Used CANDU Fuel Waste Consumed and Eliminated: Environmentally Responsible, Economically Sound, Energetically Enormous

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Abstract

The 43,800 tonnes of currently stored CANDU nuclear fuel waste can all be consumed in fast-neutron reactors (FNRs) to reduce its long-term radioactive burden 100,000 times while extracting about 130 times more nuclear energy than the prodigious amounts that have already been gained from the fuel in CANDU reactors. The cost of processing CANDU fuel for use in FNRs plus the cost of recycling the FNR fuel is about 2.5 times less on a per kWh energy basis than the currently projected cost of disposal of 3.6 million used CANDU fuel bundles in a deep geological repository.

1. Introduction

Canada's used and stored nuclear fuel waste in 2011 is expected to be about 44,000 tonnes [1]. Only 0.74% of this fuel has been converted to fission products with the concomitant yield of energy [2, p.341], while the remaining 99.26% consists of uranium and other heavy metals which together contain 130 times more untapped nuclear energy. This energy can all be extracted from the used fuel using fast-neutron reactors (FNRs). Simple calculations indicate that the gross revenue from this 99.26% would be $39.3 trillion in electricity [3] plus useful cogenerated heat.

Of the 0.74% fission products in the used fuel 70% are non-radioactive, while the radioactivity of virtually all of the remaining 30% will decay to background levels in about 300 years. Of greater concern is a small fraction of transuranics (TRUs), atoms heavier than uranium such as plutonium, americium, and curium that are created in the reactor. Although the TRUs constitute only 0.4% of the used fuel, they do not decay to background levels for about 400,000 years. Their long-term safe-keeping constitutes the major focus in nuclear waste disposal proposals.

The complete consumption of the heavy atoms, leaving only fission products, has the attractive potential of eliminating the TRUs, and with them the concern about their long-term radioactivity. The 300-year radiation hazard from the resulting fission products is a much more manageable task. After that time the fission products, atoms roughly of atomic number 35 to 65, become a valuable source of platinum-group metals, rare earths and the like, extractable by ordinary means.

To dispose of current and future used nuclear fuel waste, the world of nuclear nations, including Canada, has primarily concentrated on the potential use of deep geological repositories (DGRs)

in whatever suitable rock strata are locally available, with retrieval possibilities or with eventual permanent closure. In Canada to build such a DGR is expected to cost between $16 and $24 billion, with total life-cycle costs estimated to be from $24.4 to $40.7 billion [1; 2, p163; 20, p3].

The extraordinary potential of fast-neutron reactors to eliminate the long-term radiation hazard of used CANDU fuel and at the same time deliver about 130 times more carbon-free nuclear energy from the used fuel, begs for at least a preliminary analysis of the costs associated with such an approach in comparison to the cost of Canada's planned DGR. Both approaches are or would be self-funding via revenue-producing reactors: CANDUs or FNRs. Therefore, while some of the characteristics of an FNR facility are discussed, the emphasis will finally be on the comparative cost of the non-productive aspects: the DGR versus the fuel cycle facility that is an additional integral part of such an FNR facility.

The comparison shows that the cost of constructing and operating a fuel cycle facility for pyroprocessing of metal fuel for a sodium-cooled FNR is up to 2.5 times smaller than the cost of the planned Canadian DGR. This cost does not consider the additional benefits of the elimination of the long-term radioactive burden via the FNR facility nor of the long-term non-carbon energy potential of already-stored used nuclear fuel.

2. Evolution of Radioactivity

Figure 1 shows the radioactivity of used CANDU fuel relative to that of natural uranium over time following removal from the reactor. The evolution of radioactivity of the fission products in the fuel has two major components. Most constituents decay to background levels (and below) at around 300 years (solid green line). A few, including Zr-93, I-129, and Cs-135, together constitute a long-lived component, but are very close to background (dashed green line).

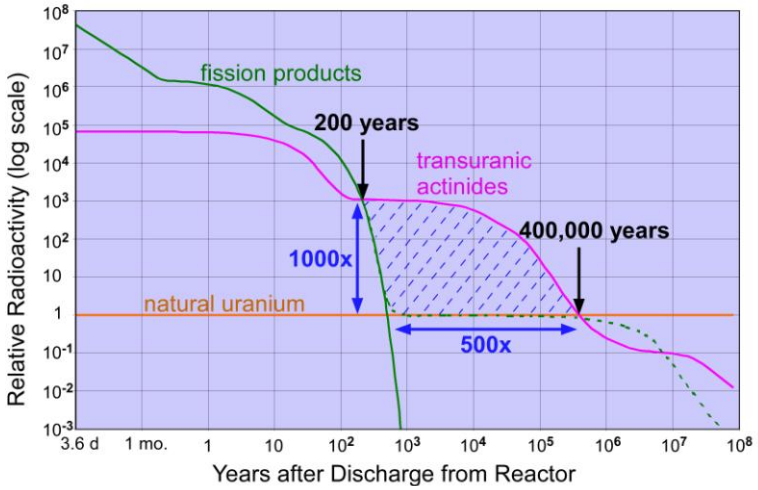

Figure 1. Evolution of Radioactivity from Used CANDU Fuel. Removal of the transuranic actinides (purple line) in fast-neutron reactors results in a 100,000 times reduction in radiation burden between 200 and 400,000 years (hatched area).

After about 200 years the major contributors to the radioactivity are the TRUs, which do not reach background levels for about 400,000 years. However, consuming the used fuel in an FNR in several fuel cycles has the consequence of eliminating TRUs and changing them to fission products (FPs). When the used FNR fuel is sent through the FNR-associated fuel cycling facility (FCF), pyroprocessing separates the fission products from the actinides (see Section 4. below), and reduces the concentration of uranium and TRUs in the FPs by a factor of 1000 [4]. This is in general sufficient to bring the TRUs to background levels after the short half-life Pu-241 isotope among the TRUs has decayed for about 100 years (Fig. 1). If further reduction of the TRU concentration is required then processes such as chromatographic separation with immobilized ligands specific for the actinides can be considered in future [5,6]. The Japanese, using amidoxime chelating groups fixed to polyethylene sheets, are extracting uranium from seawater with a concentration of 3 parts per billion [7]. This suggests strongly that lower concentrations of actinides in the extracted fission products can be achieved.

The relative benefit that can be derived from pyroprocessing alone, due to the 1000-fold reduction in TRU concentration in the used FNR fuel is very evident in the period from 200 years to 400,000 years (Fig. 1). Integrating between the original curve of TRUs and the curve for the FPs indicates that a reduction of 100,000 times can be obtained in long-term radiation burden.

3. Fuel Consumption in Fast-Neutron Reactors

3.1. Experimental Limits Achieved

Sodium-cooled FNRs in Russia (BN-350, BN-600), and France (Phenix) generally use mixed oxide fuel, and achieved 10-11% utilization of fuel. One reactor, the metal-fuelled EBR-II in the USA, regularly achieved 20% fuel utilization before refueling [8,9]. This facility has provided the greatest insight into fuel consumption, metal-fuel cycling, and FNR safety (Sections 4/5).

While, elsewhere, metal fuel was rejected early on due to swelling of the fuel under irradiation and subsequent deleterious fuel-cladding interactions at 1%-3% fuel consumption, EBR-II scientists at the Argonne National Labs systematically altered fuel charge, fuel canister, and fuel composition to achieve a safe 20% fuel consumption [8]. To avoid fuel-clad interactions the fuel slug was reduced in cross-sectional area to about 75% of the inner area of the fuel canister. To provide thermal contact between the now-smaller fuel slug and the wall of the canister, sodium, liquid at operating temperatures, was introduced as a thermal bond. When the canister burst at about 5% to 8% fuel consumption due to build-up of internal pressure from gaseous fission products, the sealed canister was provided with an empty volume (plenum) above the fuel. However, sharp indentations in the cladding, designed to prevent the fuel slug from rising into the plenum, resulted in stress failures even at 8% fuel consumption. They were replaced by spherical indentation. At the same time the steel of the canister was changed from 316 stainless steel to stronger HT9 ferritic steel. One further change was a change in fuel composition to the eventual use of 10% zirconium, to increase the melting temperature of the fuel, a

uranium/plutonium/zirconium alloy. These changes produced a fuel consumption of close to 20% without failure of any of 34,000 fuel pins used thereafter [9,10].

This suggested that the EBR-II was able to operate with fuel that before refueling contained about 20 wt% fission products.

A few fuel pins were tested to canister failure, which occurred at about 23%-25% fuel consumption. Before further improvements in fuel design were made the reactor was shut down in 1994 by edict of the US Congress [11]. Nevertheless, Argonne scientists designed and tested fuel pins with a still larger plenum for potential safe 25% fuel consumption, and tested them successfully in the French Phenix FNR starting in 2007 [12]. The pins were not tested to failure.

3.2. Calculated Theoretical Limit

The safe fuel consumption up to 25%, resulting in 25 wt% content of fission products, was not necessarily a fundamental limit. Ultimately the build-up of fission products in the fuel pin should absorb sufficient neutrons to stop the chain reaction. To reach such a limit in practice the fuel pin would need to contain a large enough plenum such that the internal pressure from gaseous fission products would not stress the steel cladding beyond its elastic limit.

On the basis of that assumption calculations were performed examining the performance of a sodium-cooled fast-neutron reactor loaded with such ideal fuel pins. As a starting point the relative volume proportions of fuel, sodium bond plus coolant, and steel were taken from the sodium-cooled GE-Hitachi S-PRISM design [13]. Neutron cross-sections for elastic and inelastic scattering, fission and radiative capture were obtained from the ENDF data base [19] for uranium and plutonium isotopes in the fuel, for 18 fission products with the highest macroscopic capture cross-sections at thermal energies, as well as for sodium, for zirconium in the fuel, and for the constituents of HT9 steel. Energy transfer matrices for inelastic neutron scattering were obtained from Yiftah et al. [14]. These transfer matrices limited the high energy calculations to 12 groups from 3.668 MeV to 9.1 keV in equal logarithmic intervals of 0.5.

The energy loss history of the spectrum of nascent fission neutrons was followed for 202 rounds of neutron interactions with reactor fuel, fission products, sodium and steel atoms. At that time the 2% of the original fission neutrons remaining were assigned proportionately to macroscopic fission and capture cross-sections of all the components. A homogeneous reactor was assumed, since mean-free-paths were larger than the fuel assembly lattice pitch of the S-PRISM reactor.

The results in Fig. 2 are for several cycles after the fuel in the reactor has reached equilibrium compositions. At this stage all of the TRUs in the reactor remain constant from cycle to cycle, except for repeating variations within each cycle. As fission products build up to a point that they absorb too many neutrons to sustain the controlled chain reaction, constant power of the reactor can no longer be maintained, and the reactor shuts down slowly. At this point the fuel assemblies would be removed and substituted with a set of suitably prepared new assemblies.

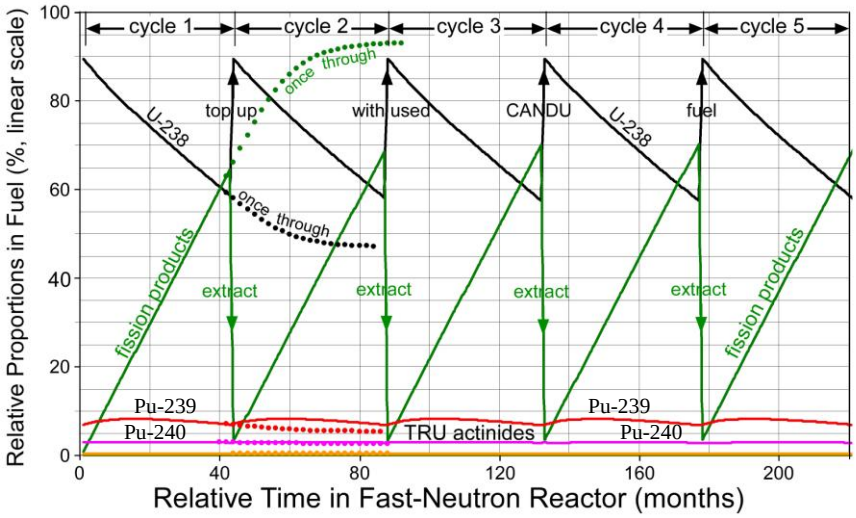

Figure 2 Consumption of uranium and behavior of other actinides

Figure 2 shows that for an ideal fuel pin the fuel charge is consumed about 35% before power is limited by fission product build-up even though the weight percent of the fissile actinides (e.g. the plutoniums) is equal to their proportion at the start of the cycle. To continue to consume the remaining actinides, the fission products must be separated from the actinides. This occurs in the fuel cycle facility (FCF) of the reactor complex by means of pyroprocessing (see Section 4).

3.3. Used CANDU Fuel Top-Up

The fission-product-free actinides from pyroprocessing, 65% of the original charge, would be topped up to 100% with used CANDU fuel which has been converted from the oxide to metal form. No separation of the CANDU fuel components is required, since the actinides from pyroprocessing contain sufficient components that are fissile at high neutron energy to restart the reactor. Moreover, the small percentage of fission products in used CANDU fuel, 0.74%, is too low to limit the reactor appreciably.

The experimental results from the EBR-II facility with 20% fuel consumption in one cycle indicate that in 5 cycles one reactor-core-full of used CANDU fuel uranium and TRUs could be consumed completely. For fuel pins built for the calculated limit of 35% fuel consumption only 3 cycles through the facility would be enough to use up one reactor-core-full of used CANDU fuel.

4. Pyroprocessing

The separation of actinides from fission products by pyroprocessing has been well-described [4,9]. In brief, used-fuel assemblies from the FNR are brought into a shielded fuel cycle facility, where all operations are sufficiently simple to be amenable to remote control. The fuel pins are disassembled, chopped into small cylinders and put into a wire mesh basket that becomes the anode of an electrolytic cell operating with molten chloride salts at about 500°C. The cell also

contains a bath of molten cadmium beneath the salt, and two cathodes suspended in the molten salt. One is solid iron, the other a vat of molten cadmium.

In the operation of the cell the various constituents in the chopped-fuel mesh basket dissolve in the salt or remain as solids that either stay in the basket or drop into the cadmium under the molten salt. These are the noble metals, such a rhodium and palladium, as well as zirconium. The actinides dissolve as chlorides in the salt bath, as do the rest of the fission products. The electrolytic process then plates out uranium on the iron cathode until a large proportion of it is removed from the molten salt. Due to the large difference in free energy between uranium and the other actinides in their chloride form, no other actinides plate out on the iron electrode.

However, this free energy difference is very much reduced when the molten cadmium cathode is put into operation. At this electrode all the actinides can form intermetallic compounds with cadmium and plate out together along with the remaining uranium.

Laidler et al. [4] indicate that less than 0.1% of the actinides remain in the salt that contains most of the fission products.

4.1. Proliferation Resistance

From the previous discussion it is clear that there is no separation of the actinides in pure form except for a large proportion of uranium. None of the plutonium is separated from the other actinides. Indeed, experimental operation of such a pyroprocessing cell on 4.5 tonnes of used fuel from the EBR-II indicates that plutonium plating out in the molted cadmium cathode contains not only the higher TRUs but also from 25% to 64% uranium [9, p189ff]. Moreover, from Figure 2 it can be seen that the plutonium is not isotopically pure, but contains about 26% Pu-240. Thus it would be difficult to utilize such material for explosive purposes, requiring large centrifugal enrichment facilities to separate the 74% Pu-239 from the 26% Pu-240.

Concerns with handling plutonium also impinge on the start-up of a fast-neutron reactor. Calculations resulting in Figure 2 indicate a requirement of about 7% Pu-239/241 to bring the reactor into neutron balance. It is conceivable that such plutonium may be obtained from dismembered-weapons stores. However, the sensitivity of such acquisition and transport may force a different start-up scenario. It is quite possible to start with a fuel mix containing low-enriched uranium with just less than 10% U-235 (see Figure 3). Figure 3a shows that fuel consumption, as represented by the usage of U-238, is not significantly different than when starting up with plutonium-enriched fuel. Figure 3b shows the detail of U-235 usage as well as the build-up in the reactor core of Pu-239/240/241, that serve to maintain neutron equilibrium. Changes in concentration of these actinides are substantial initially. However, after about two refuel cycles equilibrium of all actinides is reached at levels identical to starting with plutonium-enriched fuel. The difference is a slight remanent level of U-236 from radiative capture in the high initial concentrations of U-235.

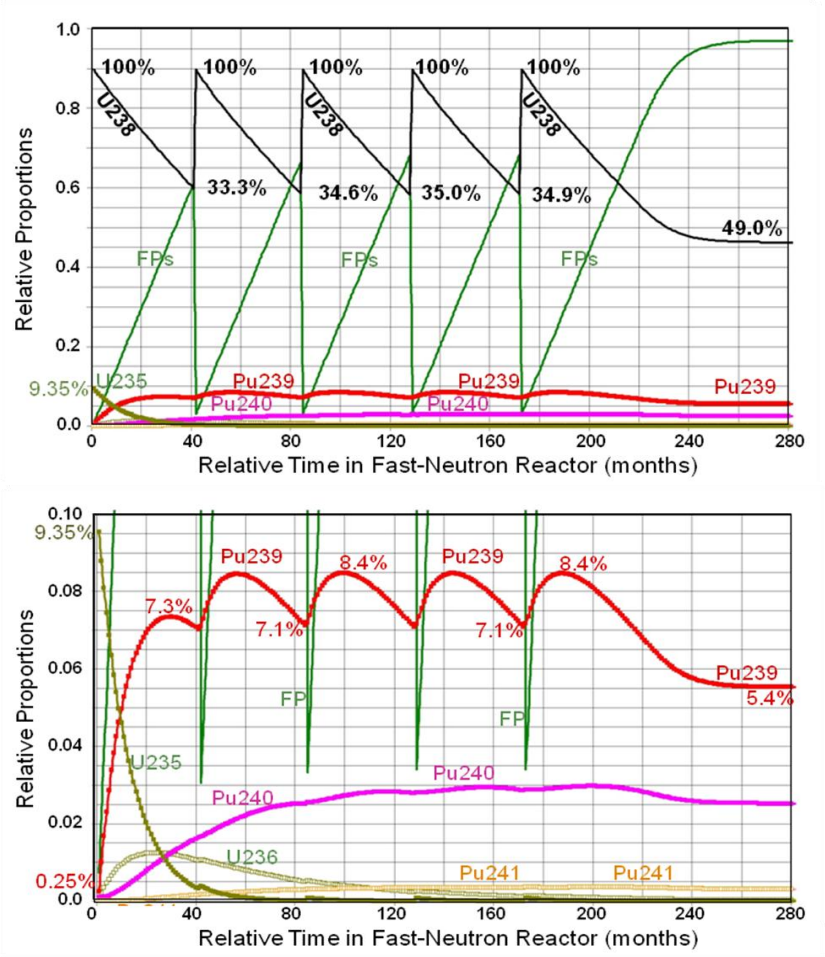

Figure 3. Fuel Consumption and Build-Up of Pu Isotopes in FNR on Start-Up with U235
Fig. 3a (top) Large scale. Fig. 3b (bottom) Expanded scale.

5. Safety

The safety characteristics of a sodium-cooled metal-fueled FNR, the US EBR-II, are described in detail by Till and Chang [9, Ch.7] and by Koch [10, p.24/25]. Only three characteristics will be mentioned here, most pertinent to the stressors of the kind that befell the reactors at Fukushima, Three-Mile-Island, and Chernobyl.

Two crucial experiments were performed in 1986 to test the passive safety characteristics of EBR-II reactor under full power that likely would not be done with thermal reactors: initiating an unprotected (no SCRAM) loss-of-flow (ULOF) situation in the core and letting the event run its course without automated or human intervention; and similarly initiating an unprotected loss-of-heat-sink (ULOHS), i.e. loss of cooling from the intermediate heat exchanger. In both cases the reactor shut down and reached a stable temperature in 300 to 500 seconds [9, p.148/150].

The third characteristic was the built-in passive cooling by two convective liquid metal heat exchangers in the reactor tank, called shut-down coolers, which carried fission product heat to

two convective atmospheric heat exchangers outside the reactor building at every normal shut-down of the reactor [10, p. 24].

While many redundant safety systems in current reactors work very well indeed, these characteristics as exemplified in the EBR-II would have provided a safe passive response to the events leading to the happenings at Chernobyl, Three-Mile-Island and Fukushima Daiichi.

6. Economics

Finally, if an FNR facility is to substitute for the use of a deep geological repository (DGR), it is important to compare the relative costs of the two approaches.

The FNR facility requires a reactor plus a fuel cycle facility. Of these the reactor cost is probably less crucial, since the FNR creates heat and electricity similar to any thermal reactor, and so would be revenue-producing. Nevertheless, such a nuclear-waste-consuming reactor should not have a completely unreasonable price. While comparisons are difficult, since FNRs have only been built so far as first-of-a-kind, and relatively few thermal reactors have been constructed lately, recent textbooks put the cost of a "mature" FNR at between 1.1 and 1.25 times the cost of a thermal reactor [15, p.42]. First-of-a-kind FNRs in the field appear to cost a low of 1.26 times the cost of established thermal reactors on the Asian continent (Table 1) to a high of 2.1 times as much (France) on a $/kWe basis. Second and further reactors of the same type are estimated to drop in cost to as low as 0.7 times thermal reactors per kWe (Table 1; Japan, Russia) to 1.05 times (France). Discounting their waste-consuming advantage, this puts initial FNRs on a relatively reachable level, with further FNRs equally or less costly than mature thermal reactors.

Table 1
Cost of Commercial Size Fast-Neutron Power Reactors versus Mature Water-Cooled Reactors

Country	Reactor		FNR Cost or Cost Ratio	Reference Reactor Cost	
France	SuperPhenix	(1,240 MWe)	2.1	1.0	French PWR
[9, p.275ff]	SuperPhenix II	(1,500 MWe)	1.05		
Russia	BN-600	(560 MWe)	1.5	1.0	Russian LWR
[9, p.276]	BN-800	(under construction)	1.35 (estimate 2011)		
	BN-1800	(under development)	0.72 (estimate 2011)		
India	PFBR	(500 MWe)	$2,500/kWe	$1,978/kWe	CANDU in China
[16]			1.26	1.0	[21]
				$1,840/kWe	PWR in China
			1.36	1.0	(Framatom ANP)
Japan	JSFR	(1500 MWe)	$2,600/kWe	$1,769/kWe	CANDU in China
[17]	Design, fist of a kind		1.47	1.0	[21]
	Nth reactor (projected)		$1,300/kWe	$1,840/kWe	PWR in China
			0.70	1.0	(Framatom ANP)

The additional expense associated with a waste-consuming FNR is the shielded fuel cycle facility (FCF) required both for processing used CANDU fuel into a metal form suitable for the FNR and for cycling the used fuel from the FNR itself. From their experience at the FCF associated with the EBR-II in the Integrated Fast Reactor facility, which is still operating and processing fuel from the EBR-II after the reactor was decommissioned, Till and Chang [9, p292] estimate that for a 1000 MWe reactor the associated shielded FCF would cost about $100 million.

Their breakdown of the fuel cycle cost components is, in mill/kWh:

Capital fixed charges at 15%	1.90
Operating and maintenance	1.27
Process consumables, etc.	0.76
Disposal fee (300 year storage)	0.50

The total is 4.43 mill/kWh with construction capital charges, and 2.52 mill/kWh without.

Till and Chang [9, p.292] have assumed a disposal fee for storage of fission products from the FNR of 0.5 mill/kWh, half the 1 mill/kWh for planned repository disposal in the USA. Studies in Japan indicate that partitioning of fission products and transmutation of minor actinides can reduce the disposal repository size by a factor of 4 to 5; moreover, additional separation and storage of cesium and strontium can reduce the size by more than a factor of 100 [15, p.19].

The numbers by Till and Chang permit a comparison on a mill/kWh basis with the costs of disposal of used CANDU fuel in a planned Canadian deep geological repository (DGR) in the Canadian Shield at the site of a willing host community [1, p.15]. It is estimated that the total lifetime cost of this DGR, a first-of-a-kind facility, will be $24.4 billion for the Adaptive Phased Management option adopted [2, p163]. Under consideration currently is increasing the capacity of the DGR from 3.6 million bundles of used CANDU fuel to 7.2 million bundles [1, p.104]. This produces some economies of scale, with an increase of only 67% in cost for doubling the capacity [20, p3]. The cost of either DGR is to be met by the tithing of the revenue from the consumer of electricity produced from nuclear energy on a mill/kWh basis. The initial charges are to be low, but are to ramp up later, in the years closer to the construction of the DGR [18].

In 2010 the total used CANDU fuel accumulated at various reactor sites in Canada amounted to 2.2 million used fuel bundles [1, p11]. On the basis that 6.25 bundles would have produced 1MW-year of electricity [2, p351], the electrical energy produced by those fuel bundles is 3.07×10^{12} kWh. The funds collected from fees on this electricity, and put in trust accounts, were $2.28 billion, averaging to 0.74 mill/kWh up to the present.

To meet the total lifetime-cycle costs of the smaller DGR, to be filled with 3.6 million used fuel bundles, $24.4 billion have to be tithed in total, requiring a further $22.12 billion above the current $2.28 billion already in trust. This amount has to be obtained from 1.4 million bundles above the currently accumulated 2.2 million bundles. If the DGR is built to accommodate 7.2 million bundles, its total life-cycle cost in proportion to the 67% increase of the repository cost

Table 2
Future Average Mill Rate Required to Meet DGR Costs

Total Life Cycle Cost	Outstanding Balance	DGR filled with 3.6 million bundles tithe from 1.4 million* or 1.96×10^{12}kWh** Avg. mill rate needed	DGR filled with 7.2 million bundles tithe from 5.0 million* or 6.99×10^{12} kWh** Avg. mill rate needed
$24.40 billion	$22.12 billion	11.29	
$40.75 billion	$38.47 billion		5.50

* 2.2 million bundles have been tithed to date
** Electrical energy yield calculated on basis of 6.25 bundles per 1 MW-year (2, p351)

would be $40.75 billion, requiring the collecting of $38.47 billion above the $2.28 billion currently accumulated. This can be levied on 5.0 million future bundles. Table 2 shows the average mill rates required from now on, going forward, to meet the costs of the DGR under these conditions. Since the effective mill rate for 72,999 used fuel bundles in 2010 [1, p197] was 1.46 mill/kWh, a planned slowly escalating mill rate to the start of construction of the DGR will result in much higher final rates in the later years than the averages shown in Table 2.

Comparison between these costs and the cost of processing and cycling used CANDU fuel for use in an FNR, above, indicates that the processing costs of 4.43 mill/kWh are a factor of 2.5 lower than the average future mill rate for a DGR accommodating 3.6 million used CANDU fuel bundles, and still 24% lower than the costs of a DGR accommodating 7.2 million used fuel bundles. However, a similar saving from the economies of scale as for a larger DGR likely applies to the fuel cycle facility which can as readily be scaled for several FNRs on the same site.

7. Summary and more

Fast-neutron reactors based on the design of the US EBR-II have the capability of consuming used nuclear fuel waste from CANDU reactors and other water-cooled reactors, reducing the long-term radioactive burden about 100,000 times, and shortening the radioactive lifetime of the current used fuel from 400,000 years to reach background to as little as 300 years. As an added bonus, such FNRs can extract about 130 times more energy from used CANDU fuel than the prodigious amounts of nuclear energy that have already been extracted. Since the used fuel already exists, stored at reactor sites, the energy can be considered to be carbon-free, devoid of fossil fuel energy associated with the mining and manufacturing of fresh fuel.

A fast-neutron reactor, required for the consumption of used fuel waste, has an additional advantage to those mentioned above. It can readily adjust power. This makes it possible to follow the variations in energy demands during any given day, and also to partner in the mix of non-

carbon renewable energies that are sporadic, such as wind or solar. These reactors, in contrast to thermal reactors, do not exhibit a xenon effect nor samarium poisoning that cause a delay in start-up or else require large compensating positive reactivity insertions. This is clearly understood from the radiative capture cross sections for high energy neutron interactions (Fig. 4) of xenon (major blue line), samarium, indeed of all fission products. Such cross sections are low at energies of nascent fission neutrons, and all are clustered below the fission cross sections of all of the actinides. Thus the decay of one fission product into another, e.g. Te-135 into I-135 into Xe-135, that is of great consequence at thermal energies, has little effect on total neutron absorption at high energies. This is a very useful characteristic that permits ready load-following by FNRs.

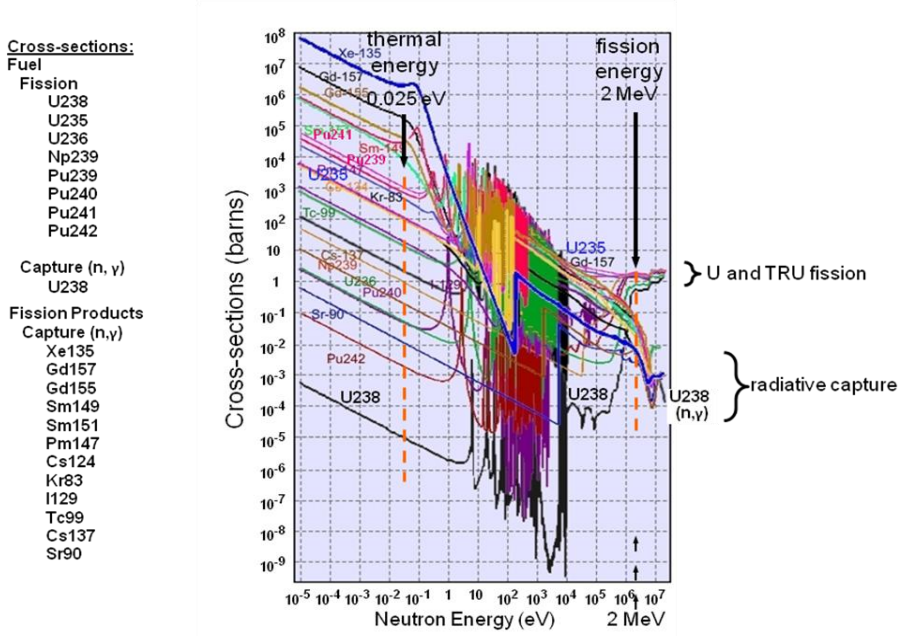

Figure 4. Combined Radiative Capture and Fission Cross Sections of Selected Isotopes [from 19]. Nascent fission neutrons have an average energy of 2 MeV.

Processing of the used CANDU fuel is required to convert it to the metal form for consumption in an FNR, and also for fuel cycling by pyroelectroprocessing in molten salts. However, the analysis above shows that the cost associated with such processing is up to 2.5 times less than the currently projected costs of disposal of used CANDU fuel in a deep geological repository.

Building and operating a DGR in a willing northern community is expected to provide local jobs for three to four decades, primarily in service-related employment [1, pp.53/56/90]. In comparison, an FNR facility built at that site would additionally provide energy to use and to sell, with an associated stimulation of the local economy and a raised standard of living for centuries.

Therefore taking advantage of a nuclear-waste-consuming fast-neutron reactor facility to consume CANDU nuclear fuel waste instead of disposing the used fuel in a DGR is economically sound and has huge societal and ecological benefits.

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