NWMO ADVISORY COUNCIL DISCUSSION PAPER

CANDU Spent Fuel: a Waste or a Resource?

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Summary: this paper has been presented to the NWMO Advisory Council for discussion. It offers some technical insight on the disposal of spent CANDU fuel from the perspective of the isotopic composition, and focuses on the economic incentives for reprocessing. The data used in the paper is mostly generic (i.e. not specific to a particular power plant) and was taken from lecture notes prepared by the author for a graduate course in Nuclear Fuel Management at École Polytechnique.

Introduction

As an integral part of its study, the NWMO has taken numerous steps to engage the public in the elaboration of a proposed management option, while exploring the social and ethical aspects as well as the scientific/technical considerations. *Making the complicated scientific and technical issues surrounding nuclear fuel cycles more understandable to the layman is of particular importance* if he is to be part of the discussion (as he must).

Accessibility

It has been reported that one of the key findings of the CPRN study was that citizens want to make sure the nuclear waste is accessible to future generations that may have new knowledge and technology to either manage the waste or re-use. This is not all that surprising. When ordinary citizens were consulted, they were presented with seemingly well established facts such as:

- the wastes are there, and we are talking *millions* of spent fuel bundles; the volume of wastes is not negligeable, and it is going to take years to collect it.
- they constitute an unprecedented hazard to the environment (or to mankind) in view of their extreme radiotoxicity and longevity, and must be closely guarded;
- using the best current scientific know-how, it seems essentially impossible to demonstrate the viability of any approach over thousands of years (that is of course assuming we *had* a plan to manage them).

Citizens were then asked *'What should we do?'* and the answer was: *'What do I know? Who can we trust? Whatever you do, don't hide them away'*.

Indeed, looking at Canada's history regarding used fuel management, the NWMO president said recently *(NucNet US, Sept.3 news brief)* that the original AECL geologic disposal concept was "essentially a proposal that didn't accommodate the concept of accessibility". We can therefore expect that the concept of accessibility will play a key role in the NWMO final recommendations.

However, ***a minimal understanding of the fuel isotopics is essential if we are to make informed judgments on the fuel management options.*** With this in mind, one can start by asking the question: *should the used fuel be considered a waste or should it be considered a resource?* The answer to this question will have a great influence on the waste management strategy and in particular on the sitting requirements for centralized storage/disposal.

In either case, waste or resource, serious socio-economic considerations are implied. The answer to these is far from obvious, but the economic potential should be addressed first, since it can greatly influence both the quantity and the degree of toxicity of the wastes we will ultimately have to deal with. For this, we need to look at the spent fuel composition.

CANDU Used Fuel Isotopic Composition

Do CANDU spent fuel bundles really have no economic potential? What about the Light Water Reactor (LWR) spent fuel, which represents more than 80% of the world's high level wastes? And what about isotopes other than uranium in the spent fuel. Do they present sufficient economic interest to justify safekeeping the spent fuel for future generations, until technological and scientific advances as well as resource depletion make reprocessing economical?

The quantity of wastes

In 2001, there were 438 nuclear reactors in operation around the world with an installed capacity of 351 GW.[1] This compares to a total capacity of 109 GW in Canada from all sources in the same year. Of these, 347 are of the LWR type, producing over 93% of the world nuclear electricity. The energy produced per unit fuel (fuel burnup) in these reactors ranges from 32-40 GWd/t, while the fresh fuel enrichment in U-235 varies from 3.2% to 4%. CANDU reactors on the other hand use natural uranium and achieve on average a discharge burnup of only 7.5 GWd/t.[2] Taking an average enrichment of 3.5% and an average burnup of 35 GWd/t for LWR's, this implies that approximately 8400 t of spent LWR fuel is produced each year, compared to 1760 t of spent CANDU fuel.[3] Since fuel enrichment for LWR's also produces about 6 times more depleted uranium, nearly 50 000t of depleted uranium is accumulated every year at enrichment plants around the world. The overall material flow can be summarized as follows:

Table 1 World Nuclear Fuel Consumption (per year)

	Installed capacity	Spent Fuel (per year)	Recoverable Uranium
CANDU	15 GW	1760 t	1720 t
LWR	335 GW	8400 t	58100 t*
Total	350 GW	10169 t	59820 t
%CANDU	4.3%	17.3%	2.9%

* including 50000 t of depleted uranium (99.8% U-238) from enrichment tails

We observe:

- CANDU, with only 4% of the energy, produces 17% of the highly radioactive wastes
- all the existing power reactors are 'thermal' reactors, in the sense than the neutron chain reaction is sustained with the aid of a 'moderator' material which slows the fast fission neutrons down to 'thermal' energies before they are absorbed in fuel (to cause additional fissions). The most common moderator materials are light water, heavy water and graphite. These reactors are of second generation, and many have had their life cycle extended to 50y-60y since construction.
- any nuclear expansion scenario for the future will ***necessarily*** involve the following steps:

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[1] OECD Nuclear Energy Agency, Nuclear Energy Data - 2001

[2] This is a conservative estimate. Factors affecting the average discharge burnup are mostly related to neutron economy in the reactor (the presence of control rods, the size of the reactor which influences the rate of neutron leakage, etc…).

[3] Assumptions : 85% average Capacity Factor, 35% thermal efficiency of the turbine, 200 MeV per fission.

  - next generation reactors (**Generation III**) are about to be brought to market, when and if the market opens up. These reactors are essentially ready to be built. For Canada, these include the Advanced CANDU Reactor (ACR), under development at AECL for the past 5 years. Compared to the current technology, this reactor will be more compact (with slightly enriched fuel), will be less expensive to build (by 30-40%), under a significantly reduced construction schedule (reduced from 72 months to 48 months), and with significantly more inherent safety features (as opposed to engineered safety). With a burnup above 20GWd/t, the number of the spent fuel bundles will also be 3 times smaller. These new reactors could be built during the period 2010-2025 in Canada, the U.S., the U.K. and elsewhere.[4]
  - It may take 15-20 years of R&D to bring another generation of advanced reactors to market (**Generation IV**). The scientific and engineering basis for these reactors is already well established, some going back 40 years when prototypes were built. But major R&D programs will still be needed to improve the designs and make them viable for commercialization. Under the initiative of the U.S. DOE, a 10-nation cooperative effort (including Canada) has produced a Generation IV technology Roadmap, selecting the six most promising reactor designs.[5] These new reactors will have a much higher thermal efficiency (>45%) and will open up new avenues for the direct use of nuclear heat (for example high temperature processes for the production of hydrogen). With large scale reprocessing and recycling, these reactors will also allow to 'close the fuel cycle' and extend uranium resources far into the future. These advanced 'breeder' reactors will essentially consume U-238, by converting it to fissile Pu-239 and recycling. Alternative fuel cycles are also possible using thorium fuel, which is as abundant as uranium.
- 50 000 t per year accumulated world-wide between 1975 and 2025 equals **2 500 000 t of depleted uranium** (chemically pure and no more radioactive than natural uranium). If 50% burnup is eventually achieved with Generation IV reactors, this would represent a total of 12 000 000 TWh, i.e. **850** times the current total world electricity consumption.

Clearly, there is *an immense potential for energy production from the used nuclear fuel*. However, this enormous potential will become available ***only if there is an expansion of nuclear power***, and this will not occur unless proliferation and environmental issues surrounding reprocessing and waste management are resolved. This may take a while.

Spent fuel composition

The question is then, will Canadians <u>ever</u> be interested in recovering the uranium content of the CANDU spent fuel? We shall see that this is very doubtful when we consider the relative composition of the spent fuels from both CANDU's and LWR's.

The majority of LWR reactors are of the Pressurized Water Reactor type. (>75%), so PWR composition is taken as typical for all LWR's. Obviously, the detailed composition of the spent fuel can vary somewhat from bundle to bundle, or from one PWR fuel assembly to the next, depending on the particular power history of the individual fuel elements. The numbers below were calculated for typical CANDU-6 fuel bundles and for standard PWR fuel (17x17) in typical reactor assemblies using sophisticated computer codes developed at École Polytechnique. Details have been reported in the open literature.[6]

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[4] The prospect of an early introduction in the US market has been delayed by a recent decision by Dominion Resources of Richmond, Virginia, to withdraw from its cooperative effort with AECL to test the U.S. NRC process for obtaining a combined Construction and Operating license for the ACR in the U.S.

[5] *A Technology Roadmap for Generation IV Nuclear Energy Systems*, GIF-001-00, U.S. DOE, March 2003.

[6] D. Rozon, W. Shen, *'A Parametric Study of the DUPIC Fuel Cycle to Reflect PWR Fuel Management Strategy*,' Nucl. Sci. Eng., **138**, 1 (2001)

Table 2 The composition of used nuclear fuel

isotope	***half-life***	***CANDU***		***PWR***	
		fresh	used	fresh	used
U-238	4 400 000 000 y	99.28%	98.58%	96.8%	93.79
U-235 (fissile)	***710 000 000 y***	***0.72%***	***0.23%***	***3.2%***	***0.91%***
U-236	23 000 000 y	-	0.07%	-	0.40%
Pu-239 (fissile)	***24 000 y***	***-***	***0.25%***	***-***	***0.59%***
Pu-240	6 600 y	-	0.10%	-	0.23%
Pu-241 (fissile)	***14.3 y***	***-***	***0.02%***	***-***	***0.08%***
Pu-242	360 000 y	-	0.01%	-	0.05%
fission products and minor actinides	variable	-	0.74%	-	3.21%

A number of observations can be made.

Nuclear reactors

A nuclear reactor requires a finite (critical) mass of fissile material to be able to sustain the chain reaction and function properly. As fission occurs, producing power, there is a slow depletion of fissile isotopes in the fuel which must be compensated by regular refueling to maintain criticality. The resulting fueling rate determines the degree of exposure of the discharged fuel (i.e. the fuel burnup). As opposed to light water, heavy water absorbs very little neutrons, and as a result, a much lower inventory of fissile material is required to achieve criticality in a CANDU reactor. This explains why CANDU reactors can operate with natural uranium (with a fissile content of less than 1%), and why fresh fuel needs to be enriched in U-235 in LWR's. In addition, with on-power refueling, CANDU reactors can maintain a minimum of excess fissile material in the core. On the other hand, LWR's need to be shut down for refueling about once a year; they therefore require more excess fissile material to carry over the year-long production campaigns. As a result, the fuel resides longer in the reactor and the fuel discharge burnup is higher in LWR's.

Recovered Uranium (RU)

There is a worldwide market for natural uranium and for uranium enrichment services. Commercial reprocessing of used fuel from LWR power reactors is carried out routinely in Europe, with an installed reprocessing capacity of nearly 3000 t/y (see the Jackson background paper, Table 6). As can be seen in table 2 above, if we ever decide to reprocess and partition the used CANDU fuel, the fissile content of the recovered uranium (i.e. the U-235 content) would be only 0.23%. *This is about the same level as the depleted tails assay in uranium enrichment plants*. Also, Canada is a leader in uranium mining and Canadian uranium reserves are far from being depleted. The cost of reprocessing is quite high and is not about to be exceeded by the cost of mined natural uranium. Obviously, to obtain uranium of the same quality as the recoverable uranium, it would always be much cheaper to simply purchase the non-radioactive depleted uranium from the vast inventories available at enrichment plants rather than reprocess CANDU spent for its uranium content. And of course, if you wanted to recycle the RU from CANDU spent fuel, you would need to separately acquire *additional fissile material* to be able to run the reactors.

There is therefore very little incentive to reprocess used fuel in Canada anytime in the future if the sole purpose is to recover the uranium

For LWR's, the residual fissile content is much higher, *exceeding that of natural uranium.* There is therefore some incentive to recycle the RU from LWR's, saving both in natural uranium feed and in enrichment services to fabricate new fuel. There is a complication however. Although U-235 is fissile, fission does not always occur when a neutron is absorbed. About 15% of the time, transmutation occurs, creating U-236 (non fissile).

The residual U-236 in the used fuel becomes significant if the RU is to be recycled in LWR's because it will then interfere with the enrichment process when the RU is re-enriched to reactor grade, requiring additional separative work for the same output. As a result, most of the uranium recovered at the existing reprocessing facilities is currently stockpiled, to be used when the economic conditions become more favorable (higher natural uranium prices).

Since the residual enrichment of the RU from LWR's is higher than that of natural uranium, it would be much simpler to use it to fabricate CANDU fuel and recycle the recovered uranium from LWR'S in CANDU reactors. This could add about 50% to the energy recovered from the original uranium, and would avoid the re-enrichment step. This is a good example of a ***tandem fuel cycle***, involving two different types of reactors, yet achieving a gain in overall resource utilization. Since there are relatively few CANDU's around the world, such tandem fuel cycles would not have much effect on the global uranium utilization.

Plutonium recovery from CANDU spent fuel

We note in Table 2 that a seemingly small amount of Plutonium-239 is created by neutron capture in U-238. As it turns out, Pu-239 is fissile, and a fraction of this plutonium undergoes fission in situ, contributing significantly to the total energy production of the reactor (nearly 40% in CANDU). As with U-235, fission does not always occur in Pu-239 when a neutron is absorbed. About 25% of the time, neutron capture leads to the formation of Pu-240. The Pu-240 is non fissile but it strongly absorbs neutrons, leading in turn to a smaller quantity of Pu-241. **The used fuel thus contains diminishing amounts of Pu isotopes starting with Pu-239, and only a fraction of these are fissile isotopes**.[7]

Because of the parasitic absorption of Pu-240, weapons-grade plutonium requires concentrations of Pu-239 higher than 90%. In the table above, we note that the fissile fraction of the recovered plutonium is only 70-71% in both CANDU and LWR fuel. This means that spent fuel from power reactors is far from ideal for nuclear weapons production. In fact, my understanding is that IAEA concerns about proliferation are primarily aimed at the nuclear facilities themselves, which could be used to divert and prematurely recover spent fuel with a higher fissile plutonium ratio. In other words, the spent fuel, *particularily in un-reprocessed form,* does not pose a major proliferation risk.[8]

We can easily calculate the amount of fissile Pu contained in the spent fuel inventory: (1760 t/y spent fuel)x(0.27% fissile Pu) = 4752 kg fissile Pu per year. Over a 50 y period (expected life cycle of existing reactors), this represents a total of nearly **240 t of fissile Pu**.

What is the economic potential of the plutonium in CANDU spent fuel? To answer this we must look at the alternatives. We can either dispose of the plutonium directly by fission, without creating any new plutonium, or we can recycle the plutonium mixed with uranium, in which case additional plutonium will be created.

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[7] The used fuel also contains trace amounts of Pu-238, due to the decay of other actinides.

[8] Of course, security threats are another matter.

Plutonium burner (actinide burner)

It has been shown that CANDU reactors could in fact be used as 'Plutonium Burners', *with fuel containing no uranium*, but only the plutonium and the other actinides in an *inert matrix*. Calculations show that 80% Pu destruction (by fission) per pass could be achieved.[9] It is quite unlikely that alternative means of disposing of the plutonium (including intense neutron sources with accelerators), would ever be more economical and as effective as Plutonium Burner CANDU reactors for one simple reason: the existing CANDU reactor designs, with known costs and reliability, are readily adaptable for this function. Most of the R&D would be in the fuel development area. Development of new fuel involves many experimental irradiations in research reactors over a number of years. Such reactors could probably be built within 10-15 years notice anytime in the future (when and if the CANDU reprocessing route is adopted).

Conservatively assuming a depletion of 80% by fission of the total plutonium inventory, the energy release represents a ***potential of 1890 TWh of electricity***, or the equivalent of 3.25 years of total electricity production in Canada. At a cheap 0.05$/kWh, the potential revenues are of the order of **100 B$.**

How does this compare to the ***cost of reprocessing***? I really don't know. However, it should be less expensive to reprocess CANDU spent fuel than to reprocess LWR spent fuel because of the lower burnup and the much lower radioactivity level. On the other hand, the concentration of Pu is 3.5 times lower in spent CANDU fuel. Assuming an optimistic cost of the order of 500 $/kg for reprocessing CANDU fuel[10], recovering the plutonium from 88 000 t of CANDU spent fuel (1760 t per year for 50 years) would accumulate to approximately **44 B$**. This is a very crude estimate, which does not account for the fuel fabrication costs, the transportation costs and the important reactor construction and operating costs. It is therefore dubious that a plutonium burner reactor would ever be economical.

On the other hand, the Pu inventory also has ***strategic value***, because much more energy could ultimately be derived from the recovered Pu if it were used as a basic **fissile feed material to start up more advanced fuel cycles**, such as thorium fuelled CANDU reactors or fast breeder reactors (Generation IV reactors).

Plutonium recycle

Recycling the recovered plutonium from CANDU reprocessing could be achieved in a number of ways:

- it could be recycled as **MOX** CANDU fuel, mixing the plutonium oxide with quantities of uranium to fabricate CANDU fuel bundles. This is not very interesting because it only extends the resource by maybe 50%, and requires many reprocessing cycles to recover all the energy. In other words, we could save as much as 50% of the natural uranium supply, but we would end up with as much wastes 25 years down the road.
- it could be more efficient to use the plutonium as fissile feed material in **thorium** fuelled CANDU reactors.[11] A once-through cycle is most economical, but by varying the initial plutonium content, and allowing for additional reprocessing of the thorium used fuel (new technology), self-sufficient fuel cycles could be achieved (i.e. the used thorium fuel would contain as much U-233 as it did when it was fresh). This option requires the development of new fuel and of new reprocessing technology. A period of 10-20 years would likely be needed for the R&D

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[9] P.G. Boczar et al., *'Plutonium Dispositioning in CANDU'*, AECL-11429

[10] compared to approx. 1000$US/kg for reprocessing LWR spent fuel

[11] Natural thorium is mostly made up of Th-232. Like U-238, it is non fissile. Neutron capture in Th-232 leads to the formation of U-233, another fissile isotope of uranium (half-life 150 ky).

The presence of plutonium in the used fuel obviously adds value to the used fuel. There is no incentive to reprocess at present, because current fuel costs represent less than 5% of the total costs in a CANDU. *Nevertheless, there appears to be some economic potential for plutonium recovery from spent CANDU fuel.* Economic conditions could be much different in 25 y or 50 y.

The waste management approach should therefore ensure accessibility for a sufficiently long time for a decision on commercial reprocessing to be made by future generations. ***This decision will obviously be linked with energy policy and the future role of nuclear power.***

Used fuel viewed as a waste

An economic potential may thus very well exist for reprocessing CANDU spent fuel and for recycling the recovered plutonium. We should therefore think twice before disposing of the used fuel and depriving future generations of the potential benefits. However, after considering the relative impact of alternative solutions, it may be decided on balance to limit or even phase out nuclear power over the next 25 years. The existing spent fuel should then obviously be viewed as a waste.

In order to leave future generations with a real choice, we should now find a site for deep geological disposal and provisions should be made to pay for it from the current revenues of nuclear electricity. There are two possibilities for the long-term management options:

- if the post-closure performance of a particular site is deemed adequate for deep-geological disposal of the ***unreprocessed*** fuel bundles, the strategy should then be the final burial of the wastes in a repository, after proper packaging. The AECL disposal concept, or a variation thereof, could serve as a model;

- if on the other hand, no site can be found for which it can be shown that very long-term releases from the repository would satisfy regulatory limits in a credible scenario (and thus pose an unacceptable risk to future generations), then extended centralized surface or near-surface storage may become necessary. *Retrievable storage* would thus need to be planned for a sufficiently long period so that ***reprocessing facilities*** can be built *on the chosen repository site*. After partitioning, transmutation of selected fractions of the high-level wastes could then be achieved using nuclear reactors or accelerator-driven intense neutron sources so as attenuate or mitigate the impact of long-term releases. These reactors would also have to be built on the same site. The final high-level wastes (with reduced half-lives) could then finally be conditioned (vitrified) for deep geological burial.

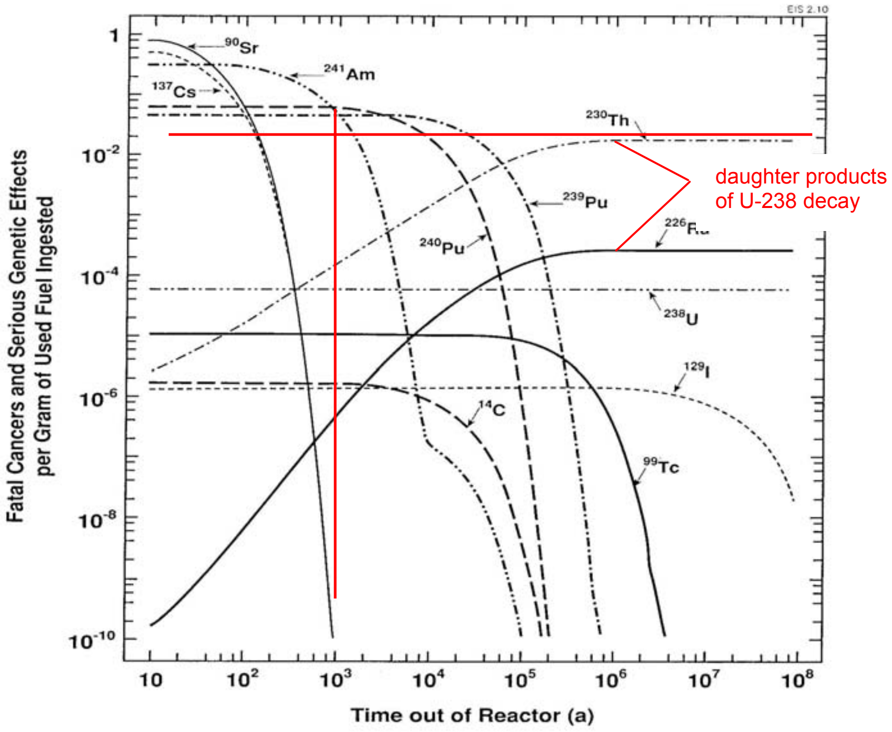
It is stated in the Jackson background paper (p7) that

> *'Based on the variety of actinides present, it can be inferred that attempts to reduce the quantity of actinides by the absorption of the relatively slow (thermal) neutrons found in LWR and CANDU power reactors would just produce even more actinides and little progress would be achieved in destroying them.*

This is particularly true if the fuel contains uranium. As I mentioned above, it is also possible to fabricate CANDU fuel containing only the plutonium and the other actinides in an inert matrix (**Plutonium Burner**). Removal of the uranium from the fuel reduces significantly the absorption of neutrons in the reactor, and criticality can then be achieved with relatively small amounts of fissile material. Almost 80% of the plutonium could be destroyed by fission, transforming the long-lived

Pu-239 and Pu-241 into short-lived fission products. Even if new actinides are created, there would still be a very significant reduction in long-term radiotoxicity.

This is made clear when we look at Fig 3. of the Jackson background paper:



The following observations can be made:

- this graph measures the contribution of individual isotopes to the radiotoxicity of the used fuel.
- while it is true that it will take one million years for the ***radioactivity*** level of the used fuel to drop the level of uranium ore (see figure 2 of the background paper), the ***radiotoxicity*** of the used fuel drops more rapidly and reaches uranium ore levels much sooner (in less than 100 000 y)
- fresh fuel is less radiotoxic than natural uranium ore which contains Th-230 and Ra-226, the daughter products of the very slow decay of U-238. In the spent fuel, it takes about one million years for these products to build up again and reach a secular equilibrium.
- a reasonable objective would be that the wastes should be confined until the radiotoxicity reaches levels comparable to natural uranium ore. That level is indicated by the Th-230 line in the figure above.
- we observe that ***between 2 000 y and 100 000 y, the radiotoxicity of the used fuel is essentially determined by the presence of Pu-239*** (with a half-life of 24 000 y)

It would then seem that if we must reduce the radiotoxicity of the spent fuel by transmutation, the best way to proceed is to eliminate as much of the Pu-239 as possible. This could reduce the horizon for confinement of the wastes to a more manageable 1000 to 2000 years and actually reduce the toxicity far below that of uranium ore for a few hundred thousand years…

In conclusion, ***the only way to actually reduce the toxicity level of the nuclear wastes via transmutation is through reprocessing and partitioning***, followed by fuel fabrication and recycling in a nuclear reactor[12]. It is an illusion to believe that future discoveries will allow us to treat the bulky unpartitioned wastes with a mysterious ray that will magically transmute the nuclides and reduce the half-life. This expectation is akin to the old alchemist dream of the middle ages. No future scientific discovery can ever contradict what has already been established and verified in nuclear physics in the past 100 years, any more than Einstein's relativity theory or quantum mechanics contradicted Newton's laws or all of classical mechanics.

When addressing the social/ethical issues surrounding the management of used nuclear fuel, one must not confuse Peter's Principle or Murphey's Law with the laws of physics. In neglecting important scientific/technical considerations, legitimate concerns have been turned into articles of faith by some interveners. It seems somewhat paradoxical that while some believe scientists are generally wrong about the future (a complete lack of trust in the science-based models), the same people trust them to find a magical solution to our currently insurmountable problem. I call this is the 'science fiction' syndrome.

Daniel Rozon
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[12] A critical reactor or even an accelerator-driven subcritical assembly