THE TRAVESTY OF DISCARDING USED CANDU FUEL

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Abstract

The current plan worldwide for virtually all used nuclear fuels is costly deep burial to attempt to isolate their long-term radiotoxicity permanently. Alternatively Canada's 50,000 tons spent CANDU fuel, of which only 0.74% of the heavy atoms have been fissioned to extract their energy, could supply 130 times more non-carbon energy using proven economical recycling and fast-neutron technologies. The result in this country alone would currently be $65 trillion of reliable electricity on demand created without greenhouse gas emissions. It would avoid adding 470 billion tons CO_2 to the atmosphere compared to the use of coal, to mitigate climate change. Worldwide recycling of stored spent nuclear fuel and replenishing with depleted uranium in fast-neutron reactors could avoid emitting over 20 trillion tons CO_2, or over six times the current total atmospheric CO_2 content. As added bonuses the long-term radiotoxicity of the used CANDU fuel is effectively eliminated, and even the shorter-lived radioisotopes become valuable stable atoms and minerals that would fetch $3.1 million per ton. That alternative is worth pursuing, instead.

1. Introduction

Consider burning only the tinder-like bark off the birch logs in your wood-burning stove or on your campfire, then discarding and burying the solid logs unburned because they are slightly charred and "contaminated" with charcoal, soot and a few ashes. That, in effect, is analogous to the *modus operandi* of our nuclear industry with respect to the use of uranium fuel in all of our thermal reactors, be they CANDUs or light-water reactors (LWRs).

In today's global ambience of "reduce, re-use, and recycle", it seems very surprising that the nuclear industry is operating nowhere near such a maxim. After extracting only 0.74% of the nuclear energy in natural uranium via its CANDU reactors, Canada, primarily Ontario, is on course to discard the remaining fuel as waste permanently in a deep geological repository (DGR) and will pay $24.4 to $40.7 billion to do so [1, p.163; 2; 6, p.3].

Light-water reactors worldwide fare even worse. They extract only about 0.5% of the nuclear energy of mined uranium, leaving the rest as fuel waste and as depleted uranium (see below).

Yet, similar to our knowing how to burn wood completely to ashes, the nuclear community knows how to consume 100% of the uranium fuel, not just the less than 1%, and has done so safely for decades by cycling uranium fuel through fast-neutron reactors [4].

This flagrant inefficiency has been accepted for at least three reasons. First, fresh uranium fuel is still relatively inexpensive, costing Ontario Power Generation (OPG) only 0.54 ¢/kWh currently

[5], plus, depending on the calculation, an additional 0.2-0.4 ¢/kWh for disposal [2]. Second, the nuclear energy extracted from even as little as 0.5% - 0.74% of this non-carbon fuel is huge. It requires only 925 grams of the 125 kg uranium charge in 6.25 CANDU fuel bundles to produce 1 megawatt-year (8.76 million kWh) of non-carbon electricity in a CANDU reactor [1, p.351]. This is enough electricity for 900 households for a whole year [6]. To produce the same amount of electricity would take 2,800 tons, or 28 hopper rail cars, of coal [7], with the emission of 8000 tons of CO_2 [8]. The equivalent use of methane gas would emit 4700 tons CO_2 [9].

That's a lot of energy from very little uranium fuel. But compared to the 925 g, the remaining 124 kg fuel in the same 6.25 used fuel bundles, if consumed completely by cycling through available fast-neutron reactors [4,10] would produce yet 134 times more non-carbon electricity, or 134 MW-years, and avoid the emission of 1,000,000 tons of CO_2 from coal.

Thirdly, it is often assumed that recycling of used fuel brings with it an increased risk of nuclear weapons proliferation. Yet in over 60 years of commercial nuclear power worldwide no proliferation of nuclear weapons has occurred as a result of commercial fuel cycling. The used fuel constituents from commercial reactors are very poor weapons materials. Moreover, in recycling, as below, they are not separated. Canada has had the wherewithal to build such weapons since the 1940s and '50s even without recycling, but has chosen not to do so. There is no reason to believe adopting a closed fuel cycle will change Canada's weapons philosophy.

Thus the wastefulness with respect to uranium fuel is our choice rather than a technological necessity. It appears that only our own inertia and our short-term thinking stand in the way of doing better. Indeed, we are willing to spend $24 to 41 billion to bury Canada's current 50,000 ton energy resource of used CANDU fuel that over centuries could instead create non-carbon electric power currently worth ~$65 trillion.

2. The root cause of uranium fuel inefficiency

The problem with uranium fuel inefficiency is inherent in the nuclear reactors that we now use, be they heavy-water-cooled CANDUs or light-water reactors (LWRs). Both types are "thermal" reactors, using slow neutrons. Such neutrons very efficiently fission, or split, only a small isotopic component of the uranium, the 0.72% U-235 in mined uranium, leaving the much larger component, the 99.28% U-238, virtually untouched. A little of the U-238 is converted into heavier elements, some of which can also be consumed. (In the jargon of the field these properties make U-235 "fissile" and U-238 "fertile".) However, once most of the fissile U-235 is consumed the reactors have to be replenished with fresh fuel.

With the massive power output of the reactors often on the order of 1000 megawatts, the public and even many workers in the nuclear industry have been blinded to the huge fuel inefficiency of the heat-producing process. Yet no other fuel source is used so wastefully, exploited to less than 1%, whether coal, oil, gas, hydraulics or even photovoltaic conversion of sunlight [11].

3. An analogous inefficiency: building of a simple log fire

A familiar analogy may drive this nuclear wastefulness home, using the burning of birch wood logs, about the size of CANDU fuel bundles, in a fireplace, or on a campfire.

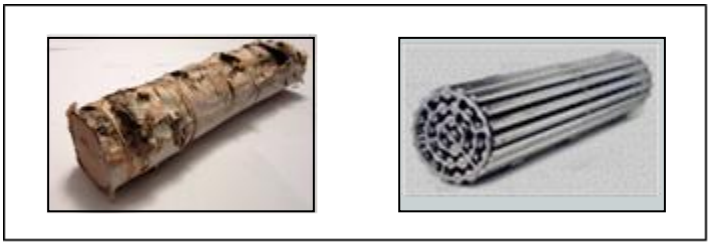

Figure 1. Analogy between a birch log (left) and fuel bundle of natural uranium (right). The easily lit thin layer of birch bark is excellent tinder, and is analogous to the isotope U-235, the small fissile component in uranium fuel, while the main constituent of the log, the heartwood that has to be "coaxed" to burn, would correspond to fertile U-238.

A birch log, in analogy to uranium (Fig. 1), also consists of two main fuel components, the bark and the heart wood. Birch bark is excellent tinder, since it will stay dry even through heavy rains and therefore will light and produce an initial flame under most circumstances. It could be considered analogous to U-235 in the nuclear field, which is fissile to neutrons of any energy. The much more massive heartwood is difficult to ignite directly, analogous to U-238.

3.1 CANDU analogue

A beginner might light the partly peeled bark of a loosely-laid single birch log (Figure 2a) and be rewarded with a jaunty flame (Fig. 2b), only to find that the initial promising flame soon dies

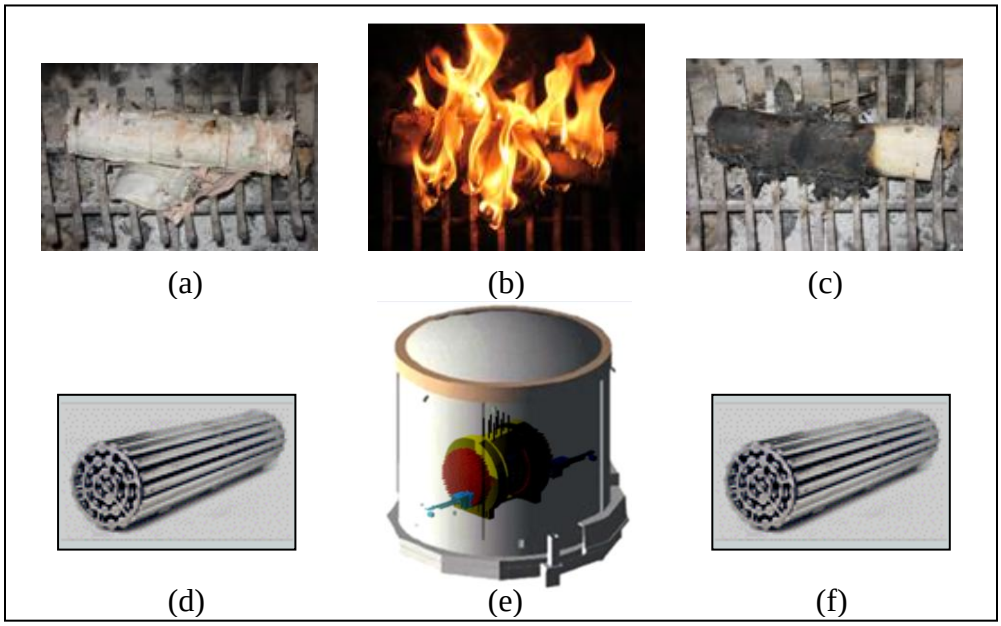

Figure 2. Analogy between birch log in fireplace and uranium fuel in CANDU reactor
(a) A fresh birch log with loose bark tinder, in analogy to fresh uranium fuel bundle with U-235 (d). (b) Lit bark tinder around log burning on grate in fireplace; (e) analogously U-235 in fuel bundle is consumed partially in the pressure tubes of the CANDU reactor (Note: the uranium is split to produce heat, but there is no fire). (c) Charred log incompletely burned. (f) Spent fuel bundle, mostly U-238, with U-235 reduced from 0.72% to 0.23%, with 0.74% fission products ("ashes") and with 0.4% TRUs (invisibly inside the zirconium cladding of fuel rods)

Figure 3. Incomplete Consumption of birch fuel log.
Incomplete combustion consumes the birch bark tinder to ash and chars the log (left) but leaves the heartwood virtually untouched. Analogously, but unseen in the zirconium casings of the fuel bundle (right), use of CANDU fuel consumes most of the 0.72% fissile U-235 to fission products ("ash"), converts a small amount of the U-238 ("heartwood") to heavier atom transuranics ("char").

when the bark is consumed (Fig. 2c). Left behind is a slightly charred log plus a small amount of wood ash (Fig. 3). Nevertheless, in the meantime an encouraging amount of useful heat was produced. Consequently, a fresh birch log would be requisitioned, and the process repeated with similar results. With such a scenario, the slightly charred used logs, blackened with charcoal and tars, would pile up (Fig. 4), with some concern perhaps as the pile grows bigger that chemical poisons produced by incomplete combustions, such as cancer-causing dioxins, might leach out of the blackened log surfaces into the ground and affect the biosphere.

Figure 4. Representative pile of slightly burned, charred birch logs (left) and analogously accumulating used fuel bundles stored dry in concrete castors (right).

This scenario in all aspects is effectively the sequence of natural uranium fuel use in what I consider the most efficient current thermal reactor design, the CANDU reactors. Once most of the small amount of fissile U-235, the easily lit nuclear tinder, is consumed in this reactor, a new fuel bundle is requisitioned to replace the old, leaving behind equivalent amounts of fission products ("ashes"), and virtually all of the fertile U-238 of which only a small mount has been converted to highly radioactive transuranics (TRUs), atoms heavier than uranium (equivalent to the charred portion of the heartwood with its poisonous and carcinogenic dioxins). The TRUs, consisting of fissile and fertile atoms, are partly used up, like uranium.

This process has not changed in principle since the first CANDU reactor was built, with used fuel bundles now amounting to a "pile" of 50,000 tons in pools and in dry storage (Fig. 4).

3.2 LWR analogue

A cleverer approach to extract more energy from one fuel load might be to use the bark stripped from several logs (Fig. 5) to try to get a single log to burn more. In this case more tinder would be burned and a greater amount of the heartwood is charred and consumed partially before the fire goes out. In this example the charred log and ashes as well as 6 to 10 fresh logs stripped of their bark are discarded and accumulate as waste. This is analogous to the light-water reactor, for which fuel is enriched in U-235 to 3-5% (cf. bark from several logs), depleted uranium is discarded (cf. stripped logs), and so is the used fuel of about 95% uranium, 1% - 1.5% TRUs, and ~3% - 5% fission products (cf. charred log and ashes). Yet in totalling the amount of energy in comparison to the overall uranium used and discarded in the process, this approach is even more wasteful than the simple use of natural uranium in a CANDU reactor, above.

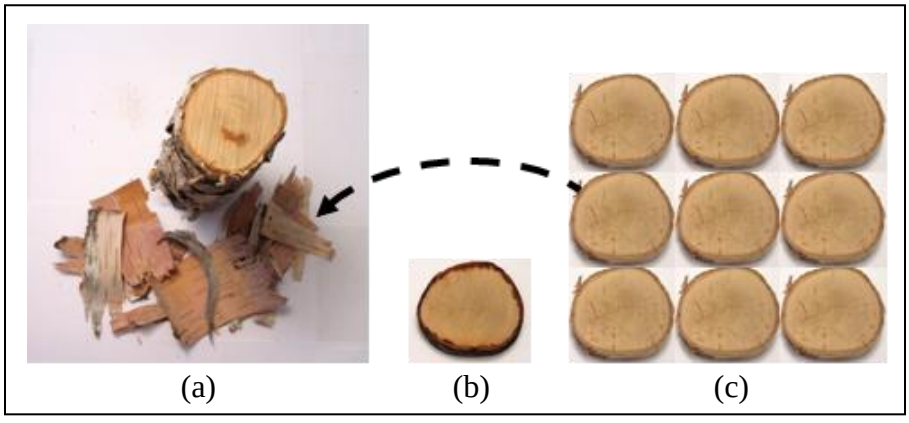

Figure 5 Analogy for LWR fuel: use of birch bark tinder "enrichment".
To yield a greater energy output per fueling, a single birch log (a) is complemented with the bark from 8-10 equivalent logs (arrow from (c) to (a)) that are then discarded without being used as fuel (c). The result in the single log is a greater conversion of heartwood to char and greater consumption of some of the latter (b). Analogously uranium fuel used in LWRs is enriched with the U-235 from 8 to 10 volumes of natural uranium, with the latter, now largely depleted of fissile atoms, accumulating and discarded as relatively benign waste. The fissile U-235 in the fuel is consumed, while converting a somewhat greater amount of U-238 into to fissile and non-fissile transuranics than occurs in CANDU reactors.

3.3 Partial fuel cycling: France

In order to derive more energy from the fuel passing through its LWR reactors France uses a modified PUREX process to extract close to 1% plutonium among the TRUs created in their LWR reactors. About two thirds of this plutonium is fissile and therefore useful for further energy extraction when reintroduced into LWRs as mixed oxide fuels (MOX) with uranium. This results in an improvement of about 30% in total energy yield from the fuel; but the process is still quite inefficient, in total using approximately as much (or as little) as the much simpler fuelling technology of CANDU reactors. It still leaves behind the initial 6 to 10 volumes of depleted uranium, the cycled used fuel, the uncycled non-Pu TRUs, the fission products (FPs), as well as radioactive chemical process wastes that increase the disposal burden of the used nuclear fuel.

Even this technology has its birch log analogy, in which the charred heartwood has a portion of its charcoal surface scraped as shown in Figure 6 (cf. the plutonium portion of the TRUs) to be reused as additional tinder in lighting and consuming a fresh log or even one that has been largely stripped of its bark (cf. natural or depleted uranium). However the remainder of the charred wood and the ashes would still accumulate along with the pile of stripped heartwood.

Figure 6 Analogy to Pu extraction from used LWR fuel: charcoal scraped off partly burnt log.

4. Fuel inefficiency of thermal reactors

The upshot in all of these analogous processes would be an ever-increasing pile of heartwood, some coated with charcoal and poisonous tars, and ashes. At the same time the forest reserves of economical birch wood stands would be cut down (cf. economical uranium mining), with the prospect in a few decades of having to cut birch saplings and shrubs at less yield and greater expense to obtain the bark as tinder.

Such methods would certainly not be accepted except as a last resort in dire need.

We do, however, accept all of these inefficient practices in the nuclear field as normal. CANDUs leave over 99% of their uranium fuel as heavy atom “waste”. LWRs leave over 95% heavy atoms in their used fuel and from six to ten times as much again as U-235-depleted but 100% uranium fuel atoms. Thus the total percentage of heavy atoms that yield their nuclear energy in LWRs is

about 99.5%. France's reactors with a single round of plutonium extraction from their used LWR fuel do about 1% better in terms of uranium used in their reactors. But in France also, piles of depleted uranium from U-238 enrichment are mounting, unused.

4. Fuel efficiency using fast neutrons

Clearly we have not accepted this type waste scenario for wood. We have perfected efficient woodstoves, as well as learned to lay logs effectively for roaring campfires or economical cooking fires, utilizing every bit of fuel from log to scrap. Even the wood ash was used by our forebears to make soap, and is still used as a source of potassium in our gardens.

A similar efficient scenario also exists in the nuclear field. It was started in the 1950s, fleshed out in greatest depth in the USA in the '80s and '90s, and is now largely forgotten, overlooked, or taught as historical footnote: fast-neutron technology with fuel cycling [4]. Few know that it was a fast-neutron reactor, the EBR-I, that produced the first nuclear-generated electricity in 1951.

The EBR-I successor, the EBR-II fast-neutron reactor (FNR), sodium-cooled to prevent thermalization or moderation of neutrons, was the test-bed from 1964 on for increased fuel utilization and for demonstrating passive safety of the technology [4, p.138ff]. It achieved 20% "burn-up" safely in the '80s, compared to 0.74% in CANDUs today. On EBR-II's decommissioning in 1994, fuel tests in the French Phenix FNR provided proof that 25% burn-up at least was possible.

These results indicated that not only was the physics of fast neutrons capable of delivering much higher fuel efficiencies than the physics of thermal neutrons, but also that the materials, the steels of the fuel rods and assemblies, were capable of withstanding the increased neutron fluxes at higher energies and at such high levels of fuel utilization.

Even the 25% fuel burn-up found experimentally in FNRs is not the theoretical limit, a limit that would ultimately be determined by excessive neutron absorption by fission products (FPs) that build up in the fuel as more and more heavy atoms are split to extract their energy. That limit is closer to 35% (Fig. 7). To continue fission of heavy atoms beyond this limit it becomes necessary to extract the FPs, i.e. undertake fuel recycling. Such fuel cycling was put in place in conjunction with the EBR-II reactor, recycling about 35,000 fuel pins or five times the load of the reactor [12]. For a 20% fuel utilization achieved in that reactor, five such cycles would utilize 100% of a single reactor load, leaving nothing but FPs.

4.1 More fast-neutron advantages

4.1.1. Maintenance of fissile content

A particular designed advantage of such a fast-neutron reactor is its internal maintenance of the level of fissile atoms required for operation. Thus after the 20% fission products are removed from the fuel at the end of one fuel cycle, the fuel has only to be replenished with any source of fissionable atoms that includes primarily fertile U-238, such as depleted uranium or used CANDU fuel. No additional fissile components are required. At the end of each such cycle,

except for some initial equilibration, all of the fuel isotopes, uranium and transuranics, reach the same level as at the end of the previous cycle (Fig. 7). This includes the minor actinides such as Am-241 and Am-242, etc., as well as even-numbered major actinide isotopes such as Pu-240 and Pu-242 that accumulate in the used fuel of all thermal reactors. It is this property of FNRs that would make it possible to consume all of the heavy atoms in used CANDU fuel.

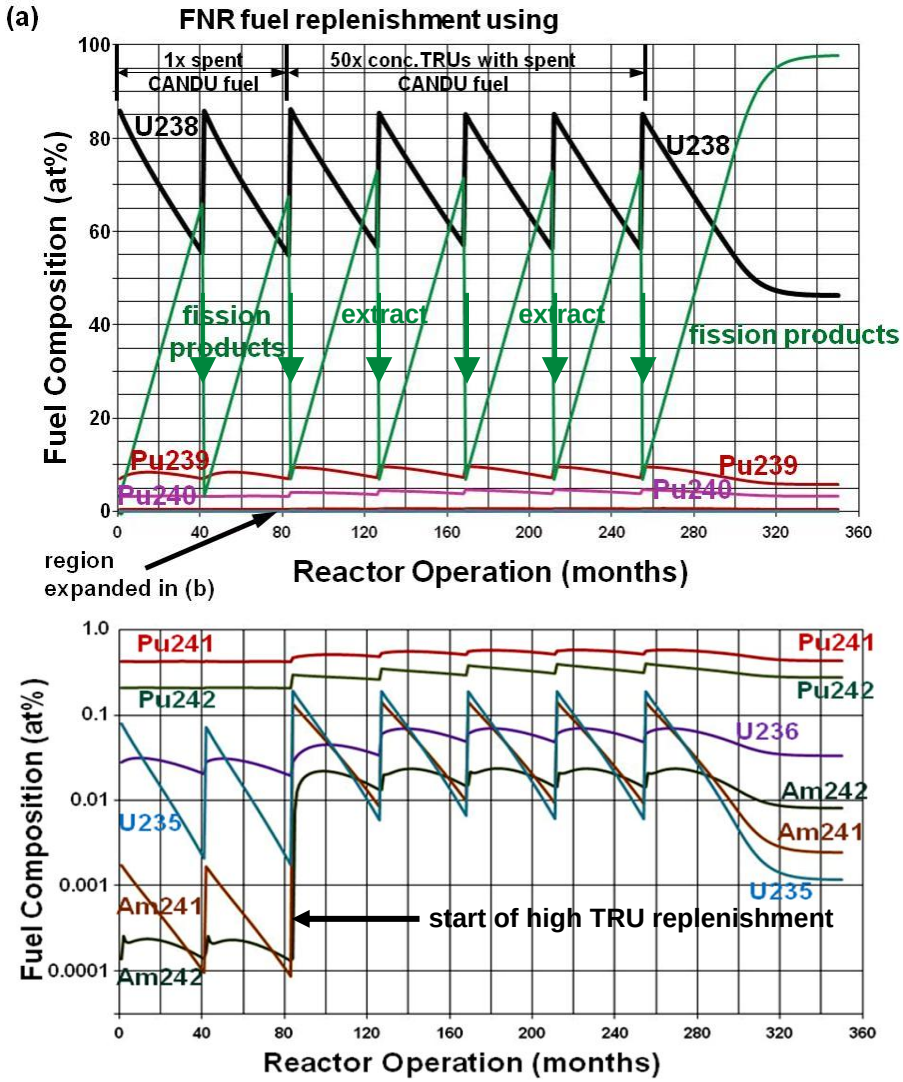

Figure 7 Fast-Neutron Reactor Fuel Behaviour under Replenishment with Two Forms of Used CANDU Fuel

The first two cycles show FNR fuel replenished with used CANDU fuel at months 1 and 41. Subsequent cycles starting at month 82 have FNR fuel replenished with used CANDU fuel from which 90% of the uranium has been extracted to increase the concentration of transuranic actinides (see text).
Panel (a) shows the major isotopes and FPs on a linear scale. Panel (b) is shows an expanded view of the lower 1% of Panel (a) with a logarithmic scale to accommodate the large variations in concentrations of the minor isotopes.

As one crucial outcome, as the heavy atoms are fissioned, whether uranium or TRUs, they cease to exist and their long-term radiotoxicity ceases to exist. Short-lived FPs remain (see Section 5).

4.1.2 No xenon poisoning: load-following

Additionally, one practical operational advantage accrues from the fact that at high energies neutron absorption of any fission product, including Xe-135, is extremely low. At 1 MeV, absorption of Xe-135 is only 0.01 barns versus 2.6 million barns at thermal energies [13]. Therefore xenon poisoning, a problem caused by massive absorption of thermal neutrons by the build-up of Xe-135 via decay from I-135 after reactor shut-down, virtually does not exist at high energies (more accurately, the effect is reduced by a factor of 2600). Such reactors can therefore have their power levels changed at will. There would be no delay in powering up or restarting even after longer shut-downs that normally prevent the restart of thermal reactors until xenon absorption has decayed enough, after several 9.2 hr Xe-135 half-lives. Therefore load-following of the daily variations in demand of energy could be readily accommodated by such reactors, opening the possibility of supplanting much GHG-emitting gas-fired electrical generation. (Note added: a referee rightly suggested that "no-xenon-effects" would likely have helped avoid the Chernobyl disaster. The strong positive feedback on Xe-135 burnout from a power increase would not have occurred in an FNR with high energy neutrons; they cause no xenon effects).

5. Elimination of long-term radiotoxicity.

Long-term radiotoxicity is the prime reason cited for discarding used CANDU fuel and to sequester it from the biosphere for the requisite several hundred thousand years (Fig. 8). As pointed out above, using all of the heavy atoms and particularly the transuranic actinites (TRUs) as fuel in fast-neutron reactors would eliminate the major portion of that future radiotoxicity.

However, since the practical limit of heavy-atom fuel burn-up in FNRs is not 100% but closer to 15-20% [10], the fuel must be recycled, with fission products being removed periodically. In such a process the efficiency of separating fission products cleanly from radiotoxic heavy atoms becomes important, as does the purity and volume of used chemicals after such separation.

5.1 Aqueous processing: PUREX and variants

The classical "recycling" procedure is the separation of fissile materials, specifically plutonium, using aqueous chemistry such as PUREX (Plutonium URanium EXtraction) or its modifications. Facilities used at La Hague in France, and Sellafield in the UK, for example, are huge, leave large volumes of radioactive liquids, and are expensive - the still unfinished Rokkasho (Japan) facility already cost $25 billion [19]. Moreover, with the focus on recovering and cycling only fissile isotopes, the amounts of radiotoxic materials are only minimally reduced.

5.2 Non-aqueous recycling: pyroprocessing

A better approach, specifically designed for recycling used nuclear fuel in fast-neutron reactors, is non-aqueous pyrometallurgical electrolytic separation, or "pyroprocessing", developed at the Argonne National Laboratories, and is still operating [4, p.181]. A full sized facility is estimated at $ 0.1 billion [4, p.291]. This procedure produces three fractions from the fuel: 1) pure

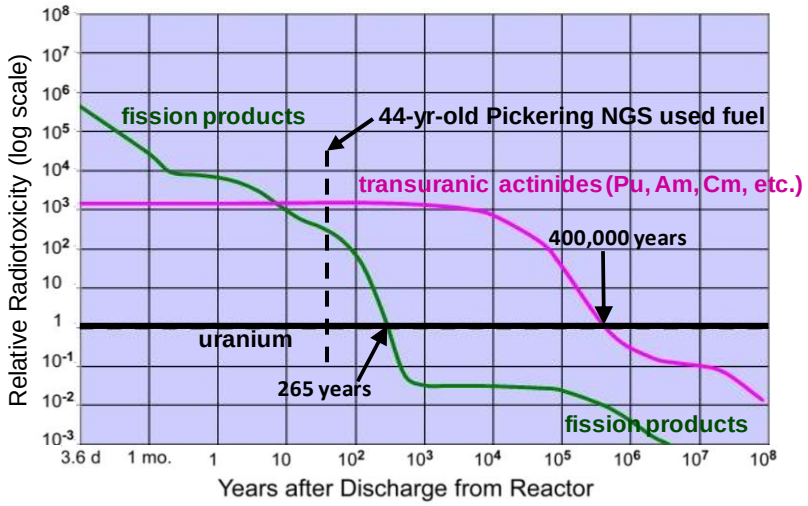

Figure 8 Evolution of Radiotoxicity from Used CANDU Fuel Components Relative to Natural Uranium.

Elimination of the transuranic actinides in fast-neutron reactors and extraction of uranium from the used fuel waste shortens the time of decay to background levels of natural uranium from 400,000 years to 265 years, and results in a reduction in radiotoxicity of used fuel waste of about 42,000X at 1000 years. After 265 years the radiotoxicity of the fission products is lower than that of the natural uranium from which they are created in the reactor. Broken line indicates current radiotoxic state of oldest used fuel (44 years) at the Pickering nuclear generating station. Note the log scales of both axes.

uranium, 2) a mix of all TRUs plus any remaining uranium, and fraction 3): fission products (FPs). There is no separation of plutonium, or any other TRU. Both fraction 1) and 2) are heavy atoms that are recycled in FNRs. Fraction 3), the FPs, are put into medium-long storage to decay further for a maximum of 300 years (Fig. 8), although most become stable after a few decades. Useful stable or radioactive isotopes can be extracted, and many are already for sale [14]. Zirconium, even long-lived radioactive Zr-93, can be recycled into FNR metal fuel, while activated iron casings are compacted and treated like FPs, for decay. Molten salt electrolytes are recycled, leaving effectively no process wastes for disposal.

Still, this non-aqueous separation process is not perfect. Laidler et al. [15] indicate that pyroprocessing achieves a separation of better than 99.9% of the actinides from the fission products. This does not effect complete elimination of the long-term TRU radiotoxocity in fraction 3), but the degree of separation would lower the toxicity of the TRUs over 1000-fold to the level of the original natural uranium or below (cf. Fig. 8).

This TRU level among the fission products may be considered sufficient to obviate the need for a long-term DGR. If not, one has effectively 300 years to search for a better separation method before those remaining TRUs would become the dominant radiotoxic component of fraction 3).

5.3 Accelerated detoxification of TRUs

One additional advantage of the non-aqueous pyroprocess is its potential to accelerate the long-term detoxification of existing used nuclear fuel. Since one fraction, fraction 2), contains the long-lived TRUs, this fraction can be preferentially cycled back into FNRs as fuel, eliminating the TRUs without a major effect on the operation of the reactors. This is demonstrated in Fig.7a

and 7b, which from month 82 on depict the replenishment of FNR fuel using a 50-fold increase in the use of pyroprocess fraction 2), i.e. the mixture of all TRUs plus some uranium.

This approach can be remarkably effective and quick. It can be calculated that with such a procedure it would take only ~25 years to eliminate the small (0.4%) long-term radiotoxic TRU component from the 15,000 tons of used CANDU fuel that would be stored at the Pickering nuclear reactors after their shutdown in 2020 [17]. Thereafter the FNR fuel would be replenished for centuries with stored pure depleted uranium from fraction 1). The requirement would be a fleet of FNRs equal in output to the current Pickering plant along with a fuel cycling complex that included a 50 ton/year pyroprocessing facility and a feeder pretreatment plant to extract 90% pure uranium from the stored used CANDU fuel. Both components of the fuel cycling complex would be about 10-fold smaller than the existing PUREX plants mentioned above.

6. Long-lived fission products

As is evident in Fig. 8, beyond 300 years, fission products contain isotopes that are also long-lived. Even though their total radiotoxicity is well below that of the uranium from which they were formed, concern exists about specific isotopes that are biologically important and might require special handling. The treatment and elimination of only 7 isotopes, either early or perhaps more easily after 300 years, would bring the toxicity level to about 1 million times lower than the toxicity of natural uranium. Here only Iodine-129 ($T_{½}$ = 17 M y), with a toxicity level ~40,000 less than natural uranium, will be mentioned, since it could accumulate in human thyroid tissue.

The pyroprocess of fuel recycling runs at about 500ºC. At this temperature iodine is volatile and consequently I-129 is captured as a vapour in cold traps. At a neutron absorption of 30 barns it can be relatively easily transmuted to I-130 in a thermal reactor. I-130 decays with a half-life of 12.3 h to stable Xenon-130. Thus a 17 M-year concern is changed to a 12.3-hour non-problem.

The other isotopes require different approaches for isolation, transmutation, or possible re-use such as zirconium, including Zr-93, as alloying element in FNR metal fuel (section 5.2).

7. Cost of recycling

Till and Chang have estimated the cost of recycling FNR fuel at 0.44 ¢/kWh, including capital, operation and storage of fission products; Bushby at the Canadian Nuclear Laboratory estimates a mid-price of 0.66 ¢/kWh for recycling alone [4,p.292; 16]. This suggests that FNR fuel cycling is similar in cost to purchase by OPG of fresh fuel plus its cost for disposal (0.5 + 0.2 ¢/kWh [18]).

8. Conclusion

In today's climate of reduce, re-use, and re-cycle, along with a strong worldwide emphasis on increasing the use of carbon-free energy, it seems utterly wrong to focus on the permanent disposal of Canada's used CANDU fuel which can currently provide about $65 trillion of non-carbon electrical energy over centuries. Its use can avoid the emission of about 275 billion tons of CO_2 to the atmosphere compared to the use of natural methane gas (470 billion for coal). That fuel resource requires no mining, and is already stored at reactor sites ready to be exploited.

If there were no other choice, and no further energy could be easily extracted by known means, one could condone burial of the used fuel, since it is highly radioactive and will be for millennia. But we, the nuclear establishment, know how to eliminate the long-term radiotoxicity by recycling the fuel through energy producing FNRs; and have done so. We therefore have that choice. Neutron physics created the radioactive material, and neutron physics has the ability to destroy it. It has been proven and has been accomplished safely -- with the help of Canadians [4].

Surely this is the path we should follow. It is a path where we can lead the world. Now.

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