BENEFITS OF FAST NEUTRONS: CANDU FUEL CREATION, WASTE ELIMINATION AND CLEAN POWER

F.P. Ottensmeyer
University of Toronto, Ontario, Canada
peter.ottensmeyer@utoronto.ca

Abstract

At the current rate of mining and exploitation of Canada's uranium resources, our economical uranium (< US$120/kg) as fuel for thermal reactors, including CANDUs, will be exhausted in about 35 years. At the same time it is estimated that Canada's highly radioactive long-term nuclear fuel waste will approach 100,000 tonnes of used CANDU fuel. However, the addition of fast-neutron reactors (FNRs) to our CANDU fleet as small modular FNRs or larger plants with recycling facilities can re-utilize this waste as fuel to extract over 100 times more energy from it. At the same time this approach can eliminate the long-term radiotoxic transuranic heavy atoms from the stored waste and produce fissile fuel for the CANDU fleet for many centuries. With recycling through FNRs no further long-term transuranic waste is created, leaving primarily short-lived fission products as the only fuel residue.

1. Introduction

The CANDU reactor, a heavy-water-cooled and –moderated nuclear power plant, is one of the most efficient thermal reactors, extracting about 0.74% of the energy in natural uranium fuel. Other thermal reactors, cooled by normal (light) water lose more neutrons to that coolant/moderator with the consequence that the overall fuel use from mined natural uranium drops below that of the CANDU reactor.

However, at 0.74% efficiently or less, thermal reactors are rather wasteful of uranium fuel. It is only the very high energy density in nuclear fuel, uranium in this case, that has us accept such a profligate approach to creating energy.

Nevertheless, there will be an end to this practice as economical world uranium reserves dwindle. The World Nuclear Organization in 2017 indicated that Canada's economically mined uranium reserves (below US $120/kg) stood at 494,000 tonnes [1]. Since Canada uses less than 2000 tonnes annually to fuel its current fleet of 18 CANDU reactors, this reserve appears to be ample for a couple of centuries. However, CAMECO and AREVA were mining, using and exporting the uranium from Canada at a rate of 14,022 tonnes in 2016. At that rate the Canadian economical reserves will be exhausted in 35 years, or by the year 2051.

Economical world reserves of uranium will last a few decades longer. Shortening of that time will be caused by an increasing world hunger for energy as well as the recent emphasis on shifting from CO_2-emitting fossil fuels to non-carbon sources of energy, including nuclear power.

These scenarios seem to presage the demise of the nuclear industry for lack of future fuel. Even an expensive effort of extracting the remaining fissile portions of used fuel for further use in thermal reactors would provide but a percentage increase in the time of available nuclear power. However, shifting to a complementary nuclear technology, fast-neutron reactors (FNRs), can multiply the available energy by a factor of one hundred or more. FNRs require enriched fuel to start, but they can subsequently without further fuel enrichment derive energy from the re-use and recycling of existing used thermal reactor fuel as well as from the masses of depleted uranium left behind during the enrichment of fuel for world’s light-water reactors.

2. Historical Neglect

It was a fast-neutron reactor that created the first electricity from nuclear energy, with power from the EBR-I in Arco, Idaho, lighting up four 100 watt light bulbs on December 20, 1951 (Fig. 1) [2]. The next day the reactor powered the entire building.

However, there was a desire by the US Navy to use a nuclear power plant to propel its submarines. Since nuclear plants do not require air to produce energy, a submarine with such a power plant could stay submerged and hidden for weeks or months. Among the still experimental nuclear plants being tried at the time, the admiral, Rickover, very logically chose a water-cooled reactor for his ships. For such applications fuel efficiency and nuclear fuel waste were not a primary concern for the military.

The rest is history. The military developments of the water-cooled reactors were taken up commercially, and water-cooled reactors became the dominant form of nuclear power on land as well, worldwide.

Figure 1. String of 100 watt light bulbs powered by first electricity produced by nuclear power on December 20, 1951 using the EBR-I fast-neutron reactor
https://www.flickr.com/photos/argonne/8167845201/in/photostream/

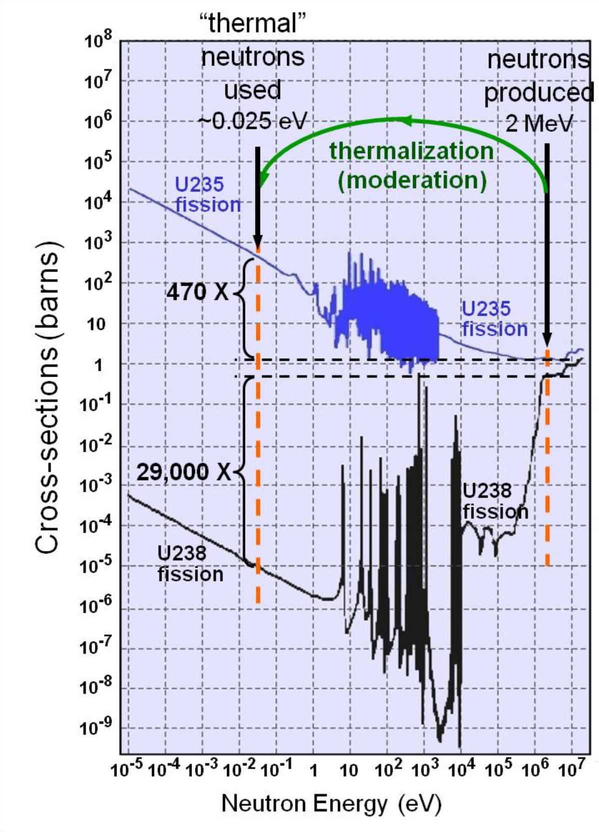

Figure 2. Cross sections for fission of U235 and U235 by neutrons of different energies as the latter are slowed from their initial high energy (20,000 km/sec) to about 2 km/sec at thermal energies [5].

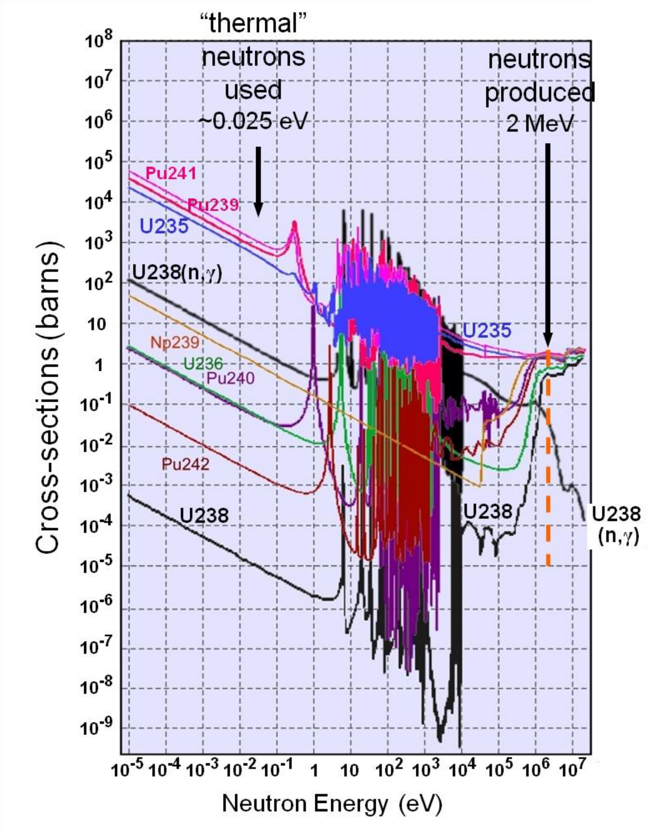

Figure 3. Cross section for fission of transuranic isotopes of neptunium (Np) and plutonium (Pu) produced by the absorption of neutrons by U238 (shown as U238(n,γ)) [5].

Fast-neutron reactors (FNRs) in most countries were developed and operated as experimental reactors, of smallish size such as the 20 MWe EBR-II in the USA, or of a fair size, such at the 250 MWe Phenix reactor in France which produced power from 1973 to 2009. Russia produced commercial FNRs, such as the 350 MWe BN-350, operating from 1972 to 1994, the BN-600 from 1980 to the present, and recently added its largest FNR to the electrical grid, the BN-800 in 2016.

In the USA and Canada the almost singular focus on water-cooled reactors had the consequence that education and maintenance of knowledge of fast neutron physics decayed. By 2005 only one university on North American continent, the University of Wisconsin in Madison, still had a course on FNRs.

Elsewhere fast neutrons were considered a hindrance to the effective extraction of energy from heavy nuclei by slow or thermal neutrons, and the focus was on efficient thermalization of fast nascent or primary neutrons produced by the fission reaction in the core of the reactor [3,4].

Statements on fast neutrons in specialized lecture notes today include such misinformation gems as: "primary neutrons have too much energy for neutron scattering" [3], or "a neutron moderator reduces the velocity of fast neutrons, thereby turning them into thermal neutrons capable of sustaining a nuclear chain reaction" [4]. Both statements are clearly not true in fact or inference. Primary fast neutrons must scatter to become thermal neutrons, and they certainly sustain chain reactions as well or the EBR-I fast-neutron reactor, above, would not have produced the first electricity ever generated by nuclear power.

Two somewhat historical texts exist on the EBR-II and one update from 1980 on FNRs [9-11]. Therefore it seems necessary to resurrect some ancient knowledge in order to explain how fast-neutron reactors can re-utilize used CANDU fuel, extract over 100 times more energy from that fuel by consuming it completely and produce fissile fuel for our CANDU reactors. Such are the beneficial characteristics that such a fast-neutron reactor can even load-follow, being immune to xenon poisoning experienced by thermal reactors. Of necessity, only a few main points can be covered in the discussion.

3. Neutron Characteristics

Neutrons as part of a fission event in a reactor are born “fast”, with a speed of about 20,000 km/sec (light is 300,000 km/sec), or an average energy of about 2 MeV (Fig. 2). The Figure shows the huge variation of the probability of such a neutron causing a further fission reaction with either uranium isotope U235 or U238 as the neutron loses energy and slows down [5].

At its nascent energy of about 2 MeV the neutron fissions U235 only slightly better than U238, with a cross section of ~1 barn (10^{-24} cm^2) versus 0.4 barn, respectively. Slowing down the neutron to “thermal” (ambient) energies of 0.025 eV increases its probability of causing fission in U235 almost 500-fold. This huge increase is the raison d’être for thermal reactors.

What is tacitly overlooked is that U238, the 99% of uranium that fissioned only slightly less than U235 at high energy, now is split about 30,000 times less often at thermal energies. Some U238 does absorb neutrons of any energy, becoming U239, which within about 3 days decays via Np239 to Pu239. The Pu239 so created, fissions like U235 and produces almost half the energy generated in the CANDU reactor. This is perhaps a small compensation for the huge loss in fission probability by U238 itself.

3.1. Transuranic elements

The absorption of a neutron by U238 starts the creation of a whole chain of transuranics, elements heavier than uranium, such as neptunium, plutonium, americium, curium, etc., each of which can absorb further neutrons or be fissioned by them. Figure 3 shows the fission cross sections of a few major isotopes created in this way. What is remarkable is that at thermal energies some of these fission as well as, or better than, U235, e.g. Pu239 and Pu241, while others, such as Pu240 and Pu242, fission rather poorly and would accumulate in the thermal reactor core fuel.

On the other hand, all of the transuranic elements created fission virtually equally well with fast neutrons at high energies around 2 MeV indicating that none would accumulate in the reactor.

3.2 Fission products

Fission products (FPs) created in the process do not fission further, but they do absorb neutrons to differing degrees. This is shown in Fig. 4 for a dozen of the major fission products that are generated in the CANDU reactor. At thermal energies neutron absorption shows a huge variation among the fission products from a low of 0.01 barn for strontium-90 to a high of 2 million barn for xenon-135, a range of 200 million. On the other hand, at high energies, around 2 MeV, virtually all of the fission product have similar absorptions within a factor of 10, and all lower than the fission cross sections of fuel atoms.

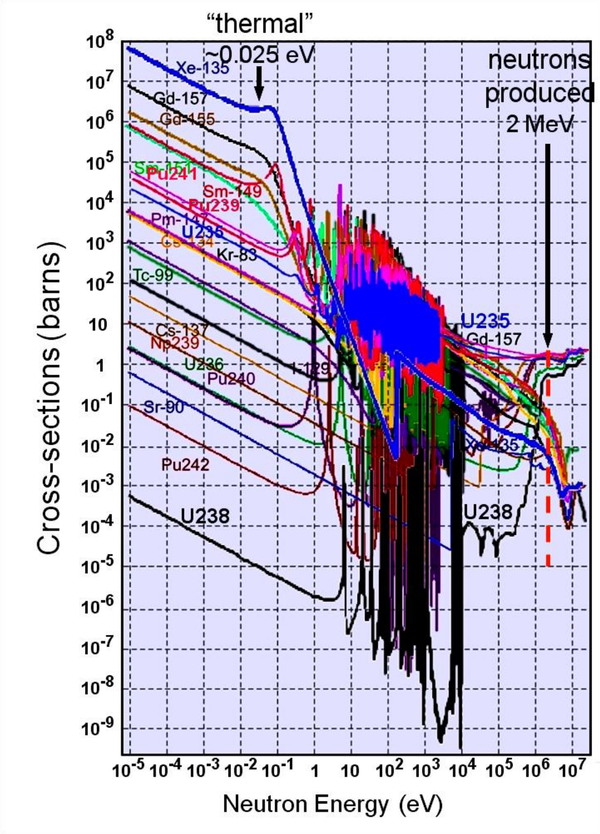

Figure 4. Cross sections for absorption (radiative capture) of neutrons of different energies by fission products created in the fuel in the reactor core [5].

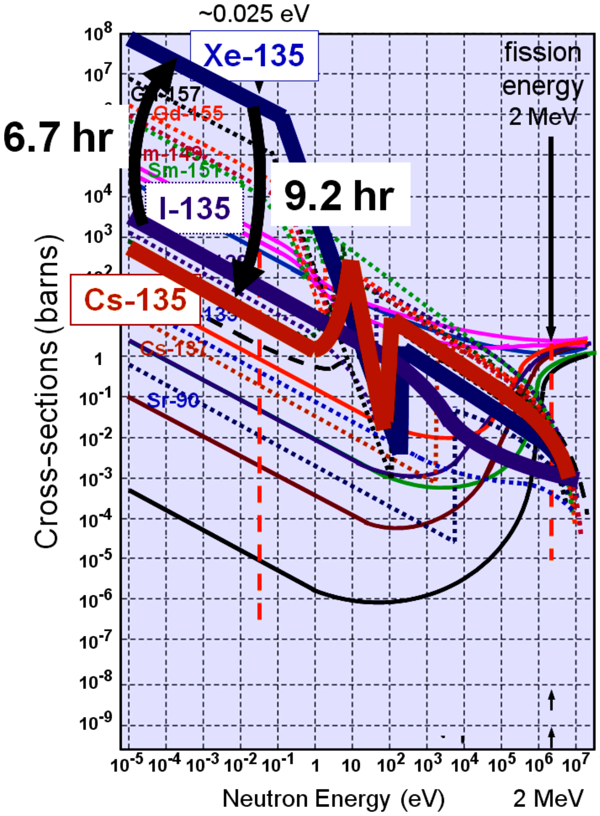

Figure 5. Schematic cross sections for fuel isotope and fission product absorption (cf. Fig. 4) with I-135, Xe-135, and Cs-135 high-lighted in bold, and with decay half-lives for I-135 and Xe-135 indicated.

The low absorption makes it possible to get higher fuel burn-up, while the clustering in absorption cross sections has major beneficial consequences on the control, shut-down and start-up of FNRs.

3.3 Xenon "poisoning"

One of the major fission products created in the fuel is iodine-135. It has a half-life of 6.7 hours, decaying to xenon-135, which in turn decays with a half-life of 9.2 hours to cesium-135. Cs-135 is effectively stable, with a half-life of 2.3 million years. When a reactor is shut down, any accumulated I-135 continues to decay to Xe-135. As seen in Figs. 4 and 5, Xe-135 has a neutron absorption cross section at thermal energies about 10,000-fold higher than I-135. It therefore heavily absorbs thermal neutrons that are needed to restart a thermal reactor such as a CANDU. After a shut-down of a few hours a restart is difficult until the I-135 has decayed and no longer replenishes the Xe-135, and until the Xe-135 in turn has decayed away sufficiently. This process, dubbed "xenon poisoning", lasts several combined half-lives of the two isotopes, delaying start-up by up to two days.

On the other hand, at high neutron energies, the swing in neutron absorption from I-135 to Xe-135 and on to Cs-135 is relatively small (Fig. 5). Therefore a fast-neutron reactor does not experience any xenon effect (more exactly the effect is over 2000 times smaller than for thermal reactors).

4. Inelastic Scatter

In thermal reactors one aim is the quick and effective reduction of energy of newly created fission neutrons. Elastic scatter in water and heavy water are good for that. In fast-neutron reactors the aim is the opposite: to maintain the neutron energy as high as possible. Elastic scatter from liquids of heavy atoms helps keep the energy high. However, what reduces the energy of fast neutrons is inelastic scatter, something that does not occur at thermal energies.

Inelastic scatter occurs when a fast neutron enters a nucleus, excites that nucleus, and, after the nucleus has relaxed by emitting a gamma ray, leaves the nucleus at a corresponding lower neutron energy. Each isotope has its own pattern of excitation and de-excitation that was measured starting in the early days of fast-neutron research [6]. Inelastic scatter is crucial in understanding the behaviour of FNRs.

Figure 6 is a visual excerpt of 12-group calculations [6] of over 200 sets of energy distributions of elastic, inelastic, fission and absorption (radiative capture) scattering by nascent fission neutrons as they

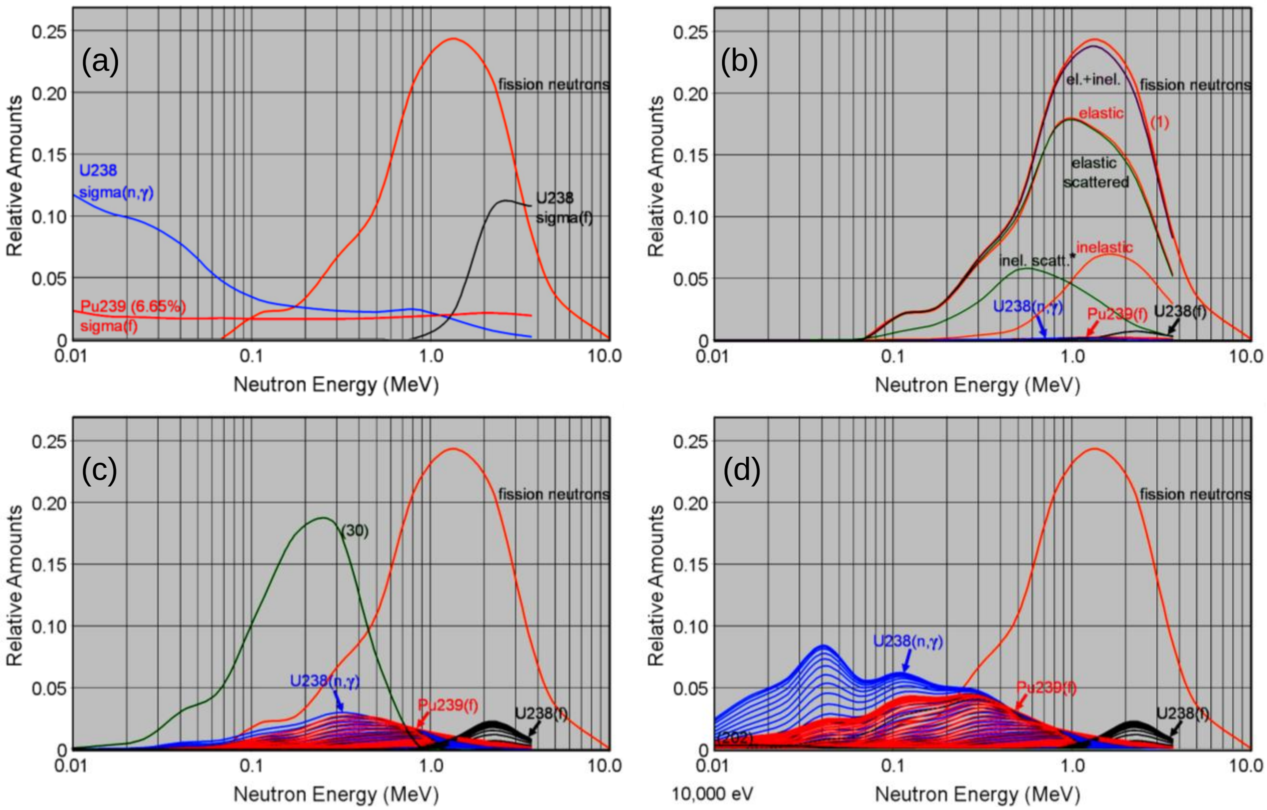

Figure 6. Elastic and inelastic scattering plus absorption and fission via the interaction of a shower of nascent high energy fission neutrons with fuel, coolant and structural components of a PRISM-like FNR. For clarity only U238(f), U238(n,γ), and Pu239(f) are shown in black, blue and red. (a) Nascent neutrons and cross sections prior to interaction. (b) First scatter showing entering and emerging neutron energy distribution for elastic and inelastic interactions. (c) State after 30th scatter showing a rising distribution of fission and of neutron capture. (d) After 202nd scatter with final distributions of fission and neutron capture. Multiple coloured lines indicate levels after intermediate scatter states. Constant original neutron distribution shown in orange in each panel.

interact proportionately with fuel (heavy atoms and fission products), liquid sodium and structural steel components of a PRISM-like fast-neutron reactor [7], and lose energy gradually in the process. The scattering interactions were followed until 99% of the neutrons were either captured or had produced fission events.

The major energy losses were via inelastic interaction. None of the neutrons reached thermal energies.

5. Fuel Characteristics

The detailed distributions of neutrons interacting with reactor core components provide relative macroscopic cross sections that permit the calculations of fuel behaviour in representative fast-neutron reactor configurations.

While actual FNR operations are constrained by the strengths of materials under irradiation, such as the HT9 steel used in the fuel canisters, computer calculations have no such limits. Fig. 7 shows idealized burn-up behaviour of U238 for a PRISM-like FNR started with sufficient fissile Pu239 to achieve initial neutron equilibrium. Refuel points for different realistic reactor types are indicated.

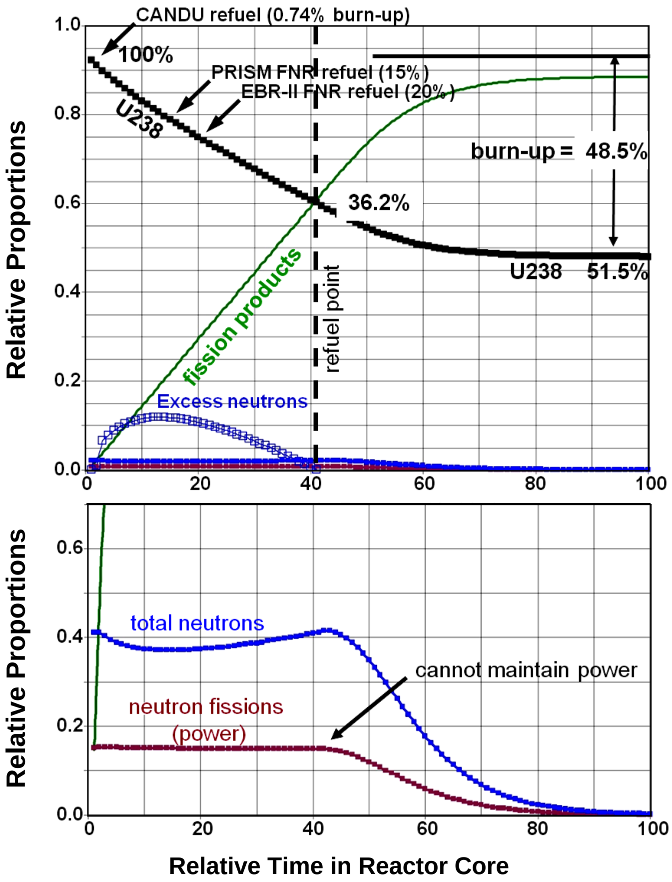

Figure 7 Fuel behaviour (U238 and fission products) for ideal FNR. Details of neutron accounting is shown in expanded scale in lower panel

Power in a reactor can only be controlled while excess neutrons are available. After that, power, or neutron fission, is slowly lost. While power is maintained, U238 is consumed both by direct fission at high neutron energies (Fig. 6) and transmutation to and fission of Pu239, Pu240 and higher transuranics (see Fig. 6).

The time point 41 (Fig. 7) when excess neutrons are no longer available is best considered the end of one fuel cycle. At this point in the ideal reactor about 35% of the U238 has been consumed. The fuel is removed, fission products are extracted, after which the heavy atoms and their remaining and sufficient fissile content are returned to the reactor. The heavy-atom deficit, equal in mass to the extracted FPs is topped up with used CANDU fuel or with fuel containing more concentrated transuranics if the latter are to be more quickly eliminated [12,13].

As shown in Fig. 8, the cycle repeats, each time with the same proportions of fuel feed being consumed.

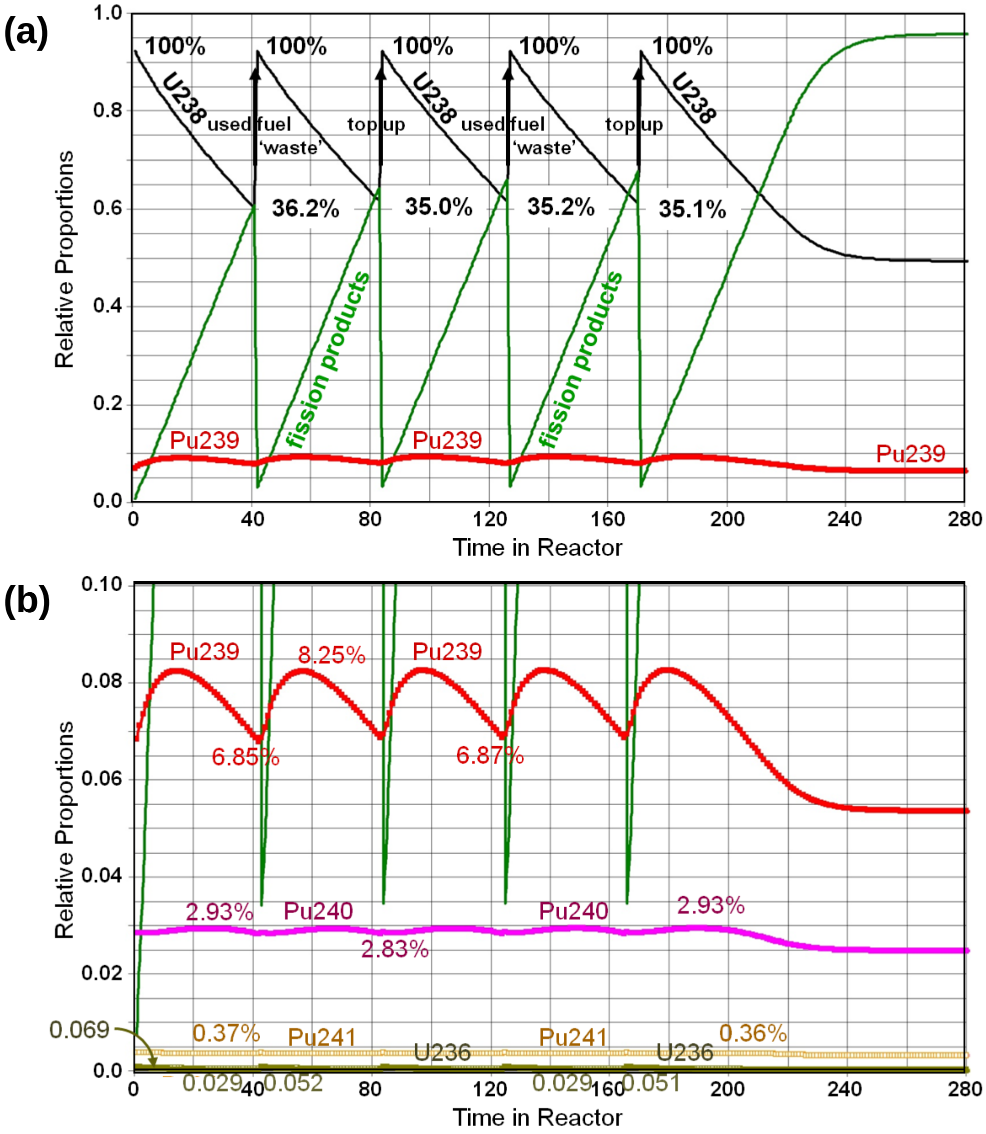

Figure 8. Fuel Cycling in Fast-Neutron Reactor.
(a) Cyclic behaviour of U238 consumption, fission product build-up and creation and use of Pu239. With 35% fuel burn-up per cycle, only three cycles would be required to consume 100% of the uranium fuel charge. (b) Cyclic behaviour of heavier transuranics and of U236.

6. Converter, Break-even, Burner: The Same Reactor

What is seen in Fig. 8a, and accentuated in Fig. 8b, is that the Pu239 concentration is augmented when the U238 content is high. As more U238 is consumed, Pu239 reaches a maximum near the middle of the fuel cycle. After that point Pu239 decreases as it and other transuranics are proportionately consumed as fuel.

A similar behaviour is seen with Pu240, a transuranic isotope that accumulates in thermal reactors. In the core of the FNR a small build-up is seen within the fuel cycle, caused by the increase of its parent isotope Pu239. However, this is followed by a decrease along with the decrease in Pu239 before the cycle ends. It is seen that over several cycles there is no increase in Pu240, in contrast to the behaviour in thermal reactors.

Other calculations, not shown here, indicate that the level of each transuranic tends towards its own equilibrium. Thus an excess feed of transuranics, such as Pu239, causes a reduction in the concentration of that isotope at the end of a cycle, while a dearth results in an increase towards equilibrium.

This behaviour indicates that if the fuel cycle is segmented into sections, then in the first segment a fast-neutron reactor can convert U238 to fissile transuranics that are useful as CANDU fuel. It has been shown previously that one FNR equal in power to a CANDU reactor can provide sufficient fissile fuel to power that CANDU, while the CANDU can provide used CANDU fuel to replenish the FNR. Only enough supplementary uranium is required to replace the fission products created by both reactors [8].

In the second segment of the cycle the FNR becomes a “break-even” reactor that can maintain its fissile content indefinitely, with the fuel being replenished from any source of U238, from used fuel, to natural uranium to depleted uranium, to replace the fission products created.

In the third section of the fuel cycle the reactor becomes a “burner”, utilizing excess uranium or excess transuranic elements from used fuel as it approaches a limiting concentration of these isotopes from above.

The sole output in any of these modes consists of fission products.

7. Summary

A fast-neutron reactor is a versatile and efficient power plant. Coupled with regular recycling of its fuel to remove “ashes” of fission products, it can utilize effectively 100% of its uranium fuel.

A fast-neutron reactor can manoeuvre. It is not affected by xenon poisoning and can therefore “load-follow” as well as shut down and start up at will.

A fast-neutron reactor, able to consume 100% of its fuel, can therefore re-use current used CANDU fuel of which only 0.74% of the energy has been extracted in our CANDUs.

A fast-neutron reactor can consume transuranics, and needs those as part of its starting fuel. It can therefore very quickly eliminate the long-term radioactive heavy atom content of Canada’s existing stockpile of used CANDU fuel.

8. References

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