

A Partial Critique
of the
MoE
Feasibility Study on the Recycling of Used CANDU Fuel, April 2016
by the
Canadian Nuclear Laboratories

http://ontarioenergyreport.ca/pdfs/MOE%20-%20Feasibility%20Study_Used%20Fuel%20Recycling%20-%20June%202016.pdf

Used Fuel Recycling:
An Opportunity for the Economy, the Environment, and Energy
using missed savings of \$ 30 to 60 billion

July 15, 2016

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The Feasibility Study

outlines options for partial recycling 103,000 tons of used CANDU fuel
but

- 1) misses the possibility of eliminating a long-term DGR, saving an accumulating \$ 20-40 billion
(with \$ 9 billion already in trust)
plus a further \$ 10-20 billion in foregone projected recycling disposition costs
- 2) overlooks the possibility of continuous recycling to consume all 103,000 tons with an
additional long term yield of over 960,000 TWh in non-carbon electricity
worth > \$ 130,000,000,000,000 (\$ 130 trillion) over 8000 years

These opportunities are detailed in the Critique

Feasibility Study on the Recycling of Used CANDU Fuel, April 2016

A Partial Critique: Summary

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The CNL Feasibility Study on the Recycling of Used CANDU Fuel, April 2016, commissioned by the Ontario Department of Energy (herein called “the Study”) provides a reasonable basis for discussion of three approaches (Options) to the further use of part of the 99% energy content in the used fuel. The Study provides an estimate of the costs of recycling and disposal of various components of the chosen technologies. And the Study itemizes the components that the study team on superficial examination considers necessary to be sequestered permanently in a currently proposed deep geological repository (DGR) for high level long-term CANDU fuel “waste” as defined by the federal Nuclear Fuel Waste Act of 2002.

There are several major flaws in the Study, plus too many minor errors and omission to itemize here.

Section 3.1 of the Request for Bids by the Ontario Ministry of Energy asks for “a feasibility study to identify candidate technologies which could provide incremental savings to Ontario relative to the current strategy for management of used CANDU fuel”.

1. The Study does not examine whether it is possible to reduce the use of the DGR or to eliminate it entirely, with corresponding savings in cost on the basis of the different approaches to recycling the used CANDU fuel. Nor does it attempt to optimize combinations of technologies that would have an impact on reducing the cost of recycling.

The lack of a critical examination of the need for a DGR in each Option is the major short-coming of the Study. As a result, major savings and a huge potential for non-carbon energy are missed.

2. The Study also does not invoke exit strategies from nuclear power at the end of the sequences of reactor generations, strategies that would minimize the radioactive fuel components such as the in-reactor fuels that would still need sequestration. Instead, the simple solution chosen is the general disposal in a DGR that is assumed as a *sine qua non*.
3. Furthermore, by choosing fast-neutron reactors with conversion ratios (CRs) of 0.5, the Study is blind to the potential of continued use of what it calls RU (reprocessed uranium), the major 90,000 ton pure uranium component of the used fuel, which can be consumed in fast neutron reactors with CRs of 1.0. Instead, the Study team disposes of the RU component entirely unnecessarily in the proposed DGR. Their choice results in an electricity yield of only 5400 TWh, missing a 175-fold greater 960,000 TWh of electricity from the RU component of the fuel.

This examination of the Study summarizes the study findings, and analyses some of them in graphic form for an augmented understanding.

It calculates the amounts and evolution of various fuel component under Option 1 (“once-through” recycling in oxide-fuelled CANDUs) and Option 3 (“once-through” **plus** continuous recycling in metal-fuelled fast-neutron reactors (FNRs)), options which differed most in fuel processing methodologies (aqueous versus non-aqueous) and in reactor character (thermal neutrons versus fast neutrons).

It specifically examines the potential impact on the need for a long-term high-level deep geological repository (DGR).

In brief: the examination, and analysis beyond the Study, show

1. that reuse of the fuel in any reactor extracts a further portion of its potential nuclear energy. The second “once-through” approach gains only an additional 30 % in CANDUs, whereas continuous cycling in FNRs with conversion ratios (CRs) of 1.0 can gain a factor greater than 100.
2. that only a simple 2nd once-through use of the fuel, as done in France, requires a DGR for the resulting used fuel residues, whether such a single additional fuel pass is carried out in a thermal reactor or in a fast-neutron reactor. Savings are achieved in the volume of the resulting used fuel versus the current used fuel, but in requiring a DGR either way the cost savings are relatively minor, since volume costs are a relatively small component of the total DGR effort.
3. When continuous recycling is carried out with fuel passed through an FNR only two used fuel components constitute the residue: fission products and RU (pure reprocessed uranium). While these are disposed of in a DGR by the Study team, it is argued here that these component require no permanent disposal in a long-term high-level DGR.
4. Nuclear exit: For an FNR with a CR=0.5 a used fuel amount from within the reactor core of 300 tons results from 32 reactors (versus ~1200 tons for 12 CANDU reactors). This is disposed of by the Study team in a DGR. However, a simple nuclear exit expedient of cycling this fuel in successively smaller numbers of FNR reactors, and separating the RU component, can reduce the long-term used fuel to that for a single reactor (~3 tons, or a volume of a 52 cm cube).
5. Converting the reactors with CR=0.5 to a fuel configuration with CR=1.0 before all the TRUs are consumed, permits the use as fuel of the 90,000 ton RU component, resulting in a pure FP residue plus an additional electricity yield of 960,000 TWh. Again, it is argued here that no long-term DGR is required for the resulting fission products.

Summary concluding statement:

Continuous recycling of used CANDU fuel in FNRs (fast-neutron reactors) with a conversion ratio of 1.0 creates over 175 times more non-carbon energy from that fuel and avoids the need for a permanent long-term DGR. The latter saves \$ 20 - 40 billion in DGR costs up front, while FNRs with CR=1.0 will create an additional gross revenue of \$ 127 trillion at \$ 13 billion per year.

A Pragmatic Approach: an Early Start

The Study makes it clear that many advantages are to be gained by Ontario's early entry into recycling of used fuel. To quote (p. 1-7 (18)):

1. *A reliable source of baseload nuclear electricity, without further depletion of the Canadian uranium reserves.*
2. *Potential for less long-lived and/or less long-term radiotoxic nuclear waste destined for disposal.*
3. *Growth in Ontario's nuclear knowledge and supply chain, including potential for international exports of certain components produced by the Canadian supply chain.*
4. *Opportunities for Canada to participate as a leader/world expert in international deployments of similar recycling systems.*

Research and development opportunities with the potential of export of know-how and materials would be additional advantages.

An entry into this area requires a commitment both in terms of reactor technology and associated recycling capabilities. The Study's choices for reactors and associated recycling technologies are two systems:

1. the future French ASTRID 1500 MWe reactor design using oxide fuel, associated with an aqueous processing technology called GANEX. The Study indicates that the technology will be available around 2050.
2. the current US GEH PRISM design of 380 MWe using metal fuel, associated with non-aqueous pyro-electrolytic refinement (pyroprocessing). Contrary to the Study estimate, GEH indicate that, post-paperwork, the factory-built modular PRISM can be on site and ready in 36 months. It has already passed Pre-application Safety Review by the US Nuclear Regulatory Commission (NUREG-1368).

Choice #1 involves an immediate large commitment, since not only is the reactor large, but any aqueous processing of enriched fuel requires large facilities to achieve dilutions required to counteract the neutron-moderating influence of the aqueous (hydrogen-containing) working fluids. It is *per force* expensive, and will remain so, and does not offer economies gained from a learning curve of many units.

Choice #2 offers entry with a minimum up-front cost by starting with a small number of smaller reactors and achieving economies of a learning curve with the addition of later units. Similarly, pyroprocessing is of necessity a batch process with compact modular electrorefiner units (see images overleaf) and equally small fuel fabrication units, etc., that can be replicated as needed for further reactors. The compactness in part is predicated by the non-aqueous technology that permits work with more concentrated enriched fuels.

The additional advantage is that with the reactor ready for licensing review, preparations can start immediately to offer Ontario an early advantage, worldwide.

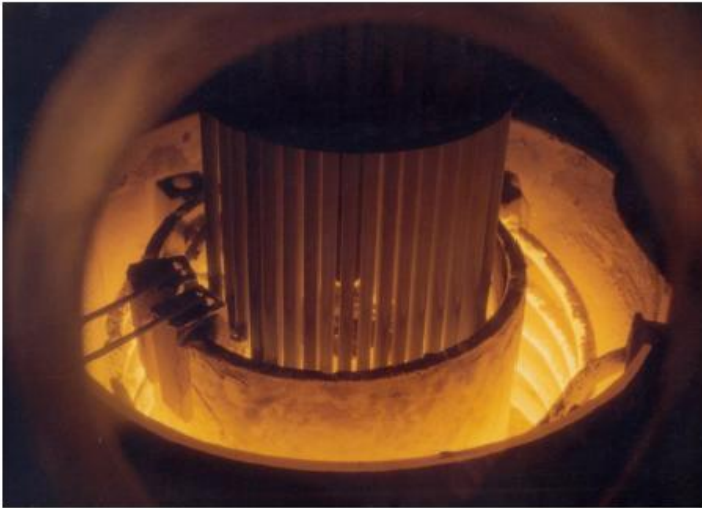
Savings:

Either choice of fast-neutron reactor saves the real expense of a long-term DGR (\$20-40 billion), and also saves the projected expense of the preparation and burial of used fuel components in such a facility (estimated to be \$ 11-35 billion in the Study; see page 7).

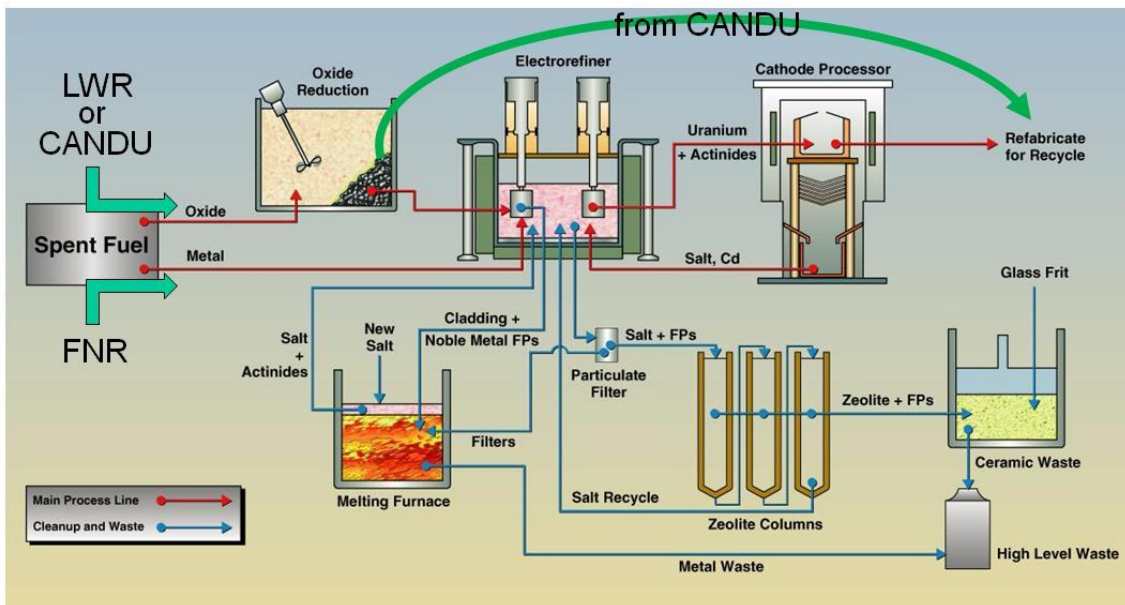
(A) Modular Pyro-Electrorefiner Unit
Argonne National Laboratories
Chicago



(B) Vacuum Injection Fuel Casting Unit
(The injection tubes are about 1 cm in diameter)
Idaho National Laboratories, Idaho Falls



(C) Pyroprocessing Flow Sheet
Courtesy of Yoon I. Chang
ANL Chicago



Cost Savings on Reprocessed Uranium (RU) and High Level Waste (HLW) Fission Products

It is assumed that the non-carbon production of electricity from uranium will require nuclear reactors of one kind or another. Therefore, while such reactor costs are very important, purchase/construction, refurbishment and eventual decommissioning are not part of the consideration of expenditures or savings between recycling costs and/or disposal costs of existing and future used fuel.

A relevant comparison between the current approach of disposal in a deep geological repository (DGR) at a cost of \$20-40 billion versus recycling of the used CANDU fuel through fast-neutron reactors (FNRs), Option 3, is shown in Table A, as modified from Table 1-3, Table 11-7 and Table 11-8 of the Study.

Savings 1: \$ 20 Billion

Table A. Costs and Savings between Study Option 3c (revised) and Deep Geological Repository (DGR)

	RU*	RepHLW** mid-range	Total
	(M\$)	(M\$)	(M\$)
Reference (DGR)	0	24,670	24,670
Option 3c (FNRs)	11,205	23,633	34,838
Option 3c (revised)	1,600***	3,000 [‡]	4,600
<u>Savings via Option 3c (revised)</u>			<u>\$ 20,070 million</u>
* RU = Reprocessed Uranium			
** RepHLW = reprocessing High Level Waste (fission products, small traces of U and TRUs)			
*** conversion to oxide only; no DGR			
[‡] shielded storage for FPs (300-year above-ground containment)			

Savings 2: \$ 130 Trillion (9.2 million per person in Ontario)

Preservation of Reprocessed Uranium (RU) as Fuel in Fast-Neutron Reactors

Reprocessed uranium (RU) can be used as sole fuel replenishment in fast-neutron reactors with conversion ratios of 1.0 (these reactors maintain their “catalytic” fissile content). Therefore the 90,000 tons of RU extracted under Option 3c (revised) should not be buried in a DGR as proposed in the Study, but be used instead to produce non-carbon energy of more than

960,000 TWh electricity worth **\$ 130 trillion** (at a current **\$ 15.4 billion/year**)

Analysis

The Study is a good compendium of the technologies that would make it possible to recycle used CANDU fuel, to extract more of the energy in that used fuel and to diminish the long-term radiotoxicity of that fuel. In the Study any and all remaining fuel components are disposed of in an already planned deep geological repository (DGR) costed elsewhere at \$ 20-40 billion (see <https://www.nwmo.ca/>).

A major flaw of the Study is its lack of a greater discussion of the elimination, diminution, or change in character of such a DGR in light of the different scenarios or options. The assumed use of such a DGR, whose purpose is the permanent sequestration of very slightly used CANDU fuel, precludes the realization of the very positive impact of that non-carbon energy resource on the economy of Ontario.

This positive potential is highlighted in the brief descriptive discussion of Options 1 and 3 below (Option 2, using oxide-fueled FNRs, is not discussed in detail here, since it is very similar in principle to Option 3).

In brief, that analysis shows that a long-term permanent DGR is only needed if the used CANDU fuel is not recycled continuously. With continuous recycling through fast-neutron reactors, the \$20-40 billion DGR becomes unnecessary. Instead of using a DGR, the separated fuel components should be placed in readily accessible secure retrievable storage and in equally secure shielded storage. Those components are comprised of

- 1) extracted pure uranium (called RU in the Study),
 - 2) fission products (FPs),
 - 3) a mixture of all transuranic actinides with some uranium (TRUs),
- and
- 4) zirconium cladding from fuel bundles.

Components 3 and 4 would be of immediate use as starting fuel and as fuel replenishment in the fast-neutron reactors discussed in the Study. Component 1, RU, is a long-term fuel replenishment of very low radioactivity/radiotoxicity. Component 3, the FPs, decays in storage to valuable atoms and minerals that include rare earths and immediately non-radioactive platinum group metals. (The radiotoxicity of some of the constituents will be touched on below.)

Basic Fuel Use Data from Options of the Study.

It is clear from the outset that further use of the irradiated fuel will bring with it a major increase in energy yield, since every heavy fuel atom that is fissioned, be it uranium or the transuranic actinides from Pu to Am and beyond, will provide about 200 MeV energy. It is the type of reactor, using either thermal (slow) neutrons or alternatively fast neutrons, that determines how much of the fuel can be turned into energy and how much of a long-term radioactive residue remains. It is the modes of fuel processing that not only make it possible to recycle the used fuel, but that also bring with them a greater or lesser environmental risk and also different potentials for nuclear proliferation. These aspects are summarized in light of the data presented in the Study, and are augmented where data is lacking.

In this analysis thermal and fast reactor approaches in Options 1 and 3 will be discussed first, adding a missing nuclear exit and a discussion of the long-term advantage of recycling for millennia. This is followed by an exposition of the need for a DGR, specifically highlighting the properties of reprocessed uranium (RU) and of extracted fission products. Finally, the analysis touches on safety, environmental Impact, non-proliferation concerns and the relative maturity of the technologies.

For all approaches the starting material consists of 103,000 tons of used CANDU fuel accumulated after a future shut-down of all existing CANDU facilities at the end of their current round of refurbishment.

Option 1 – Recycling Used Fuel “Once-Through” in Re-refurbished CANDU Reactors

For this option, stored CANDU fuel is treated with an aqueous chemical extraction procedure called COEX. This aqueous procedure separates fission products from a stream containing most of the uranium and minor actinides, which are separated in subsequent steps. A second, separate uranium stream contains plutonium. This second stream provides the needed augmentation of fissile components which are then diluted to the concentration suitable in fuel for refurbished CANDU reactors.

The effectiveness of this approach is shown graphically in Figure 1 to supplement Tables 1-3 and D-3 of the Study. For simplicity, to avoid complex time gaps during additional refurbishment, all refurbished CANDU reactors were considered to operate proportionately throughout the time period in the Study. Since the new MOX fuel (mixed oxide of Pu and U) required 1.1% fissile Pu compared to the 0.27% content of used CANDU fuel (UCF), about 4 volumes of UCF were required to create one new volume of MOX fuel. The excess processed uranium (RU) is stored. By the time all of the UCF is processed and its transuranics (TRUs) are passed through the CANDUs again, the material balance shows

0 tons used CANDU fuel	>>>>	no DRG required
~70,000 tons RU (pure reprocessed uranium)	>>>>	to storage
~700 tons extracted fission products (FPs)	>>>>	to shielded storage
~25,000 tons used MOX (used mixed oxide fuel).	>>>>	to DGR

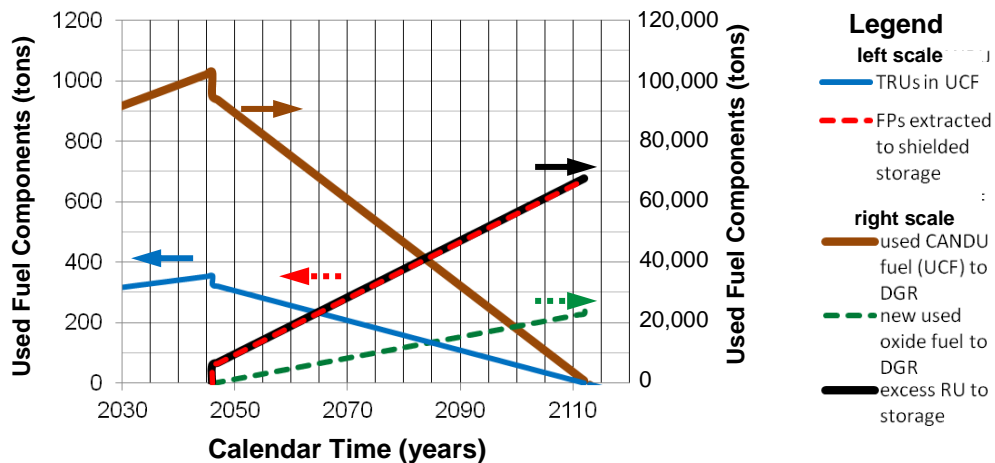


Figure 1. Graphical representation of the effect of one cycle of re-using UCF in CANDUs. Note different scales (arrows) for major amounts of used fuel (right axis) versus smaller amounts of FPs and TRUs (left).

Since the non-fissile component of the plutonium (Pu240/242) increased in this cycle relative to the fissile Pu239/241, further recycling is a process of diminishing returns. Without further recycling the 25,000 tons long-lived MOX UCF created would require long-term secure sequestering, i.e. a long-term DGR. The savings in terms of DGR space requirements are of the order of 25,000 tons compared to the original 103,000 tons. However the savings in costs would be a much smaller ratio. The potential fate of 70,000 tons of RU and 700 tons of fission products, not requiring a DGR, will be discussed below.

Option 3 -- Recycling “Once-through”, then Continuously in Metal-Fuelled Fast-Neutron Reactors

Since Option 2, using an oxide-fuelled fast-neutron reactor (FNR), is similar in principle to Option 3, a metal-fuelled FNR, only Option 3 will be discussed here in detail.

Used CANDU fuel, after conversion to metal form is processed via non-aqueous metallurgical electro-refining in molten salt (pyroprocessing for short) to produce four product streams:

- 1) highly pure uranium (at ~ 1 part in 10^6) constituting about 90% of the uranium (RU)
- 2) fission products (FP) with a purity of less than 1 part in 10^3 of heavy atoms
- 3) a mixture of all TRUs along with the remainder of the uranium
- 4) zirconium from the cladding in CANDU fuel bundles

Fraction 3) is quite concentrated in terms of TRUs, easily satisfying the fuel requirement in FNRs of TRU concentrations of 20%-25%. The appropriate concentration is achieved by dilution with part of the pure uranium in fraction 1). About 10% zirconium is added to the metal fuel to raise the melting temperature of the fuel alloy and provide a ~ 200 C cushion above the highest operating temperature in the FNR.

An identical pyroprocess is used for spent (used) FNR fuel during continuous recycling of fuel in those reactors with the zirconium cladding being replaced by steel in the canisters for FNR fuel rods.

*{Note: **Potential cost saving.** Since used CANDU fuel has very few TRUs and FPs, the pyroprocess is lengthy and more costly than needed, processing mostly pure uranium. Pre-extracting much of the uranium by a more economical dissolution in nitric acid and purification by crystallization [Hart et al., Prog. Nucl. Energy, III, p. 544, 1958] would leave a much smaller more concentrated TRU-containing volume for pyroprocessing, and require a 10 times smaller and less costly pyro-facility to do so.}*

In the Study, Option 3 was considered to proceed in two stages, though the reasons for this were not given. In the first stage the stored used CANDU fuel is processed in a large pyro-electrorefining facility (see potential cost saving in preceding paragraph) to extract TRUs and RU as starting fuel and as fuel replenishment for 8 FNRs of the 380 MWe¹ PRISM-type reactors from GE-Hitachi during the 60-year life of those reactors. The reactor configuration chosen was one with a conversion ratio of 0.5, i.e. at the end of any fuel cycle (~ 4 -6 years) only 0.5 of the total fissile components in the reactor fuel remained compared to that present at the beginning of the cycle.

¹ Note: in the Study the material balances appear based on the 311 MWe PRISM, while the Study power calculations are based on a 380 MWe PRISM output. Making this consistent will change the time scale.

The second stage applied pyroprocessing to the used FNR fuel created in the first stage and to a small amount of used CANDU fuel remaining. The higher TRU concentration in the used FNR fuel permitted a much smaller pyroelectric-facility. The FNR fuel emerging from the reactors was then continuously recycled through 32 PRISM-like FNRs, again with a fissile conversion ratio of 0.5. The effectiveness of these two sequential approaches is shown in Figure 2.

The first stage is similar to Option 1 in that the 103,000 tons used CANDU fuel are processed almost completely during the 60-year life of the FNRs, whereas the used MOX FNR fuel from the reactors builds

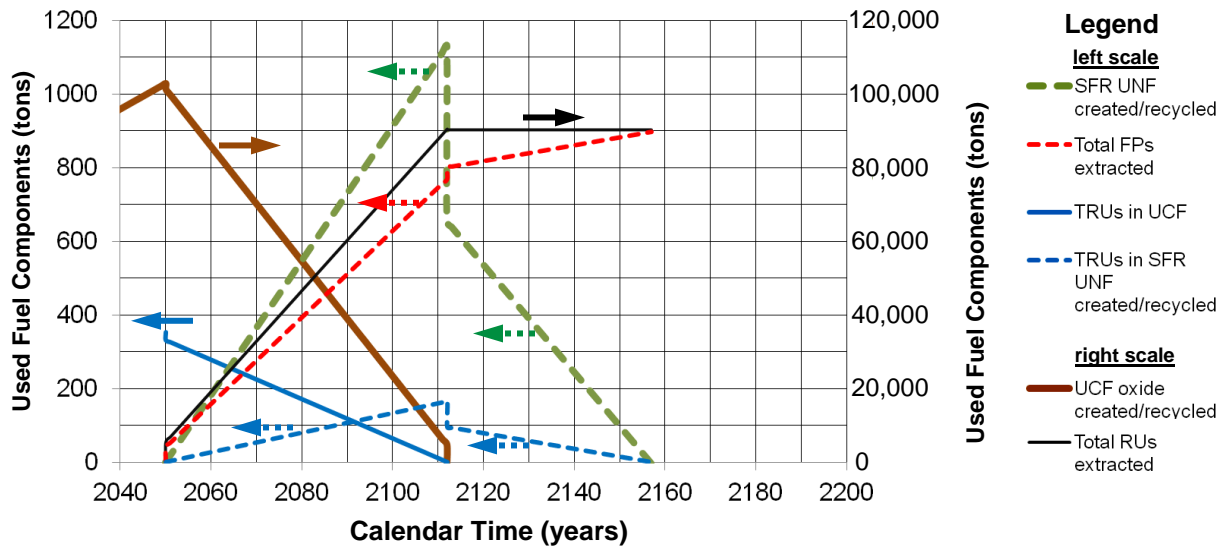


Figure 2. Graphical representation of Fuel Components in Option 3. Note different scales (arrow directions) for major amounts of CANDU used fuel and RU (right axis) versus smaller amounts of FPs and FNR used fuel (left).

up in quantity to 1130 tons. By year 2112, 800 tons fission products are extracted, as are just over 90,000 tons RU. At the end of this stage the used nuclear fuel that requires long-term sequestering in a DGR is reduced almost a factor of 20 from 103,000 to 5900 tons.

Mass balance at end of stage 1

Used CANDU fuel	4770 tons	>>>>	to DGR
Used FNR fuel	1130 tons	>>>>	to DGR
RU (reproc. uran.)	90306 tons	>>>>	to storage
Fission products	770 tons	>>>>	to shielded storage

This by itself is a substantial saving in DGR volume. However, the second stage of this process reduces this volume even further by cycling the used MOX FNR fuel through the 32 PRISM reactors mentioned above.

About one third of the accumulated 1130 tons used MOX fuel is required immediately to furnish starting fuel for the 32 FNRs (the major drop in the dashed green line at year 2112 in Fig. 2). The remainder of the used MOX fuel, plus any used MOX fuel emerging from the reactors, are recycled. After about two cycles all of the TRUs, both plutonium and the minor actinides reach a dynamic equilibrium at the end of each cycle, i.e. all added TRU isotopes are consumed in proportion. In about 45 years, around year 2155, the TRUs external to the reactors are eliminated.

The mass balance state of reactor fuel at the end of Stage 2 is the following:

0 tons used CANDU fuel	>>>>	no DGR needed
0 tons used FNR fuel	>>>>	no DGR needed
90300 tons RU	>>>>	to storage
900 tons fission products.	>>>>	to shielded storage

Neither of the latter should be placed into a long-term DGR. However, within the 32 reactor cores there is used fuel amounting to 216 tons RU and 86 tons long-lasting TRUs. This fuel requires a better nuclear exit (see next paragraph).

Potential nuclear exit

If no further nuclear power were desired, these TRUs can be reduced further in several fuel cycles, taking advantage of the 0.5 conversion ratio (CR) of the reactors. At the end of each cycle there is only enough TRU material to refuel half of the remaining reactors. Thus the number of reactors would be reduced to 16, then 8, 4, 2, and 1, leaving only 3 tons of TRUs in the last reactor, equivalent to a volume of 52 cm cubed, for final disposal.

In this Option, with a suitable nuclear exit, the DGR would not be required at all. As a consequence, this Option would result in a real saving of \$20-40 billion for the cost of such a DGR, and of a further of ~\$11-30 billion as budgeted for disposal of RU and high level waste (HLW) in Table 1-3 of the Study. Only funds for conversion of metal RU to oxide would be needed, and funds for safe shielded shorter term storage of FPs.

Long term advantage -- 8000 years of non-carbon power with existing fuel

The Study did not address a major advantage of FNRs at all, that of being able to consume the accumulated 90,000 tons of RU as fuel replenishment with a saving of 8000 years of uranium stock.

With some foresight the configuration of an FNR similar to the PRISM reactor can be changed from a conversion ratio (CR) of 0.5 to a CR of 1.0 without a redesign of the reactor core (see ref. 92 in Study: E. A. Hoffman, W. S. Yang and R. N. Hill, "Preliminary core design studies for the advanced burner reactor over a wide range of conversion ratios," Argonne National Laboratory, Argonne, 2006.). The change is accomplished by adding more uranium to the core, changing some peripheral reflector steel assemblies to RU-containing units, and by adding RU-containing rods above and below the fuel rods in the more central driver assemblies. In the vernacular of the field these changes add radial and axial "uranium blankets".

An additional change is a small absolute increase in fissile TRUs, although, as a ratio of the total core mass, the percentage fissile content is in fact lower. Nevertheless, it is important that this change occur while enough TRUs remain to satisfy these requirements. For Option 3 as is, the latest time is year 2147.

These changes are indicated in Fig. 3 by the “jog” in the “in-reactor” fuel components (thin dashed lines) at year 2147. Subsequent to this change the “In-reactor” TRU content and composition remain identical

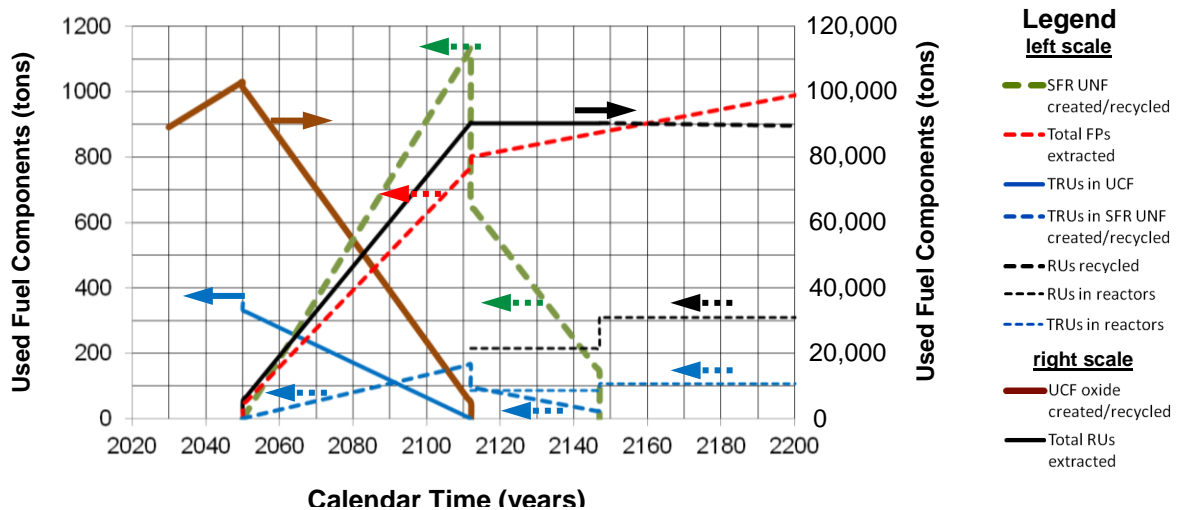


Figure 3. Graphical representation of Fuel Components in Option 3 with reactor configuration changed from CR=0.5 to CR=1.0. Note change of “in-reactor” fuel composition in year 2147.

at both the beginning and end of a fuel cycle. Fuel is cycled to extract FPs, but is replenished only with RU, which is continually changed to TRUs in the reactor as fast as reactor TRUs are fissioned.

The change in RU levels as it is used as fuel replenishment in FNRs with a CR=1.0 goes almost unnoticed at the time scale in Fig. 3 (dashed medium black line), since it is an annual change of only 14.8 tons in 90,000 tons per year. However, this change becomes quite obvious when the scale is expanded to many centuries (Fig. 4).

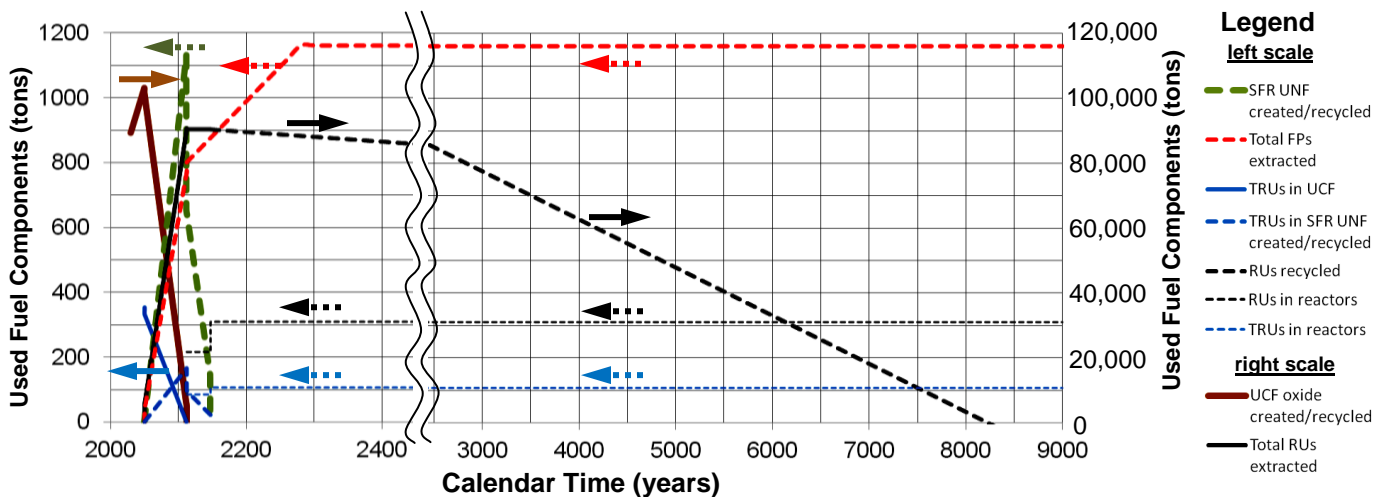


Figure 4. Graphical representation of Fuel Components in Option 3 with reactor configuration changed from CR=0.5 to CR=1.0 in year 2147. (extended time scale)

It is clear that the 90,000 tons of RUs could furnish over 8000 years of non-carbon power at a rate of ~116 TWh per year, at ~13 GW of electrical power using the equivalent of the 32 FNRs in the Study.

This would create at least 960,000 TWh additional electrical energy.

To cover the 8000-year time, many generations of fast-neutron reactors would be needed, each passing its fissile content to the next. For the configurations with CR=1.0 no further TRUs would be required.

As an important ecological alternative approach, these 90,000 tons of RU, with the addition of 6 times the reactor power, could power all 70 GW of current Ontario fossil fuel energy requirements in the transportation, industry and residential sectors for over 1500 years.

Total Fission Product Mass Limits: 1200 tons

What is noticeable as well in Fig. 4 is the attainment of a constant level of fission products of just under 1200 tons close to year 2300 even with the power-creating production of new fission products. Since fission product radiotoxicity decays and attains background levels of uranium toxicities close to 300 years after discharge from a reactor (Fig.5), any fission products of that age can be treated effectively as

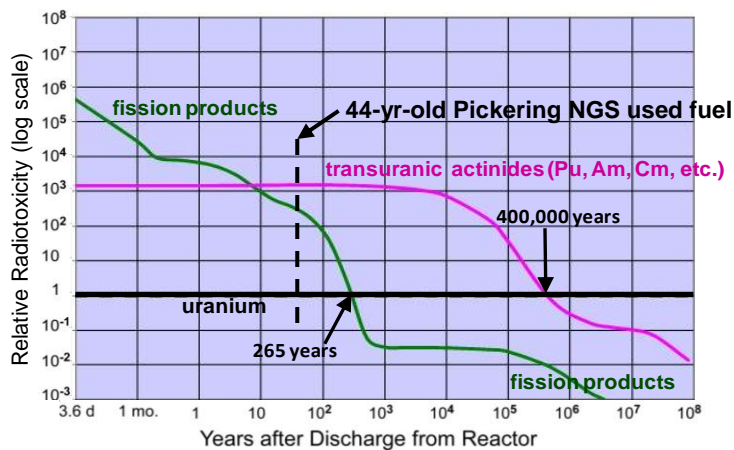


Figure 5. Evolution of Radiotoxicity from Used CANDU Fuel Components Relative to Natural Uranium.

Elimination of the transuranic actinides shortens the time of decay of radiotoxicity to background levels of natural uranium and lower from 400,000 years to 265 years. The broken line refers to the oldest used fuel (44 years) at the Pickering nuclear generating station (NGS). Note the log scales of both axes.

stable atoms and minerals. Thus any amount of new fission products discharged into shielded storage from any reactor at that time would be matched by an equal amount of fission product being released from storage as “non-radioactive”.

Therefore the shielded storage required should not increase beyond this 1200 ton limit.

Requirement, or not, for long-term high-level DGR

The plans for the construction and use of a deep geological repository for used CANDU fuel were predicated on the current habit of using natural uranium to fuel CANDUs in a “once-through” fuelling regime. Without recycling the used fuel, the constituent long-lived transuranics (TRUs) would require safe sequestering for many millennia.

The recycling options considered in the Ministry of Energy Recycling Feasibility Study suggest that additional “once-through” fuel cycles do not alter the requirement for a DGR substantially. However, with continuous recycling through fast-neutron reactors all of the long-lived TRUs can be eliminated.

During recycling the major, pure uranium constituent, called reprocessed uranium (RU) in the Study, is extracted. As indicated in Fig. 4, it is a ready fuel constituent in FNRs with a CR=1.0. Therefore due to its value in energy production RU should not be disposed of permanently. Contrary to this conclusion, the CNL Study suggests that RU should be placed in the planned DGR, since it contains traces of fission products and U232, whose decay daughter Tl208 emits a 2.6 MeV gamma ray. Data below on RU and on fission products indicate such sequestering is not necessary.

U232 in Reprocessed Uranium (RU):

Work by W.B. Arthur on “Uranium-232 production in current design LWRs” shows the level of U232 in fuel irradiated at various doses (<http://www.osti.gov/scitech/servlets/purl/5963522>). From that data it can be seen that for a typical energy yield for used CANDU fuel irradiated to 5.7 MWd/kg, the level of U232 in pure extracted RU is 2×10^{-9} atoms per atom of U235. This would lead to a dose of 0.3 mSv per year for 20 kg uranium (= 1 CANDU bundle) at 1 m. This is less than 1/10 of the North American annual background dose.

The CNL Study indicates that MAGNOX fuel at a low dose level of 0.2×10^9 Bq/tonne is consigned to landfill at Clifton Marsh, UK, [p. 5-3, (35); ref[19]]. This corresponds to 0.56 mSv/yr for 20 kg uranium at a distance of 1 m. Therefore consigning as valuable a non-carbon energy resource as RU to a permanent DGR would be hugely irresponsible overkill.

Fission products in RU:

In irradiated CANDU fuel, the FPs in the fuel decay to a radiotoxicity level 1000 times uranium background in 10 years after leaving the reactor (Fig. 5), with Cs137 being the only gamma-emitting FP isotope after 30 years. Pyroprocessing provides uranium purities of the order of 1 part in 10^6 with respect to plutonium, with thermodynamics making it even more difficult for fission products to deposit with uranium (Till and Chang, Plentiful Energy, Amazon 2011, p. 194). Extraction of uranium in the various chemical processing schemes in the Study indicate high degrees of uranium purity as well. Therefore fission product impurities are expected to be small in RU. As a result “trace amounts” (p. 11-10 (108)) need to be defined in the Study before they can or should be used as a reason to discard a resource such as RU out of hand, with a cost of \$ 20-40 billions and a loss of many trillion dollars.

Extracted Fission Product Values

Fission products are in general considered to have a lifetime of the order of 300 years (Fig. 5), requiring safe shielded storage before that. They consist of elements in the central portion of the periodic table and are created in the reactor with a mass distribution shown in Fig. 6. They contain many valuable elements such as rare earths and platinum group metals of which rhodium and rubidium are stable non-radioactive elements immediately. Shown as well in Fig. 6 are the current values associated with the individual elements in proportion to their mass, as obtained from international vendors on the Internet.

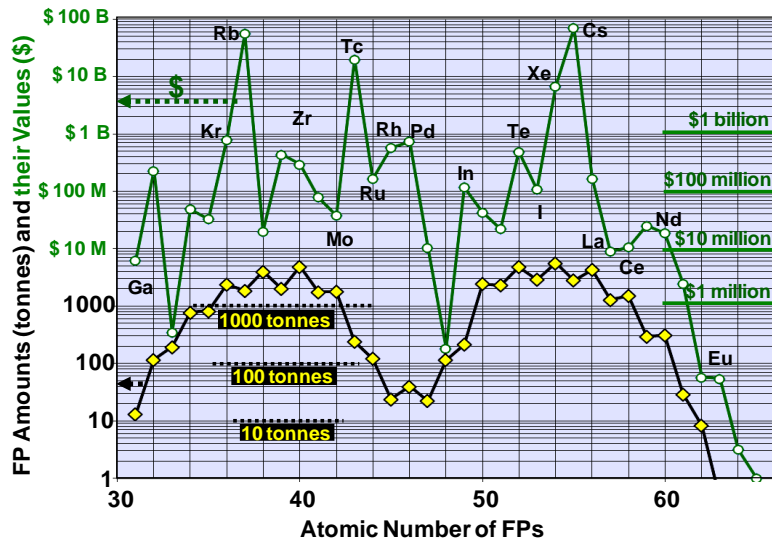


Figure 6. Distribution of amounts (black line) and values (green line) of fission products resulting from consuming 50,000 tons of nuclear fuel completely.

The amounts add to about \$ 3 million per ton. After 300 years such elements can be extracted by ordinary means.

While the values in Fig. 6 refer to gross revenue, it is highly probable that extraction of elements will be a profitable endeavour. As an example, the platinum group metals, Rh, Rb, and Pd are at a higher concentration among the FPs than they are in the best ores in Africa; and FPs do not have to be mined.

It is therefore incumbent that the FPs not be sequestered permanently in a long-term DGR, but be stored safely and accessibly, and in a form that makes such future extraction economic. At a recent COG-OPG workshop on used fuel recycling Max Fratoni of the University of Berkeley suggested a solid salt form in stainless steel cylinders. This is preferable to the ceramic sodalite waste form suggested by the Argonne Laboratories that makes the fission products chemically virtually inaccessible (Till and Chang, p. 183).

Long-lived Fission Products

Figure 5 shows that some fission products at very low radiotoxicity have very long half-lives, of the order of a million years or more. These are shown in greater detail in Fig. 7. Of these, iodine-129 (I-129) with a 17 million-year half-life may be of biological concern even though its toxicity levels are almost 40000 times less than uranium, since ingested iodine can concentrate in the thyroid gland about 10000-fold [A. Taurog et al., J. Biol. Chem. 171, 1947, 189].

However, this particular concern can be alleviated as part of normal FNR fuel cycling. During pyroprocessing at 500°C, and again during casting and refabrication of new FNR fuel rods at about 1500°C, iodine, mainly I-129 and stable I-127, is volatile and is removed via condensation in cold-traps. I-129, with a neutron cross section of 30 b, can be transmuted in reactors to I-130, which has a half-life of only 12.3 hours, decaying to stable Xe-130. Some of the stable I-127 would become I-128, which decays in 25 minutes to stable Xe-128. Similarly, other iodine isotopes formed, e.g. I-130 to I-131, decay

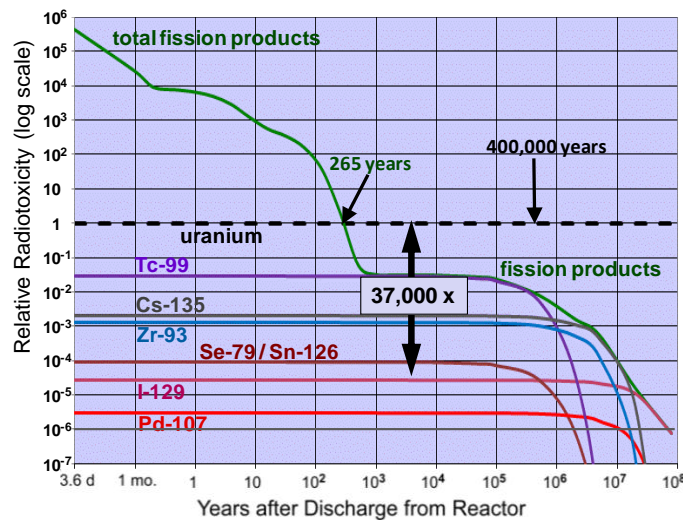


Figure 7. Radiotoxicity of seven very-long-lived CANDU fuel fission products after discharge from reactor

rapidly to their stable xenon analogues. In this way a million-year concern becomes a 12.3-hour non-problem transmuting into a valuable noble gas.

Mentioned earlier was the re-use of the pure zirconium from CANDU fuel bundles as a 10% alloying element in the metal fuel rods of PRISM-like FNRs. Although zirconium is activated in part to Zr-93 in the CANDU fuel bundle, this has no bearing at all on its function nor does its radioactivity matter in the high flux of the reactor core. Among the fission products (Fig. 7) it segregates in pyroprocessing as a noble metal, and can be extracted and reused in the metal fuel.

It is anticipated that during the 300-year decay of the other fission products, a technology will be developed to extract and possibly find applications for the other long-lived fission products.

Additional Points of Discussion

Safety

This appraisal will be brief, since the safety of current reactors has been covered in the Study. However, a word on FNRs, particularly the sodium-cooled metal-fuelled model such as the GEH PRISM design. To my knowledge its prototype, the US EBR-II, is the only reactor which was tested close to 50 times with sudden loss of cooling under full power with deliberately inactivated control rods. The reactor shut itself down without human or automated intervention and maintained cooling under no-power conditions indefinitely (Till and Chang, p. 147ff). This type of reactor would have sustained no damage under the conditions that caused failure in Fukushima, Three-Mile-Island, and even in Chernobyl.

Recycling and Environmental Criteria, GHG Emissions

The discussion of environmental aspects in the Study leaves much detail too opaque to assess the concluding rankings. For instance, Table 5-4 ranks Option 3 worst on the basis of 10,606 tons of high level waste left (HLW). The origins of that number are not at all clear. The mass balance analysis related to Fig. 2 (page 11, above), indicates 90,000 tons RU, 900 tons fission products, not counting the material

left inside the 32 reactors, which is about 300 tons of mixed RU and TRUs. Nothing comes close to 10,600 tons high level waste.

Furthermore, while potential spills and environmental contamination from aqueous processing in Option 1 and 2 are mentioned, little credit is given to the non-aqueous processing of Option 3 in which cooling of the molten salts immediately renders a solid non-movable form for potential clean-up. Thus discharges to local waters, feared with aqueous techniques, are very much less of a concern with non-aqueous pyroelectrolytic processing. Since the Study team partners, the National Nuclear Laboratories in the UK, were part of the development and testing of the proposed aqueous GANEX technology, it is understandable that there should be a natural bias towards aqueous chemistry.

One aspect not discussed is the impact that nuclear power has on avoiding emission of greenhouse gases. While this is well known in nuclear circles, even there it is not realized that the complete use of the 103,000 tons of used CANDU fuel, as depicted in Figure 4, can avoid the emission of 980 billion tons of CO₂ compared to the use of coal. That amounts to 32% of all the CO₂ currently in the entire atmosphere.

Non-proliferation

The Study ranks Option 3 lowest in the basis that the processing separates fission products leaving the heavy metal plutonium un-self-protected. In fact all separation technologies in all options equally separate fission products in the first separation step. The important aspect is that plutonium is not purified in any of the techniques. Indeed, great pains were taken in the pyroprocess to make it impossible to derive pure plutonium, but always to have it mixed with some uranium and with all of the other transuranics (Till and Chang, Plentiful Energy, Amazon, 2011; many discussions, p. 111, 167ff, 244ff). Contrary to fears quoted in the Study (p. 8-3,4 (58,59)), the physical parameters are not easily adjusted.

On the other hand, in the aqueous chemical technologies for recycling, the a change in suitable chemicals in either COEX or GANEX as proposed for Options 1 and 2 changes those processes to the military PUREX form to separate plutonium, using the same chemical separation equipment throughout.

Therefore electrorefining of Option 3, a physical technology far removed from chemical PUREX, is preferable.

Maturity of the technologies

None of the three non-PUREX processing technologies, COEX, GANEX, and pyroprocessing, has been commercialized. However, in contrast to the other technologies, pyroprocessing has been applied since 1994 to process the used uranium from the core of a metal fuelled reactor, the EBR-II. All of the driver assemblies from that reactor have been processed and three times as many blanket assemblies (Till and Chang, p.181). Thus pyroprocessing is likely the MOST MATURE of the technologies.

Summary

A summary is given in pages 3 to 7 at the beginning of this exposition.