TWO UNAVOIDABLE ESSENTIAL REQUIREMENTS FOR A CANADIAN LONG-TERM WASTE-FREE NUCLEAR FUTURE: RECYCLING OF USED FUEL AND FAST-SPECTRUM FISSILE-BREEDER SMRs

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Abstract

The development of CANDU technology with natural uranium as fuel 65 years ago was a brilliant step in utilizing nuclear energy. Nevertheless, although offering an industry-best energy yield of 0.74%, this technology still leaves the remaining 99.26% heavy atoms unused, all being valuable non-carbon fuel. Refurbished CANDU technology continues fueling with now rapidly exhausting Canadian uranium reserves. Therefore it is crucial also to adopt two Canadian-developed methodologies that permit the extraction of 130 times more energy from the same fuel, enough energy for millennia: 1) fuel recycling technologies, e.g. pyroprocessing developed at the Argonne National Laboratories (ANL) by Saskatchewan-born Charles E. Till, and 2) fissile-breeding fast-spectrum reactors, such as first built by Kitchener-born Walter Zinn, also at the ANL, to recycle existing used-CANDU-fuel components. Such recycling provides Canadian-grown HALEU-equivalent enriched starting fuel for all SMRs. Crucially, it avoids dependence on foreign fuel suppliers for Canada's future nuclear energy needs. Moreover, it effectively eliminates the million-year radiotoxicity of heavy atoms in Canada's used fuel stockpiles, substituting stable or short-term-radioactive fission products, e.g. useful medium-sized valuable platinum-group-metal catalysts, scarce rare earth atoms, or costly noble gases. Such a beneficial about-turn questions the current design, or even need, of the World's and Canada's deep nuclear fuel waste repositories.

1. Introduction – the Canadian nuclear milieu

Canadian-designed CANDU technology has provided Canada, primarily Ontario, with nuclear electrical power since its beginnings in June 1962 with the horizontal on-power fueling of the heavy-water-cooled and -moderated 20 MWe demonstration reactor, the NPD. This was followed in overlapping time frames of construction and commissioning of the 200 MWe Douglas Point reactor between 1958 and 1968, and the Pickering 500 MWe reactors 1 to 8 between 1966 and 1986 [1-3].

The Bruce Nuclear Generating Station with CANDU reactors of about 800 MWe, was constructed and commissioned in two units. Bruce A with four reactors was built and commissioned between 1971 and 1979, while Bruce B, also with four reactors was built and commissioned between 1978 and 1987. Darlington Nuclear Generating Station with four CANDU reactors of about 880 MWe was built and commissioned between 1982 and 1993.

Two other generating stations with the CANDU-6 design were built, Gentilly-2, at Becancour in Quebec built at commissioned between 1974 and 1983 and decommissioned in 2012. and the Point Lepreau in New Brunswick, built and commissioned between 1975 and 1983. both with a power of around 670 MWe.

Counting the 200 MWe Douglas point reactor Canada built 23 power reactors in a 35-year span, between 1958 and 1993, an average rate of one reactor every 1.5 years.

Currently 19 of the 23 power reactors are still operating, with a total power of 13,750 MWe.

All Canadian reactors combined have produced about 60,000 tonnes of used natural uranium fuel. At about 20 kg of fuel per CANDU fuel bundle, that corresponds to 3,000,000 used fuel bundles, in round numbers.

Since it takes about 6.25 fuel bundles to produce 1 MW-year of electricity in a CANDU reactor [4, p.351], those 3,000,000 fuel bundles have produced 480,000 MW-years of electricity from nuclear energy, or 4.2 billion MW-hours (4.2 trillion kWh).

If, for simplicity, one assumes the cost to the electricity consumer is 10 ¢/kWh, then the gross revenue from nuclear electricity over the years has been $ 420 billion.

A less visible benefit of this nuclear electricity, a virtually carbon-free source of power, is the now normally smog-free environment in Ontario compared to the many smog days prior due to the refurbishment of four Bruce Power reactors that permitted the elimination of coal-fired electricity-generating plants in 2014. An even less obvious benefit from Canada's reactors has been the avoidance of the emission of 3.6 billion tonnes of CO2 into the atmosphere [5].

2. An untapped nuclear potential

Generally overlooked among the obvious and large benefits of nuclear power is the fact that the massive amount of non-carbon energy created so far is the result of consuming only 0.74% of the natural uranium in CANDU-type reactors. Non-CANDU reactors (light water reactors) have exploited even less of the mined uranium, at about 0.5%. Most of the fuel consumed is the relatively rare fissile U-235 isotope that constitutes 0.72% of uranium, augmented in part with fissile Pu-239 and Pu-241 transmuted internally in the reactor core from U-238, the major uranium isotope that is 99.26% of the mined element.

Yet each of these heavy atom isotopes, not just the fissile U-235s, Pu-239s, Pu-241s, but also the fertile Pu-240s, Pu-242s, etc., and especially the huge amounts of U-238, can deliver about 200 MeV per atom, either directly or after transmutation. That's about 130 times more energy from the used CANDU fuel than the 4.2 Trillion kWh already extracted from the current 60,000 tonnes of it.

In terms of electricity at $ 0.10 / kWh this amounts to $ 56 trillion in revenue, or about $ 1.5 million of electricity for every person in Canada [6]. The amount of CO_2 emissions that could be avoided in the process is 480 billion tonnes [5]. That amount of CO_2 is equivalent to about 15 % of the CO_2 currently in the atmosphere of the entire Earth [7,8].

For perhaps a more typical comparison, Alberta estimates that the reserves of oil sands in total amount to 165 billion barrels [9]. Converted to energy, this resource would deliver a total of 274 trillion kWh of heat. The current 60,000 tonnes of used CANDU fuel could deliver 563 trillion kWh of electricity, or alternatively 1690 trillion kWh of heat (arbitrary conversion efficiency of heat to electricity of 0.33). Thus Canada's current used CANDU fuel alone can deliver over 6 times as much energy as all of the oil sands reserves in Alberta.

However, in Canada (and in a number of other nuclear nations) the remaining unused fuel, still containing over 99% of the potential nuclear energy, is slated to be discarded in permanent deep geological repositories, because, exiting from reactors, it is now highly radioactive [4]. Worldwide, an even greater portion of the uranium is languishing above ground at enrichment facilities as U-235-depleted uranium, about 99.8% U-238, often in the form of UF_6.

Much effort and money has been expended in such nuclear nations, including Canada, on planning and the construction of millennia-safe underground repositories, to spare future generations the hazards of the long-term radiation that is part of current used nuclear fuel. Comparatively miniscule monies, not to say no funds nor efforts, have been applied to the alternative of recycling and reusing the used fuel to extract that "lion's share" of its energy, from its U-238. The inadvertent result of this approach will be the deprivation of future generations of a huge resource of carbon-free energy, and ultimately the demise of nuclear power itself in less than 100 years.

Clearly a different mind-set to energy generation from nuclear fuel must be adopted to extract more, ideally all, of the over 99% still-available carbon-free energy in the used fuel.

Such an approach can be and must be part of the changing nuclear landscape in Canada, and indeed in the World. Small modular reactors are being introduced as part of the fleet of Canadian nuclear reactors. The variety of such reactors currently seems overwhelming, ranging from smaller versions of existing light- and heavy-water-cooled reactors, to liquid-salt-fuelled and/or liquid-salt–cooled reactors, to liquid-metal-cooled versions. Fuel types range from the current standard uranium oxide, to uranium metal, uranium salts, and even uranium mixtures with thorium and/or plutonium.

3. A clear and present danger

In general there is no discussion of the efficiency of utilization of the mined fuel, or of Canada's loss of fuel independence or long-term fuel security. Thought processes are guided by fears of not meeting promises of constructing a new SMR on time and on budget.

Reactor physics considerations are at the level of statements of the fissile enrichment requirement and a touting of the percent fuel consumption or burn-up, i.e. fission product creation before fissile fuel replenishment becomes necessary. While these are necessary economic concerns, they are not sufficient to reveal the long-term consequences of such characteristics.

As an example, from a gross operational viewpoint, fuel with an enrichment at the upper limit of HALEU (high assay low enriched uranium) at 19.9% fissile that might result in a "deep burn"

down to 5% before fuel replenishment is needed, appears excellent with a 15% burn-up (15% fission products created).

However, to produce the core fuel at 19.9% U-235 requires close to 40 core volumes of mined uranium since enrichment delivers only 0.5% additional U-235 for each volume of such uranium (i.e. 0.22% tailings left in the process). Thus the 15% burn-up of the "in-reactor" fuel is actually only 15/40%, or a measly 0.375% utilization of the energy potential of mined uranium.

The result of this emphasis that is focused virtually solely on the small 0.72 % U-235 component of uranium as fissile fuel is the "imminent" exhaustion of the World's uranium fuel supply. The World Nuclear Association indicates that Canadian uranium reserves are currently at 270,000 tonnes for "proven and probable" reserves, and a possible high of 670,000 tonnes if less certain "measured and indicated resources" are included [10].

While this seems more than adequate for Canadian needs, which currently are of the order of 2,000 tonnes per year for our CANDU requirements, Canada mines and exports uranium commercially at much higher levels. In years 2015 – 2017 Canada excavated and sold an average of 13,500 tonnes uranium per year [11].

While subsequent years indicated a smaller yield, a recent statement by Cameco in Port Hope (CANDU Fuel Conference, August 2022) indicated with some pride that this year Cameco was processing 21% of the World's 67,500 tonne annual requirement of uranium, or 14,200 tonnes. With the recent rise in the World price of uranium, coupled with the anticipated added uranium demand for SMRs, one would not soon expect a lowering of such a level of mining and export.

The concern with Canadian uranium exploitation of 14,000 tonnes per year is the prospect of exhausting Canadian assured reserves of 270,000 tonnes in as few as 19 years (by 2041), or of depleting the more tentative 670,000 tonnes in 47 years (by 2069). This is of the order of only one operating lifetime of recently refurbished CANDU reactors. Canada would subsequently be dependent on foreign sources of fuel for its own nuclear energy needs.

Fortunately the World reserves are larger, at 6,147,800 tonnes. But even at only the current annual requirements of 67,500 tonnes they too will be exhausted in 91 years [12]. If World nuclear power expands with new SMRs and additional full-sized reactors, World uranium fuel exhaustion will occur sooner. Sadly, new uranium discoveries have not kept pace.

Fuel dependence on foreign nations is a much more imminent possibility of our Canadian future with the introduction of SMRs into Canada. All such reactors require enriched fuel, either LEU (low enriched uranium) up to 5% U-235, or HALEU, up to 19.9% U-235. Canada by historical choice has no enrichment facilities, eschewing the technology generally used for nuclear weapons production in favour of CANDU reactor technology which can use natural uranium as fuel. Therefore even the first such SMR requiring enriched U-235 triggers that fuel dependence.

4. Recycling to the rescue?

Can recycling of used fuel provide an exit out of this dilemma? Such efforts are indeed the only solution, but not the way they are currently applied.

Current recycling efforts, such as those in France which use PUREX (Plutonium and URanium EXtraction, an off-shoot of their military efforts), focus only on extracting and re-using pure plutonium as part of MOX (Mixed OXide) fuel in their existing light-water reactors. As a result they miss the main target: the massive amount of U-238, and its large potential for augmented transmutation with fast neutrons. Not enough U-238 transmution occurs in water- or carbon-moderated reactors with their thermalized low-energy neutrons.

It can be calculated that for a single round of such recycling in thermal reactors the rcycled Pu will increase the heavy-atom utilization by 20%, from 0.55% of mined uranium to 0.65% [13; p.147]. A second cycle of Pu-extraction/recycling increases the yield minimally, to a total of 0.70%, or almost equivalent to a single cycle through a CANDU-like heavy-water reactor which already extracts 0.74 % of the potential nuclear energy. Yet such results pale in comparison to the potential of extracting over 130 times more energy from U-238, i.e. an improvement of over 13,000%, by cycling the used fuel through fast-spectrum reactors (see below).

Recycling is important, but to be efficient it must be coupled with a type of reactor that maintains (breaks even) or even augments (breeds) the fissile complement of its fuel rather than merely consuming that rather vital fuel component. In such reactors the fissile complement acts somewhat like a catalyst. The fissile isotopes, while providing energy via fission, are also the potential providers of sufficient numbers of fast neutrons that transform U-238 into new fissile components at a rate at least as fast as those isotopes are consumed due to fission.

As is shown below, this does not occur in any existing thermal reactors fuelled with a uranium concentration as low as 0.72 % U-235 of natural uranium, with uranium enriched to 5 % U235, to 20% HALEU, or to the higher military U-235 concentrations used in submarine reactors.

Even for fast-spectrum reactors, maintenance and augmentation of fissile components is not a given. Those characteristics depend on a sufficiently high proportion of fertile U-238 in the fuel.

Thus both components, recycling and the use of fast-spectrum reactors, are crucial and necessary, but they are also sufficient. Those two interrelated technologies can assure Canada's continuing nuclear energy independence and sovereignty even without enriched U-235.

5. Canadian Enriched Fissile Fuel

Recycling of Canada's current 60,000 tonnes used CANDU fuel can provide about 240 tonnes of transuranic actinides (TRUs) that were created in the fuel during its sojourn through the CANDU reactors. Those actinides contain about 70% fissile TRUs, or 170 tonnes [4, p.341], that can serve as starting fissile fuel for all SMRs to a combined total power of close to 24,000 MWe.

Such TRUs can be extracted in impure form (see below) that still results in a large enough fissile enrichment while at the same time preserving the non-proliferation aspect of the current used CANDU fuel. The CANDU-produced isotopic composition of plutonium in commercial used CANDU fuel is not suitable for weapons production, and is further diluted by extraction via pyroprocessing (see below) with other TRU elements (Np, Am, Cm) and with fission products.

However, at only 170 tonnes such an accessible Canadian fissile reserve is a limited quantity. As a cautionary example, a single thermal CANDU reactor fuelled with about 100 tonnes natural

uranium consumes the equivalent of 0.74 tonnes of fissile material annually. Twenty such CANDUs would use up the 170 tonnes of fissile TRUs in about 11 years. SMRs are smaller, but the equivalent limitation applies, if such SMRs are thermal reactors.

Certainly, continued operation of our CANDU reactors and creation of used fuel will slowly increase this potential 170 tonne amount of fissile transuranics. But without maintaining this fissile resource Canadian fuel sovereignty and long-term fuel security would still be in jeopardy.

6. Choice of reactor

The solution to overcoming this limitation is an SMR, or larger reactor, that can internally maintain or even augment (breed) the fissile content of its fuel charge.

The main criterion currently assigned to reactors is the controlled continuous creation of energy, i.e. neutron equilibrium in the face of neutrons absorbed by fuel, by structural elements, by coolant/moderator and by control elements in the core, balanced by neutrons created due to fission of fuel isotopes.

Transmutation of fertile fuel elements to new fissile fuel components is in general not considered a positive criterion, since it lowers the immediate number of neutrons available for energy production by fission, even if after about 3 days (U-238 cycle) it results in an increased number of fissile Pu-239 fuel isotopes, or after about 30 days (thorium cycle) in some fissile U-233s.

While other factors go into the choice of a reactor, here only the interaction of neutrons of different energies with different compositions of fuel isotopes of the uranium cycle in the reactor core will considered.

7. Creation of new neutrons

The primary concern, sufficient neutrons or more for a prolonged yield of nuclear energy, can be represented by the ratio between the new neutrons created by fission and the neutrons used by the fission process and by all other absorptions in the core and losses from the core. This is the New Neutron Ratio (NNR), given in equation 1.

$$\mathbf{NNR} = \frac{\mathbf{N_5 \times \sigma_5(n,f) \times \overline{\nu_5} + other\ fission\ events}}{\mathbf{N_5 \times \sigma_5(n,f) + N_5 \times \sigma_5(n,\gamma) + N_8 \times \sigma_8(n,\gamma) + other\ absorptions/losses}} \quad (1)$$

where $\mathbf{N_i}$ is the concentration of a particular isotope "**i**" (the subscripts 5 and 8 in Eq. 1 refer to U-235 and U-238 respectively), $\boldsymbol{\sigma(n,f)}$ and $\boldsymbol{\sigma(n,\gamma)}$ are the fission and radiative capture cross sections [14] for individual fuel isotopes or other absorptive isotopes in the core, and $\overline{\nu}$ (nu-bar) is the neutron yield per fission.

For pure U-235, the new neutron yield per fission is 2.44 at thermal energies. However, since U-235 also absorbs neutrons to become U-236, not all neutrons are available for fission, and the NNR for the pure isotope is reduced to 2.09. This NNR for U-235, or for any pure isotope at thermal energies, is also known as the thermal fission factor, or more generally as the neutron reproduction factor for the isotope. When the absorption of structural core components,

coolant/moderator and neutron losses from the core are considered for individual reactor cores, an even smaller percentage of neutrons results in fission, and the NNR decreases further.

For the CANDU reactor using natural uranium (0.72% U-235) at thermal energies the NNR becomes 1.34. This is still sufficiently high above neutron equilibrium (NNR = 1.0), that losses of neutrons to used fuel in neighbouring fuel channels are acceptable, changes in power levels are accommodated, and so is a slow decline in the concentration of U-235 during on-power operation of over a year. Only then does refuelling with fresh natural uranium become necessary.

If it is desired to achieve or maintain the NNR above 1.0 for longer, the concentration of the fissile U-235 can be increased by U-235 enrichment, as is necessary in pressurized and boiling water reactors due to a greater absorption of neutrons by the light water coolant/moderator used.

The NNR, calculated above at thermal energies, can be determined using Eq. 1 for any neutron energy, since the relevant neutron parameters are available [14]. Figure 1 shows the obvious, that at thermal energies (0.025 eV), and also up to neutron energies of 1.0 eV, thermal reactors such as the CANDU using 0.72% U-235, as well as reactors light water reactors with fuel at U-235 enrichment from 2.0% to HALEU near 20% U-235 are all capable of operating with NNRs above 1.0 (with a controllable positive reactivity).

However, at higher neutron energies, in the regime of fast-spectrum reactors (e.g. between 100,000 eV and 1 million eV), a functioning reactor core (NNR ≥ 1.0) is possible only if the fissile fuel enrichment is around 8% U-235 or higher (Fig. 1; black and purple solid symbols).

8. Replenishment of fissile isotopes

In all operating reactor cores that contain fertile fuel isotopes such as U-238, the ambient neutrons will transmute some of those fertile isotopes into fissile fuel elements. The degree to which this occurs depends on the relative fuel isotope composition and the corresponding

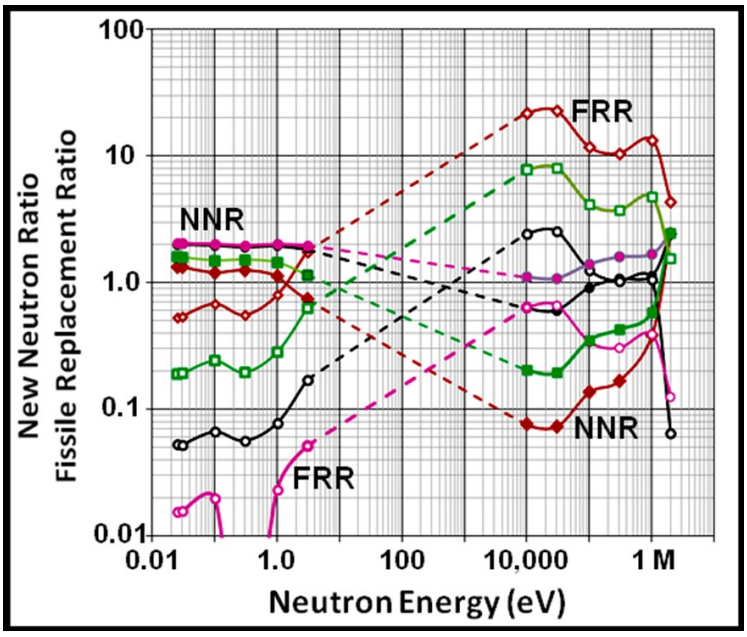

Figure 1. Dependence of New Neutron Ratio (NNR; closed symbols) and Fissile Replenishment Ratio (FRR; open symboles) on energy of interacting neutrons and on the relative concentration of fissile U-235 in uranium fuel composition. CANDU reactor: 0.72% U-235; PWR: 2.0%, 8.0% and 20.0% U-235. Dotted lines indicate the resonance region (not calculated).

radiative capture cross sections at the specific neutron energy. This effect is solely a function of the fuel isotopes. The effect can be captured by the ratio between proportion of neutrons used for transformation of fertile isotopes and the proportion used in fission and transformation of fissile isotopes, to provide a Fissile Replacement Ratio (FRR) as shown in Equation 2.

$$\mathbf{FRR} = \frac{\mathbf{N_8 \times \sigma_8 (n, \gamma) + other\ fertile\ transmuted}}{\mathbf{N_5 \times [\sigma_5 (n, f) + \sigma_5(n, \gamma)] + other\ fissile\ consumed}} \qquad (2)$$

The meaning of the symbols is the same as for equation 1, above.

For a CANDU reactor with a U-235 concentration of 0.72% and U-238 at 99.28% the FFR initially is equal to 0.54. During operation of the reactor the FFR rises, since the level of U-235 decreases, while the rate of production of Pu-239 via transmutation from the high levels of U-238 remains relatively constant. The overall result at the end of the CANDU fuel cycle is an integrated FFR equal to the conversion ratio (CR) of the CANDU or about 0.7.

It is obvious from equation 2 that a core fuelled with a greater enrichment of U-235 would have a lower rate of fissile replacement by transmutation, since a greater proportion of the available neutrons is used in fission. Thus for a PWR with an enrichment of 2% U-235, the fissile replacement ratio would drop to 0.19 at thermal neutron energies. This effect is shown above in Fig. 1 above (open symbols) for different U-235 enrichments at the low energy extreme of the curves.

What is remarkable in Fig. 1 is that at low neutron energies one can construct an operating reactor at virtually any reasonable concentration of U-235, but there seems to be no combination of fuel isotopes that permits the replacement of U-235 with enough fertile U-238 transformed to fissile Pu-239.

Curiously, at high neutron energy almost the reverse is true. This can be explained by a large change in the relative fission and radiative absorption cross sections of U-235 and U-238 with a change in neutron energy.

As an example, at 8% U-235 and 92% U-238 the combination of cross sections at thermal energy delivers a relative fissile replacement of 92 x 2.72 barn, or 250 b, and a relative consumption of fissile U-235 of 8 x (600 b + 100 b), or 5,600 b, for an FRR of 0.045. This reactor would be a major “burner” of fissile fuel.

However, at a neutron energy of 300,000 eV the fissile replacement is 92 x 0.117 b, or 10.8 b, while the relative consumption of fissile U-235 has a value of 8 x 1.26 b, or 10.1 b, resulting in an FRR of 1.07. Such a reactor would augment the fissile fuel content in the core during operation.

It is important to note that for “ideal” operation both ratios, the NNR and the FRR, should be equal to or greater than 1.0. An NNR < 1.0 shuts down the reactor, while an NNR ≥ 1.0 permits a longer controlled operation. An FRR > 1.0 not only permits a greater utilization of the large U-238 isotope component of uranium, but also delivers an excess of fissile material that permits the fuelling of additional reactors that require enriched fuel.

Figure 1 indicates that such an "ideal" scenario for uranium-fuelled reactors occurs only in the regime of fast-spectrum neutrons, and only in a limited range of fuel enrichment. Too low an enrichment results in a non-functioning reactor (NNR < 1.0). Too high an enrichment results in a "burner" of fissile fuel (FRR < 1.0), one that neglects the energy value of U-238.

9. Recycling

However, even with ideal fuel isotope concentrations and the use of fast-spectrum neutrons to maintain a constant or increasing concentration of fissile components upwards, the benefit of efficient fuel utilization will not be achieved without recycling of the fuel. This limitation is imposed by the accumulation in the fuel of neutron-absorbing fission products, which have to be removed periodically and replaced by an equivalent mass of fertile fuel isotopes, e.g. U-238.

In the absence of recycling there will be continuous reliance on fresh U-235, which even in natural uranium is a rapidly decreasing resource for Canada, and in enriched form is only available from foreign nuclear weapons states.

But can such recycling be accomplished economically, cleanly and with minimal proliferation concerns?

There are three main approaches to recycling, historically: aqueous PUREX processing mentioned earlier, fluoride volatility approaches, and electro-refining in molten salt (pyroprocessing). Cost estimates for each are shown below. The first two were developed in large part for the production of nuclear weapons and other military uses, directly or indirectly; but civilian commercial users have benefitted from excess military capacity. The third, pyroprocessing, was specifically developed for recycling used fast-spectrum-reactor fuel directly at reactor sites, addressing costs, rapid turn-over, as well as proliferation concerns [15; p.167ff].

9.1 PUREX

Plutonium Uranium Extraction is an aqueous procedure designed to extract pure plutonium, specifically weapons-grade Pu-239 from military reactors. The plutonium from the used fuel of civilian commercial reactors is a non-separable plutonium isotope mixture of Pu-239 through Pu-240, Pu-241 and Pu-242 which makes even pure plutonium unsuitable for nuclear weapons. Even so, since water is an excellent moderator of neutrons, the aqueous processing has to be sufficiently dilute to avoid inadvertent criticality concerns. As a result the PUREX facilities are large and expensive and leave behind large quantities of radioactive working fluids for subsequent treatment and disposal. It is therefore an expensive technology, even in many modified forms, e.g. GANEX, UREX, etc., and it is often used as a *bête noire* to squelch any overtures towards recycling, particularly since major disposal requirements are not eliminated.

On the positive side, a number of the chemical purification steps of the procedure are familiar to CAMECO in Canada, being used as part of its purification of uranium.

9.2 Fluoride Volatility Methods

The constituents of used fuel can be separated in part by chemical conversion into compounds which individually or as small groupings become solid, or liquid, or gas. As a result they can then be physically isolated on the basis of their change of state.

For example one of the major steps involves the partial or complete fluorination and volatilization of uranium, the transuranics, and some of the fission products, depending on the temperature. A reduction in temperature then causes the condensation or sublimation of various individual constituents at different temperatures. For instance, upon hydrofluoridation at 400°C followed by flame fluorination of a mixture of used fuel components at 500°C, AmF_3 and CmF_3 will precipitate as solids, followed by condensation of BrF_3 on cooling to 100°C; then a mixture of UF_6, PuF_6, and NpF_6 separates out in liquid form at 40°C, and at -15°C MoF_6, IF_7, and WF_6 precipitate out as solids.

While Canada has no experience with such a total system, CAMECO uses hydrofluoridation and flame fluorination to produce pure UF_6 for export to U-235 enrichment facilities.

9.3 Electro-refining in molten salt (pyroprocessing)

Pyroprocessing was developed at the Argonne National Laboratories in the 1980s specifically for the used metal fuel of the EBR-2 fast-spectrum reactor that operated from 1964 to 1994. In the electrolytic refinement the used metal rods, chopped into short cylinders, fill a wire mesh anode basket. Molten chloride salts of sodium, potassium and lithium, with small amounts of salts of the heavy metal actinides are used as conducting electrolyte. A current at about 1 Volt dissolves the used fuel metal and transports it to two different cathodes. An iron cathode is used to plate out the bulk of now very pure uranium, while a liquid cadmium cathode is used subsequently to plate out a mixture of the transuranics and the remaining uranium. Fission products (FPs) accumulate in the liquid electrolyte, and are periodically removed chromatographically by passing the FP-laden electrolyte through Zeolite columns. The cleaned electrolyte is returned to the electro-refiner. The fission products can be eluted from the Zeolite columns and put into chloride form for storage and eventual refinement as their radioactivity diminishes in days, weeks, months or a few years. Only two significant FPs, Cs-137 and Sr-90, have longer half-lives, of 30 years each (however, these radioactive elements are also considered valuable as industrial gamma sources and long-term thermoelectric devices in arctic or space applications).

10. Recycling Costs

Table 1 (below) shows the cost of recycling by the three technologies above, normalized to the cost of treating a single used CANDU fuel bundle. It is clear that if an inappropriate methodology is chosen, e.g. PUREX, or PUREX modified to avoid purification of plutonium, the cost is not only about twice that of disposition in a Deep Geological Repository (DGR), the radioactive residues still require accommodation in a DGR.

The other two technologies are both less costly than a DGR by a factor close to three. Of the two, electro-refining is particularly suitable for metal fuel, as used in the EBR-2 reactor and proposed for the 100 MWe ARC-100 currently being considered by NB Power in New Brunswick, and for

Table 1. Used CANDU fuel recycling cost estimates in comparison to funds committed for a Deep Geological Repository

Methodology	**Cost per used CANDU fuel bundle**	
	Our work	**Others**
Modified PUREX (gold standard; military origin)	$ 7,430	$ 8,460*
Fluoride volatility methods	$ 1,110	$ 1,730**
Electro-refining (pyroprocessing)	$ 1,370	$ 1,690*
Deep Geological Repository	**Funds per CANDU fuel bundle**	
	$ 3,654	

* [15; p.274ff], **[16]

fuel in the 300 MWe PRISM fast-spectrum reactor by GE-Hitachi. Electro-refining takes the impure used metal rods directly as input and delivers a pure metal as output to be recast into new fuel rods.

Metal fuel in the EBR-2 reactor has been shown to permit a burn-up of about 20% while maintaining its high fissile content. This is particularly attractive for recycling, since recycling requires the treatment of the total fuel load. Compared to a 5% burn-up, the cost of recycling fuel with a 20% burn-up would be reduced by a factor of 4.

11. Summary

This short examination of a potential future for Canada's nuclear industry has indicated a scenario which maintains Canada's nuclear energy sovereignty and fuel security far into the future.

The prospect requires a coupling of two interrelated processes. One is the operation of energy-producing fast-spectrum reactors (FSRs) in a mode that as a minimum maintains the fissile fuel content of the core by transmutation of constituent fertile fuel isotopes. The second is a suitable recycling methodology whose initial function is the electrolytic separation of used CANDU fuel into pure fractions of uranium and of fission products as well as of impure proliferation-resistant fissile transuranics for reactor starting fuel. The continuing recycling function is the periodic electrolytic removal of neutron-absorbing fission products (FPs) from subsequent used fuel. Their replacement with an equivalent mass of fertile isotopes, primarily with the abundant U-238, effectively reconstitutes the FSR fuel for subsequent fuel cycles.

Suggested is the use of fast-spectrum reactors fuelled with conducting metal uranium-plutonium rods. Such dense fuel not only fosters high efficient burn-up, but its electrical conductivity also facilitates recycling and purification by electro-refining. Virtually the only residue of the process consists of fission products with their relatively short radioactive half-lives.

The economic analysis indicates that the cost of such an approach is not only about one third of disposing of Canada's used fuel, but that it also delivers over 130 times as much energy as has been extracted from the used fuel over the last 60 years. The already existing used fuel can therefore power all of Canada for centuries.

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