## Accelerated Reduction of Used CANDU Fuel Waste with Fast-Neutron Reactors: Fuel Cycle Strategy Cuts TRU Waste Lifespan from 400,000 Years to Less than 80 Years

Peter Ottensmeyer University of Toronto, Ontario, Canada (peter.ottensmeyer@utoronto.ca)

#### Abstract

Canada's 45,000 tonnes of nuclear fuel waste contain over 99% heavy atoms whose nuclear energy can provide \$50 trillion of non-carbon electricity in fast-neutron reactors (FNRs), equivalent to 4000 years of nuclear power at present levels. FNRs can utilize the 98.9% uranium in CANDU fuel waste and also the 0.38% transuranic actinides which impose its 400,000-year radiotoxicity. Separation of uranium from CANDU nuclear fuel waste would permit refueling of FNRs primarily with transuranics, hugely accelerating the elimination of the long-term radiotoxicity of the CANDU fuel waste. Practicable separations of uranium would result in the complete elimination of the transuranics in about 80 years using FNRs at current Canadian nuclear energy output, while ideal separations could lower this to 16 years.

#### 1. Introduction

The million-year radiotoxic hazard of used nuclear fuel waste [1, p. 344] is a major worldwide concern that is generally being addressed with plans for sequestration of the material in deep geological repositories (DGRs). The 95% to 99% of this material from light-water and heavy-water reactors is still potential heavy-atom fuel which can be used up completely in fast-neutron reactors (FNRs) to produce 100 times more non-carbon energy than has already been extracted. For Canada's 45,000 tonnes of currently stored used nuclear fuel this would amount to \$50 trillion of electricity at current mid-peak time-of-use costs of 9.9 cents/kWh to the consumer in Ontario [2], plus useful cogenerated heat. This is so vast an energy that at current power production levels it would take about 4000 years to use up this resource [3]. Unless the lifespan of the hazard can be shortened, a 4000-year safe storage would still be required. We examine here the potential to accelerate the elimination of the long-term hazardous components of the used fuel by extracting the relatively benign uranium, and consuming initially the remainder of hazardous heavy atoms in fast-neutron reactors.

### 2. Used CANDU fuel

Canada's CANDU reactors create their massive power from the fission of natural uranium, in the process splitting just 0.74% of the heavy atoms into smaller atoms, the fission products (FPs). At the same time, neutron absorption changes some of the uranium atoms. They transmute into heavier elements, the transuranic actinides (TRUs) neptunium, plutonium, americium, curium, etc., that together constitute 0.38% of the used fuel. [1, p. 341].

The rather small percentage of fuel used is the result of the interaction of slow neutrons that are used in virtually all commercial nuclear reactors, including CANDUs. Slow neutrons very effectively fission the isotope U235, a 0.72% component in natural uranium, as well as a few TRU isotopes created in the reactor, such Pu239 and Pu241, the fissile isotopes. Slow neutrons do not fission the predominant isotope, the 99.28% U238, its fission cross section being about 50 million times smaller than that of U235 at low neutron energy. Nor do they fission TRU isotopes such as Pu240, Pu242, Am241, etc., causing such TRUs to accumulate in the fuel of the reactor.

The neutron interactions in the reactor are finely balanced. New, fast neutrons, are created with each fission of a heavy atom These fast neutrons are slowed down by heavy water surrounding the fuel bundles. They then very efficiently fission the few fissile isotopes U235, Pu239, Pu241, etc. to extract energy and create fission-product atoms (FPs) and new neutrons. In the process all excess neutrons are absorbed in the control rods, in heavy atoms in the fuel without causing fission, in the water, in the reactor structure, and in the growing numbers of FPs. Even though some fissile isotopes are created from U238 to augment the fission process, the FPs eventually absorb too many neutrons to maintain the reaction. In the CANDU reactor this occurs at 0.74% fission of heavy atoms, equivalent to the creation of 0.74 wt% fission products. The used fuel then has to be replaced with fresh fuel. In water-cooled reactors such used fuel is considered nuclear fuel waste. It is highly radioactive from its content of FPs and TRUs.

The radioactivity of the fission products consists primarily of beta rays (emission of electrons) and gamma rays (X rays emitted from the nucleus). The TRUs predominantly emit alpha rays (particles that are helium nuclei) as well as gamma rays. Of these emissions, alpha particles are about 20 times more injurious than either electrons or gamma rays of the same energy [4]. Adjusted for both energy and biological effect, Fig. 1 shows the radiotoxicity of the fission product component and the TRU component in used CANDU fuel relative to that of natural uranium. The fission product radiotoxicity decays to levels below that of natural uranium in less than 300 years, whereas the radiotoxicity of the TRUs starts at about 1000 times that of uranium and remains above uranium levels for about 400,000 years. This long-term radiotoxic hazard of the used nuclear fuel is a major worldwide concern.

Interestingly, the heavy atoms, including the hazardous TRUs, can all be fissioned in fast-neutron reactors (FNRs) and converted to fission products. Therefore FNRs provide a means to eliminate the long-term radiotoxic hazard of the TRUs, reducing the radiotoxic lifespan of the resulting waste from 400,000 years to the 300 year lifespan of the fission products (Fig. 1).

# **3.** Deep geological repositories

The world of nuclear nations, including Canada, has primarily concentrated on the potential use of deep geological repositories (DGRs) to sequester current and future used nuclear fuel waste for up to 1 million years [1, p.344] in whatever suitable rock strata are locally available. In Canada the task of nuclear waste disposal has fallen to the Nuclear Waste Management Organization (NWMO), following the Nuclear Fuel Waste Act [1, p. 330]. According to the



Figure 1. Evolution of Radiotoxicity from Used CANDU Fuel Components Relative to Natural Uranium.

Elimination of the transuranics (TRUs) in fast-neutron reactors and extraction of uranium (dashed horizontal line) would result in a huge reduction in radiotoxicity of used fuel waste, about 42,000X per unit time at 1000 years, and also shortens the time of decay to background levels from 400,000 years to 265 years. After 265 years the radiotoxicity of the fission products is lower than that of the mined natural uranium from which they are created in the reactor. The suggested 25-year point for processing of used fuel is indicated by the dotted line (see text). Note the log scales of both axes.

NWMO, to build such a DGR will cost between \$16 and \$24 billion depending on the capacity of the DGR, with total life-cycle costs estimated to be \$24.4 to \$40.7 billion [1, p.163; 5; 6, p.3].

The DGR costs would add close to 1  $\phi$ /kWh to the price of generating nuclear electricity [7]. More worrisome, the loss of just the currently stored used CANDU fuel in a closed DGR would mean about \$50 trillion in forgone non-carbon electricity [8]. It is worth looking at a realistic alternative to a DGR: a fast-neutron reactor facility.

# 3. The fast-neutron reactor and its fuel cycle

Research FNRs have been built since the 1950s in England, France, Germany, Russia, India, Japan and the US. Russia also has commercial FNRs. Past and present FNRs predominantly used uranium/plutonium oxide fuels, achieving a fuel utilization of 10 to 11 per cent [9-11]. Used fuel was normally not recycled, since fresh uranium fuel was and remains relatively inexpensive. Moreover, used fuel waste volumes were so small at the time that management and disposal were not a major concern then. This has changed, with current world estimates of used fuel waste being upwards of 350,000 tonnes, including Canada's 45,000 tonnes [5].

However, recycling of all of the used fuel is possible in FNRs, and has been proven safe. One FNR, the sodium-cooled metal-fuelled EBR-II in the U.S.A. recycled about 34,000 used fuel

pins, equivalent to five complete reactor refuelings during its 30 years of safe and uneventful operation ending in 1994 [12]. With its fuel canister design, the EBR-II regularly achieved 20% fuel utilization. A modification of that design achieved a 25% burn-up in the French Phenix reactor [13]. Our calculations based on the PRISM reactor design indicated that a 35% burn-up is theoretically possible before fuel replenishment is required [2,7,8].

A reactor modeled after the successful U.S. EBR-II is now commercially available, the GE-Hitachi PRISM reactor [14,15], and was recently offered to the UK for disposing of that country's excess plutonium as fuel. Moreover, FNR-associated fuel cycling facilities (FCFs) have been operating since 1996 to process the core fuel of the now-decommissioned EBR-II [16, p. 181]. Based on that experience, the cost of fuel cycling for a 1 GWe FNR facility was estimated at 0.443 ¢/kWh [16, p. 292]. These estimates have led to the conclusion that cycling used CANDU fuel in an FNR facility would be more economical than the non-productive disposal of that fuel "waste" in the currently planned DGR in Canada [7].

# 3.1. The fast-neutron reactor: replenishment with used CANDU fuel

Once initiated with enriched fuel, a fast-neutron reactor can replenish its used-up heavy-atom fuel complement effectively indefinitely directly from unenriched sources. There is no further need for enrichment of fissile material, since this is maintained internally by transmutation of U238 and other fertile isotopes (atoms that become more fissile by the absorption of a neutron). Mere extraction of the fission products would permit the fission reactions to continue, while concurrent heavy atom replenishment would reset the cycle. Such replenishment can come from natural uranium, depleted uranium, or, very advantageously, from already-stored used fuel from thermal reactors, including the CANDU reactors in Canada. The results of replenishment via the direct utilization of used CANDU fuel have been presented previously [2,7,8].

# 4. The long-term conundrum

The capability of FNRs to use and split of all uranium and TRU atoms would, at present nuclear power levels in Canada, result in a 4000-year fuel supply solely from currently stored used CANDU fuel waste [2]. This would suggest that a 4000-year safe storage might still be required before the current fuel waste has all been consumed. However, 98.9% of the "waste" is relatively benign uranium. Therefore by separating that uranium from the hazardous TRUs and using the latter in FNRs could greatly reduce the time needed to eliminate the long-term hazard that the TRUs constitute. In ideal circumstances that time can be as short as 16 years rather than 4000. This possibility as well as a practicable approach, are examined in the sections below.

# 5. Idealized accelerated consumption of used CANDU fuel "waste"

Used CANDU fuel consists of 98.88 wt% uranium, 0.38 wt% TRUs, and 0.74 wt% FPs [1, p.341]. Of these components the relatively innocuous uranium, if separated, could be stored safely indefinitely with minimal shielding. This uranium component is similar to natural uranium

but has been depleted of about two thirds of its fissile content of U235, rendering it unusable as a source of fuel in any thermal reactor, including the CANDUs. The uranium radioactivity, rather sparse alpha emissions, is dominated by U238 with a half-life of 4.5 billion years.

If all of the uranium were to be extracted cleanly from the used CANDU fuel, then, ideally, only the remaining 0.38 wt% heavy-atom TRUs could serve to replenish the fuel of the FNRs. The FP component would be removed later in the cycling of used FNR fuel by pyroprocessing described below [16,17]. The uranium would be stored as future FNR fuel for use after the hazardous TRUs in the stored CANDU fuel "wastes" have been consumed as immediate FNR fuel.

Under such idealized conditions a limit can be calculated of the time it would take to use up the contents of TRUs in the currently stored 45,000 tonnes of CANDU fuel "waste" if Canada's current nuclear power levels were furnished by FNRs. At the present annual use of ~1400 tonnes of fuel [5], the 45,000 tonnes could have accumulated in about 32 years, although in actual fact it took over 60 years since the beginning of nuclear power in Canada on June 4, 1962 [3, p.18]. The energy output achieved during that time is represented by the percentage of fission products created from the equivalent weight in heavy atoms, 0.74 wt% [1, p.341]. During the same time 0.38 wt% heavy-atom TRUs in the spent fuel were created in the CANDU reactors [1, p. 341].

If the FNRs were refueled only with the heavy-atom TRUs to create the same annual energy output as the CANDU reactors, the FNRs would have to fission a quantity of TRUs equal to the 0.74 wt% in heavy atoms, or 1.95 times the 0.38 wt% of TRUs from the stored fuel "waste". Thus under these idealized conditions all the TRUs, along with their long-term hazard, in the 45,000 tonnes of stored CANDU waste would be eliminated in the FNRs in a remarkably short 32/1.95 = 16.4 years. After that time the FNR fuel would be replenished for over 4000 years from the relatively non-toxic stored pure uranium extracted from the current fuel "waste".

### 6. Realistic accelerated consumption of used CANDU fuel "waste"

Current methods of extraction of uranium cannot provide such an idealized separation of uranium from the TRUs. However, a combination of at least two available extraction techniques described below, uranium nitrate crystallization and pyroprocessing, permits a more realistic assessment of the accelerated timeframes in which the TRUs could be eliminated from the stock of stored used CANDU fuel. Neither process purifies plutonium nor specific atomic isotopes, making them effectively proliferation resistant. The methods are likely not the optimum combination, nor already the most economical. However, they are indicative of what is possible.

Pyroprocessing is considered as the normal fuel cycling process for spent FNR fuel in metal form to separate FPs from the remaining heavy atoms [16, p.111]. It separates a large proportion of uranium from the other actinides and so could extract uranium from used CANDU fuel as well. However, the rather large volumes of used CANDU fuel containing relatively small percentages of TRUs suggest that crystallization of uranium in nitrate form is perhaps a better initial method of extracting uranium and thus also concentrating the TRUs among the remaining actinides.

- 5 of total pages -

## 6.1 Uranium nitrate crystallization for uranium extraction

The primary step of this procedure changes the oxide form of used CANDU fuel into the corresponding nitrate of all of the constituents. This process is very much akin to extraction of uranium from underground ore bodies by *in situ* leaching [18] in which acid administered via boreholes is used to dissolve the uranium oxide, which is then pumped out for further processing.

In the case of used fuel, all operations would be carried out remotely in shielded facilities. Nevertheless, some advantage could be gained in radiation protection and shielding costs by starting with used fuel that has exited the CANDU reactors 25 years ago or earlier. At 25 years the fission product radiotoxicity would have decreased over 1000-fold to a level that is close to 100 times the background of natural uranium (Fig. 1).

In brief, the used CANDU fuel, after being made accessible by shearing the fuel rods of the fuel bundle, would be dissolved in nitric acid [19]. The resulting uranyl nitrate dissolves readily in hot aqueous media (~600 g/L, [20, p. B-173]), less so in cold. Thus at 20°C about 65% of the uranium crystallizes out, increasing to 90% at 0°C, 96% at -10°C and 99% at -30 [21,22].

For simplicity it is assumed here that 90% of the uranium is extracted at 0°C, leaving close to 9% of the uranium, the TRUs and fission products in solution. Experiments have indicated that about 1% of the fission products would be entrained in the crystal mass [21]. As seen in Figure 1 for 25-year-old used fuel, this would make the overall level of radiotoxicity of the FPs in the crystal mass almost equal to that of uranium. Nevertheless, since uranyl nitrate is readily dissolved in hot water, re-crystallization would very much reduce the FPs further. These small amounts of FPs and any still-dissolved uranyl nitrate would be recovered simply by evaporation of the water and recycled or added to the 10% U/TRU/FP component for further processing.

The uranyl nitrate crystals can be converted to a suitable form, metal or oxide, for bulk storage. Then no precautions other than those now used for mined and purified uranium need be taken.

The 10% fraction of uranium/TRU/FP-nitrate can be converted to oxide, then to metal form [23], as input into the replenishment stream of the FNR fuel cycle pyroprocessing facility. In principle this 10% fraction, with the TRU/FP component concentrated 9 times, consists of the original 0.38 wt% TRUs, the 0.74 wt% FPs, and only 8.88 wt% uranium. The proportion of FPs would likely be lower, since the volatile FPs would be largely removed and captured in processing.

### 6.2 Pyroprocessing for FNR fuel cycling and uranium extraction

A fuel cycling facility using pyroprocessing has been developed at the Argonne National Laboratories in Idaho (now Idaho National Laboratories) for the extraction of FPs and the refabrication of fuel rods from the EBR-II used fuel. Even though the reactor was shut-down in 1994, the fuel cycling facility has continued to process the fuel from the core of reactor since 1996 [16, p. 181]. Moreover, designs and economic analyses exist for a similar cycling facility for a 1000 MWe FNR fuelled with metallic U/Pu/zirconium alloys [16].

The pyroprocess is the electrolytic separation at  $\sim 500 \text{ C}^{\circ}$  of uranium, the TRUs, and the FPs in spent metal fuel. It is carried out in shielded cells as a continuous remotely controlled operation. It is described in detail in "Plentiful Energy" by Till and Chang [16] as well as in the entire 1997 issue of Progress in Nuclear Energy [24].

In brief, the electrolytic process is divided into two major steps necessitated by the free-energy differences between uranium and the TRUs. The common start is the dismantling and cleaving of used fuel assemblies, with the resulting small metallic pieces filling an anode basket in the electrolytic cell of molten chlorides primarily of sodium, potassium and lithium.

In step one, a steel cathode is used to plate out only the uranium, after the latter dissolves as uranyl chloride along with the other actinides. Fission products either do not dissolve or remain dissolved in the molten salt. The difference in free energy between uranium- and plutonium-chlorides at equilibrium imposes a separation factor between the two elements at the steel cathode of  $1.2 \times 10^6$ . Therefore the plutonium does not plate out [16, p. 194]. Other TRUs behave like plutonium. Thus uranium is preferentially extracted at this cathode.

Step two is the extraction of a mixture of plutonium, the other TRU actinides, and the remaining uranium. The fission products remain in solution. This is accomplished by a second cathode in the form of a small vat of molten cadmium submerged in the molten salt. The molten cadmium drastically reduces the free-energy difference between uranium and plutonium/TRUs. Even so, plutonium and the other TRUs will not start to plate out and dissolve in the molten cadmium cathode until the concentration of uranyl chloride in solution has been reduced to about one third to one quarter of that of plutonium chloride via extraction of uranium at the iron cathode. Now solid uranium forms in the cadmium bath while plutonium forms an intermetallic compound,  $PuCd_6$ , until all the cadmium is used up, necessitating a replacement with a fresh liquid cadmium cathode. The other TRUs behave like plutonium [16, p. 355]. To recover the heavy metals from the cadmium cathode, the cadmium is distilled off at high temperature [16, p. 176].

Throughout this process the fission products either stay in solution or remain as solids. They are separated and free from actinides by factors that bring the actinide concentrations to radiotoxicity levels equivalent to natural uranium or lower [17].

It is clear from the above discussion that plutonium by itself is never separated, but that plutonium and the other TRUs are extracted together, along with a concentration of uranium determined by the functioning of the cadmium cathode. Since the cathode begins to function when the concentration of uranium is about one third to one quarter of the concentration of plutonium, this ratio can be taken as an initial estimate of the relative amounts of these two actinides extracted at the cadmium cathode. Experimental results indicate that in general the final concentration of uranium is higher than this [16, p.201].

- 7 of total pages -

For the sake of calculation, a 15% plutonium isotope content in the used FNR fuel is assumed here, which results in a corresponding ~4% uranium content in the Pu/TRU extracted at the cadmium cathode as suggested by the required Pu/U ratio of three to four, above.

## 6.3 Combined uranium extraction and FNR fuel replenishment

If used FNR fuel had a fuel burn-up of 15%, the extracted 15 wt% FPs could simply be replaced with an equivalent weight of used CANDU fuel from which 90% of the uranium has been extracted by crystallization, as described above. However, pyroprocessing, in which uranium is separated from the other used fuel constituents, offers a further enhancement in TRU usage.

FNR fuel at the end of its cycle would contain 70 wt% uranium, 15 wt% plutonium/TRUs, and 15 wt% FPs. In separating the Pu/TRU fraction during pyroprocessing, about ~4 wt% uranium was co-separated at the cadmium cathode, whereas the remaining 66% uranium was obtained in pure form at the steel cathode. This pure uranium could be stored, and replaced in the FNR fuel with more of the concentrated TRU-containing fraction from used CANDU fuel. Thus a total of 66% plus the 15% FPs, or 81% of the FNR fuel could be so replenished.

With this approach, the FNR fuel at the beginning of any subsequent FNR energy cycle would consist of 15 wt% of its original Pu/TRU content plus 4 wt% uranium co-extracted in pyroprocessing, with the other 81% made up with 8.1 multiples of the 10 wt% TRU/FP/uranium fraction from used CANDU fuel after uranium extraction by crystallization.

The 8.1 multiples comprising 81 wt% of the new FNR fuel would be reconstituted by 3.1 wt% TRUs, 71.9 wt% uranium, and 6.0 wt% FPs, each 8.1 times their weight in the 10% fraction (0.38 wt% TRUs, 0.74 wt% FPs, 8.88 wt% U). The FPs would be removed by pyroprocessing at the end of the next fuel cycle.

Each tonne of FNR fuel prepared in this way would result in the storage of 7.29 tonnes (90% of 8.1 tonnes) of pure uranium extracted from the used CANDU fuel plus another 0.66 tonnes of pure uranium extracted from the used FNR fuel by pyroprocessing for a total of 7.95 tonnes. The normal FNR refueling procedure would have been to replace the 15 wt% fission products with 0.15 tonnes of heavy atoms directly from used CANDU fuel per tonne of FNR fuel. Using the more concentrated TRUs as proposed here accelerates the elimination of long-term radiotoxicity in stored used CANDU fuel by a factor of 53, i.e. 7.95/0.15, over using the unseparated CANDU fuel "waste" for FNR refueling.

### 7. Fast-neutron reactor response to concentrated TRU fuel replenishment

Calculations were carried out for a PRISM-like FNR with ~30% fuel burn-up operating for two cycles with fuel replenished by fuel isotope concentrations as found directly in used CANDU fuel, i.e. with 0.38% TRUs, followed by cycles of fuel replenishment with TRUs concentrated to



3.1% prepared as outlined in Section 6, above. The results are shown in Fig. 2. For a 15% burnup results are qualitatively similar, with fuel recycling occurring twice as often.



Two cycles have spent FNR replenished with used CANDU fuel directly at months 1 and 41. Subsequent cycles starting at month 82 have FNR fuel replenished with used CANDU fuel from which 90% of the uranium has been extracted to increase the concentration of transuranic actinides (see text). Note: Scale on vertical axis of Panel A is linear. Scale for Panel B is logarithmic to accommodate the large variations in concentrations of the minor isotopes. The first two cycles indicate the equilibrium behaviour after the reactor fuel has been replenished with the unaltered used CANDU fuel composition. On changing to used fuel with the more concentrated TRUs at the beginning of the 3<sup>rd</sup> cycle, no major change in behaviour is seen in panel A, with only a slight reduction in starting U238 concentration to compensate for the increased TRU levels. Panel B shows that over the transition from cycle 2 to cycle 3 and beyond the power level is easily maintained (fission, black line). The initial concentration of Pu239 is noticeably increased above the initial levels in cycles 1 and 2, resulting in a greater usage and continuous decrease of Pu239 during the cycle. Pu240 also is increased and reaches a new steady state after two cycles. Most noticeable is the effect of the increased Am241 concentration on the levels of Am242. Since Am242 levels are very low in the used fuel, the levels of this isotope increase dramatically as it is created in the reactor by neutron absorption in Am241, reaching a new equilibrium level after one cycle.

The important observation is that even in this refuel regimen all of the actinides, uranium (including U236) and TRUs reach a new constant equilibrium level at the end of each new cycle. This indicates that all of the actinides, and particularly the TRUs from the used CANDU fuel, added as replenishment at the beginning of a cycle, are used up completely at the end of each cycle.

# 9. Acceleration of reduction of long-term radiotoxicity

Since the FNR readily accommodated the more concentrated TRU components in its fuel cycles, it is clear that the approach of extracting uranium from used CANDU fuel can accelerate the elimination of the TRUs in the fuel. Ideally for a 15% burn-up of fuel in one fuel cycle and replacement of the used 15% heavy atoms with a pure TRUs from used CANDU fuel would eliminate the TRUs in the stored CANDU fuel in 16.4 year (Section 5). A realistic extraction of uranium, as outlined in Section 6, would permit a replenishment of only 3.1% with TRUs rather than 15%, or a factor of 4.8 less. Therefore with FNRs equal in power output to the current CANDU reactors the time to eliminate the TRUs in used CANDU fuel, and with them their long-term radiotoxic hazard, would increase from the ideal 16.4 years to a practicable 79 years. Event the latter is a substantial time reduction in the long-term radiotoxic TRU hazard compared to the 4000-year time frame of using spent CANDU fuel directly.

### 10. Conclusions

All of the uranium and hazardous transuranic actinides that constitute over 99% of the currently stored 45,000 tonnes of highly radioactive used CANDU fuel "waste" can be consumed completely in refueling fast-neutron reactors (FNRs) to eliminate their long-term radiotoxicity. At present nuclear power levels in Canada such FNRs could deliver over 4000 years of non-carbon electricity from the "waste" alone, worth close to \$ 50 trillion at today's mid-peak time-of-use consumer price of 9.9 cents/kWh. To avoid the need for a very long-term safe storage before this fuel is used up, the FNRs can preferentially consume and eliminate the transuranic

actinides (TRUs) that constitute only 0.38% of the fuel but effectively represent all of the longterm, 400,000-year radiotoxicity. To realize this possibility the combined techniques of uranium nitrate crystallization and electrolytic uranium extraction by pyroprocessing could with relative ease extract at least 90% of the uranium in pure form for unproblematic storage as future FNR fuel. The remainder, a concentrated mixture of the TRUs, fission products, and the residual uranium would then constitute the immediate FNR fuel. With a number of FNRs equal in power output to Canada's current CANDU reactors this approach could eliminate the long-term hazard of the current 45,000 tons of used CANDU fuel waste in 79 years. If it were possible to extract all of the uranium cleanly and economically, such FNRs could eliminate the TRUs from all of the currently stored fuel waste in as few as 16 years. The stored, almost non-radioactive, pure depleted uranium would subsequently serve as fuel for the next 4000 years.

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#### 12. References

- Choosing a Way Forward: The Future Management of Canada's Used Nuclear Fuel. Final [1] Study. NWMO, 22 St. Clair Avenue East, Sixth Floor, Toronto, Ontario, M4T 2S3 Canada. www.nwmo.ca/studyreport/?action=downloadfile&id=341
- [2] Electricity prices, Ontario Energy Board. http://www.ontarioenergyboard.ca/OEB/Consumers/Electricity/Electricity+Prices
- P. Ottensmeyer, "CANDU fuel waste re-used, recycled, eliminated: \$45 trillion of [3] carbon-free electricity via fast-neutron reactors", Engineering Dimensions, Vol. 33, July/August, 2012, pp.47-50, 2012.

http://members.peo.on.ca/index.cfm/document/1/ci id/74178/la id/1

Curtis, Robert A. "Introduction to Ionizing Radiation," United States Department of [4] Labor, 2009.

www.osha.gov/SLTC/radiationionizing/introtoionizing/ionizinghandout.html. [5] "Moving Forward Together", Triennial Report 2008 to 2010. NWMO

- http://www.nwmo.ca/uploads managed/MediaFiles/1721 triennialreport2008to2010.pdf
- M. Hung, "Financial Implications of Used Fuel", APM-REP-03780-0001 December 2008 [6] http://www.nwmo.ca/uploads managed/MediaFiles/358 FinancialImplicationsofUsedFu elVolumeVariationinLongTermManagement2008Update.pdf
- P. Ottensmeyer, "Used CANDU fuel waste consumed and eliminated: environmentally [7] responsible, economically sound, energetically enormous." Proc. 33<sup>rd</sup> Ann. Conf. Can. Nucl. Soc., Saskatoon, June 10-13, 2012.

- [8] P. Ottensmeyer, "An alternative perspective. Used nuclear fuel waste: a \$36 trillion energy resource." Canadian Nuclear Society Bulletin. Vol. 31, 2010. p. 29-32.
- [9] "BN-600 Nuclear Fuel", Elemash Joint-Stock Company. www.elemash.ru/en/production/Products/NFCP/BN600
- [10] Golan, S., J. Leduc, and H. Nakagawa, "Liquid-metal fast reactors: Technical and economic status," IAEA Bulletin, 3/1989. p. 30-35. www.iaea.org/Publications/Magazines/Bulletin/Bull313/31304793035.pdf.
- [11] Mizuno, T., "Fast Reactor Fuel Development in Japan", Japan Atomic Energy Agency, Advanced Nuclear System Research and Development Directorate, 2009.
- [12] Hofman, G.L., L.C. Walters, and T.H. Bauer, "Metallic fast reactor fuels," Progress in Nuclear Energy. Vol. 31, 1997. p. 83-110.
- [13] S.L. Hayes and D.L. Porter, "SFR Fuel Performance and Approach to Qualification," DOE/NRC Seminar Series on Sodium Fast Reactors. http://www.ne.doe.gov/pdfFiles/NRCSeminarSFRFuels.pdf
- [14] Dubberly, A.E.,Boardman, C.E., Wu, T. and Yoshida, K. "G.E. SuperPRISM Oxide and Metal Fuel Core Design," 8th Int'n'l Conf. Nucl. Eng., ICONE 8, April 2-6, 2000, Baltimore MD.
- [15] Clark, D., "Nuclear waste-burning reactor moves a step closer to reality", The Guardian, July 9, 2012. <u>http://www.guardian.co.uk/environment/2012/jul/09/nuclear-waste-burning-reactor</u>
- [16] C.E. Till, Y.I.Chang, "Plentiful energy", CreateSpace (Pub.), 2011.
- [17] J.J. Laidler, J.E. Battles, W.E. Miller, J.P. Ackerman, E.L. Carls, "Development of pyroprocessing technology", Progress in Nucl. Engineering, Vol. 31, 1997, pp.131-140.
- [18] "In Situ Leach (ISL) Mining of Uranium", World Nuclear Organization: http://www.world-nuclear.org/info/inf27.html
- [19] H. M. Mineo, H. Isogai, Y. Morita and G. Uchiyama, "An Investigation into Dissolution Rate of Spent Nuclear Fuel in Aqueous Reprocessing," J. Nucl. Sci. Technol., Vol. 41, no. 2, pp. 126-134, 2004.
- [20] Handbook of Chemistry and Physics, 58<sup>th</sup> ed., Chemical Rubber Company Press, 1978.
- [21] T. Takata, Y. Koma, Sato, Koji, Kamiya, Masayoshi, A. Shibata, K. Nomura, H. Ogino, Koyama, Tomozo and S.-i. Aose, "Conceptual Design Study on Advanced Aqueous Reprocessing System for Fast Reactor Fuel Cycle," J. Nucl. Sci. Technol., Vol. 41, no. 3, pp. 307-314, 2004.
- [22] R. Hart and G. Morris, "Crystallization temperatures of uranyl ntirate-nitric acid solutions," Prog. Nucl. Energy, Vol. III, p. 544, 1958.
- [23] Kitts, F.G., "Pilot-scale demonstration of the modified direct denitration process to prepare uranium oxide for fuel fabrication evaluation", Oak Ridge National Laboratory, Oak Ridge, Tenn., Report ORNL/TM-12726, April 1994: http://www.ornl.gov/info/reports/1994/3445603813443.pdf
- [24] Hannum, W.H., guest editor. "The Technology of the Integral Fast Reactor and its Associated Fuel Cycle", Progress in Nuclear Energy, Vol. 31, No. 1/2, p. 1-217.