5th Generation IV and Small Reactors (G4SR) Conference
The Westin Ottawa, ON, Canada, October 1-4, 2024

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WASTE BURNING PERFORMANCE OF THE MOLTEX SSR-W

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Abstract

Moltex Energy is pioneering the Stable Salt Reactor – Wasteburner (SSR-W), a fast spectrum molten salt reactor fuelled with recycled spent fuel, selected to be built at Point Lepreau by NB Power. A fast neutron reactor specifically designed as a wasteburner enables the fission of all actinides in the generation of electricity, mitigating both the problems of spent fuel waste and fresh fuel availability. Moltex is also developing the Waste to Stable Salts process, through which fuel salt is produced from existing spent fuel, and the fuel cycle is closed by repeated recycling of fuel salt that has been used in the SSR-W.

This paper presents results from modelling of this fuel cycle and demonstrates that with repeated fuel recycling an equilibrium can be reached where the concentration of all actinides is reduced during burnup in the reactor and actinide burning can continue indefinitely. The combination of this actinide burning in the reactor and the separation of most fission products in the recycling process results in a large reduction in waste volume, radiotoxicity and heat generation. The flexibility of the fuel cycle is also demonstrated, enabled by the recycling process, online refuelling and a fuel salt chemistry that allows variation of the conversion ratio.

1. Introduction

Moltex Energy is pioneering the Stable Salt Reactor – Wasteburner (SSR-W), a fast spectrum molten salt reactor (MSR) fuelled with recycled spent fuel, selected to be built at Point Lepreau in New Brunswick, Canada by NB Power. Moltex is also developing the Waste to Stable Salts (WATSS) process, which produces fuel salt from legacy spent fuel, and closes the fuel cycle by recycling of fuel salt consumed in SSR-W.

Several parties are developing molten salt fast reactors, in which the actinides and fission products form halide salts. Molten salt fast reactors are very flexible in the fuel cycle strategy adopted. The SSR-W operates as a burner in the U-Pu cycle, prioritizing fission of actinides over neutron capture, via a fuel form that gives a relatively low conversion ratio.

Recycling of spent fuel waste increases utilization of the fuel compared with the once-through open cycle, generating further electricity while also reducing the volume, radiotoxicity and heat load of the waste that requires final disposal. The environmental merits of spent fuel recycling, and especially the combination of multi-recycling with fast reactors, are highlighted in [1].

1.1 Safety and waste reduction

The safety advantages of molten salt reactors have been recognized for many years, and include the absence of significant pressurization, chemical retention of radionuclides by the salts, chemical inertness, lack of energy release or hydrogen generation in accident scenarios, large margins to phase change (boiling) and strong negative reactivity feedback due to salt thermal expansion [2].

All actinides can be recycled using the WATSS process and the reactor performance is tolerant of a wide range of actinide compositions, which means that the only waste produced is fission and salt activation products as well as any losses in recycling. This significantly reduces the waste requiring final disposal, providing a large economic benefit and minimizing the burden on future generations.

MSRs are uniquely suited to actinide burning (i.e. transmutation), because high transuranics (TRU) loading of the fuel is possible, limited only by the phase diagram. In contrast, other reactor types are limited due to the influence of high TRU loadings on the reactor performance. For example, sodium fast reactors may not be able to tolerate high actinide loading due to the resulting degradation of reactivity feedback coefficients [3]. In MSRs, the strong negative fuel expansion feedback dominates and is not very sensitive to the fuel composition.

Uranium content can also be minimized in an MSR, to maximize the destruction of TRU by minimizing the conversion of U-238 to higher mass isotopes. With solid fuels, low uranium content leads to a small or even positive Doppler reactivity coefficient [4]. There are also metallurgic challenges that would require significant R&D to achieve low uranium solid fuels.

For reduction of long term radiotoxicity it is important to recycle all actinides [5]. This can be done in MSRs but may be more complex with other fast reactors, since the fabrication of solid fuel is made much more difficult with the neutron emission from isotopes of curium in particular.

1.2 Proliferation resistance

Since the SSR-W does not require plutonium separated from other higher actinides, spent fuel recycling can be done with the application of a process derived from pyroprocessing and does not involve the low temperature solvent-solvent extraction processes typically associated with proliferation concerns.

The International Atomic Energy Agency (IAEA), the international body responsible for verifying the implementation of obligations under the Non-Proliferation Treaty, acknowledges the high proliferation resistance of pyroprocessing in contrast with conventional aqueous reprocessing [6]. It is highlighted that safeguardability is one of the advantages of pyroprocessing technologies as pure uranium and pure plutonium are not separated. Besides TRUs being recycled as a group, the contamination of the actinide product by lanthanide fission products leads to not only a strong proliferation resistance but also a high barrier of physical protection due to high levels of heat and radiation [6].

Although opponents to reprocessing in general refer to a specific report ([7]) to discredit proliferation resistance features of pyroprocessing, the content and conclusions of this study are often misinterpreted, as described in reference [8]. The 2002 report assesses the potential proliferation risk of a technology transfer to a fictional country that already has pre-existing PUREX or equivalent

capabilities as well as the resources to build clandestine plants. In sum, it assesses the potential for proliferation in a state where the proliferation risk is already elevated due to factors in place before the technology transfer under consideration. What the study highlights however is that the technical barriers to build a bomb with impure plutonium are so high that the preferred diversion route would require further chemical separation treatment, with another technology. This is the same pathway that would be applied in the absence of performing a process that led to impure plutonium [8].

More broadly, the combination of fuel recycling and transmutation provides considerable opportunities for long-term proliferation resistance, by permanently depleting the waste of its most sensitive fissile components ([8], [9]).

2. The SSR-W design

As a stable salt reactor, the SSR-W has separate fuel and coolant salts, the fuel salt being contained in pins and arranged into fuel assemblies. This provides several advantages:

- No fixed structure close to the fissioning salt, and hence experiencing high rates of radiation damage (the only non-fuel material in the active core is the metal forming the fuel assembly, which is replaced at refuelling).
- Delayed neutron precursors don't leave the active core, and hence modelling of transient neutronic behaviour is similar to other solid fuel fast reactors.
- Simplification of corrosion control.
- Simplification of primary circuit design, operation and maintenance.
- Simplification of nuclear material accountancy and inventory verification for Safeguards.

The SSR-W is refuelled at power, a single fuel assembly at a time. This means that the excess reactivity is very low, limiting the possibility for faults that increase reactivity. Between refuelling, the reactivity loss of the fuel due to burnup is compensated solely by passive temperature feedback. The strongly negative fuel feedback enables this passive control strategy. As reactivity reduces between refuelling, a small drop in fuel temperature gives a compensating increase in reactivity.

The power density of the SSR-W is deliberately low, to enhance the inherent safety, allow minimization of hazards and simplification of design substantiation:

- Noble gas solubility in the fuel can be controlled by design, to prevent production of gas bubbles in the fuel salt that could otherwise affect reactivity via changes in the fuel density.
- Fuel pin metal temperatures can be limited to a range where relevant irradiation data is available.
- Neutron flux is reduced relative to other fast reactors, reducing the impact of irradiation damage.
- There are large margins to temperature safety limits and no significant coolant void effect as in sodium fast reactors, due to the large margin to coolant boiling.

The fuel salt is $NaCl–MgCl_2–XCl_3$, where X is formed of uranium (U), transuranics (TRU) and lanthanides (Ln). Use of $MgCl_2$ allows the fuel reactivity to be controlled via either the ratio of $MgCl_2$ to XCl_3 or by the ratio of U/TRU, either of which can be varied as part of the WATSS process. This gives a high degree of fuel cycle flexibility – the conversion ratio of the fuel can be controlled by the U/TRU ratio to optimize between pure TRU burning or maximization of power generation given a

fixed quantity of TRU. For example, the start-up TRU inventory will be minimized by minimizing the uranium content of the fuel, reducing parasitic capture in uranium and giving a high rate of TRU burn. The uranium content of the fuel could then be increased over time by refuelling with a higher U/TRU ratio, to increase the utilization of actinides.

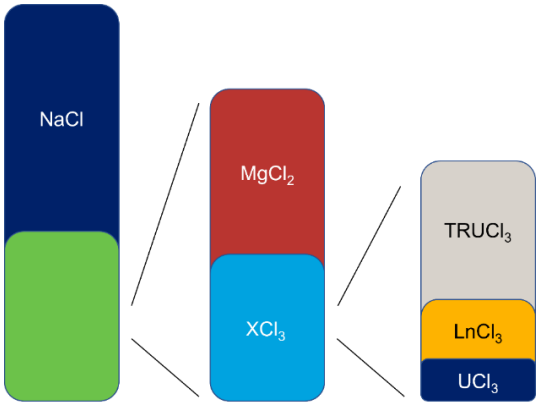

Figure 1 Components of the fuel salt for the SSR-W reactor.

The NaCl–$MgCl_2$–UCl_3–$PuCl_3$ system has been studied in reference [10]. The phase diagrams indicate freedom to vary the proportions of $MgCl_2$, UCl_3 and $PuCl_3$ components over wide ranges without significant changes in properties such as melting point.

The use of chloride salts in fast reactors often assumes the use of enriched chlorine, because the Cl-35 isotope that makes up around 75 % of natural chlorine has relatively high cross sections for neutron capture. Its presence therefore reduces the neutron economy of the reactor, which is especially problematic for breeder designs that require excess neutrons to be utilized in production of new fuel (Pu-239). The SSR-W operates as a burner, with relatively low conversion ratio, which makes the impact of Cl-35 neutron capture on neutron economy tolerable.

Actinides burning is maximized with a hard neutron spectrum, because the ratio of fission to capture cross sections tends to increase with increasing neutron energy, particularly for even mass isotopes. For example, at thermal energies neutron capture is hundreds of times more likely than fission for Pu-240, while at fast energies (around 0.1 MeV where the SSR-W spectrum peaks) the cross sections are comparable, as is shown in Figure 2.

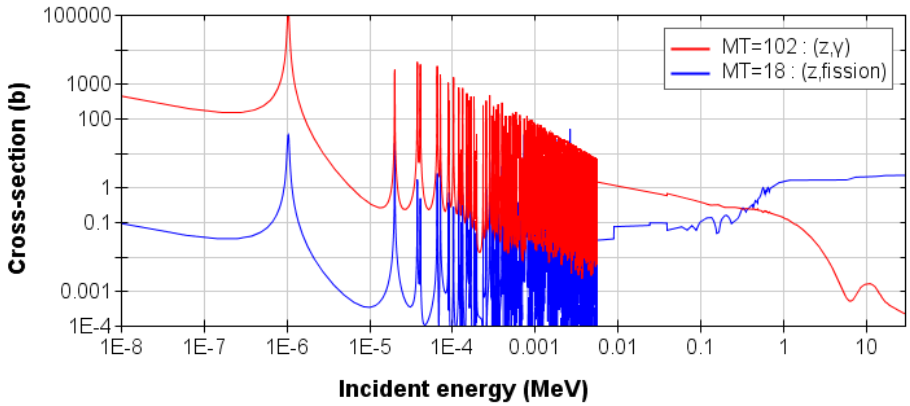

Figure 2 Neutron cross-sections for fission and neutron capture reactions on Pu-240. Data are extracted from the JEFF-3.3 library [11].

3. The fuel cycle

3.1 Conceptual description

The combined reactor and WATSS fuel cycle can be divided into two stages, corresponding to the so-called WATSS-C and WATSS-S processes (depending on the feed material being CANDU or SSR-W spent fuel).

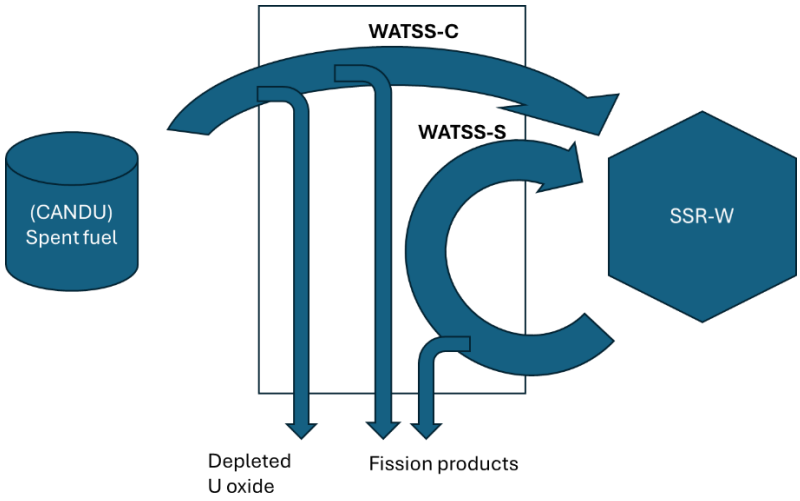

Figure 3 Schematic representation of WATSS / SSR-W fuel cycle.

3.1.1 WATSS-C

In the first stage, the WATSS plant processes spent CANDU fuel bundles to produce the first core load for the SSR-W. It also produces the fuel needed to refuel the operating reactor for a period of time.

The production of fuel by WATSS during this stage is simulated as follows:

- The isotopic composition of the spent fuel is characterized by use of ORIGEN 6.2 simulations of the fuel history [12], for a range of discharge burnups and an average power rating appropriate for the Point Lepreau CANDU, with a nominal post irradiation cooling and decay time. This gives:
  - The relative proportions of the fission products, uranium and TRU.
  - The isotopic breakdown within each component.
- The effect of the WATSS process is simulated simply via a set of extraction efficiencies, to give:
  - A combined TRU and Ln fission product isotopic vector.
  - A uranium isotopic vector. Uranium is treated independently since the WATSS process can vary the uranium content.
- The fuel salt is then formed by chlorinating the actinides and lanthanides and including the NaCl and $MgCl_2$ diluents, with the NaCl content fixed and set by the phase diagram region that gives the desired properties.
  - The uranium content is set as a fixed proportion of the actinide and lanthanide component. A minimum uranium content is determined by the WATSS process. This

provides the isotopic composition, or vector, for the actinide and lanthanide chloride component, labelled XCl_3 in figure 1.

- The $MgCl_2$ content is free to be varied in line with the constraints imposed by the phase diagram. The XCl_3 content is simultaneously varied to respect the fixed NaCl content. This degree of freedom is used to vary the reactivity of the fuel salt.

3.1.2 WATSS-S

At some point fuel assemblies will start to be transferred from SSR-W back to WATSS for recycling. The WATSS process for this fuel salt consists of the following relevant steps:

- Removal of fission products, with the exception of some lanthanides as above.
- Replacement with 'fresh' makeup feed uranium, TRU and Ln from CANDU spent fuel, via the WATSS-C process described above.
- Adjustment of the $MgCl_2$ content to achieve the required fuel reactivity (TRU content).

The uranium content (relative to TRU and Ln) can also be re-adjusted if needed. The fuel is then transferred in new fuel pins, assembled in fuel assemblies and returned to the reactor for further use.

3.1.3 The reactor cycle

The selection of the reactor fuelling strategy is a complex problem, with several variables to be optimized:

- The radial power peaking. This must be limited to achieve the target reactor power while satisfying constraints on the peak power assembly.
- The average period of time between refuels, or equivalently the average discharge burnup.
- The peak discharge burnup. This is constrained by any limits derived from fuel pin metal performance, e.g. irradiation damage or time at temperature.
- Reactivity worth of a refuel. In principle this may be limited by the temperature change (via reactivity feedback) that is acceptable given the operational envelope.

These variables can be optimized by choice of the following control parameters:

- The TRU content of the fuel in the assembly to be loaded (the XCl_3 to $MgCl_2$ ratio and/or the U/TRU ratio).
- The refuel location (and hence the power, burnup, reactivity of the fuel to be discharged).
- The time between each refuelling, which may vary.

In addition, the cost associated with the wider fuel cycle – including WATSS – must also be minimized. For example, very frequent refueling would support minimization of the reactor power peaking, but this would increase the costs associated with fuel recycling through WATSS and re-fabrication of fuel assemblies.

Work is ongoing to develop techniques to optimize this process [13], but this topic is not considered further in this paper. However, of relevance to this paper is the isotopic change of the fuel during burnup in the reactor. The TRU from the spent CANDU fuel is rich in Pu-239, relative to the higher

Pu isotopes and isotopes of Np, Am and Cm. The Pu-239 also has high fission cross section and high fission to capture ratio, which means that it is preferentially fissioned, as is demonstrated below.

Neutron capture also creates higher mass isotopes. This process occurs in any reactor, but production of higher mass isotopes is minimized by a hard neutron spectrum, because the fission to capture ratio tends to increase with increasing incident neutron energy as reported earlier. The TRU content of the fuel salt must therefore be increased in each cycle, to compensate for the degradation of the TRU vector and maintain reactivity.

Fuel assemblies that are discharged from the active core are placed in the periphery of the primary coolant tank to allow decay and cooling prior for transfer to WATSS for recycling.

3.1.4 Repeated recycling

The cycle of burning fuel in the reactor and then recycling it in WATSS can continue to be repeated indefinitely, as long as:

- Feed TRU from spent thermal reactor fuel is available.
- The phase diagram constraints allow further increase in the TRU content, if the TRU vector has not reached an equilibrium yet. If the TRU vector has reached equilibrium, no further increase in TRU content is required to maintain reactivity. Some feed recycled fuel from thermal reactor is still needed as top-up to compensate for TRUs being burnt, but this quantity will remain the same at each subsequent cycle.

At some point, the effect of burnup in the reactor and subsequent makeup in recycling gives no net change in any isotope – an equilibrium composition is reached. From this point the reactor is net burning all isotopes of all actinides and can continue to do so indefinitely as long as feed fissile material is available for recycling.

3.1.5 End of reactor life

The fuel salt from an end-of-life reactor can be returned to WATSS to create the fuel for another SSR-W. Since this fuel will already be close to, if not at equilibrium, its composition would change little when used in the following reactor, unless feed TRU with a different composition was used in the recycling process.

3.2 Modelling of burnup in the reactor and recycling

A 1,200 MWth SSR-W core model has been used for the present study.

The neutron transport code WIMS11 [14] is used to model the reactor part of the fuel cycle. WIMS has been used extensively as part of SSR-W design and is validated using sodium fast reactor benchmarks [15] and for the assessment of conventional liquid metal fast reactors such as the UK DFR and PFR, the Russian BN reactors [16] or the French Superphénix reactor [17].

A two-step process is applied, with preparation of lattice data for use in a whole core calculation. The process has been verified by comparison to reference Monte Carlo solutions using Serpent 2.2.1 [18].

3.2.1 Lattice data preparation

The WIMSECCO module of WIMS is used to calculate problem-specific resonance shielded cross sections in a heterogeneous fuel assembly model, with reflective boundary conditions. It reads library cross section data stored in 1968 energy groups, performs a flux solution with this detailed energy resolution and then condenses to 172 groups. The library cross sections are derived from the JEFF-3.3 evaluated data library [11].

The 172-group flux solution is calculated by the CACTUS module using the method of characteristics, and the fuel is then homogenized using this flux. Non-fuel data is created via further calls to WIMSECCO with a fuel neutron source. The impact of leakage, particularly important in a fast reactor, is accounted for by use of a simplified (R, Z) whole core model using the MERLIN module with homogenized fuel and representation of the non-fuel regions outside the active core. MERLIN performs a flux solution using the simplified neutron transport SP3 method. Finally, the cross sections are condensed to 33 energy groups and stored.

3.2.2 Whole core flux solution

The whole core flux solution uses the 33-energy group cross sections from the lattice data step and a 3D triangular prism geometry that models each homogenized fuel assembly, as well as the non-fuel regions such as reflectors and the radial shield. This allows explicit modelling of single assembly refuelling.

This whole core flux solution is used to determine the critical fuel composition (required TRU content).

3.2.3 Burnup

For modelling of the evolution of the fuel TRU vector, a simplified model of burnup in the reactor is used, without explicit representation of single channel refuelling. Instead, the fuel is all burnt in a single assembly calculation with fixed discharge burnup. The lanthanide content of the fuel is neglected, on the basis that the reactivity impact is small and WATSS will limit the lanthanide content.

3.2.4 Decay / cooling

A 1 year zero power step is also added to simulate the cooling time that fuel will undergo before transfer to WATSS for recycling. The decay that occurs during this time affects the TRU composition, particularly due to Pu-241 decay to Am-241.

3.2.5 Recycling

Recycling in WATSS is simulated by extracting the fuel composition after burnup and cooling, removing the fission products, carrying over the U and TRU into the new fuel salt and adding the necessary amount of makeup feed to reach the TRU content required to give a critical core at the start of the next burnup. This is performed using a system of Python scripts.

3.3 Results from repeated burnup and recycling

3.3.1 Isotopic changes in burnup

Table 1 shows the main TRU components and the changes in the mass of key nuclides when burning to 100 GWd/t and then cooling for 1 year, with fuel derived from spent CANDU fuel. Note that the relative mass change cannot be derived from the first two columns, due to the reduction in total TRU between the start and discharge (the TRU vectors sum to 100% in both cases). Minor actinides account for the sum of all isotopes of non-Pu TRUs considered (Np, Am, Cm).

Table 1 TRU composition and relative mass change after discharge with fuel directly extracted from CANDU spent fuel.

Nuclide	Starting TRU vector (%)	Discharge TRU vector (%)	Relative mass change (%)
Pu-238	0.093	0.798	666.2
Pu-239	66.7	58.8	-21.3
Pu-240	24.8	30.4	9.4
Pu-241	2.33	3.35	28.5
Pu-242	1.47	1.83	11.0
Minor Actinides	4.65	4.78	-8.4

The consumption of Pu-239 and production of higher Pu isotopes are clearly visible. Note that the high production of Pu-238 is due to Am-242 neutron capture to Cm-242, which alpha-decays to Pu-238 with a half-life of 163 days.

3.3.2 Impact of repeated recycling

After 20 burnup cycles, with recycling between, the starting TRU vector has changed, and the most important transuranic nuclides are all reduced during burnup (and subsequent cooling):

Table 2 TRU composition and relative mass change after discharge following 20th burnup cycle.

Nuclide	Starting TRU vector (%)	Discharge TRU vector (%)	Relative mass change (%)
Pu-238	4.08	4.69	2.4
Pu-239	30.6	25.3	-26.1
Pu-240	43.6	46.1	-5.6
Pu-241	6.25	6.78	-3.2
Pu-242	6.30	7.12	0.8
Minor Actinides	9.15	9.94	-3.0

The starting TRU vector corresponds to the discharge TRU composition after the previous irradiation cycle, with a top-up of freshly recycled CANDU spent fuel (with a TRU vector corresponding to the first column of Table 1).

The low uranium content of the fuel means that the rate of TRU transmutation is 0.97 g/MWth-d, very close to the maximum of around 1 g/MWth-d. This is equivalent to the destruction of around 425 kg of TRU per year of operation for the reactor power considered. This figure only changes marginally with the burnup cycle.

The transition towards equilibrium, as the fuel is burnt in the reactor and recycled several times, is highlighted in Figures 4 and 5 representing respectively the quantity of TRU in the fuel salt and its isotopic composition.

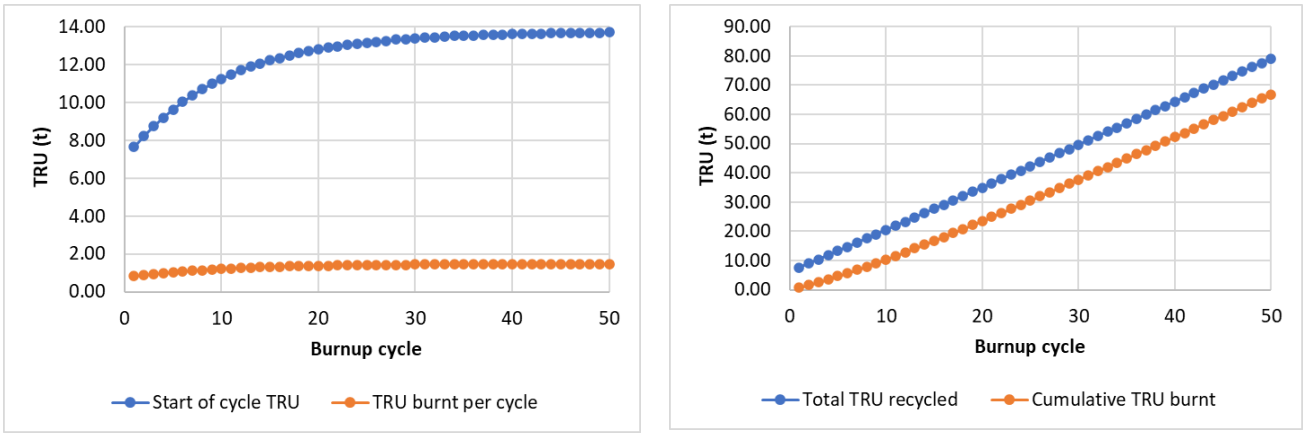

Figure 4 (left) Amount of TRU in the SSR-W core, assuming a homogeneous burnup history of fuel salt in the assemblies. The mass difference relates to an increase of the XCl_3 fraction in the fuel salt. (right) Cumulative quantities of TRU recycled and transmuted.

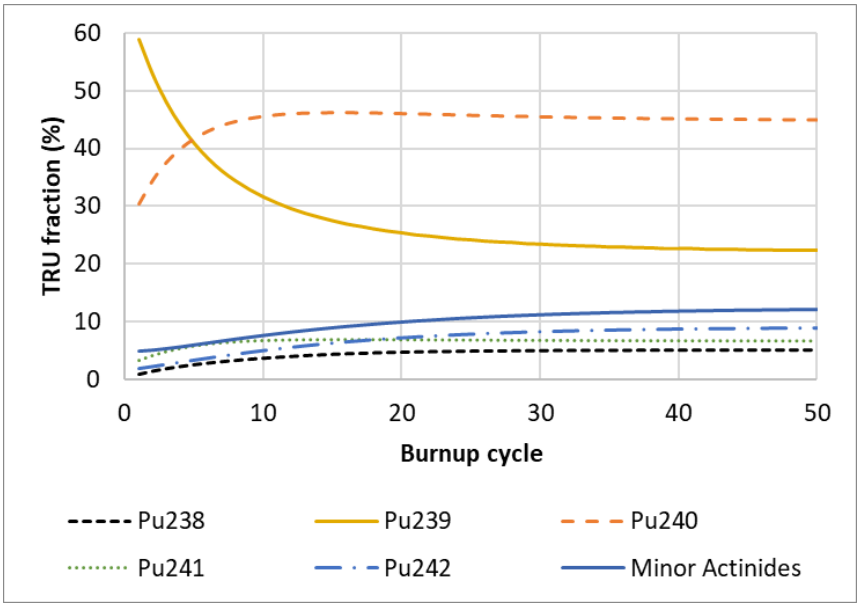

Figure 5 Discharge TRU isotopic vector after several cycles.

The higher mass elements take longest to reach equilibrium, due to the number of neutron captures required to reach them.

3.4 Fuel cycle options

The results above assume that the uranium content of the fuel has been minimized, in order to maximize TRU burning to reduce the waste that must otherwise be disposed of in a geological repository. However, the WATSS process is able to control the proportion of uranium in the fuel salt relative to TRU, hence providing a way of tuning the conversion ratio to achieve the desired balance between TRU burning and power generation given a fixed quantity of TRU. For example, if the uranium content of the fuel salt is maximized, the TRU burning rate drops from 0.97 to around 0.6 g/MWthd. The power generated per mass of TRU input is however increased, which gives a longer reactor lifetime from a fixed quantity of available TRU.

4. Conclusions

A SSR-W reactor with a thermal power of 1,200 MWth eliminates 425 kg of actinides on an annual basis, or about 25 metric tons over its lifetime, with a fuel salt composition and isotopic vector that evolves to reach an equilibrium. At this point, the required top-up of TRUs from freshly recycled CANDU fuel is constant and corresponds to the amount of TRU transmuted. The equilibrium is also visible with the isotopic vector of the discharged fuel, in which the proportion of Pu-239 is significantly reduced compared with spent CANDU fuel.

At the end of operational lifetime of the reactor, the TRU content of the last core load would be significantly lower than the quantities recycled from the spent CANDU spent fuel inventory, which would otherwise be considered as waste. Moreover, the reactor will have produced energy which, if produced by a thermal reactor, would have generated more transuranic actinides rather than a net reduction.

Finally, the end-of-life core load could itself be recycled and used as start-of-life core load for a new SSR-W, therefore fully closing the cycle.

5. List of acronyms

CANDU	CANadian Deuterium Uranium (reactor)
IAEA	International Atomic Energy Agency
JEFF	Joint Evaluated Fission and Fusion (nuclear data library)
MSR	Molten Salt Reactor
SSR-W	Stable Salt Reactor - Wasteburner
TRU	Transuranic (element / isotope)
WATSS	Waste to Stable Salt (process / facility)
WATSS-C	Waste to Stable Salt process – CANDU spent fuel as feed
WATSS-S	Waste to Stable Salt process – SSR-W spent fuel as feed
WIMS	Winfrith Improved Multigroup Scheme (code for reactor lattice cell calculation)

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