Chemical Engineering,
University of New Brunswick,
Fredericton, NB, Canada

Abstract

Disposal versus reprocessing costs for used CANDU fuel was recently discussed by *Rozon and Lister* in a report produced for the Nuclear Waste Management Organization (NWMO) [1]. Their study discussed the economic incentives for reprocessing, not for the recovery of fissile uranium but for the recovery of plutonium ash. A $370/kg break-even price of uranium was calculated, and their model was found to be very sensitive to the reprocessing costs of the chosen technology. Findings were consistent with earlier studies done by Harvard University. Various reprocessing technologies (most based on solvent extraction) have been in use for many decades, but there appears to be no conceptual engineering study available in the open literature for a spent fuel reprocessing facility – one that includes process flows, operating costs and economic analysis. A deeper engineering study of the design and economics of re-processing technologies has since been undertaken by the nuclear group at the University of New Brunswick. An improved fluorination process was developed and modeled using ASPEN® process simulation software. This study examines the impact of chosen technology on the spent fuel re-processing costs.

1. Introduction

According to the International Atomic Energy Agency [2], approximately 270 000 tonnes of spent nuclear fuel is being held in storage repositories around the world. Over 90% of this material could be considered low level waste if it were not mixed with high level waste requiring radiation shielding. Physical-chemical processes can be used to separate these materials and in fact reprocessing of spent fuel is currently carried out in France, Russia, Japan, India and the UK for the recovery of U-235. However these facilities are utilized at only 50% capacity due in part to the costs associated with reprocessing. With an increase in mining capacity over the last 3 years uranium prices have fallen, so there is general consensus in the industry that a once-through fuel model with waste stored indefinitely is more economical than a reprocessing model where mixed oxide (MOX) fuel and different reactor designs are used to extend the useful life of mined material. Less than 15% of the spent fuel generated from today's reactors is reprocessed worldwide.

There is even less incentive currently to recover uranium from spent CANDU fuel. The U-235 concentration in spent CANDU fuel (approximately 0.23%) is comparable to the waste stream emerging from a uranium enrichment facility (0.2-0.3%). The reasons for reprocessing CANDU fuel would not be to recover uranium; it would be to recover the plutonium ash and to reduce the volume of material needing long term storage. However the costs of PUREX - the most widely used re-processing technology - remain an obstacle that cannot be overcome without more engineering research and economic analysis of possible options to reduce reprocessing costs. A study done by *Bunn, et al* at Harvard University suggested that a 75% reduction in costs would be necessary for reprocessing to be considered competitive with a once-through disposal model [3].

2. PUREX: an industry standard with issues.

The process chemistry for the separation of uranium from plutonium and fission products is well-documented and the most common technology, PUREX, has been in commercial operation for over 50 years. This hydrometallurgical process is similar in principle to technologies used today by any mining industry; in fact, natural uranium is refined on the front-end of the fuel cycle using the same process. PUREX is a simple technology with high separation efficiency (greater than 95%), but like all hydrometallurgical processes it generates liquid waste. The Hanford, Washington nuclear research facility, site of the “Manhattan Project” in the mid 1940s, used solvent extraction processing similar to PUREX for plutonium enrichment. Today, the Hanford site – which is now substantially decommissioned – is the most contaminated nuclear site in the United States. A 1992 government report stated the facility was responsible for the release of “copious volumes of liquid wastes”, resulting in contamination of soil and groundwater. The report estimated a 2 billion $/year cleanup cost, making it the largest environmental clean-up project in the United States [4], and today the project has been estimated to have cost over $50 billion. This facility wasn’t the only facility to experience problems: many users of the PUREX technology have had serious environmental releases, most notably British Nuclear Fuel’s THORP facility in Sellafield, England [5]. Liquid wastes from PUREX, representing 99% of all high level waste in the spent fuel, must be vitrified into glass for disposal [6].

The other disadvantage of the PUREX process is its facility capital and operating cost. To achieve a high level of recovery, several stages of separation are necessary; large volumes of liquids (water, acids and solvents) flow through many vessels that require shielding, etc. Acids are consumable and cannot be re-used. Proprietary organic solvents are recycled through several stages of extraction resulting in the build-up of impurities – radioactive species that could follow uranium through the process into the final product or into the environment. The Nuclear Waste Management Organization (NWMO) estimates a cost of 1500 $/kg heavy metal (kgHM) for the make-up of this solvent. *Bunn et al* [3] used a speculative estimate of $1700/kgHM as a total (capital and operating) reprocessing cost in their study on the economics of reprocessing with PUREX versus direct disposal.

For environmental and operational cost reasons the nuclear research community continues to look for alternatives and improvements to PUREX. Variations involve the selective removal of actinides from the waste; studies for the DIAMEX-SANEX process (also a solvent-extraction process) are underway in France and results are promising, but they do not completely solve the issues of liquid waste, and this process does not reduce processing steps. Several other studies suggest fluorination – another front-end technology used in uranium conversion – and combinations of fluorination with solvent extraction (FLUOREX) to reduce waste volumes. However, no existing commercial scale facilities use either DIAMEX-SANEX or fluorine chemistry for reprocessing spent fuel; and there is little reason to expect any commercial facilities to be built without proof of its performance at scale and confirmation of its economics over the industry-standard PUREX process.

Only a “dry” re-processing option would completely eliminate aqueous waste. Proponents for pyroprocessing, or molten salt processing, have suggested the potential for substantial reduction in reprocessing costs over PUREX. But a subsequent review of this option by the National Academy of Sciences stated that cost estimates produced in the mid-1990s for pyroprocessing were “inexplicably low” and that it was “by no means certain that pyroprocessing will prove more economical than hydroprocessing”. More recent studies by the Nuclear Energy Agency have suggested that pyroprocessing is 2.5 times *more* expensive than hydroprocessing [7] [8].

Direct “flame” fluorination is another technology that has been considered. It has been shown, however, that direct fluorination of spent fuel results in the simultaneous generation of volatile and non-volatile fluorides of many species in the fuel bundle [9]. Table 1, below, lists some of the known stable fluorides that would be generated by fluorination of a spent CANDU fuel bundle.

Table 1- A Partial List of Fluoride Species Resulting from the Fluorination of Spent CANDU fuel.

Element	Approximate Mass fraction in Spent CANDU Fuel	Fluoride Species	Boiling point (°C)	Melting Point (°C)
U	0.997	UF_6	56.5(subl.)	64.8
Pu	0.0013	PuF_6/PuF_4	52 (subl.)	1027
Mo	217ppm	MoF_6	34	17.4
Nd	189ppm	NdF_3	2300	1374
Zr	162ppm	ZrF_4	905	932 (trip. pt.)
La	67ppm	LaF_3	---	1493
Sr	61ppm	SrF_2	2460	1447
Tc	57ppm	TcF_6	55.3	37.4
Sm	28ppm	SmF_3	2427	1306
Np	4.3ppm	NpF_6	54.4	55.2
Sn	2ppm	SnF_4	705	750 (subl.)
Cd	1.4ppm	CdF_2	1748	1110
Am	0.3ppm	AmF_3	---	1395

Volatile fluorides with similar boiling points would have to be separated by a challenging – and expensive – series of distillation steps. This limits the usefulness of this method for recovering uranium from spent LWR fuel. For spent CANDU fuel, where the ash would be the product, non-volatile solids may have to be separated from each other. Direct fluorination is also highly exothermic and requires specially designed flame reactors or fluidized beds, where temperatures are typically controlled using a solid fluidization medium such as alumina pellets that would mix with solid products such as PuF_4, further complicating the separation. In short, proposed fluorination processes have their own economic and practical problems. There is still room for improvement on reprocessing technologies.

3. INFLUX©: A Reprocessing Alternative

Research in the late 1960s and early 1970s was conducted on the use of alternative fluorinating agents for spent LWR fuels. Some of the most promising ideas came from the use of bromine pentafluoride, BrF_5, fed in nitrogen to a fluidized bed of UO_2 / PuO_2 with alumina as a heat transfer medium. *Holmes et al* concluded that BrF_5 was an effective fluorinating agent over a wide range of operating conditions, with uranium conversions exceeding 97% in semi-pilot scale trials [10]. The net reactions were found to be:

$$5U_3O_8(s) + 18BrF_5(g) \longrightarrow 15UF_6(g) + 9Br_2(g) + 20O_2(g)$$
$$5PuO_2(s) + 4BrF_5(g) \longrightarrow 5PuF_4(s) + 2Br_2(g) + 5O_2(g)$$

As with direct fluorination, this reaction provided for the immediate phase-separation of plutonium from the uranium. Holmes reported lower operating temperatures with excellent control of temperatures below 400°C, and the absence of bed sintering when using BrF_5 versus elemental fluorine. *Jarry and Steindler* modeled the kinetics of the reaction with a shrinking core model and accurately predicted the reaction rate as a function of temperature and partial pressure of BrF_5[11]. The work of *Anastasia et al* followed up this work to show how the conversion is affected by the solids concentration in the fluidized bed [12]. All of these researchers acknowledged fission products affecting the purity of the product UF_6 – the valuable component obtained when reprocessing spent LWR fuel. Complicating matters for downstream separation is the fact that *Ellis* et al., in earlier works, reported a difficult separation of bromine (boiling point 58.5°C) and UF_6 (sublimation point 56.5°C); and the existence of an azeotrope in the atmospheric UF_6-BrF_5 system at 0.1 mole fraction UF_6 [13]. So although the chemistry of bromofluorination is favourable, downstream separation of products by simple fractional distillation would not be possible. Very little work has been done on this system since the early 1970s, likely because of this.

For CANDU fuel, where the valuable component would be plutonium, volatile fluorides of fission products do not represent as much of a problem; the non-volatile solid species such as the alumina that would be present in any fluidized bed would be the issue. Fluid-bed reprocessing of CANDU fuel to obtain solid products would be very difficult. The concept recently developed by UNB Nuclear, *INFLUX*© (INdirect FLuorination Uranium oXides), is a variation on the bromofluorination scheme which virtually eliminates the complexity of downstream separation of volatile species. This patent-pending process is shown conceptually in Figure 1.

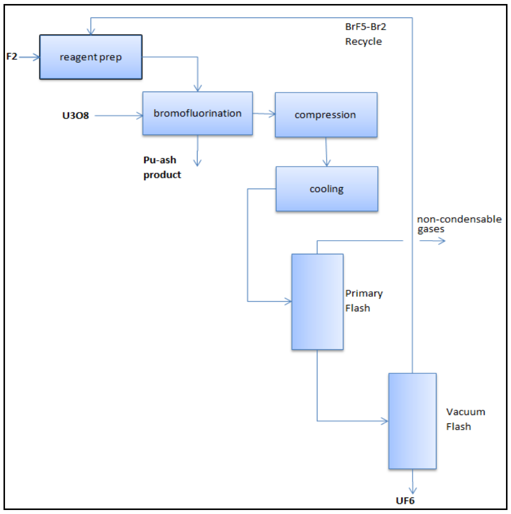

Figure 1 - INFLUX: a Proposed Alternative to Direct Fluorination

This 3-phase process takes advantage of the differences in phase equilibria of uranium hexafluoride and other fluorinated species. Unlike typical vapour-liquid separation schemes involving fluorination, two solid products emerge from the process. After de-cladding and oxidizing, the spent fuel in the form of U_3O_8 is introduced to the first stage of the process where it reacts with BrF_5. The first solid product emerges here as PuF_4, along with the non-volatile fluorides of americium, neodymium, zirconium, niobium, strontium and other minor elements, none of which are serious poisons in common MOX fuels. The gases emerging from the first stage are then compressed and cooled to remove oxygen from the system. Trace quantities of minor actinide fluorides would be removed by conventional treatment methods at this stage. In the final stage, solid UF_6 is produced by vacuum flash separation. Excess reactants are recycled back to the reagent preparation stage. The UF_6 emerging from this system could be shipped for use or, if CANDU fuel had be reprocessed, oxidized for disposal or the uranium as low level waste.

This process combines the attractive features of PUREX (low pressures and temperatures with high yield) and fluoride volatility (low environmental impact), without inheriting the drawbacks of both. In summary, INFLUX$^{©}$ would have the following advantages:

- A non-aqueous process with minimal environmental impact, and high proliferation resistance.
- Favourable reaction chemistry with high yield.
- Low operating temperatures (less than 400°C), giving easily-controllable reactions and an ability to use cheaper designs and materials.
- Simple separation of product species, eliminating complex distillation processes as used in conventional volatility separation.
- Continuous process with a small physical footprint, which would greatly reduce capital and operating costs.
- Ability to accept a variety of feeds, including thorium-rich fuels.

4. INFLUX©: Process modelling

The research work of 50 years ago on fluorine-based processes appears to have ended with the conclusion that it might be too difficult to separate many of the species by volatility differences. This separation can be optimized today with the power of commercial process simulation packages like *Aspen*, which was used in this study to develop the process around bromofluorination chemistry. Using published literature from several sources, the components involved in the reaction of BrF_5 with CANDU fuel pellets were characterized. Their physical and thermodynamic properties of these species were estimated using several semi-empirical correlations and the results were compared with experimentally-determined properties published in the literature. As an example, the results for uranium hexafluoride are shown in Figure 2 where individual property models are compared to generally accepted literature values. Between 60°C and 150°C, the thermophysical properties of UF_6 were shown to vary by 10% from published values. The result of phase diagram modelling for UF_6 is shown in Figure 3 alongside the work of *DeWitt* [14].

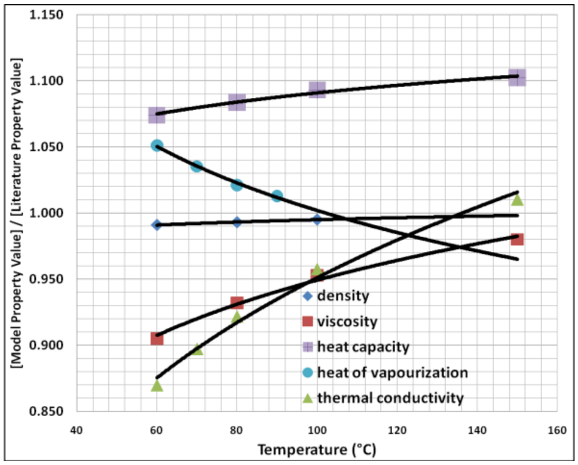

Figure 2 - Estimated Thermophysical Property Variance for Gaseous Uranium Hexafluoride

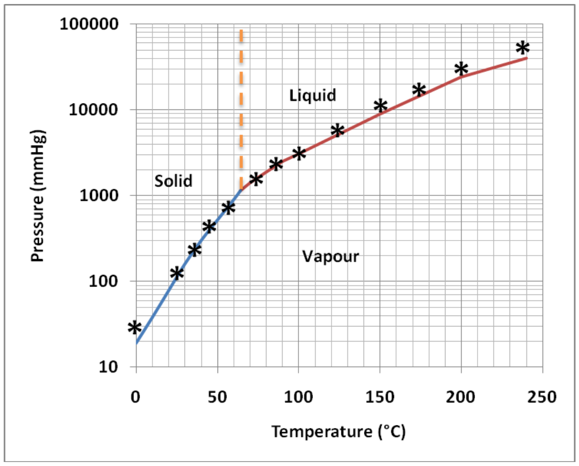

Figure 3 – Comparison of Uranium Hexafluoride Phase Diagram with the model presented by DeWitt[12]

For vapour pressure calculations, published models were used directly in the simulation. In Figure 3, for example, the sublimation and saturation lines were calculated using two separate models developed by *Llewellyn* [15]. These models were said to be accurate to within 0.5% between 0°C and 100°C. Deviations above 100°C are therefore expected but do not seem to be extreme.

All species listed in Table 1 were characterised as hypothetical components in Aspen. Predicted results were compared to literature sources where possible and, in the absence of published data, calculated properties were assumed to be reasonably accurate for this study provided their phase behaviour was correct around the normal boiling points. Physical property models showed good agreement with published literature for most species. However, when species were mixed the modelling was not able to predict some of the binary interaction parameters that would more accurately predict non-idealities. Whenever possible these parameters were forced on the system using available literature (such as for the BrF_5-UF_6-Br_2 system) [16].

On this basis, the INFLUX© process was simulated to produce a working material and energy balance of the process. Temperature, pressure and flow of process streams were varied to arrive at a continuous, steady-state system. The following conditions were found to be optimal for the recovery of plutonium ash from a spent CANDU fuel bundle:

- Bromofluorination reactor conditions:1.5 atm, 230°C.
- Gas compression/condensation stage: 6.8 atm, 80°C.
- Vacuum Flash stage: 0.8 atm, 30°C.

This combination of operating conditions achieved a 97% recovery of the plutonium ash with negligible loss of reagents and a stoichiometric amount of fluorine consumed. These operating conditions were used as a basis for estimating costs associated with a commercial–scale INFLUX© facility.

5. Cost Estimations for Reprocessing with INFLUX©

The economics of reprocessing versus direct disposal of spent nuclear fuel has been a highly contentious issue over the last decade. At one extreme, a 2003 report by researchers at *Harvard University's Kennedy School of Government* implied that reprocessing (spent LWR fuel) would be more than double the cost of direct disposal [3]; at the other extreme, a 2006 report by *Boston Consulting Group* (BCG), which was prepared for *Areva*, the owner and operator of the UP2 and UP3 facilities at La Hague, France, suggested a small 6% differential [17]. This difference results from assumptions each group made on the capital and operating costs of reprocessing and the financial models assumed for each business case. The US Congressional Budget Office (CBO) noted this when it reviewed these documents in 2007 [18]. In trying to bring each to a common basis, the CBO simply took numbers that fell between these estimates and concluded that "*the costs of reprocessing may be comparable to direct disposal under limited circumstances, but under a wide variety of assumptions, reprocessing <of LWR fuel> is more expensive...<with current technologies>".*

It would seem reasonable to assume that the most reliable cost estimations are those based on historical costs from actual facilities. However, the actual economics of reprocessing is considered proprietary so even the most recognized studies in the literature are highly speculative. The BCG study, for example, used an annual operating cost for a PUREX reprocessing facility equivalent to 6% of its total capital investment - a "rule of thumb" speculated by the Nuclear Energy Agency. In fact, there is no standard, general correlation between capital and operating costs used in engineering cost estimation. Operating costs vary widely with the application: the chosen technology, the manpower needed, the energy consumed, the reagents required and the environmental management necessary. It is curious that Areva – the licensor and operator of a PUREX facility - did not provide actual operating cost data to BCG for the study they themselves had requested; it indicates the level of propriety on operating costs for such facilities. Published estimates on the "construction costs" of *THORP* (thermal oxide reprocessing plant), the British Nuclear Fuels Limited (BNFL) facility in Sellafield, England, are equally misleading. The $6 billion price tag often reported by the media for this facility includes capital *and* operating costs accumulated over a 10 year period, where up to 15,000 workers were employed in what could only be classified as R&D and decommissioning work at multiple facilities. The actual total *capital* investment for the facility is likely much less.

For the purposes of comparison, UNB Nuclear completed its own process design and cost estimation of a typical PUREX process. However, standard chemical engineering cost estimation methods do not apply well to nuclear chemical processes because of special requirements with the latter for shielding, storage and monitoring. Since PUREX is a relatively simple hydrometallurgical process which uses standard equipment that has been modified for the nuclear industry, UNB Nuclear determined costing factors which, when used with sized equipment costs, produced capital cost numbers that were comparable to published values on the *THORP* facility. This approach is one of the top-down cost estimating methods suggested by the suggested by the NEA's *Economic Modelling Working Group for Generation IV Nuclear Energy Systems* [19]. The costing factors we derived were then used to estimate capital and operating costs for a base case of reprocessing using INFLUX. A capacity of 100 tonnes of heavy metal (HM) per year was used – a small size that would roughly accommodate the annual spent fuel discharged from a single CANDU reactor [20]. We have also assumed the facility would be 50% debt-financed and amortized over 35 years at an attractive 2% cost of capital – a case that could only be achieved if the venture were backed by a government partnership. Results for the base case are presented in Table 2.

Table 2- Capital and Operating Cost Estimation for a 100 tHM/year INFLUX Reprocessing Facility

Costs	Total Cost (millions $)
Direct Costs (purchased and installed equipment, instrumentation, site preparation and ancillary services)	112
Indirect Costs (engineering, construction, start-up)	46
Contingency (30% of Fixed Capital Investment)	48
Working Capital (10% of Total Capital Investment)	23
Total Capital Investment (excluding interim storage)	**229**
Total Annual Operating Cost (including debt repayments)	**9.1**

The operating cost of the facility was estimated from the material and energy requirements calculated by our ASPEN simulation, adding in an estimate on the manpower and overhead needed to operate the facility. On a per tonne basis, capital and operating cost figures for

INFLUX are *much* smaller than those reported by BNFL and Areva, possibly due to the advantages of this technology over PUREX as described above.

6. The Economic Comparison Model

UNB Nuclear feels it is incorrect to include R&D costs in any economic model that compares direct disposal of spent fuel to reprocessing. Neither the BCG study nor the Harvard study explicitly includes R&D costs from decades worth of research on deep geologic storage of spent fuel, yet they indirectly use them for reprocessing when they use press releases on “project costs” for the THORP facility. Indeed, significant R&D work would have to be done on INFLUX before this technology could be considered proven. Years or decades of development work could be necessary. To avoid introducing greater uncertainty into the model, we have assumed the technology is already licensable. Based on the value of the ash being $2800/tonne fob the MOX fuel site (which would be similar to the current value of enriched UO_2), the annual INFLUX reprocessing cost was found to be **$91/kg heavy metal** (kgHM). Interestingly enough, this estimate is an order of magnitude larger than costs often reported for front-end fluorination of uranium oxides to form uranium fluorides ($5-15/kgHM) [7]. Since spent fuel is more radioactive, and given the front-end conversion process includes the consumption of acids and high energy stages of evaporation and calcination, our calculated cost for reprocessing with INFLUX seems reasonable, if not conservative.

The small footprint of the INFLUX process now opens up the possibility for on-site re- (or pre-) processing for the purposes of reducing waste volumes. Two different reprocessing scenarios were compared using the base case:

Case#1 - One centralized reprocessing facility. In this model, utilities would pay to have their waste removed from the site, transported, temporarily stored and then reprocessed. A credit would be issued back to the utility for the ash extracted and the low level waste would be shipped to a repository. This model is similar to the arrangement that Areva currently has with several countries for their La Hague facility. The principal advantage of this model is centralized control of waste management.

The total cost of reprocessing for this case can be represented by:

$$C_{TRC1}\text{ (total annual reprocessing cost)} = m_{HLW}\,(C_{HLW} + C_{IS} + C_{R1}) + m_{LLW}C_{LLW} \qquad (1)$$

Where:

m_{HLW} - spent fuel from the reactor that is to be treated (roughly 19kg UO_2/bundle)
m_{LLW} - mass of low level uranium waste remaining after processing (18.2 kg/bundle assuming a 97% recovery)
C_{HLW} - cost to transport high level waste to the central facility. According to the NWMO [21] this varies per site, but assuming road transport, on average it is approximately $350/bundle (**$19/kgHM**)
C_{IS} - cost of construction and operation of intermediate storage, which would be a necessary addition to a central repository. (**$9/kgHM**) [22] [7]

C_{R1} - the unit cost for INFLUX reprocessing in Case 1(including the ash sale credit): **$68/kgHM** when scaled to accommodate 120000 bundles per year.
C_{LLW} - cost of disposing of low level waste

<u>Case#2 - *Distributed pre-processing*</u>. In this model, spent fuel would be pre-processed at each utility site. The ash product would be scavenged from the spent fuel for sale to a MOX fuel facility and the low level waste would be shipped to a LLW repository. This scheme is similar in principle to the one used by some fossil-fired power plants that remove vanadium from their fly ash. The principal advantage of this scenario is that existing on-site interim storage facilities could be utilized, and utilities could choose to build such a facility based on their own specific plant economics.

An onsite facility eliminates the need for both interim storage and the shipping of high level waste. Equation (1) becomes

$$C_{TRC2}\,\text{(total annual reprocessing cost)} = m_{HLW}\, C_{R2} + m_{LLW} C_{LLW} \qquad (2)$$

Subtracting these two equations provides a $12million/year cost differential in favour of the distributed pre-processing scheme. This savings is mainly due to additional costs associated with providing intermediate storage of the spent fuel. It should be noted that pre-processing on a plant site is not part of a utility's core business so this option might not be attractive to power plant owners; and additional investment would be necessary to secure a facility that would be producing plutonium ash. Notwithstanding, the ash would be in small quantities and would be proliferation resistant due to the presence of highly radioactive fission products.

7. Conclusions

This study has shown that bromofluorination could be a viable option in the search for alternatives to conventional aqueous reprocessing methods. Process simulations matched well to the literature, including with earlier experimental work done on process chemistry and phase separation. The innovative and simple separation scheme makes fluoride volatility a more attractive option for treating spent fuel: the costs of reprocessing are dramatically reduced over PUREX, and the potential for aqueous waste generation is virtually eliminated. A reprocessing cost of $91/kgHM was calculated using a bottom-up approach - from capital and operating cost estimates for a 100tHM/year facility ($68/kgHM for a centralized facility). This number has been shown to be consistent with estimates on conventional front-end conversion facilities that also use fluorination. These numbers are also interestingly similar to the average annual contributions currently being made by all nuclear utility owners in Canada (roughly $70/kgHM per year) to the nuclear waste management trust fund [23]. For these reasons, INFLUX will continue to be a technology of interest for Canada's nuclear industry as we continue to investigate economic and responsible options for managing our nuclear waste.

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