NWMO DISCUSSION PAPER

Reprocessing versus Direct Disposal of Spent CANDU Nuclear Fuel: A Possible Application of Fluoride Volatility

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Summary: Although there is no current perspective for commercial reprocessing of spent CANDU fuel, an application of the fluoride volatility process may offer this possibility. This paper focuses on the economic incentives for reprocessing.

Introduction

The current NWMO management approach is to consider used CANDU fuel as a waste. Although technically quite feasible, reprocessing of spent CANDU fuel on a commercial scale would lead to prohibitive costs even when considering the revenues from the additional electricity produced with recycled fuel. This is also the case for all the recycle options from reprocessing of spent LWR fuel, but to a much lesser extent. In fact, reprocessing LWR spent fuel and recycling the recovered uranium and plutonium in LWR's or future fast reactors (FBR) may become economical when and if the price of natural uranium reaches a breakeven value. A recent economic study at Harvard concluded that once-through LWR fuel cycles are likely to remain significantly cheaper than recycling for at least the next 50 years, even with substantial growth in nuclear power.[1]

For reprocessing to become a credible alternative within the next 50 years in Canada, we would soon need to find a much cheaper process for extracting the plutonium from CANDU fuel, a process that is safeguardable and specially tailored to the low burnup CANDU fuel. Recent interest in the use of fluoride volatility techniques for recycling spent LWR fuel suggests that there may be an alternative to direct disposal of CANDU fuel. It is proposed here that NWMO investigate this possibility.

The Current View on Reprocessing CANDU Fuel

In a previous discussion paper presented to the NWMO Advisory Council, technical aspects of the disposal of spent CANDU fuel were addressed from the perspective of isotopic composition.[2] It was noted that the economic incentive for reprocessing spent fuel from Light Water Reactors (LWR) is far greater than for reprocessing spent CANDU fuel because of the very low fissile content of CANDU spent fuel compared to that of LWR spent fuel. In fact, the U-235 concentration in the uranium in the spent CANDU fuel (approx. 0.23%) is comparable to the tails assay at the enrichment plants (0.2%-0.3%). Thus, it would be foolish to reprocess CANDU fuel in order to recover the uranium, since there are vast amounts of non-radioactive and chemically pure depleted uranium available from past enrichment operations all over the world (probably well over one million tonnes). The only economic incentive to reprocess spent CANDU fuel would then be to recover the plutonium. This is not entirely excluded for special applications such as the

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[1] M. BUNN, J.P. HOLDREN, S. FETTER and B. van der ZWAAN, "*The Economics of Reprocessing versus Direct Disposal of Spent Nuclear Fuel*", Nuclear Technology, Vol. 150, June 2005.

[2] D. ROZON, "*CANDU Spent Fuel : a Waste or a Resource*", NWMO Advisory Council discussion paper, January 2005.

production of fissile feed for future thorium cycles in CANDU.[3] On a large scale however, the high costs of the current commercial reprocessing technology (PUREX) and the difficulties associated with the handling of isolated plutonium lead to very high recycle costs and serious proliferation issues. This all but ensures that the current inventory of spent CANDU fuel bundles will never be reprocessed. CANDU spent fuel should thus be treated as a waste.

This is consistent with NWMO's strategy (APM) leading to the direct disposal of the spent fuel bundles in a suitable deep geologic formation. Appendix 9 of the NWMO Final Study Report contains a detailed discussion of reprocessing, including the above considerations. It is concluded that reprocessing as a management approach for used nuclear fuel is considered to be highly unlikely as a viable scenario for Canada at this time. But it is recognized that reprocessing of used fuel is potentially feasible in the future if there is a continuing nuclear program in Canada. It is also acknowledged that economic conditions could be much different in 50 or 300 years. APM provides sufficient flexibility to deal with this issue by ensuring accessibility to the used fuel for a sufficiently long time so that future generations can make decisions on the case for reprocessing CANDU fuel.

The Harvard Study

At present, approximately 1/3 of the spent LWR fuel has been reprocessed commercially around the world, the remaining spent fuel being stored on reactor sites (including all U.S. plants). The commercial reprocessing facilities use the PUREX aqueous process to separate the uranium and the plutonium from the fission products and minor actinides. Since only a small fraction of the separated plutonium is currently recycled as MOX in LWR reactors, significant plutonium stocks are accumulating worldwide, in wait for the introduction of fourth-generation fast reactors. While advanced fuel cycles and alternative reprocessing technologies are being sought to recycle the plutonium and the recovered uranium from reprocessing, the international focus is naturally on closing the LWR fuel cycle.

There is general agreement that even at today's higher uranium prices, reprocessing and recycling are more expensive than direct disposal of the spent LWR fuel. The debate is over the magnitude of the difference and how long it is likely to persist. The Harvard study looked at this problem from the point of view of an LWR operator with discharged fuel having to decide which option is less expensive: direct disposal or reprocessing.

The two options were compared, including the following cost items:
a) for direct disposal :

- interim storage of the spent fuel
- transport to a repository site
- waste conditioning
- disposal

b) for the reprocessing option :

- transport to a reprocessing plant (after on-site storage)
- reprocessing of the spent fuel
- disposal of the reprocessing wastes (on the same site?)
- recycle of the recovered uranium and plutonium, *reducing requirements for fresh fuel*.

The recycle option therefore includes a <u>credit</u> for the reduction in fresh fuel requirements, which is *a function of the price of uranium*. Assuming that reprocessing costs are higher than direct disposal costs, an increase in the price of uranium will reduce the difference. The price of uranium at

[3] Natural thorium contains mostly Th-232, which is not fissile but when used as fuel can produce *in situ* a fissile isotope of uranium, U-233. In order to start the thorium cycle, you need to add a certain amount of fissile material in the fresh fuel, in this case the Pu-239 recovered from reprocessing CANDU fuel.

which the *net present cost* of the two fuel cycles is equal is called the **breakeven price of uranium**.

Using a conservative price of 1000 $/kg for reprocessing LWR fuel (in 2003 USD), as well as other best estimates for recycle fuel fabrication and HLW disposal costs, a breakeven price of uranium of about **370 $/kg** was found in the Harvard study, i.e. about eight times the 2003 uranium price.

Even at today's higher uranium price, the margin is still quite large. The breakeven price is very sensitive to reprocessing cost assumptions, increasing to over 600 $/kg when reprocessing costs reach 1500 $/kg. Assuming that reprocessing costs will always remain high and considering the known uranium resources, the Harvard study concluded that available uranium resources are likely to be sufficient to sustain once-through LWR fuel cycles for many decades.

Fluoride Volatility

Dry reprocessing methods, as opposed to wet solvent extraction methods like PUREX, were briefly discussed in section 3.2 of the Jackson report on reprocessing.[4] Fluorine volatilization is identified as "*a promising method for the selective extraction of actinides*". We also recall that reacting natural uranium with fluorine to produce uranium hexafluoride (UF_6) is the first step in uranium enrichment. In fact, most of the Canadian uranium exports are shipped in this form.

The use of fluoride volatility techniques has also been considered in the past to reduce the cost of reprocessing. It has been proposed to use fluoride volatility to extract most of the uranium from the spent fuel and the remainder would then be treated with a PUREX process to separate the remaining uranium, plutonium and fission products. Significant savings were expected because of the large volume reduction in the plutonium extraction stage.

An alternative approach was recently proposed that would dramatically simplify the process for the reprocessing of LWR spent fuel.[5] This could be achieved by recycling both product streams from the fluoride volatility process directly into CANDU reactors, with no further purification or separation. In addition to the generation of power from recycling both streams, there are significant advantages with this approach:

- it entirely eliminates the aqueous PUREX process from the flow sheet (major cost component)
- it achieves a *high degree of proliferation resistance* because the plutonium always remains mixed with highly radioactive fission products. Also, the residual plutonium in the discharged fuel after recycling is significantly degraded (a smaller fissile component), with a net reduction in total plutonium inventory.

WIMS calculations were carried out in reference 5 for a CANFLEX fuel bundle in a CANDU lattice using sintered oxide pellets made from the fluoride-volatility ash material. The simulations showed that a discharge burnup of almost 60 MWd/kg could be achieved in a CANDU reactor with this recycled fuel. Such a high burnup seems possible because the ash fuel contains much less uranium (only 40%) and some of the fission product poisons initially contained in the fuel burn off as the fissile plutonium component is depleted. This reduces the rate of decline of fuel reactivity and prolongs the irradiation cycle, leading to a higher discharge burnup.

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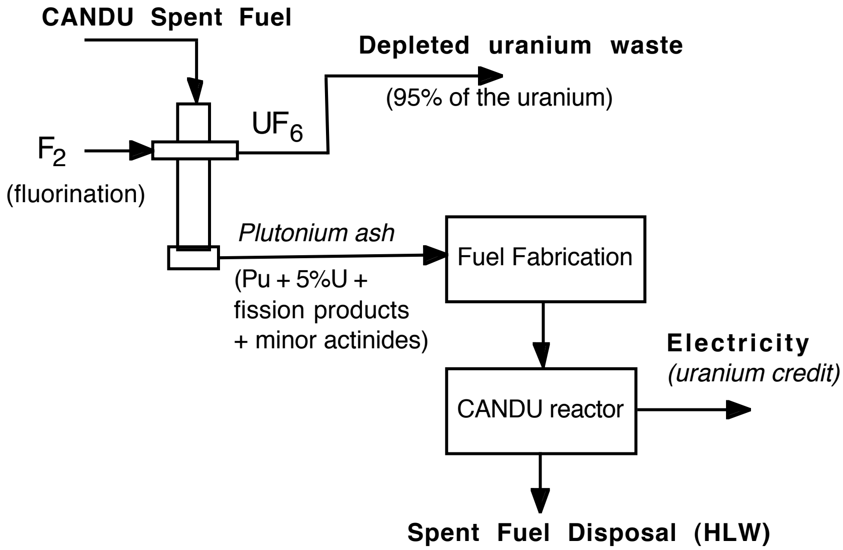
[4] D.P. JACKSON, "*Status of Nuclear Fuel Reprocessing, Partitioning and Transmutation",* Mc Master University (NWMO invited paper 2005)
[5] G.R. DYCK, T. MOCHIDA and T. FUKASAWA, "*Application of Fluoride Volatility to the Recycling of LWR Spent Fuel into CANDU*", Proc. GLOBAL 2005, Tsukuba, Japan, 2005 Oct. 9-13. (paper 493)

Application of Fluoride Volatility (FV) to Spent CANDU Fuel

Our interest here lies with spent CANDU fuel, not spent LWR fuel. Let us then consider conceptually the application of Fluoride Volatility as a reprocessing option for spent CANDU fuel. This is illustrated in the following diagram:



a) <u>Decladding and Immobilization of Radioactive Fission Product Gases</u>

The first step (not shown) is to remove the cladding on the fuel. A promising approach is *oxidative decladding*,[6] which makes use of the fact that when UO_2 is oxidized to U_3O_8, it swells and turns into a powder. The cladding is cut in a manner that allows oxygen to reach the enclosed UO_2 and react at elevated temperature to form the U_3O_8. The process causes the fuel to swell, splitting the cladding open and releasing the resulting powder. According to reference 5, more than 99% of the fuel can be extracted this way, in a granular form appropriate for the fluorination step to follow (as shown above).

The decladding process also releases radioactive fission-product gases which must be immobilized in clay filters for storage and disposal. It is noted that the small volume of HLW generated during decladding are all short-lived fission products that will decay and reduce the activity of these wastes to LLW within approximately 50 years. We also note that specific fission products (like Cesium-137) are significant components of the heat source in the spent fuel. Their removal at this early stage could reduce the activity level in the recycle material and eventually reduce recycle fuel fabrication and disposal costs.

b) <u>Depleted Uranium Wastes</u>

With fluoride volatility, most of the uranium in the spent CANDU fuel would be recovered in the form of uranium hexafluoride. Reference 5 reports an efficiency of 95% for the volatile extraction. It is important to note that the recovered uranium stream contains essentially <u>depleted uranium</u> (DU), contrary to the LWR case for which the recovered uranium contains more U-235 than natural uranium. The recovered uranium may then well be considered as a <u>low level waste</u> (LLW) if it does not contain too many F.P. contaminants. Otherwise, there may be a need for an additional purification step, which would increase the costs of reprocessing and generate a small stream of HLW that can be recycled with the Pu ash. In principle, the disposal of the depleted UF_6 could

[6] O. AMANO, G.R. DYCK, and J. SULLIVAN, "*Option of Dry Process of LWR and FBR Fuel*", Proc. GLOBAL 2005, Tsukuba, Japan, 2005 Oct. 9-13. (paper 521)

lead to a *very significant reduction in waste disposal costs* compared to direct disposal, at least for 94% of the spent fuel volume. Of course, the remaining 6% will eventually generate high level wastes after recycle and the generation of additional electricity, with very significant disposal costs.

c) Plutonium Ash

The plutonium "ash" contains most of the fission products, the residual 5% of uranium and all the other actinides. The composition of the Pu ash can be inferred simply by removing 95% of the uranium from the spent fuel. This is shown in Table 1 using the CANDU used fuel compositions found in reference 2 (average burnup of 7.5 MWd/kg). We note:

- reprocessing **16.5 kg of spent CANDU fuel** with fluoride volatility yields **1 kg of recycle fuel material** containing 82% depleted uranium, 6% plutonium and 12% fission products and minor actinides.
- with a fissile component of nearly 5% (mostly Pu-239), there is a distinct possibility that this recycle material would be sufficiently reactive in a CANDU lattice to yield a **significant discharge burnup**. *Only detailed physics calculations can confirm this assumption*.
- by comparison, LWR ash fuel has a 7% fissile component.and produces a burnup of 60 MWd/kg. Considering the lower initial fissile content and a more rapid reactivity decline, we speculate that an average burnup in the range of **30 to 40 MWd/kg** could be obtained from the CANDU ash fuel.

Table 1 Fuel composition before and after 95% uranium extraction from CANDU spent fuel with fluoride volatility (Pu ash fuel)

Fuel composition (g)	before U extraction	after U extraction	
U-235	2.3	0.12	
U-236	0.7	0.04	82%
U-238	985.8	49.29	
Pu-239	2.5	2.5	
Pu-240	1.0	1.0	6%
Pu-241	0.2	0.2	
Pu-242	0.1	0.1	
F.P. + other actinides	7.4	7.4	12%
total	1000 g	60.6 g	100 %

d) Recycle Fuel Fabrication

The flexibility of the CANDU reactor for fuel cycles based on recycled fuel materials has been demonstrated. For example, numerous studies in Korea and Chalk River have shown the feasibility of the DUPIC cycle, where spent PWR fuel is reduced to a fine powder using a dry process (OREOX) and re-sintered into new CANDU fuel pellets. Experimental DUPIC fuel was actually produced at Chalk River from spent PWR fuel, and irradiated successfully in the NRU reactor. Remote handling processes for fuel fabrication have been evaluated and the performance of the CANDU reactor with DUPIC fuel was studied in detail in Korea. Recycle of the Pu ash from the above fluoride volatility process would certainly benefit from past experience with DUPIC.

Fabricating fuel from the highly radioactive plutonium ash would undoubtedly be a challenge. It could even require a different approach other than sintering (such as vibration packing). A new fuel bundle design may also be required to ensure proper distribution of power, with the use of burnable poisons to control the reactivity and limit power peaking. We should therefore expect that fuel fabrication costs will be significantly higher than for natural uranium fuel. A factor of ten may not be an exaggeration.

Assuming than no significant change would be required to the CANDU reactor design to accept this new fuel, the recycle fuel could actually substitute for fresh fuel in an existing CANDU reactor. In reality, the more likely outcome would be that a new special purpose CANDU reactor would be built, with special fuel handling equipment designed and licensed to handle the radioactive recycle fuel.

e) Disposal of Spent Recycle Fuel

The disposal costs for the spent recycle fuel must finally be included in the total waste disposal cost. We would expect the recycle fuel disposal costs to be *higher* than the reference disposal cost (for natural uranium spent fuel), because of the significantly increased heat load associated with the higher accumulated burnup and the increased actinide concentration in the discharged recycle fuel.

The HLW waste disposal costs would therefore depend on the burnup achieved during recycle. The penalty for the direct disposal of the recycle spent fuel compared to the original spent fuel does not vary linearly with burnup. This penalty can be greatly reduced with an appropriate insertion strategy into the repository. Nevertheless, the penalty would be significant.

Preliminary estimate of the breakeven price of uranium

Breakeven between direct disposal and reprocessing can be expressed simply by the following equation:

$$C_D = C_R + f \cdot C_{LLW} + (1-f)\cdot(C_{rec} + C_{HLW}) - (1-f)\cdot\frac{B_{rec}}{B_{nat}}\cdot(C_{fab} + C_U)$$

Let C_D be the unit cost (per kg) for the direct disposal of CANDU spent fuel. All the other terms are cost items affecting the reprocessing option and are defined below. During its study, NWMO produced cost estimates for various direct disposal options. For the APM approach, which was finally selected by the Government, a total direct cost of 22 B$ was found for an inventory of 3.7 million bundles, excluding interim storage costs at the reactor sites and transportation costs to the repository (see Table 11-3 of the Final Study Report). This cost is spread over 350 y. Using a discount rate of 5.7% to cover the price escalation of materials and labor as well as a reasonable rate of return on the trust funds, an equivalent present worth of 4.6 B$ was found (in $2004).

For a more realistic comparison of the direct disposal option to the reprocessing option, the unit costs in the equation above should all be discounted to the present day, since expenditures would not be occurring at the same time in both scenarios. For example, C_{HLW} is the disposal cost of the recycle spent fuel (per kg). Disposal of the spent recycle fuel could occur much later (years) than the time assumed for direct disposal of the un-reprocessed bundles. This would tend to reduce the HLW disposal costs relative to the direct disposal costs.

For a crude comparison of the options, we will simply use direct costs, neglecting the different time frames of the two options. The following estimates will be used:

C_D At 22 B$ for 3,7 million bundles, each containing initially 19 kg of natural uranium, the cost of direct disposal (C_D) is approximately **300 $/kg**.

C_R A cost of 1500 $/kg is quoted in the NWMO study for reprocessing CANDU fuel with a commercial PUREX process. The cost of extraction of uranium from used CANDU fuel with fluoride volatility should be significantly lower, since it is a dry process and the activity is much lower than that of the LWR spent fuel. We will use for C_R a *target price* of 10-20% of the conventional process, i.e. values ranging between **200 $/kg** and **400 $/kg**.

C_{LLW} An estimate of 19 B$ was also provided in Table 11-3 of the Final Study Report for APM costs without the optional shallow interim storage, a reduction of about 20%. Providing for shallow interim storage is similar to what could be done to dispose of the large volume of low level wastes arising from the FLUOREX process (depleted UF_6). This suggests a value of C_{LLW}=0.2 C_D, i.e. **60 $/kg**. for the disposal costs associated with the depleted uranium.[7]

C_{rec} We will use a highly speculative cost of **600 $/kg** for the remote fabrication of fuel bundles with the Pu ash arising from the fluoride volatility process. This value is consistent with values found in feasibility studies for DUPIC fuel fabrication, and was chosen because of the similarities involved (OREOX process, remote fabrication of highly radioactive plutonium bearing fuel).[8]

C_{HLW} Because of the higher heat load, we will arbitrarily assume that the direct disposal cost for the recycle fuel will be 3 times higher than for natural uranium, C_{HLW}= 3 C_D, i.e. **900 $/kg**. Although C_{rec} and C_{HLW} seem high, these are unit costs and we are reminded that the volumes are much smaller (thus the factor of 1-*f* in the equation).

C_{fab} The credit for the avoided fresh fuel includes the cost of fabrication of fresh (natural uranium) fuel bundles, i.e. approximatly **60 $/kg**.

C_U The most significant component of the uranium credit is the price of uranium itself. The current spot price of uranium (Dec 2007) is around **230 $/kg** (90 US$/lb U_30_8).[9] A reasonable estimate for long-term contract prices would then be **150 $/kg**. We will use this value as a reference.

Technological Factors

f This is the fraction of the spent fuel that ends up as depleted uranium. In the example shown in Table 1, where we assumed that 95% of the uranium is extracted, *f* = **0.94**. (1-*f*) is therefore the fraction of the spent fuel contained in the Pu ash (1-*f* = 6%). With this assumed value, *a total of 16.3 kg of spent CANDU will be required to produce each kg of recycle fuel*.

B_{rec} This is the average discharge burnup that could be achieved in a CANDU reactor with the recycle fuel. Discharge burnup in a CANDU with natural uranium (B_{nat}) is approximately 7.5 MWd/kg. As noted above, we speculate that a burnup in the range of 30-40 MWd/kg could be obtained with this recycle fuel.

These two technological factors have a direct impact on the breakeven price of uranium or on the viability of reprocessing. They are closely interrelated, since a small increase in the UF_6 volatile extraction efficiency will concentrate the fissile component even more and potentially yield a higher burnup when recycled. For *f* = 0.94, we note the following features:

- assuming a burnup of 40 MWd/kg, **1 kg of recycle fuel is equivalent to 5.3 kg of natural uranium fuel** (with a burnup of only 7.5 MWd/kg).

7 We note that the depleted uranium would then be readily available for future generations as a free resource to produce vast amounts of electricity in fast-breeder reactors.
8 H. Choi, W.I. Ko, M.S. Yang, « Economic Analysis on Direct Use of Spent Pressurized Water Reactor Fuel in CANDU Reactors – I : DUPIC Fuel Fabrication Cost», Nuclear Technology, Vol. 134, Number 2, May 2001.
9 www.uxc.com

- a 1100 MWe CANDU reactor operating at 90% capacity factor could consume approximately 26 tonnes of recycle fuel per year, from the reprocessing of 16.3x26 = 424 tonnes of spent CANDU fuel.
- since current CANDU's produce about 150 tonnes of used fuel per GWe-y, one 1100 MWe Actinide Burner CANDU reactor could then recycle fuel originating from 3000 MWe of installed CANDU capacity.
- with 40 MWd/kg burnup, the current inventory of spent CANDU fuel (approx. 2 million bundles) could supply one 1100 MWe reactor for over 100 years.
- recycling would increase the energy produced from the original uranium by about **33%.**

Breakeven Price

An increase in the price of uranium does not affect any of the above unit costs, but it does increase the value of the uranium credit term, thus making the reprocessing option more viable compared to the direct disposal option. Using the above values for unit costs in the equation, we find a **breakeven price of uranium** of **393 $/kg**, i.e. *a value 2.5 times higher than the current price* (~150$/kg).

We note that the breakeven price uranium is very sensitive to the unit cost of reprocessing, as show in the following table:

Cost of Reprocessing C_R ($/kg)	**DIscharge Burnup with Recycle Fuel** B_{rec} (MWd/kg)	**Breakeven Price of Uranium** C_U ($/kg)
200	40	*88*
200	30	*137*
300	**40**	**393**
300	30	544
400	40	699
400	30	951

When the cost of reprocessing is reduced, the breakeven price of uranium is reduced and can reach the current market price. Using a current uranium price of 150 $/kg, we find that **the cost of reprocessing with fluoride volatility would need to be lower than 220 $/kg for the reprocessing option to be viable today**, assuming a burnup of 40 MWd/kg for the recycle fuel.

We note finally that the breakeven price of uranium is not quite as sensitive to the HLW disposal costs. Using a value of C_{HLW}= 2 C_D (instead of a factor of 3) brings the breakeven price of uranium down to 337 $/kg, a reduction of only 15%.

Conclusion and Recommendation to NWMO

Our preliminary examination indicates that fluoride volatility offers some potential for the viability of the reprocessing option for CANDU used fuel. The price of uranium has already increased by a factor of 6 over the past 10 years. Since increasing demand for uranium is likely to sustain higher prices, there is a significant chance that the breakeven price of uranium will be reached long before NWMO needs to proceed with Phase 3 of APM.

Economics is certainly not the only factor influencing decisions on reprocessing. A great number of additional factors would need to be considered, including environmental and strategic issues. These issues will be considered in due time, but the economics and the technical feasibility should be established early.

We recommend that NWMO carry out a more detailed study to identify the potential costs of the fluoride volatility process in the Canadian context. In particular, it would be essential to confirm that appropriate values for the uranium extraction efficiency (f) can be achieved with the FV process. WIMS calculations are needed to estimate the discharge burnup of the recycle fuel (B_{rec}) in a CANDU lattice. These lattice calculations could also suggest additional volatility extraction steps to remove specific fission products from the plutonium ash to extend the burnup.

Although reprocessing of used fuel in Canada is not considered economical at the present time, it may become an interesting alternative to direct disposal in the future and NWMO should be prepared for this eventuality. However, ***speculation on the future reprocessing of CANDU spent fuel should not interfere with the site selection process for deep geological disposal.***

Indeed, whatever the outcome of the NWMO studies on fluoride volatility, the used fuel currently residing at reactor sites will need to be transported to a centralized location. The chosen site should be able to accept all the spent fuel, with a deep geological formation suitable for the confinement of all the radionuclides contained in the used fuel. Reprocessing of the used fuel and disposal of the HLW on the site will not increase the burden of the repository. It may in fact reduce it, if an actinide burner reactor is also build on the site.

The safety case for the future repository should therefore consider both direct disposal of spent fuel and disposal of HLW from reprocessing. Reprocessing should not pose a social acceptability problem inasmuch as all the HLW originate from used fuel taken from Canadian reactors. With current attitudes, it is doubtful that people would accept taking in spent fuel or HLW from other countries.