SOVEREIGN NUCLEAR CANADA AT A CROSSROADS: RECYCLE OR DIE

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Abstract

For over 60 years Canada has relied on its mined natural uranium to power its CANDU reactors. At current rates of mining and exporting, Canada's 500,000 tons of economical uranium reserves will be exhausted by 2050. The addition of small modular reactors (SMRs) will only quicken the reduction of this resource, particularly since the SMR requirement for enriched fuel calls for manifold the volume of natural uranium. However, recycling of Canada's current stockpiles of used CANDU fuel can furnish starting fuel enriched with fissile transuranics for SMRs with a total power of 24,000 MWe. Moreover, for SMRs that can maintain and augment their fissile components, the fuel can be replenished for centuries solely with U238 or depleted uranium, while such SMRs provide fissile fuel for our CANDU reactors at equivalent power. We are at a crossroads. We can either do nuclear right, or it dies.

1. Introduction

To paraphrase George Santayana, "those who fail to learn from history are doomed" [1].

At the beginning of the age of nuclear power generation uranium was a precious commodity. Reactors that were being built relied for their operation on a minor component of uranium, the isotope U-235 that was present at a level of only 0.72 % in natural uranium. Moreover, since so few uranium ores had been found, and most were used for the war effort, this rather promising energy-producing technology was virtually still-born.

Two approaches were undertaken as a result; both of them successful. One was the creation of a technology that would utilize more or all of the natural uranium as fuel. The other was the search for and successful discovery of rich pockets of uranium ores in many countries, Canada among them.

1.1 Transmutation of U-238

Enrico Fermi, who built the first nuclear reactor in Chicago [2], considered the physics of the nuclear reaction and opined that the current reactors were but a stepping stone from the fission of the U-235 isotope to the eventual use of the remainder of the 99.28% of natural uranium that is in the form of the isotope U-238. As a result over 100 times more energy would be harvested from the same fuel. After all, the splitting of every heavy atom isotope yields an energy of about 200 MeV, not just U-235, but U238, or even isotopes of Np, Pu, Am, etc., heavier atoms produced by transmutation in the reactors.

Of these heavier isotopes Pu-239 was a natural product of the absorption of a neutron in the major uranium isotope U-238, initially producing U-239 which via two nuclear electron emissions (beta-decays) converted to Np-239 within a few minutes and then to Pu-239 in just over two days.

Pu-239 behaves like U-235 in that it can be readily split to yield those precious 200 MeV of useful energy. Moreover, the fission of Pu-239 produces more nascent neutrons, fast neutrons, than the fission of U-235, especially if the fission is triggered by neutrons of higher energy. Indeed, as an added bonus, at high neutron energies virtually every heavy atom can be fissioned to produce energy and to produce more neutrons.

Fermi judged that such new neutrons might be sufficient in number to maintain a chain reaction for energy production and also to continuously convert the major uranium isotope U-238 to Pu-239 and heavier isotopes [3]

One of Fermi's co-workers and successors, Walter Zinn, a Canadian from Berlin (now renamed Kitchener) proved that this concept was feasible by constructing and operating a sodium/potassium-cooled reactor, the EBR-1 [4], at the Argonne National Laboratories in Idaho. That reactor, in contrast to water-cooled or graphite-core reactors, preserved the high velocity of nascent fission neutrons until they had found their target U-238 isotope to convert as well as to split a sufficient number of the other heavier fuel isotopes.

Rather interestingly, this reactor, a fast-neutron reactor, was the first to produce electricity by nuclear power, a few days before Christmas on December 20, 1951 [3].

This development was followed in a decade by larger reactors with similar technology [5], which were able to consume up to 20 % of the U-238 in the fuel, and, with recycling to remove the resulting fission products [6,7], effectively utilize all of the heavy atom isotopes in the fuel.

Thus these reactor/recycling developments solved two major challenges already early in the days of nuclear power generation. First, they very effectively maximized the energy yield from uranium fuel by demonstrating that all heavy atoms in the fuel could be fissioned in a controlled manner.

Second, and crucial to changing our current nuclear waste disposal approach, by splitting all the heavy atoms, including the transuranics Np, Pu, Am and heavier isotopes, these developments eliminated the long-term radiotoxicity of any such atoms that were produced by transmutation in the reactor core.

1.2 Uranium Discoveries

These reactor advances as well as subsequent recycling technologies were side-tracked by the discovery of masses of uranium in many countries [8]. Today over 20 countries supply the world's needs of about 60,000 tonnes of uranium annually [9], of which Canada lately furnishes about 16,000 tonnes each year [10]. Canada's economically mined reserves are estimated at 514,400 tonnes [8].

In total 2,802,267 tonnes of uranium have been mined worldwide up to 2018, with Canada contributing 531,608 tonnes to that total [8].

This glut of uranium has resulted in the proliferation worldwide of nuclear reactors which are unfortunately some of the least fuel-efficient technologies for energy production. The Canadian heavy-water-cooled CANDU reactors utilize only 0.74% of the mined natural uranium that constitutes CANDU fuel. The remaining 99.26% of the heavy atoms in the fuel is considered high-level nuclear waste.

Light-water reactors (LWRs) that are common in the rest of the world fare even worse. Although they utilize about 3 to 5 % of their type of fuel, that fuel is enriched from the natural level of 0.72% U-235 in mined uranium to a level of 3 to 5 % U-235 to compensate for the loss of neutrons in the light-water coolant/moderator. Since each additional 0.5% enrichment level requires the processing of an additional reactor core load volume of mined natural uranium, a 5 % enrichment requires almost 10 times the volume of mined uranium to charge up a single volume of an LWR core. The result is a net fuel utilization of only about 0.5 % of the mined natural uranium.

What makes such profligacy possible and still economical is the huge density of energy in nuclear fuel. Each fission of a single heavy atom produces about 200 MeV of energy, of which about 180 MeV is captured for our use. In contrast each single carbon atom combined with two oxygen atoms when burned produces only about 5 eV of energy. Therefore the single fission event of one uranium atom produces close to 36 million times more energy. Thus even if only 0.5% of the natural uranium fuel is used, the energy yield ratio is still 180,000 times in favour of uranium over the equivalent number of carbon atoms. So why worry?

The consequence of this inefficient use of fuel is three-fold, 1) the production of many tonnes of very slightly used highly radioactive uranium fuel as “waste”, 2) the production worldwide of about 2.5 million tons of apparently useless depleted uranium, and 3) the spectre of exhausting even the current relatively large finds of uranium ore.

To date the world has accumulated about 340,000 tonnes of highly radioactive used fuel, with the order of 60,000 tonnes in Canada [11]. While such used fuel is officially designated as “waste”, this term is really a gross misnomer, since less than 1% of its nuclear energy content has actually been extracted.

Nevertheless, nuclear nations are engaged with plans to find suitable geological strata for the permanent disposal of this so-called “waste”, rather than to treat this material as the huge non-carbon energy resource that it still is. In Canada a deep geological repository (DGR) is being planned in Precambrian or Ordovician rock, with a potential 22 DGR sites now having been narrowed down to two [12]. Such a Canadian DGR was recently re-estimated to have a lifetime cost $18.3 to $28.4 billion [13], with over $10 billion of that already accumulated in trust funds, tithed from the electricity bills of consumers using nuclear power [14-17].

2. Uranium: Limited by Wastefulness

The uranium finds in various countries of the world suggest that there is an abundance of mineral available for many years. However, unless major discoveries are made in the near future the survival of the nuclear industry as it exists today is again in jeopardy. Even now the world’s economical reserves have plateaued while the cost of exploration has increased four-fold [8].

A simple examination of the uranium reserves drives home the concern more strongly, with Canada faring worse than the world at large. The world reserves of proven, probable, “measured and indicated” resources stand at 6,142,600 tonnes [8], of which the actual “proven and probable” reserves are about half. The current annual production and requirements of fuel for world reactors amount to some 60,000 tonnes per year. This provides enough uranium fuel for between 50 years (proven and probable) and 100 years with less certainty. If nuclear output is increased in the near future due to pressures for carbon emission reductions in relation to climate change, then each of these estimates has to be reduced.

In Canada the picture is not as rosy. Canada's proven and probable reserves of uranium stand at 228,000 tonnes [18], while adding the category of more questionable "measured and indicated" reserves increases Canada's potential uranium cache to 531,608 tonnes [8]. Canada's current requirements for its CANDU reactors are only approximately 2,000 tonnes of natural uranium per year, suggesting well over 250 years of potential reserves. However, the continued profitable exploitation at recent levels of mining and export of almost 16,000 tonnes of uranium annually by Cameco and Orono in 2015 (15,709 tonnes), 2016 (16,541 tonnes) and 2017 (15,467 tonnes)[8], will exhaust Canada's potential uranium reserves within 33 years, or by the year 2053, while the proven and probable reserves of 228,000 tonnes would be exhausted already by 2034.

3. History to the Rescue: Fast-Spectrum Reactors and Recycling

3.1 Fast-Neutron Reactors and Other Small Modular Reactors

The pressure is particularly acute on Canadian uranium fuel reserves. At current rates of mining and exporting, Canadian proven reserves will be gone within 15 years, and even if measured and indicated reserves come on line, they too will be gone in three decades. That is less than the life extension of the CANDU reactors being currently refurbished. Canada therefore faces the prospect of losing its intrinsic uranium fuel independence, its nuclear energy sovereignty, even for its natural-uranium-fueled CANDU reactors, facing reliance on the good will and acceptable economics from other countries for a large portion of its energy budget, especially in Ontario.

The recent emphasis on the introduction of small modular reactors (SMRs) into Canada's nuclear energy mix only exacerbates the prospects of uranium strictures placed on this country in the current scenario. All SMRs require enriched fuel, some as high as 20% fissile U-235, which, as indicated above, requires many times the volume of mined uranium in order to fill the reactor cores with starting fuel. Thus the Canadian uranium reserves would be depleted even faster.

Most of these SMR designs, e.g. NuScale [19], are the equivalent of smaller versions of the existing thermal light water reactors, while some are cooled with molten salt or an inert gas. These all function in a once-through fuel mode, expecting third-party used-fuel disposal for the fuel waste products of the operation.

However, there are several designs that have the capability of maintaining their fissile content throughout their operation. This is especially true if the starting fuel is a mixture of transuranics plus uranium, specifically U-238, that contains sufficient fissile Pu-239 and Pu-241 to achieve neutron equilibrium. This characteristic would require new fissile fuel only at the start of operation, with subsequent fuel replenishment requiring only U-238 at a level equal merely to the fission products produced during energy production in the reactor.

Some of them produce sufficient excess fissile material to be able to provide fuel continuously for CANDU reactors of equivalent power [20].

All such reactors are fast-neutron reactors (FNRs, alternatively also called fast-spectrum reactors (FSRs)) cooled with heat transport media that do not moderate or slow down the nascent fast neutrons that are the result of a fission reaction. Existing design examples would include the 300 MWe PRISM reactor from GE-Hitachi in the USA, the US 100 MWe ARC-100 from Advanced Reactor Concepts, and the 3 – 10 MWe SEALER from LeadCold in Sweden [21-23]. The first two are based on the design of the EBR-2 reactor [5] that operated flawlessly in the USA from 1964 to 1994, being only shut down by act of Congress.

3.2 Mixed Fuel Behaviour in a Fast-Neutron Reactor

As an example of the mixed fuel behaviour in such reactors, the PRISM reactor was modelled theoretically on the basis of the sPRISM design detailed by Dubberly et al. in 2000 [24]. The results are shown in Figures 1 and 2.

It was assumed that the reactor was not limited by the strength of structural materials in the core during irradiation in order to follow the theoretical operational reduction or increase of various fuel isotopes. The end of any fuel cycle in this case was triggered when the level of excess neutrons in the core

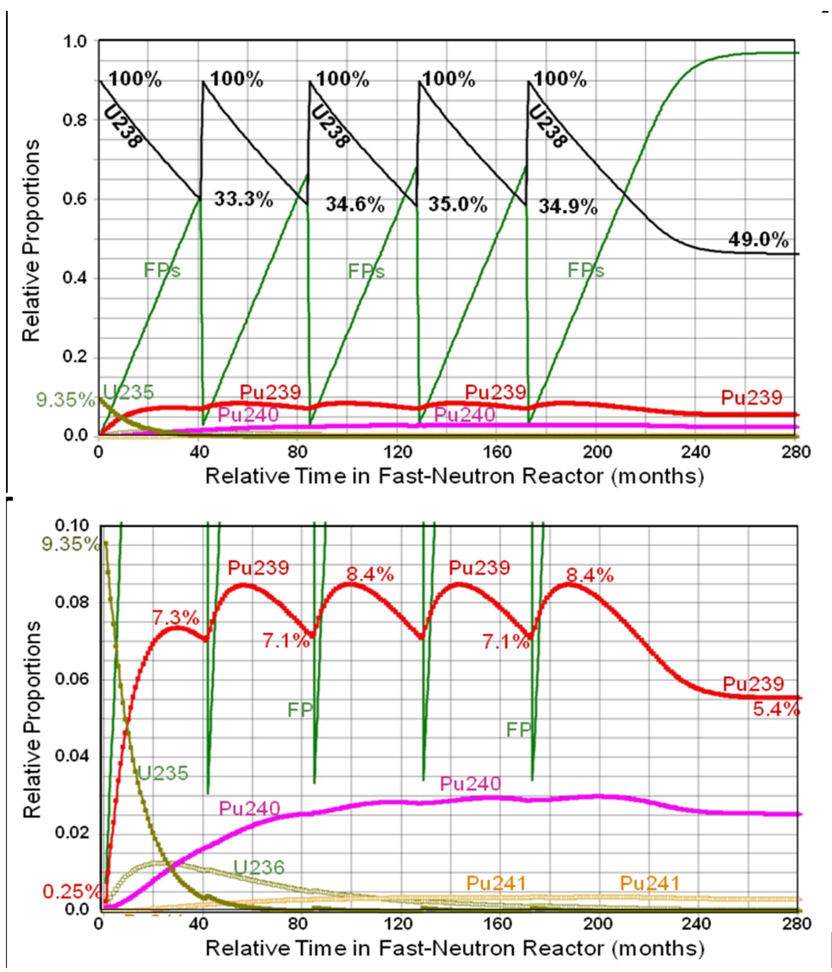

Figure 1. Fuel Consumption and Build-Up of Pu Isotopes in FNR on Start-Up with U235
Fig. 1a (top) Large scale. Fig. 1b (bottom) Expanded scale.

approached zero (a certain degree of excess neutrons is required to control the power level in the reactor core). Under these circumstances the reactor achieved about 35% fuel burn-up (reduction of U-238; Fig. 1a) before fuel replenishment was required.

For the data in Figure 1 the reactor core was loaded initially with uranium enriched with sufficient U-235 (charteuse colour in Fig. 1) to achieve neutron equilibrium in the functioning core. With time the major effects are the decrease of U-235 and the build-up of Pu-239 (red), the latter mirrored by the decrease in U-238 (black) (Fig. 1a). The fission product build-up (green) is virtually linear, indicative of a controlled constant power level.

Lesser effects, more easily seen in Fig. 1b, are the build-up and eventual decrease of U-236 (gray-green) from the absorption neutrons in U-235, and the transmutation of some of the Pu-239 into Pu-240 (purple) and the subsequent creation of Pu-241 (yellow).

The time scale is somewhat arbitrary, since it is inversely related to the power levels chosen.

When the excess neutron levels reached zero, the calculations were reset, e.g. at time =41, 82, etc., with a theoretical extraction of fission products to about 4% and a replenishment of U-238 to the original levels, arbitrarily utilizing used CANDU fuel and adjusting the levels of other isotopes to reflect the addition of the low levels of U-235 and of transuranics in that used fuel. This type of resetting of levels and adjustments corresponding to incoming used CANDU fuel compositions was repeated for all cycles.

No additional fissile components were needed, since the loss of U-235 during the cycle had been compensated by the creation of Pu-239 (and some Pu-241). Moreover, the extraction of fission products reduced extraneous neutron absorption somewhat.

During the first cycle the concentration of Pu-239 initially increases due to neutron absorption and transmutation of high levels of U-238. There was fission of Pu-239 as well, increasing in proportion as levels of U-235 dropped. This is indicated by the continuously decreasing slope of the Pu-239 levels. However, as U-238 levels decrease the creation of Pu-239 decreases as well, with the total amounts of Pu-239 levelling off approximately in mid-cycle and then decreasing as more fission of Pu-239 is needed to keep power levels constant.

At the beginning of the next cycle the replenished levels of U-238 immediately enhance transmutation, with Pu-239 levels rising again to a maximum near mid-cycle before decreasing again with decreasing U-238 levels.

After several such fuel cycles all major and minor actinide levels followed a constant pattern during each subsequent cycle. If no replenishment of U-238 was undertaken then power decreased as neutron equilibrium could not be maintained, and levels of all transuranic isotopes dropped as long as any neutrons were available to split them (right side of Fig. 1, after 4th refuel cycle).

3.3 Fuelling with Extracts of Used CANDU Fuel

Canada has no enrichment facilities for U-235. Therefore Canada would depend on the good-will of other countries to obtain enriched fuel for its energy needs, countries such as the USA, the UK,

France, Russia, China, etc., with excess enrichment capacities beyond their requirements for nuclear weapons.

However, the data in Figure 1 indicates that after several cycles of fuel replenishment, primarily with U-238 in used CANDU fuel, such a fast-spectrum reactor operates effectively by using and maintaining a dynamic equilibrium level of Pu-239 and its higher transuranics, with any fissile fuel used being topped up simultaneously by transmutation from U-238, i.e. no additional fissile isotopes are required for continual operation.

Such starting fuel, effectively ignited and catalysed by transuranics, can be obtained from Canada's stockpile of used CANDU fuel. While the transuranics are at low concentrations in such used fuel, they do not have to be extracted and purified, only provided in more concentrated form within bulk uranium, since a relatively high level of U-238 is needed for transmutation as an intrinsic part of FSR fuel.

Removal from used CANDU fuel of sufficient uranium, a substance with relatively low radiotoxicity, is all that is required to concentrate the transuranics to a suitable level.

Canada's current 60,000 tonne stockpile of used CANDU fuel can furnish enough transuranics to start, and via recycling maintain, FSRs such as the PRISM with a total power level of about 24,000 MWe, more than twice the present nuclear power level in Canada.

Similar to removing only part of the uranium, not all of the fission products (FPs) have to be extracted from the concentrated used CANDU fuel. Fig. 1 indicates that neutron equilibrium can be maintained for operation of the fast-neutron reactor up to FP levels of over 30 wt%. This is due to the fact that at the high neutron energies used in such reactors the neutron absorption by fission products is uniformly low. This is true even for such isotopes as Xe-135. Whereas in the thermal regime Xe-135 has an absorption cross section of 2.8 million barns, the cross section for 100 keV neutrons is only 0.031 barns, and at 2 MeV of nascent fission neutrons it is a mere 0.008 barns [25].

Figure 2 shows the data for the same PRISM-like reactor of Fig. 1 fuelled from the outset with transuranics concentrated from used CANDU fuel rather than using enriched U-235. That is in principle similar to starting after several fuel cycles in Fig. 1.

The linear scale in Fig. 2a has been expanded to a log scale in Fig. 2b to be able to provide detail the behaviour of low-level actinides such as Am-241 and Am-242 on the same graph.

For the first two fuel cycles in Fig. 2 the fuel replenishment is with standard used CANDU fuel. For subsequent cycles the replenishment is with used CANDU fuel in which uranium and fission products have been theoretically extracted over 20-fold, i.e. the transuranics in the used fuel have been concentrated in order to see how such higher transuranic levels affect the reactor.

The immediate effect is the establishment of a new dynamic equilibrium for the transuranics at a higher concentration for the subsequent fuel cycles. Nevertheless, no major disruptions in fuel behaviour or power levels (fission product production) are observed at all, suggesting that such a reactor is rather resilient and forgiving to fuel isotope changes.

Interestingly, as a consequence of the higher added Pu levels, none of the Pu isotopes show the pronounced growth and decrease during the cycle as they did in Fig. 1, but merely decrease from each higher Pu isotope level at the beginning of each new cycle. This suggests that the Pu concentration is higher than needed for operation, turning the reactor from a "break-even" mode to an actinide "burner" regime.

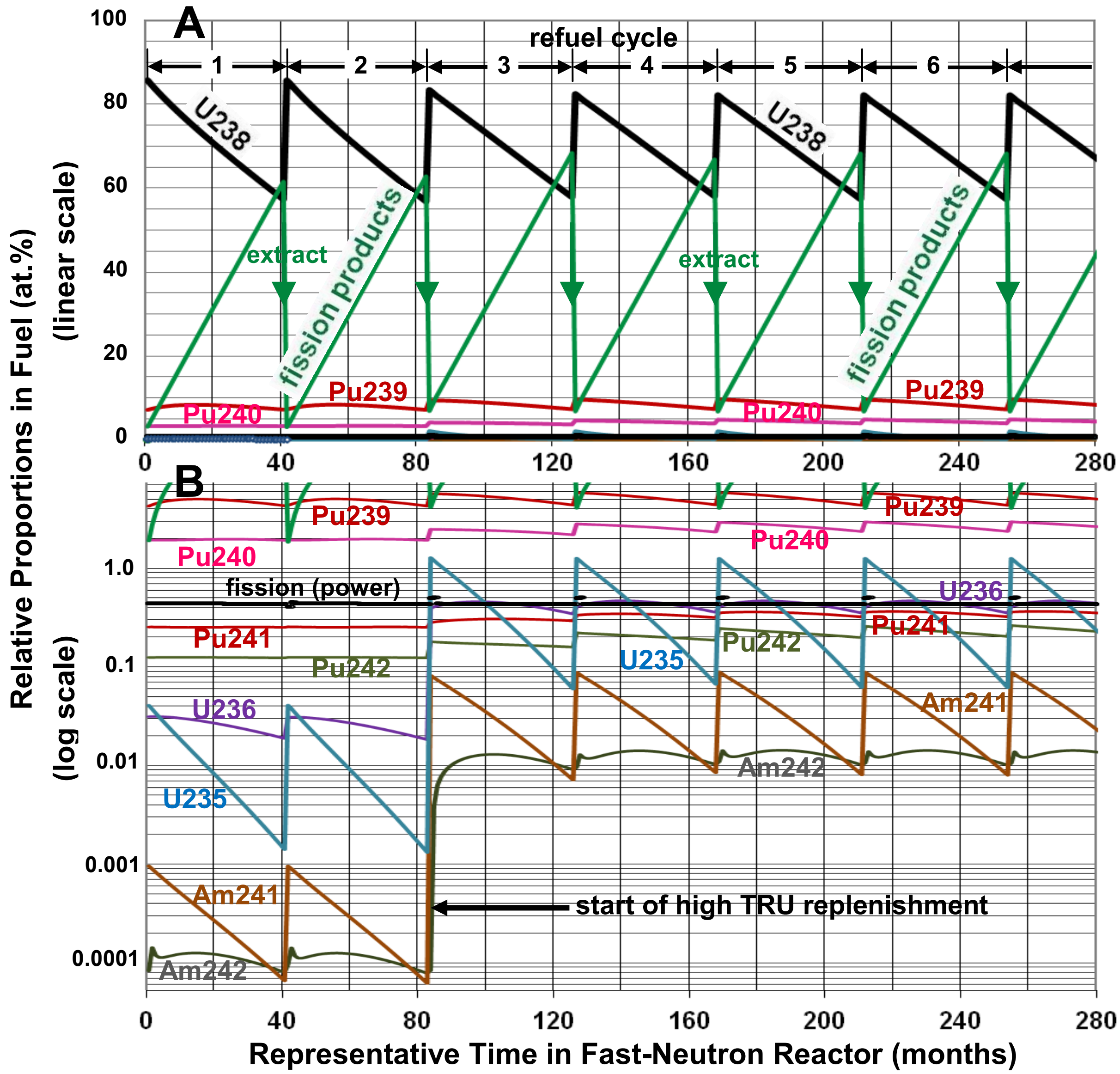

Figure 2. Fast-Neutron Reactor Fuel Behaviour under Replenishment with Two Forms of Used CANDU Fuel
Two cycles have spent FNR replenished with used CANDU fuel directly at months 1 and 41. Subsequent cycles starting at month 82 have FNR fuel replenished with used CANDU fuel from which over 90% of the 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.

One crucial and important observation in Fig. 2, both in 2a and 2b, is the fact that the patterns of behaviour for all of the transuranics from Pu to Am, after an initial adjustment to input concentrations, remain at the same levels from cycle to cycle. There is no increase in such levels. Thus a fast-neutron reactor both in "break-even" and "burner" modes does not produce additional long-lasting transuranics that need to be sequestered safely for hundreds of thousands of years.

Indeed, as an additional advantage, should nuclear power by fission be superseded in future by a better modality, there is a ready exit strategy that relatively quickly reduces the total in-reactor transuranics by suitable deliberate reduction in the number of reactors [26, section 12].

4. The Economies of Recycling, or Not

Can used CANDU fuel be recycled economically and converted to fuel for small modular reactors of the FNR type?

This was examined using three approaches to fuel recycling [26], each of which by design did not produce pure plutonium as a prerequisite to avoid nuclear weapons proliferation concerns:

1) a modified PUREX approach (aqueous methods)
2) an electrorefining approach in molten salt (non-aqueous pyroprocessing)
3) a fluoride volatility approach (non-aqueous, similar to uranium refining)

Table 1 from that study [26] is reproduced here in part.

	Modified PUREX	Electrorefining (Pyroprocessing)	Fluoride Volatility
Capital - overnight cost (land, constr., license, indir. costs + contingencies ~30%)	186.9 M$	80.2 M$	11.5 M$
Total annual cost (OM&A, taxes, 5% interest for 40yrs)	37.2 M$	11.0 M$	5.6 M$
Annual thru-put (bundles)	5000	8050	5000
Cost per bundle	$ 7,432	$ 1,368	$ 1,114

As a financial comparison the current funds in trust for nuclear waste management as tithed from the users of nuclear electricity up to 2017 stood at CAD 10.125 billion [14-17]. The number of used CANDU fuel bundles at that time was 2.771 million [27]. This suggests that the 2017 cost to the user for nuclear waste management via the planned DGR was

$3,654 per used CANDU fuel bundle.

This comparison indicates that used CANDU fuel recycling via at least two methodologies, electro-refining and fluoride volatility, are less costly than burying the used fuel in a planned DGR. Aqueous methodologies, such as a modification of the canonical PUREX method for purifying weapons plutonium, are more expensive.

Furthermore, as a comparison with purchasing fuel enriched with U-235, a fuel charge of a 20-tonne reactor including 4 tonnes of U-235 to provide a 20% enrichment would cost about $150 million

[28, 29, 26 (Table 5)]. Alternatively, the same reactor-full of TRU-enriched fuel via electro-refining of used CANDU fuel from 37,500 fuel bundles would cost about $75 million, [26].

The DGR approach provides no benefit in terms of offering a future non-carbon fuel resource, provides no alternative energy, nor does it alter the trajectory of natural nuclear decay of the used fuel from the current lifetime of several hundred thousand years before its radioactivity is near background levels again. Neither, as an unproven approach, can it provide the assurance that over a hundred years, or even several thousand years, the radiotoxic material will not re-emerge. No such successful DGR exists today; indeed internationally there have been at least three failed attempts caused by either fire, an explosion, water ingress, or political pressure [30-32].

On the other hand, recycling used CANDU fuel provides Canadian reactors with sovereign Canadian fissile materials. It opens the door to re-utilizing our 60,000 tonnes of very slightly used CANDU fuel to provide over 100 times more energy from the same fuel that has served Canada since its inception of nuclear power generation. More exactly, since only 0.74% of the energy has been extracted from that fuel, one can obtain 99.26/0.74 = 134 times more energy from the same fuel.

If one estimates that at constant current rates of Canadian nuclear energy generation the 60,000 tonnes of used fuel would have accumulated in about 40 years, then recycling that same fuel through fast-neutron reactors would create the same non-carbon power for a further 134 x 40 = 5360 years.

That can be considered Canadian energy independence and Canadian energy security for a very long time, even without further Canadian in-ground uranium reserves.

5. Summary and Conclusions

With current rates of mining and exporting of Canadian uranium reserves Canada faces a scarcity or even lack of Canadian uranium fuel for its own non-carbon nuclear energy generation in 15 to 30 years from today, depending on how optimistically our present reserves are calculated. Subsequently Canada would have to rely heavily on the good will of other nations even to fuel its CANDU reactors with natural uranium, never mind fuelling imminent small modular reactors that require enriched fuel for their operation.

However, a change in Canada's nuclear technology would retain Canada's nuclear fuel independence, its nuclear energy sovereignty and an independent Canadian long-term energy security. This change in technology has to include the recycling of Canada's 60,000 tonne stockpile of used CANDU fuel to furnish Canadian fissile fuel via the transuranic components of that used fuel. And it would need to adopt the use of reactors such as fast-neutron reactors that maintain their fissile fuel component indefinitely. Fuel replenishment is then by massively available U-238 that is nearly 100% of the recovered uranium in the stockpiles, or over 99% of any remaining mined natural uranium.

This approach applied to the 60,000 tonne stockpile of used CANDU fuel would provide Canada with over 5,000 years of nuclear energy at current nuclear power output. Alternatively, if such nuclear energy were to substitute also for all of Canada's fossil energy needs in transportation, heating and industry [33] the used fuel stockpile alone could support all of that energy for some 580 years.

As a bonus, since early in this process all long-lived highly radioactive transuranics in the stockpile are removed as starting fuel and converted to short-lived or stable fission products, the need for a long-term deep geological repository for fuel "waste" is eliminated.

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