Synergy Between CANDU® and Fast-Neutron Reactor Technologies

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ABSTRACT

Natural Resources Canada estimates that economical conventional Canadian sources of uranium will be used up in about 40 years at the current rate of Canadian use and of exports. However, if the CANDU® technology is combined with nuclear technology using fast neutrons, then the currently stored spent CANDU® fuel used as replenishment of the fuel needs of fast-neutron reactors can at the same time furnish fissile fuel for our fleet of CANDU® reactors as well as for potential future CANDU®s concentrating on the thorium cycle. This combination of technologies would supply Canadian nuclear fuel needs for many centuries with a fuel source stable both in availability and price. Nevertheless, with the long lead-times currently envisioned to bring any nuclear technology on line, it is imperative that this combined path be examined and initiated before a lack of an economical fuel supply becomes the driver of cost considerations. This paper is intended to foster such an examination.

1. INTRODUCTION

The CANDU® reactor moderated with heavy water is arguably the most efficient among thermal reactors in its use of uranium fuel. Through the use of heavy water its neutron economy is such that, in comparison to light-water reactors, that require fuel enriched in fissile uranium-235 (U235), CANDU® reactors can produce power from natural uranium with a fissile U235 content of only 0.72%.

As a result of the high neutron efficiency in CANDU® reactors there are two natural consequences for spent or used CANDU® fuel that in some eyes might be considered drawbacks. First, the fissile content left in the spent CANDU® fuel waste is low (~0.23% U235 and ~0.27% Pu239/Pu241), too low to be considered economically extractable for further use in thermal reactors [1]. In contrast, LWR spent fuel with ~1.4% U235/Pu239/Pu241 could be reused directly in CANDU® reactors or have its plutonium extracted for reuse, as is done by AREVA in France. Second, the volume of spent CANDU® fuel waste per GW-hour of energy produced is about 7 to 10 times larger than that of spent LWR fuel waste. This latter property paradoxically is a result of the enrichment of LWR fuel to 3.5% or 5% U235 with the simultaneous creation of 7 to 10 times the volume of depleted uranium which is not considered to be nuclear "waste".

These factors have combined to leave Canada, primarily Ontario, with about 47,000 tons of spent CANDU® fuel waste stored predominately at the reactor sites where the spent fuel was produced. The amount increases by about 1,400 tons annually. This is not a large volume, fitting into the proverbial six hockey rinks from the ice surface to the top of the boards [2, p.20].

[1] CANDU® is a registered trademark of Atomic Energy of Canada Limited

However, the concern is the long-term radiotoxicity of the spent fuel. While much of the spent fuel radioactivity, primarily from the fission products, decays relatively quickly, heavy atom constituents such as plutonium, americium, curium, etc., the transuranic actinides (TRUs) transmuted from uranium in the reactor, remain 1000 times more radiotoxic than their natural uranium source for well beyond 10,000 years (Figure 1), reaching background levels equivalent to uranium only about 400,000 years after leaving the reactor core.

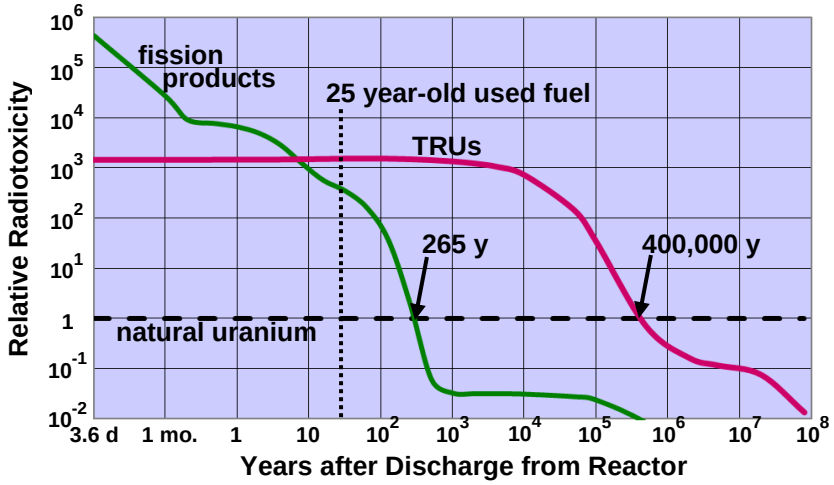

Figure 1. Evolution of Radiotoxicity from Used CANDU® Fuel Components Relative to Natural Uranium.

Elimination of the transuranics (TRUs) in fast-neutron reactors and extraction of uranium (dashed horizontal line) would result in a huge reduction in radiotoxicity of used fuel waste, about 42,000X per unit time at 1000 years, and also shortens the time of decay to background levels from 400,000 years to 265 years. After 265 years the radiotoxicity of the fission products is lower than that of the mined natural uranium from which they are created in the reactor. The suggested 25-year point for processing of used fuel is indicated by the dotted line (see text). Note the log scales of both axes.

These heavy atoms emit primarily highly energetic alpha particles (helium nuclei). While these emissions are easily shielded, ingestion or inhalation of the TRUs constitutes a serious health hazard. Consequently, virtually all nations with nuclear power reactors are considering interring this nuclear waste in as yet unproven deep geological repositories (DGRs), hopefully safely so for several hundred thousand years.

In Canada the estimated life cycle cost of such a repository is $24 to $40 billion depending on the capacity of the facility [1, p.163; 3]. This cost amounts to roughly 0.5 cents per kWh of electricity produced from the fuel bundles, with the cost borne by the consumer (Table 1). Currently only about 0.15 cents per kWh are being levied, and only $2.9 billion of the required $24 billion has been accumulated from the electricity of 2.5 million fuel bundles. This number is already more than half of the capacity of the DGR of smaller capacity at 3.6 million fuel bundles. The $24 billion cost could now only be met by an average charge of about 1.34 cents/kWh on the

electricity of the remaining 1.4 million fuel bundles, mitigated somewhat by the current rather low interest on the accumulating funds.

Table 1
Future Average Rate Required to Meet DGR Costs

(rate for 2014 = 0.151 ¢/kWh)[2]

Planned DGR Capacity (bundles)*	Total Life Cycle Cost	Outstanding Balance**	Going forward Avg. rate needed*** ¢/kWh
3.6 million[1]	$24.4 billion	$21.5 billion	1.34
4.6 million[2]	$29.8 billion	$26.9 billion	0.90
7.2 million[3]	$40.8 billion	$37.8 billion	0.57

* 2.46 million bundles have been proportionately charged to December 2013[2]
** $2.92 billion have been accumulated in trust
*** Electrical energy yield calculated on basis of 6.25 bundles per 1 MW-year [1, p351]

2. THE FAST-NEUTRON REACTOR ALTERNATIVE

This approach of burying the spent fuel appears to be folly in light of the fact that used CANDU® fuel is still more than 99% heavy atoms all of which, uranium and the TRUs, can produce energy when fissioned in proven and available fast-neutron reactors (FNRs). In FNRs each ton of used CANDU® fuel can produce electrical energy worth about $1.2 billion at today's mid-point time-of-use rates [4]. Moreover, as the heavy atoms are split to extract their energy, they cease to exist as actinides and as a result their long-term radiotoxicity is eliminated as well, obviating the need for a million-year sequester in a DGR. The fuel emerging from the FNRs is merely cycled via pyroprocessing [5] to remove the resulting fission products, most of which decay to valuable non-radioactive minerals in a few decades, with two stragglers, strontium-90 and cesium-134, lasting for almost 300 years before they too decay to radiotoxic levels below that of uranium from which they were created.

This latter process was demonstrated in the US Argonne National Lab EBR-II sodium-cooled fast-neutron reactor that operated without incident for three decades to its decommissioning in 1994 with the complete reactor/fuel-cycling process put together in 1984 as the so-called Integrated Fast Reactor (IFR) complex [6]. The fuel cycling facility is still operational and is being developed further to service FNRs of 1000 MWe capacities.

The EBR-II reactor could consume about 20% of the fuel in one cycle, with the burn-up limited by the build-up of internal pressure inside the sealed fuel canisters from the gaseous fission products created in the fission process. Calculations show that for an idealized larger or stronger

fuel canister the fuel burn-up reaction could proceed to about 35 wt% before neutron absorption by the increasing amounts of fission products would stop the chain reaction (Figure 2(a)) [7,8]. At the end of such a cycle, or total fuel dwell time before replenishment, the fission

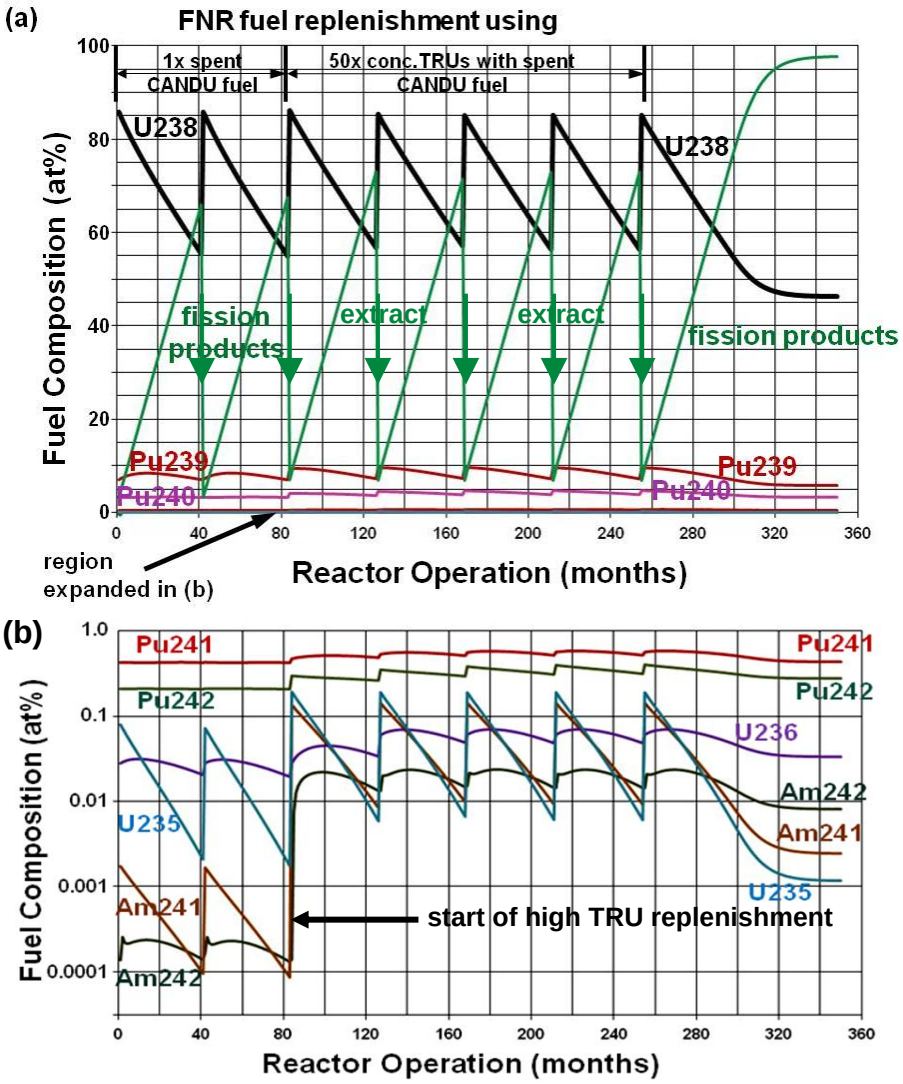

Figure 2. Fast-Neutron Reactor Fuel Behaviour under Replenishment with Two Forms of Used CANDU® Fuel

Two cycles have spent FNR replenished with used CANDU® fuel directly at months 1 and 41. Subsequent cycles starting at month 82 have FNR fuel replenished with used CANDU® fuel from which 90% of the uranium has been extracted to increase the concentration of transuranic actinides (see text). Note: Scale on vertical axis of Panel (a) is linear. Scale for Panel (b) is logarithmic to accommodate the large variations in concentrations of the minor isotopes.

products would be extracted by pyroprocessing, an electrolytic process in molten salt electrolyte [5], with the heavy atoms, which include sufficient fissile material, returned to the reactor and topped up with additional heavy atoms. This top-up heavy atom source could be either natural uranium, depleted uranium, or more interestingly spent nuclear fuel from CANDU® reactors. No additional fissile material is required since the required fissile level is maintained in the fuel of the FNR by being transmuted internally from uranium-238 to Pu-239 equivalent to the amount of Pu-239 used up.

A commercial design based on the EBR-II is now available as the GE-Hitachi PRISM fast-neutron reactor (Figure 3), currently being offered to the UK to help eliminate their excess plutonium [9]. It is designed for a fuel burn-up of about 15%, suggesting that this 15% could be replaced with spent CANDU® fuel waste to replenish the FNR fuel at the end of each cycle.

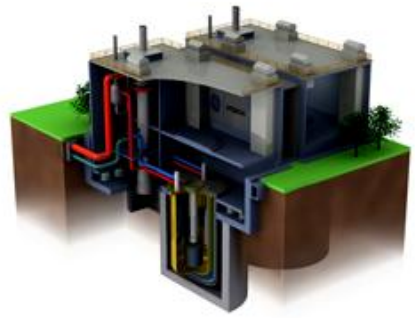

Figure 3. 300 MWe Fast-Neutron Reactor PRISM by GE-Hitachi in the USA.

The reactor is one of three designs currently considered by the UK Nuclear Decommissioning Authority [9] to use up the UK stockpile of civilian plutonium and to eliminate its proliferation concern.

http://www.nrc.gov/reactors/advanced/prism.html

Such an approach is exceedingly useful to make major inroads into the elimination of Canada's stored spent CANDU® fuel in a rather productive manner. On top of that there are further advantages that make it possible to synergize our fleet of CANDU® reactors and its need for fissile fuel. This arises from three complementary features: 1) electrolytic pyroprocessing of necessity separates a large pure uranium fraction during used FNR fuel cycling, 2) spent CANDU® fuel has a rather low content of fission products, and 3) the FNR maintains its fissile content required for operation and can even increase it. This interplay will be discussed below using the characteristics of the PRISM reactor with its 15% fuel burn-up as an instructive example.

3. FUEL UTILIZATION IN A PRISM-LIKE FAST-NEUTRON REACTOR

3.1 Characteristics of fuel cycling by pyroprocessing

The pyroprocess of separating used fast-neutron reactor fuel components is an electrolytic procedure carried out remotely in shielded facilities, that separates spent FNR fuel into three fractions: 1) a pure uranium component free of actinides and fission products; 2) a mixture of uranium and transuranic actinides (TRUs) that contains sufficient fissile material to restart the FNR; and 3) fission products virtually devoid of heavy-atom content [5].

3.1.1 Fraction 1: Pure Uranium (~60%)

An iron cathode is used initially to plate out most of the uranium as it dissolves as UCl_3 into the salt melt. The other fuel constituents, except for the noble metals, dissolve similarly. The free-energy difference between uranium chloride and the chlorides of the TRUs as well as the chlorides of the dissolved fission products assures that only uranium is plated out on the iron anode. For instance, plutonium is held to less than one part per million. Some salts are entrained among the crystals of uranium as this metal plates out.

3.1.2 Fraction 2: Uranium/TRU mixture (~25%)

When the uranium concentration has decreased sufficiently, to about one quarter of the TRU concentration, a second cathode is activated which consists of liquid cadmium in a ceramic crucible suspended in the molten salt. This cathode plates out the remaining uranium plus the mixture of all TRU elements, which form intermetallic compounds with cadmium. This process substantially decreases the free energy difference between TRUs and uranium to permit the joint electrolytic removal of the actinides from the salt melt. The metallic uranium/TRU as an unseparated mixture is won by evaporation of the cadmium.

3.1.3 Fraction 3: Fission Products (~15%)

During the operation of the electrolytic cell, fission products (FPs) accumulate in the salt, or, if noble metal FPs such as rhodium and ruthenium, drop into a molten cadmium layer at the bottom of the electrolytic cell. Due to their radioactive decay the FPs eventually increase the heat load of the molten salt to the permissible limit of the equipment. At that point any remaining TRUs are recovered by drawing down electolytically, while the salt and metal phases are removed to extract the fission products for 300-year storage. In this process the FPs are about 99.9% free of heavy metals, or better [5]. The salts and cadmium are reused.

3.2 Fuel reconstitution with uranium

The metallic uranium (Fraction 1) and the metallic uranium/TRU mixture (Fraction 2) are melted together under partial vacuum, along with heavy metals from a source such as depleted uranium to make up the fuel deficit (~15% in the case of the PRISM reactor design [10]) due to the extraction of the fission products (Fraction 3). Fertile isotopes such as U238 are sufficient, since the uranium/TRU fraction 2 mixture retains all of the required fissile components to permit the reactor to resume power and achieve neutron equilibrium. New fuel pins are simply cast from the melt into molds to the required tolerance, accomplished remotely in shielded facilities [6, p.134].

3.3 Fuel reconstitution with spend CANDU® fuel

A major advantage in terms of elimination of stored spent CANDU® fuel "waste" can be gained if the 15% heavy atom deficit in the FNR fuel, above, is filled by spent CANDU® fuel which is constituted to over 99% of uranium and atoms heavier than uranium, the latter being primarily plutonium and smaller amounts of other transuranic elements. Table 2 column (2) shows the isotopic composition of the heavy atoms for spent CANDU® fuel that has been stored for about 26 years during which time 75% of the Pu241 (half-life 13.2 years) has decayed to Am241.

Composition in Weight-Percent of Spent CANDU® Fuel at Two Times After Exit from Reactor (adapted from [1, p.341])

	(1)	(2)	(3)
		T = 26.4 y**	
U235	0.72*	0.23*	0.207*
U236	0.0	0.07	0.063
U238	99.28	98.58	88.53
Pu239	0.0	0.25*	2.5*
Pu240	0.0	0.10	1.0
Pu241**	0.0	0.005*	0.05*
Pu242	0.0	0.01	0.1
Am241	0.0	0.015*	0.15*
Fission Products	0.0	0.74	7.4
*Fissile content	0.72	0.50	2.757

** Pu241($T_{½}$ = 13.2 y)

Table 2. Isotopic composition of fresh CANDU® fuel (column 1), spent CANDU® fuel about 26 years (2 half-lives of Pu241) after exiting from the reactor (column 2), and spent CANDU® fuel from which 90% of the uranium has been extracted by a technique such as uranium nitrate crystallization [11,12] (column 3).

A further advantage in nuclear waste management is gained if the spent CANDU® fuel has about 90% of the pure uranium removed from it and only the 10% residue that is a mixture of mostly uranium and all of the transuranic actinides and fission products is used to replenish the used FNR fuel during pyroprocessing and fuel refabrication. As has been reported previously, this would accelerate the elimination of the long-term radioactivity of spent CANDU® fuel [13].

Moreover, as part of the FNR fuel refabrication, it is possible not only to replace the extracted fission products with such treated spent CANDU® fuel but also the 60% pure uranium that is separated in the pyroprocess.

4. EXCESS FISSILE MATERIAL FOR USE IN CANDU® REACTOR FUEL

Figure 4 shows that during the proposed refabrication of FNR fuel using treated spent CANDU® fuel as replenishment and replacement of pyroprocessing fractions (1) and (3), not only is 75% of the used FNR fuel replaced, but fissile material is brought in with the treated CANDU® fuel in excess of the fissile content needed to maintain neutron equilibrium in the FNR. Since this material is not required it is possible to take an equivalent part of the FP-free uranium/TRU mixture (fraction 3) from the pyroprocess separation and create fuel for CANDU® reactors.

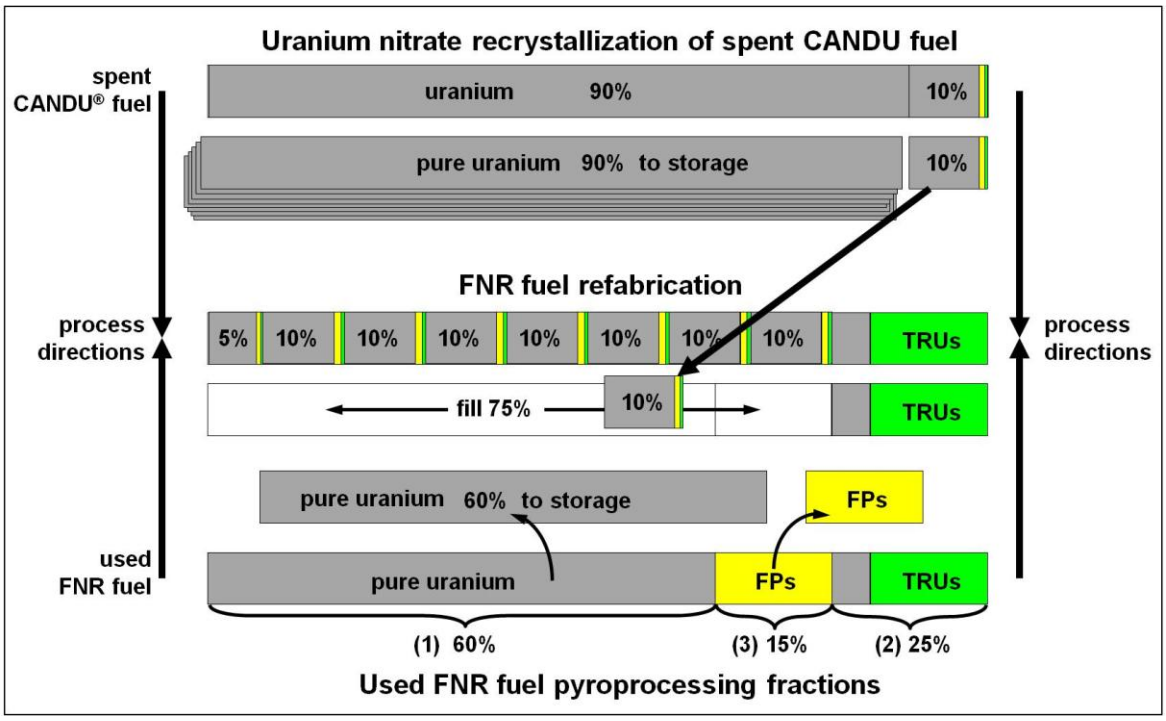

Figure 4. Schematic of the processing of both used FNR fuel (from the bottom) and spent CANDU® fuel (from the top) during cycling and re-fabrication of FNR fuel.

Fission products (FPs) are represented proportionately by yellow rectangles, the transuranic actinides (TRUs) that include fissile and non-fissile Pu, Am, Cm, etc. are shown proportionately as green rectangles, while uranium, largely depleted of the U235 isotope, is shown as gray rectangles. Note that within each of the fractions the constituents are homogeneously distributed and that in reconstituting the FNR fuel all of the constituents are melted together.

A quick calculation of the fissile content of the 10% residue of the treated spent CANDU® fuel would be 2.76% with a fission product content of 7.4% (Table 2, column (2). Since the refabricated fuel would only contain 75% of this (Figure 4), the excess fissile content from the spent CANDU® fuel would only be 2.07%.

A single PRISM reactor has a core load of about 9180 kg of heavy atoms [10], suggesting that the excess fissile load brought in via treated spent CANDU® fuel would be 190 kg. This amount would be removed from fraction (3) of pyroprocessed used FNR fuel, which contains a mixture of uranium and TRUs, Since this mass from fraction (3) is also replaced with treated CANDU® fuel, the total fissile amount that can be taken from fraction (3) is close to 197 kg. This material, taken via the pyroprocess at the end of the fuel dwell time of 5.75 year in a PRISM reactor, contains effectively no fission products and is therefore relatively easy to use in comparison to the treated spent CANDU® fuel.

A near comparison of excess fissile availability and make-up fissile requirements at equal power outputs would match a CANDU® reactor at the Darlington Nuclear Generating Station with an output of 890 MWe with three PRISM units with a combined output of 933 MWe [10]. Table 3 details some of the relevant fuelling characteristic between the two types of reactors and including fissile conversion ratios that help determine the fissile excess and deficit.

Table 3. Comparison of Characteristics between CANDU® and PRISM reactors

	1 PRISM**	**3 PRISMS**	**CANDU®**
Output	311 MWe	933 MWe	890 MWe
Estimated core load (HA)	9.18 tons	27.6 tons	120 tons
Fissile load (BOFDT)*	1.43 tons	4.30 tons	0.865 tons
Fissile load (EOFDT)*	1.43 tons	4.30 tons	0.601 tons
Fuel dwell time (FDT)	5.75 years	5.75 years	1 year
Fissile conversion ratio	1.23	1.23	0.69
Fissile excess (deficit) at FDT	329 kg	988 kg	(264 kg)
Fissile excess (deficit) per year	57.3 kg	172 kg	(264 kg)
Excess fissile (replenishment with spent CANDU fuel / year)	33.0 kg	99.0 kg	
Total excess fissile per year	90.3 kg	271 kg	

*Beginning (BOFDT) and End (EOFDT of Fuel Dwell Time in reactor
**PRISM data from [10], CANDU data from [14]

The above result of a fissile excess of 197 kg from FNR fuel replenishment with treated spent CANDU® fuel at the end of a fuel dwell time of 69 months (3 fuel shift cycles for the PRISM [10]) translates into 33 kg/year of available fissile material for CANDU® fuel, or 99 kg from three PRISM reactors. At the same time three PRISM reactors with a conversion ratio of 1.23 produce a further 172 kg fissile material that is mixed with an additional 40% non-fissile uranium and TRUs. However, the yearly combined 271 kg of available fissile excess virtually completely matches the annual fissile deficit of 264 kg of one Darlington CANDU® reactor.

5. CANDU® – FNR SYNERGY AT ONE-TO-ONE OR BETTER

The discussion of the process above suggests that a fast-neutron reactor such as the GE-Hitachi PRISM, coupled with a CANDU® reactor of equal size can synergize the fuel requirements of both reactors. The fuel of the FNR can be replenished with currently stored and future spent CANDU® fuel, while fissile deficit of the CANDU® reactor fuel can be replenished with the excess fissile material produced during pyroprocessing and refabrication of that FNR fuel. Currently the ratio between these types of reactors has to be one to one for reactors of equal power. However, in future it may be possible to increase the conversion ration of the FNR such that one FNR could provide fuel for two CANDU® reactors of equal power each. Alternatively, should CANDU reactors be fuelled with thorium such that the conversion ratio of the CANDU reactor is substantially raised from 0.69 (Table 3) towards 1.0, say 0.95, then even the current PRISM FNR fuel cycle using spent CANDU fuel with a combined fissile excess of 0.36 would cover the fissile deficit of about seven such reactors of equal power to the PRISM reactor.

6. POSTSCRIPT

While there are other scenarios that are possible depending on how comfortable one feels with the handling and remanufacture of fuel that is much more highly radioactive than natural uranium, the process outlined above builds quite logically on current thinking of manufacturing CANDU® fuel using the DUPIC approach, natural equivalent uranium or recovered uranium. It is clear that with the projected end of economically available uranium in Canada in about 40 years the refuelling of CANDU® reactors has to consider the existing stocks of spent fuel on the one hand and the creation of fissile material via fast-neutron reactors on the other. Both will require a more intensive involvement with highly radioactive raw materials.

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