SOME IMPLICATIONS OF RECYCLING USED CANDU FUEL IN FAST REACTORS

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Abstract

This paper presents the potential implications of adopting an advanced nuclear fuel cycle where used CANDU fuel is reprocessed and supplied to fast reactors designed to burn actinides. The analysis assumes that used CANDU fuel is reprocessed to recover uranium, plutonium and other actinides, which are then used to fabricate fresh fuel for the fast reactors. Once in operation, the used fast reactor fuel is reprocessed and recycled together with makeup from used CANDU fuel to produce energy. Pyroprocessing is assumed for reprocessing of CANDU and fast reactor used fuels.

Deployment of fast reactors in the nuclear energy system is assumed as a method for waste management and for electricity production. With respect to waste management, mass flow calculations estimate that the reduction in mass of transuranics for disposal would be accompanied by a larger increase in the mass of fission products. The time required to consume most of the transuranics in the used CANDU fuel is also estimated.

Understanding the long-term hazard of the wastes from an advanced fuel cycle is important to assess options for their long-term management. Estimates of the radioactivity, radiotoxicity, thermal power and unshielded dose from a reprocessing/fast reactor wasteform and from used CANDU fuel are presented.

The long-term safety of the fast reactor waste is also addressed. Two options are considered, placement in a deep geological repository, and placement after 300 years decay in a near surface landfill. The analysis estimates that the dose consequences as a result of surface disposal of reprocessing wastes could exceed regulatory limits over long periods of time. That is, even after a few hundred years of decay, the fast reactor wasteform is sufficiently radioactive that it would require appropriate long-term management, such as in a deep geological repository.

1. Introduction

Research and development studies of advanced nuclear fuel cycles are being pursued by a number of countries through national and international collaborative projects to examine the waste management, resource use, economics and proliferation resistance of different types of fuel cycles using thermal reactors in combination with fast spectrum reactors or accelerator-driven systems. Typically these studies focus on recycling used fuel from the light water reactors. Recently, some assessments on recycling CANDU used fuel in fast reactors have been published [1, 2]. This paper presents a high level analysis of some of the implications of adopting an advanced fuel cycle where used CANDU fuel is reprocessed and supplied to fast reactors designed to burn actinides [3, 4].

2. Nuclear fuel cycle

A closed nuclear fuel cycle is considered using fast reactors designed to burn actinides. These reactors require a continuous external source of fissile material in addition to uranium, and the external fuel could include transuranic (TRU) elements from the used fuel of a thermal reactor. Fast reactors operating in this mode have been proposed for waste management purposes. This analysis assumes that used CANDU fuel is reprocessed to recover the uranium, plutonium and minor actinides such as neptunium, americium, and curium, which are then used to fabricate the fresh fuel required for starting and operating a fleet of burner fast reactors (see Figure 1). Once in operation, the used fast reactor fuel is reprocessed and recycled continuously together with make-up from used CANDU fuel to produce energy.

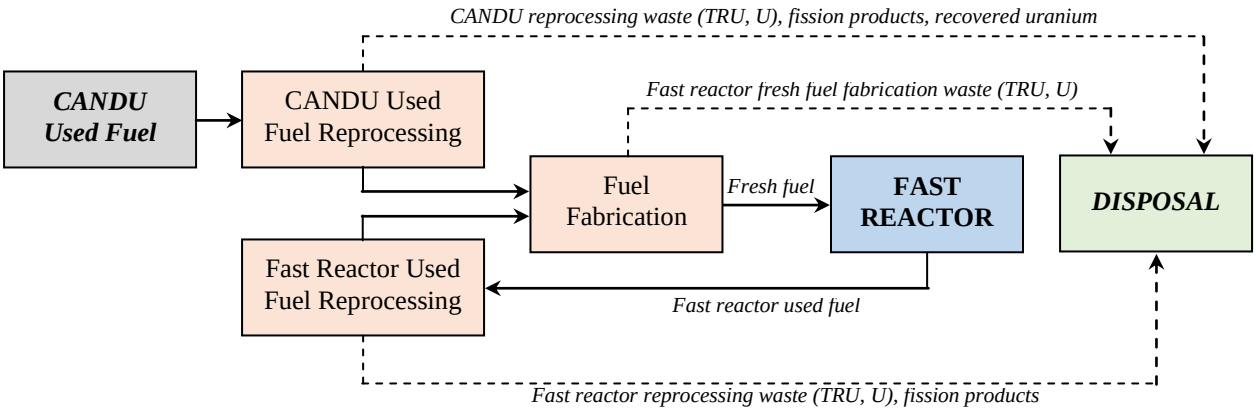

Figure 1 Nuclear fuel cycle considered.

2.1 Reprocessing and fuel fabrication

Reprocessing is a key process in a closed nuclear fuel cycle. Various technologies are presently being considered, such as aqueous, pyro, and fluoride volatility processes. They are at various stages of development, ranging from conceptual phase to industrial scale operation. The technology considered in this analysis for reprocessing both CANDU and fast reactor used fuel is pyrometallurgy reprocessing ("pyroprocessing"). This technology has been implemented on a small scale for the Experimental Breeder Reactor-II fast reactor spent fuel [5]. Laboratory scale tests of reprocessing used light water reactor fuel, an enriched-uranium oxide fuel, have been completed [6].

The present analysis assumes that pyroprocessing can be successfully adapted to reprocess CANDU as well as fast reactor used fuel on a commercial scale. The pyroprocessing rate is assumed to be sufficient to meet the fuelling requirements of the fast reactors.

A range of recovery efficiencies have been reported or estimated for pyroprocessing. For this study, during reprocessing a 99.5 wt% efficiency is assumed for recovery of TRU elements, 100 wt% for the separation of fission products, and 99.0 wt% for the recovery of uranium [3]. A further 99.9 wt% overall fabrication efficiency is assumed for the TRU elements and uranium during fuel fabrication [3].

2.2 Fast reactor

The fast reactor considered in this analysis is based on the preliminary design of advanced burner reactor developed at the Argonne National Laboratory (ANL), which is based on the 380 MWe SuperPRISM (S-PRISM) reactor designed by GE Hitachi Nuclear Energy, with a 38% thermal efficiency [7].

The reactor is assumed to operate with different core configurations, each with the same power output but with different conversion ratios[1] (CRs) less than one. From a waste management perspective, a fast reactor with a very low conversion ratio, for example as low as 0.25, would be advantageous in a nuclear fuel cycle as it would require a larger amount of external fuel, therefore maximizing the burnup of TRUs from the wastes. A low conversion ratio however would require a high TRU enrichment, beyond current irradiation experience with fast reactor fuels. Based on current technology, the conversion ratio would be more likely in the range of 0.5-0.6 [9]. This paper assumes a range of 0.25-0.75 to estimate the overall system inventory, and further discusses the nature of hazard and long-term safety of the fast reactor waste for a system employing fast reactors with a favourable (for actinide burning) very low conversion ratio of 0.25.

The reactor is assumed to operate with a capacity factor of 85% for all core configurations. The capacity factor would ultimately depend on the conversion ratio as the refuelling frequency, number of assemblies replaced per outage, and unplanned outages related to fuel failures, will differ from one core configuration to another.

3. Mass flow assessment

Mass flow calculations are performed for two scenarios considering the deployment of fast reactors only (i.e., no further CANDUs) in the nuclear energy system as a method for waste management and for electricity production. One scenario considers operation of a block of two S-PRISM type fast reactors (0.76 GWe) as a deliberate method for long-term waste management of all of the used CANDU fuel in Canada's reactors. This generates some electricity in parallel, but that is not the primary intent. The second scenario considers the current CANDU nuclear fleet would be replaced with fast reactors producing the same amount of electricity. In this case, 36 S-PRISM type fast reactors would be required to operate producing about 13.7 GWe annually. This is an energy supply scenario; however the analysis in this paper is with respect to the implications of this scenario for waste management. All reactors are assumed to start operation simultaneously in year 1 and operate continuously for 60 years. The current analysis provides an overall mass balance perspective, however, it does not comment on the practicality of these fast reactor related technologies, the deployment of fast reactors, or the specific isotopic and reactor physics implications of these fuel cycles.

In all cases, it is assumed that the used CANDU fuel is available to be reprocessed into TRU and uranium (U) streams to support operation of the fast reactors. The amount of used CANDU fuel

[1] "Conversion (breeding) ratio" is defined as the number of fissionable atoms produced to the number of fissionable atoms consumed in a reactor. If the ratio is less than 1, it is referred to as "conversion ratio". If it is greater or equal to 1, it is referred to as "breeding ratio". [8]

considered to be available is 103,000 tonnes of (initial) heavy metal. This is the estimate for a high scenario of projected nuclear fuel waste from the existing reactors, based on approximately 5.2 million bundles, assuming that most of the current reactors are refurbished [10].

The fuel characteristics are derived from reference data or estimates from literature. The used CANDU fuel composition is based on the radionuclide inventory for actinides and fission products in CANDU fuel at a reference burnup of 220 MWh/kgU [11]. The fissile plutonium content (Pu-239 and Pu-241) is about 66 wt% of the total TRU content in the used CANDU fuel, where the TRUs include plutonium, neptunium, americium, and curium. The compositions of the fresh and used fast reactor fuel are estimated from the ANL data [7]. The present mass flow assessment focuses on the TRU content as an aggregate, and does not specifically consider the isotopic content. In practice, a certain amount of more fissile isotopes would be required in the core. The fissile plutonium content derived for the fast reactor fresh fuel is between about 39 wt% (CR=0.25) to 54 wt% (for CR=0.75) of the total TRU content [7]. Since the fissile plutonium content in TRU from used CANDU fuel is higher than this, it is assumed that the CANDU TRU would be suitable for direct use in these fast reactors. However, it may be that some adjustment of the fuel may be needed, in order to make the reactor physics work.

The total amounts of TRUs and fission products that would constitute high level waste from the advanced nuclear fuel cycle are estimated for both scenarios, i.e., the waste management scenario with two fast reactors, and energy supply scenario with 36 fast reactors, for different conversion ratios.

An illustration of the results obtained assuming the favourable very low conversion ratio of 0.25, starting from 103,000 tonnes of used CANDU fuel, is presented in Figure 2. With one power block of two S-PRISM type fast reactors (0.76 GWe) in operation, it is noted that the yearly consumption of the TRUs is very small (a similar trend was observed for higher conversion ratios of 0.5 or 0.75). Extending the analysis from Figure 2 for two fast reactors, it is estimated that it would take almost 1,000 years to consume the entire amount of TRUs in 103,000 tonnes of used CANDU fuel. However, as the operating life of a fast reactor is assumed to be 60 years, this means that approximately 15 generations of 2 fast reactors (or 30 fast reactors) would be needed to burn the entire amount of TRUs in the used CANDU fuel. During this period, there would also be continuous production of fission products.

If the current nuclear fleet is replaced with fast reactors, it would require 36 S-PRISM type fast reactors to produce 13.7 GWe annually. If all these reactors were assumed to start operation simultaneously at year 1 with a favourably low conversion ratio of 0.25, it would take about 50 years to burn the TRUs in the 103,000 tonnes of used CANDU fuel. Although there would be insufficient TRU amounts in the remaining unreprocessed used CANDU fuel to continue operation of all the fast reactors after slightly over 40 years, as noted in Figure 2, there would still be significant TRU amounts in the fast reactor cores. This could be consumed by continued longer operation of one or two fast reactors. For the same scenario (36 fast reactors) and considering more practical conversion ratios (0.5 or 0.75), the CANDU TRU inventory would not be consumed within the assumed 60 years life of these fast reactors.

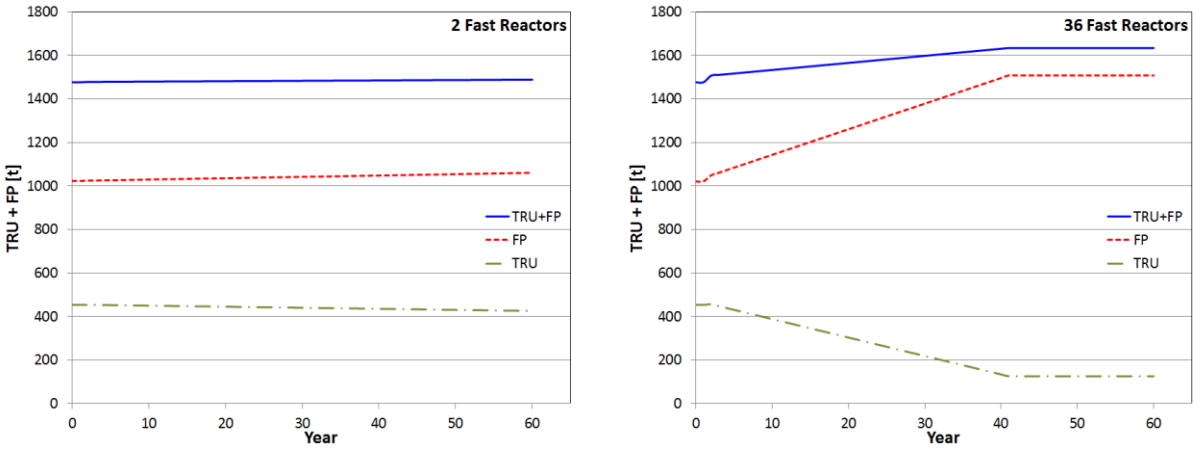

Figure 2 Total TRU and fission products (FP) system inventory for scenario with fast reactors with a very low conversion ratio (CR=0.25).

For both scenarios and for all conversion ratios, the total amount of TRU plus fission products that would have to be managed as high level waste in Canada increases with time, reflecting the steady production of fission products resulted from the fission of TRUs and uranium at a rate that is faster than the TRU consumption. The nature of the high level waste will change over time as the TRU fraction decreases, and becomes more fission product based. The preliminary implications of this on long-term safety and heat generation are presented in the following sections of this paper.

With respect to uranium, the results indicate that there is little reduction in the total amount of uranium in the system inventory that would need to be managed, for any of the conversion ratios considered. A large amount of uranium would be recovered during the reprocessing of the used CANDU fuel, which could be either stored for future re-use as make-up in the fast reactor or sent for disposal.

4. System inventory and waste form

The total amounts of uranium (U), TRUs, and fission products (FP) in the used CANDU fuel available in the system initially, at startup of the fast reactors, as well as their remaining quantities after operating the fast reactors for a number of years, are provided in Table 1. The results are presented for the scenario with 36 fast reactors and 103,000 tonnes of used CANDU fuel, for all conversion ratios.

Table 1 shows that reprocessing of the used CANDU fuel and the fast reactor fuel will generate radioactive waste products. Assuming the used fuels are pyroprocessed as described in Section 2.1, waste salt from the electrorefining process will contain significant quantities of radionuclides for disposal. A ceramic waste process [5], in which the waste salts are blended with zeolite and borosilicate glass and then consolidated into glass-bonded zeolite ingots, produces a stable wasteform.

Table 1 System inventory assuming 36 fast reactors and 103,000 tonnes of used CANDU fuel

		Initial	Final		
			CR=0.25 (41 yrs) (1)	**CR=0.5** (60 yrs) (2)	**CR=0.75** (60 yrs) (2)
Unreprocessed used CANDU fuel [tonnes]	U	101,524	714.4	6,786.8	47,990.5
	TRU	454	3.2	30.4	214.6
	FP	1,022	7.2	68.3	483.3
Fast reactors core(s) [tonnes]	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 [tonnes]	U (3)	0	1,018.5	980.4	587.3
	TRU (4)	0	8.7	10.7	9.1
	FP	0	1,485.8	1,655.4	1,245
• Recovered uranium [tonnes]	U (5)	0	99,540.2	93128	51,986.6
Total U [tonnes]		101,524	101,366	101,112	100,929
Total TRU [tonnes]		454	125	146	323
Total FP [tonnes]		1,022	1,509	1,742	1,748

Notes:

1) At the end of year 41, there will be insufficient TRU in the unprocessed used CANDU fuel to continue operation of 36 fast reactors with CR=0.25.
2) It is assumed that the operating lifetime of the fast reactors is 60 years.
3) Uranium losses from reprocessing of CANDU and fast reactor used fuel, plus losses from fuel fabrication.
4) TRU losses from reprocessing of CANDU and fast reactor used fuel, plus losses from fuel fabrication.
5) Uranium recovered during reprocessing of used CANDU fuel which could be stored for future use or sent for disposal.

5. Nature of the hazard

This section compares the potential hazard posed by wasteforms resulting from a once-through CANDU fuel cycle (i.e., used CANDU fuel bundles) with wasteforms produced as a result of adopting a fast reactor based fuel cycle (i.e. glass-bonded zeolite wasteforms). For a conversion ratio of 0.25, roughly 1,486 tonnes of fission products, 1,019 tonnes of uranium and 8.7 tonnes of TRUs will be sent for disposal. Based on trial fabrication reported in the literature, the fast reactor wasteforms are assumed to have a mass of 400 kg [5] and contain 8 wt% waste products [12], resulting in 78,553 waste packages. This would mean a total of 31,421 tonnes of waste, about one third of the initial 103,000 tonnes of used CANDU fuel. Assuming the fission products, uranium and TRU wastes are evenly distributed amongst all the waste packages means each package will contain roughly 18.9 kg of fission products, 13 kg of U and 0.11 kg of TRU.

Used CANDU fuel and fast reactor fuel will also contain small amounts of light element activation products from impurities in the fuel [11], most notably C-14. Assuming the total inventory of radioactive light element activation products from reprocessed CANDU fuel and fast reactor fuel is evenly distributed amongst the waste packages results in each waste package containing approximately 0.0096 kg of light element activation products.

To estimate the potential hazard posed by the two wasteforms, one has to consider the radionuclide makeup of the fission products, uranium, TRUs and light element activation products. For the CANDU fuel bundles these data are taken from Reference [11], assuming a burnup of 220 MWh/kgU. For the fast reactor wasteform, the radionuclide makeup is assumed to be the same as used CANDU fuel with a burnup of 220 MWh/kgU [11]. That is, the amount

of a radionuclide per kg of fission products would be the same as in CANDU fuel. This is a reasonable assumption because roughly two thirds of fission products and nearly all the uranium, which make up the bulk of the radionuclides in the waste stream, are from the reprocessing of the used CANDU fuel. The radionuclide profile in fast reactor used fuel will be different because of the different initial fuel composition, different neutron spectrum, and the higher fuel burnup. However, a relevant fission product inventory data for the fast reactor used fuel for this scenario is not currently available.

It should be noted that the 99,540 tonnes of uranium recovered from reprocessing as listed in Table 1 are not included in the fast reactor wasteform described above. The potential hazard of this surplus uranium is compared to that of the fast reactor wasteform and used CANDU fuel below.

Figure 3 compares the total radioactivity (in Bq) of the entire inventory of the base case of 103,000 tonnes of used CANDU fuel bundles with that resulting from reprocessing and re-use of this CANDU fuel to produce more electricity – about 31,000 tonnes of fast reactor wasteforms and 99,540 tonnes of surplus uranium. The radioactivity of the fast reactor wasteform is dominated by fission products for all times after discharge while the radioactivity of the CANDU fuel bundles is initially controlled by fission products but beyond a few hundred years the total radioactivity is controlled by the actinides. Initially the radioactivity of the fast reactor wasteforms is higher than the CANDU fuel bundles due to the larger inventory of shorter lived fission products.

Figure 4 compares the radiotoxicity (in Sv) of the total inventory of used CANDU fuel, fast reactor wasteforms and surplus uranium. The radiotoxicity is calculated by multiplying the radioactivity of each radionuclide by that radionuclide's corresponding ingestion dose coefficient [13]. Figure 5 shows the thermal power (in W) for the total inventory of used CANDU fuel, fast reactor wasteforms and surplus uranium. Both the radiotoxicity and thermal power of the fast reactor wasteform and CANDU fuel follow similar trends. For times less than a few hundred years fission products are dominant, after which the actinides control the radiotoxicity and thermal power. Due to the larger inventory of actinides, the used CANDU fuel has a higher radiotoxicity and thermal power than the fast reactor wasteform beyond a few hundred years. Figure 4 and Figure 5 also show that the long-term management of the surplus uranium is a factor that needs to be considered in evaluation of future scenarios.

Figure 6 presents the calculated annual dose rate per kg of wasteform to an unshielded person at 10 m distance from a fast reactor wasteform and a CANDU used fuel bundle. Initially the dose rate (per kg) from the fast reactor wasteform is about six times higher than the CANDU fuel bundle; however, after 1,000 years decay, the fast reactor wasteform is comparable to a CANDU fuel bundle. Beyond 100,000 years, the dose rate for the fast reactor wasteform remains dominated by fission products but starts to decrease as the dominant species (Sn-126) decays. The CANDU used fuel dose rate remains relatively flat at long times due to the ingrowth of uranium decay chain daughters.

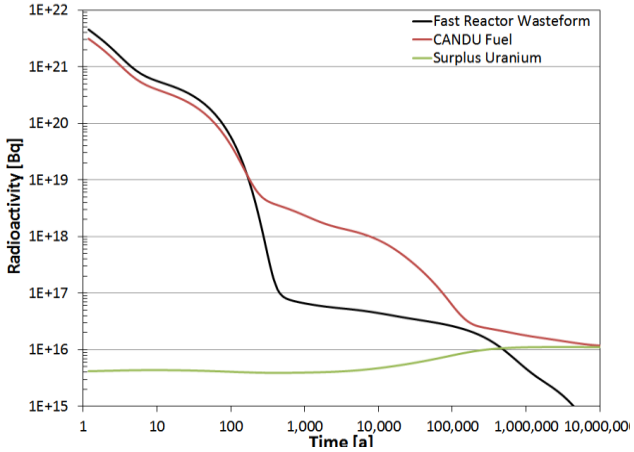

Figure 3 Wasteform total radioactivity.

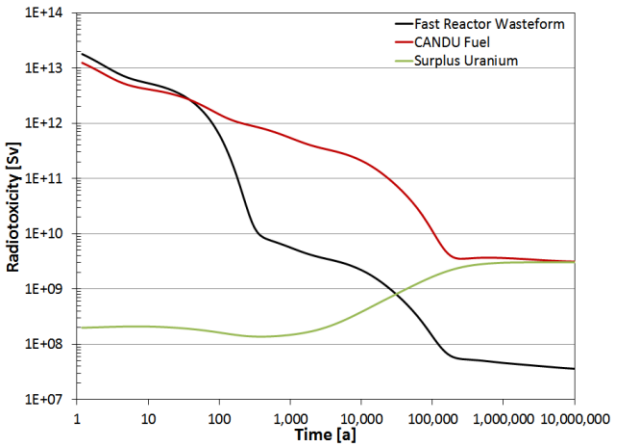

Figure 4 Wasteform total radiotoxicity.

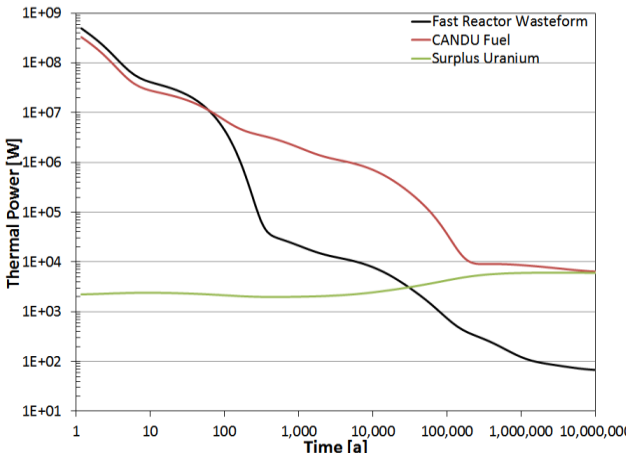

Figure 5 Wasteform total thermal power.

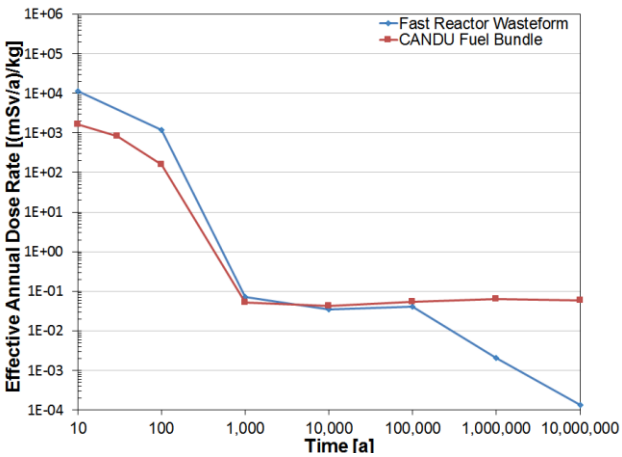

Figure 6 Unshielded dose rate at 10 m.

6. Long-term management of reprocessing waste

Section 5 shows that the fast reactor wasteform from reprocessing and the CANDU fuel are broadly similar especially initially in terms of radioactivity, radiotoxicity, thermal power and unshielded dose rate. After several hundred years the fast reactor wasteform is less hazardous than the comparable amount of CANDU used fuel. Two potential long term management options for disposal of the fast reactor wasteform are discussed below, that is: disposal in a deep geological repository and disposal in a near surface landfill site.

6.1 Deep geological repository

The purpose of a deep geological repository is to safely isolate used nuclear fuel or nuclear wastes from the surface environment through a number of passive barriers. In the case of used CANDU fuel, these barriers consist of the used fuel bundle, a steel and copper container, bentonite clay, and hundreds of meters of low permeability rock. It is intended that these barriers will remain intact essentially indefinitely. Postclosure safety assessments [14, 15] show such a facility could meet regulatory requirements for the protection of people and the environment. In these assessments, the significant dose contributors are either fission products or light element

activation products. The most influential radionuclide is I-129, a mobile (soluble and non-sorbing), long-lived fission product. Despite uranium and TRUs having long half-lives and making up the bulk of the used CANDU fuel, these species do not contribute significantly to the total dose due to the insoluble and immobile (insoluble and high sorbing) nature of these species.

If a parallel is drawn between a repository containing used CANDU fuel and one containing fast reactor waste from reprocessing, one would expect higher dose consequences from the fast reactor wasteforms (in a similar container and geological setting) given that it would have a higher fission product inventory. Obviously, the dose consequences would depend on a number of factors including the degradation rate of the wasteform. However, it is likely that a deep geological repository could be designed to safely store the glass-zeolite wasteforms from reprocessing.

6.2 Surface disposal

The radioactivity and radiotoxicity of the fast reactor wasteforms drop significantly over the first 300 years after discharge. It is therefore sometimes implied or inferred that beyond 300 years, this material need no longer be considered (long-lived) nuclear waste. If that were correct, then by implication the post-300 year material could be disposed in a near surface landfill facility. In reality a number of nuclear regulations would apply to the fast reactor wasteform given the residual level of radioactivity. However, in this assessment these requirements are not considered and the potential implications of near surface disposal are evaluated.

After the 300 year cooling period, the entire inventory of fast reactor wasteforms are assumed to be placed in a modern near-surface (but non-nuclear) landfill. Over time, as the wasteform degrades, radionuclides can migrate via advection and diffusion through the gravel, high density polyethylene liners, clay, and attenuation layers assumed to be present in the landfill. Eventually some radionuclides enter an aquifer below the landfill, and a fraction of these are captured by a well assumed to intercept the aquifer 100 m from the landfill site. The only dose pathway considered in this assessment is from drinking water from the well near the landfill.

Figure 7 shows the ingestion dose results for three assumed wasteform degradation modes: (1) leaching only; (2) a short leaching phase combined with slow congruent degradation of the wasteform, and (3) a short leaching phase with slow congruent dissolution and solubility limited release of radionuclides. Figure 8 shows the ingestion dose results for other sensitivity cases.

The results in Figure 7 and Figure 8 show that the dose consequences are quite dependent on some uncertain model parameters and assumptions, but within these uncertainties the dose consequences from disposal of reprocessing waste in a near surface facility after 300 years could exceed regulatory limits. In cases considering solubility limited release from the wasteform, the doses are dominated by fission products and light element activation products (I-129, C-14 and Cl-36). In other cases the dose rate is controlled by the uranium daughters (notably Ra-226).

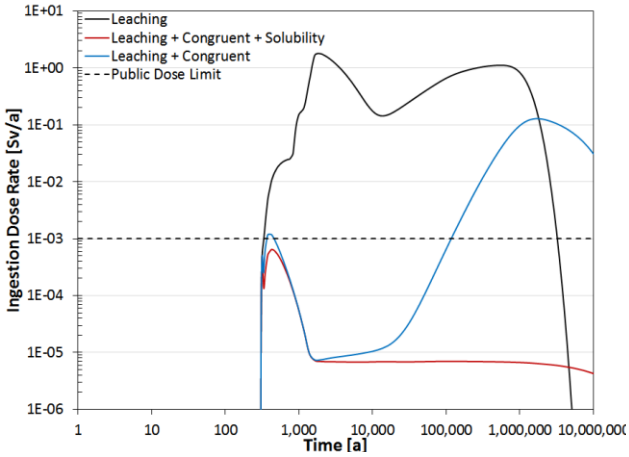

Figure 7 Effect of wasteform degradation.

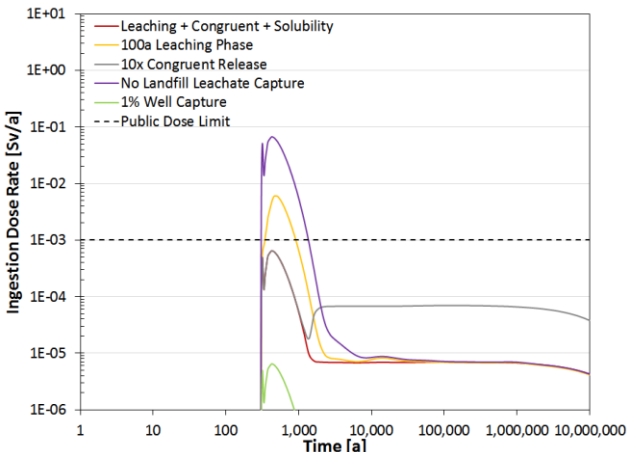

Figure 8 Effect of model assumptions.

7. Discussion

This paper documents a high level analysis of an advanced nuclear fuel cycle where the TRUs from the used CANDU fuel are assumed to be burned in a S-PRISM type fast reactor. Mass flow calculations and a preliminary hazard assessment have been performed to estimate the impact of such a nuclear fuel cycle from a waste management perspective. Scenarios considered the deployment of fast reactors only (i.e., no further CANDUs) as a method for waste management and for electricity production, with all reactors assumed to start operation simultaneously.

With one power block of two S-PRISM type fast reactors (0.76 GWe) in operation, the yearly consumption of the TRUs is very small, regardless of whether the fast reactors are operating with a conversion ratio of 0.25, 0.5 or 0.75. For two fast reactors operating with a favourable very low conversion ratio of 0.25, it was estimated that it would take almost 1,000 years to consume the entire amount of TRUs in 103,000 tonnes of used CANDU fuel. However, as the operating life of a fast reactor was assumed to be 60 years, this means that about 15 generations of 2 fast reactors (or 30 fast reactors) would be needed to burn the entire amount of TRUs in the used CANDU fuel. During this period, there would also be continuous production of fission products.

If the current nuclear fleet is replaced with fast reactors, 36 S-PRISM type fast reactors would be required to produce 13.7 GWe annually. If all these 36 reactors are assumed to start operation simultaneously at year 1 with a favourably low conversion ratio of 0.25, it would take about 50 years to burn the TRUs in the 103,000 tonnes of used CANDU fuel. For the same scenario (36 fast reactors) and considering more practical conversion ratios such as 0.5 or 0.75, the CANDU TRU inventory would not be consumed within the assumed 60 year lifetime of these reactors.

In all cases, it was observed that the total amount of TRUs plus fission products from the advanced nuclear fuel cycle increases with time, reflecting the steady production of fission products resulted from the fission of TRUs and uranium at a rate that is faster than the TRU consumption. That is, neither of these fast reactor scenarios results in a net loss of the hazardous components of used fuel. For clarity, as more power has been generated from the recycled used CANDU fuel, there is less total waste per amount of power produced, but more total waste.

The nature of the resulting high level waste would change over time as the TRU fraction decreases, and becomes more fission product based. Therefore the hazard associated with waste produced by reprocessing used CANDU fuel and burning this in fast reactors was also estimated. The results of this assessment indicate that the reprocessing and fast reactor wastes from 103,000 tonnes of used CANDU fuel could be stabilized in about 31,000 tonnes of a glass-zeolite fast reactor wasteform (and about 99,000 tonnes of uranium). These wasteforms could have a level of radioactivity, radiotoxicity, thermal power and unshielded dose rate broadly similar to that of a CANDU fuel bundle - higher initially, lower after about 300 years with the differences quantified in this paper.

A simplified postclosure assessment assumed disposal in a modern surface landfill of the fast reactor wasteform after a 300 year decay period. The results show that the dose to a person drinking water from a well that intercepts an aquifer near the landfill could exceed public dose limits. It is recognized that this analysis has made several simplifying assumptions; however, it is likely that wastes from the reprocessing of CANDU and fast reactor fuel would still be sufficiently radioactive to be considered as long-lived nuclear waste. Such wastes would remain hazardous for very long times, would need to be carefully managed and would ultimately need to be disposed of in a safe manner similar to the existing inventories of used CANDU fuel.

8. References

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