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Fusion Driven Transmutation of Transuranics in a Molten Salt

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A first set of computational studies of transmutation of spent nuclear fuel using a compact tunable 14 MeV D-T fusion driven neutron source is presented. Where we study the controllability, time evolution, as well as spatial distribution of the neutronics in the transmutation in the subcritical operations regime of a transmutator, in which our neutron sources are small, distributed, and can be monitored. The monitoring condition is included through the use of a molten salt fuel carrier which can also allow for in line chemical processing. In order to carry out these studies we put together a linkage code using the Monte Carlo neutron transport and material depletion code OpenMC with self-developed python based artificial intelligence (AI) tools. The obtained results show (i) possible pathways to efficient steady state waste transmutation; and (ii) the feasibility of using neural nets to provide real time control of the system.

Keywords: Fusion, Neural Net, Neutronics, Radioactive Waste Disposal, Molten Salt

1 Introduction

The utilization of neutrons of fusion has substantial roots in the fusion research. In a fusion reactor in which the energy production is the ultimate goal, a substantial energy fraction of neutrons may be directed to its energy conversion, where the Q-value (the energy production divided by the input energy to trigger fusion) be greater than unity. Meanwhile, if the goal is not to make a fusion reactor, this constraint may be relaxed. Here we consider a transmutator of radioactive spent nuclear fuel (SNF) driven by fusion-derived neutrons. In such a transmutation, therefore, the Q-value condition (Q > 1) is not the primary constraint. Many authors suggested [1–6] using DT fusion neutrons to transmute SNF. Besides the easier threshold not to reach Q > 1 condition, the fusion neutron energy that exceeds MeV plays an important role in transmuting minor actinides MA) (as part of SNF), where MA's are a subset of the transuranic elements (TRU) are the longest lived and most toxic elements in SNF, as we discuss below. There have been plenty of beam-driven fusion reaction schemes. The role of the beam may be considered as a more straightforward access to its directed energy to induce high energy thresholds of nuclear reactions both of triggering fusion and transmutation of nuclei, circumventing the large fraction of thermal plasma population. These include: the beam-fusion [7], the beam-driven muon fusion [8]. In addition there have been a class of works on the fusion-reactor-generated

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neutrons [9,10] that could pave the way for transmutation. There is another class of neutrons that are created by high energy proton/deuteron beams in a process called neutron stripping and spallation [11–13].

As mentioned above, the fusion neutron energy (such as 14 MeV neutrons from the DT fusion reaction) has an advantage in transmutation. The reason is the following. As shown in **Fig. 1**, the cross section of neutron capture by Americium (Am-241) is exceeded by fission cross section if the energy of neutrons are sufficient high (such as MeV), where the fusion-produced neutron (and its immediate slowdown) energies are in.

In addition, this helps to overcome small dips in the fissionability that would otherwise be steadily increasing from an increasing nucleon count as seen in **Fig. 2**. The result is the transmutation of these nuclides, such as Americium and other Minor Actinides (MA), that accumulate in these fissionability dips from repeated captures of the uranium fuels. This would diminish the loss of potentially fission causing neutrons needed to continue the secondary fission chain reaction. A properly rebalanced system would not only stop the accumulation but remove existing MA which are the heart of the nuclear waste issue.

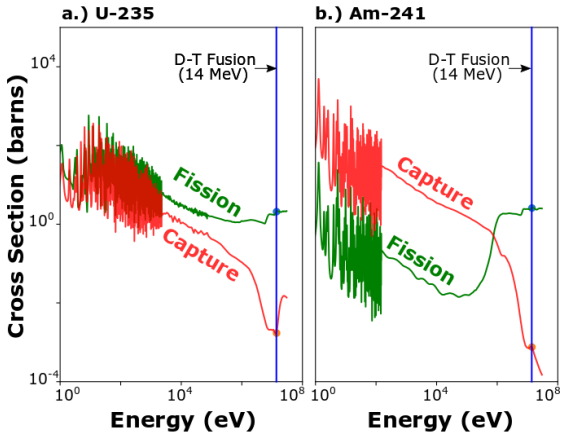

Fig. 1 *Comparison of cross sections of the fission induced by a neutron and the neutron capture by* ^{235}U *and* ^{241}Am

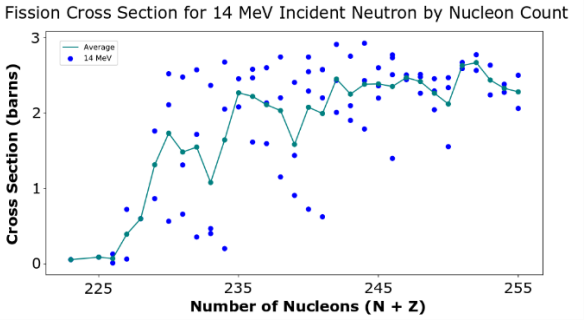

Fig. 2 Increasing fission cross section showing ease of fission following nucleon count. Uses nuclides with greater than 224 nucleons from ENDF/B-VIII.0 library with 900K temperature profiles.

In the present paper we present three main pillars of the principles of how best we operate the D-T fusion-driven neutrons to induce transmutation of transuraniums. The first pillar of the idea is to induce transmutation in the molten salt [14]. This liquid operation [15], as opposed to the usual solid rod operation of nuclear reactors, has the following distinct advantages: liquid operation allows much wider stability (and thus safety) and flexibility (as we discussed in more details later), along with the convergence of the operational timescales, which also helps in our system, as we see below. The second major feature of our system has the distributed neutron sources. This distributed nature of the source, again, contributes to the safety and flexibility of the operation as well as the cost (We can have an innovative laser-driven neutron sources [16], [17] or a more conventional linear accelerators [18]). The third major innovation in our operation is the ability of feedback control and its operation by using the artificial intelligence (AI) such as genetic algorithm (GA) [19,20] and neural network (NN) [21–25]. Because our system operates in the molten salt liquid, we converge many time scales into somewhat overlapping time scales of various physical, chemical, and operational processes. In addition, the introduced liquidness allows many of the feedback processes are permitted and transparent (such as the laser or other photon diagnoses are accessible). Note also

that many physical processes are too fast for human interventional time scales and only accessible via electronic systems of detection, processing, and feedback controlled computational and transmission time scales. In other words, our fundamental three elements of the major features of our transmutator are intertwined.

We discuss our motivation and the basic ideas in Sec. 2, while in Sec. 3 we survey the physics of subcritical systems. Our processing control simulation works are shown in Sec. 4, based on the introduced ideas. We further discuss on active control issues of a molten salt transmutation by artificial intelligence in Sec. 5. The summary is collected in Sec. 6.

2 Motivation

Here we sketch some of the preliminary modest results for a proposed Transmutator using fusion driven neutrons as described here [26]. This includes a novel fusion driven neutron source, a molten salt fuel carrier, and a proposed AI control system working together to perform transmutation of nuclear waste. The main purpose of these results is to show how control systems for the two disparate time scales in nuclear process to make transmutation in this manner an effective means of waste reduction. The first being the long time scale which is on the order of weeks to years, showing how even simple control of in line chemical processing can create an effective and sustainable solution for further waste processing. The second much shorter time scale, in the range of nanoseconds to microseconds, can show how using the molten salt and multiple tunable fusion sources can allow for increased safety and efficiency with the use of NN in an active control setup.

2.1 Fusion Driven Neutron Source

The shell added stability is relatively small compared to the overall increase in fissionability for higher nucleon counts and requires only slightly more energetic neutrons to fission. This can be seen in [**Fig. 1**] which shows the cross section as a function of neutron energies. Artificial transmutation can aid natural tendencies by helping overcome this shell stability through the use of an external neutron source, such as the 14 MeV neutrons of D-T fusion. This fusion driven transmutation is being explored following recent developments of a novel neutron source [15] that utilizes laser driven acceleration techniques to drive fusion neutron production in a compact, and production rate tunable way. The source is also relatively cheap allowing for many, even thousands of, points of entry into the transmutator. The source is also directly tunable in both intensity and timing by control of the driving laser. While fusion neutrons have long been used in fission experiments and considered for ADS style transmutation systems, there use here is the only one the author is aware of that use multiple controlled sources and AI control to drive waste burning transmutation. This would be on a short time scale similar to NN's employed for fusion in [23,24] for tokamaks and FRC, respectively.

This method is particularly useful if the distinction of source neutrons and secondary chain reaction fission neutrons can be accounted for well enough to drive effects relating to their different energies. This would pair with differing isotopic evolution pathways that would result to create a guided pathway that can result in efficient and thorough waste burning. These pathways could for example take advantage of breeding making the loss of some neutrons to capture in fertile waste to increase the chance that they will produce a sufficient amount of neutrons to cover this loss from increased fissionability in the future. All operations would ideally be in the subcritical range to keep chain reactions limited by a dependency on the source neutrons, which would further increase control.

2.2 Molten Salt

To develop the transmutator as presented in [15] also implies a detailed neutronics study for a system using molten salts with dissolved transuranic wastes. With the addition of a TRU dissolving molten salts [14,27–29] acting as a fuel carrier, the liquid properties would allow transmutation to be done in a safer manner while providing additional opportunities for control. This would be through both inherent liquid properties, and the specific properties of the molten salt. These factors together make the adoption of a liquid core a key step to address attempts to bridge the nuclear processes with the rest of the processes (chemical and macroscopic).

In regards to existing utilizations of molten salts; there are some similar systems that have been considered such as Molten Salt Fast Reactors (MSFR), which include Russia's MOSART [14,30] and Europe's eponymous MSFR [30,31], in addition to other ADS such as MYRRHA [32,33], which is currently under development in Belgium and is more similar in operation. Proton spallation ADS is also similar in its use of a subcritical core due to the importance of the source in its operation. However, there are key differences that do not allow for a straightforward transfer of sufficient existing information to this system. In particularly there are differences in the insertion of the nuclear waste as an aqueous additive to the molten salt, which has been considered but we could find no information on existing systems that incorporates it. Additionally there is the use of many small placeable sources rather than a single large source. This large source would also necessarily come with a large spallation target used to ensure impact within a reasonable path length, and thus neutron production, from the high energy protons.

The molten salt advantages come from several points, which include its liquidity, laser transparency, its low neutron capture cross section, and most importantly its added safety. The aqueous mixture allows for keeping the whole of the system in a near homogeneous mixed state [34] even if the reactions taking place are not uniform throughout the system. Combined with a high vaporization temperature, additional thermal displacements through convection, and the negative temperature coefficients of, FLiBe and FLiNaK, the molten salts being considered here [14,30,31,35] safety is increased. This is accomplished through easier to control density perturbations and the easier prevention of voiding that must be accounted for with much greater care in the widely used solid fuel systems. Additionally, monitoring [36] through the optically clear salts provide a possible low latency feedback vital for short time scale control. While modification of the molten salt solution through removal of fission products (FP) can be done in parts [14,30,31,34,35,37] and possibly in-line as the liquidity and homogeneity allows for partitioning of batches without major disruptions in the overall process. Finally, there is a decrease in the typical density for fissile targets from solid fuels, this density can also be varied to a degree by adjusting the concentration of dissolved nuclear waste.

The lower density, while not significant, changes the typical time scales of action for prompt and high energy neutrons while the tunable laser driven source with possible optical monitoring vastly increase the response time for a control system. Molten salt's liquid nature also allows for additions and removals to extend otherwise very transient mixtures. This action response cycle is orders of magnitudes faster than for traditional nuclear systems, which are typically done by thermally or thermal neutron speed relevant actions only. The "thermal" processes used include shifting capture to fission cross section ratios by: neutron moderation, mechanical addition of thermally capturing isotopes, Doppler broadening in the resonance regions of the thermal sections of the cross sections, and density controls through cooling. These could be supplemented by much faster response by electrical and computational control of the source, which would now be possible.

2.3 AI Control

The combined use of a molten salt fuel carrier, the small tunable fusion driven neutron sources, and subcritical operation allow for the use of a NN to provide an active control method to the transmutator. Feedback control of a nuclear system is not normally considered possible due to the very short time scales for nuclear systems measured in terms of power deposition or neutron population. This would especially hold true for fast spectrum systems where a prompt neutron generation time (I_{Prompt}) can be on the order of 10^{-7} to 10^{-9} s. These faster spectra would be the case considered here because of the higher energies needed to transmute the MA waste. Because of this perceived difficulty, this paper is to our knowledge the first attempt to use a NN for control in a fission system.

Despite the short time scales feedback control may be possible if a controller can reactively adjust the system's conditions in a short enough time and to a degree that the systems output stays within an acceptable bound. To see the effectiveness of the feedback we need to know how fast the system can react (latency) and what level of change this response can cause (local and total reactivity insertion).

There are five main parts to this:

1.) The time for a system state to be relayed to a controller (feedback signal) by sensor medium.
2.) The sensor response time.
3.) The controller response time
4.) The time to enact this correction. (output to reactivity injection)
5.) The ability for this bounded input to keep the system within a bounded output envelope (BIBO).

The first four parts are time scales have a lower bound dictated by the physics of the sensors and current technological computing capabilities.

1.) The optically passable molten salt allows for light based sensor readings such as velocimetry which could reveal information such as temperature. The sensor medium time (T_{medium}) would then be the distance from a point of interest (D_{sensor}) to the sensor divided by the speed of light (c).
2.) The response time of senor (T_{sensor}) can be improved by not doing calculations at this point and passing inputs directly into an NN. This can be done through spatially parallelized and gated CCDs and can poll as fast as a single sensor clock cycle frequency (S_{cf}) per bit resolution (R_{sensor}).
3.) For controller response time to meet the short time scale requirements and only consider feasibility, only pre optimized computational systems will be considered here. A highly parallelized NN's computational time ($T_{controller}$) can thus be primarily described by its depths (N_{layers}) [21] and the complexity of its neutron activation function in terms of how many computational instructions it requires ($I_{activation}$) as limited by its systems computational cycle frequency (C_{cf}). If an application specific integrated circuit (ASIC) is used we can assume that many of the conventional overhead issues can be ignored. We can also pipeline weighting values to prevent memory latency issues. This could make the computational speed equivalent to 1 instruction per cycle (IPC) to best match its computation time in terms of clock cycles. The weighting process can be done by a multiple accumulate operation which can be a single instruction or a single clock cycle if done through a single rounding in a fused accumulator (FMAC).
4.) The time for response ($T_{response}$) is assumed here to be a direct response to output values modulating an input power

level for an already timed input. Such as a modulated 100 kHz pulse. This repetition rate would be a limiting factor for response rate. However, having multiple sources can allow for interleaving of the repetition times so as to achieve much higher input rates.

All of this together adds to the total input output lag or latency for the control system

$$T_{lag} = T_{medium} + T_{sensor} + T_{controller} + T_{response} = \left(\frac{D_{sensor}}{c}\right) + \left(\frac{R_{sensor}}{S_{cf}}\right) + \left(\frac{N_{layers} I_{activation}+1}{C_{cf}}\right), \quad (1)$$

for a simple system using the simplest activation function which is a single instruction. If we for simplicity assume reasonable values such that $\left(\frac{R_{sensor}}{S_{cf}}\right) = \left(\frac{1}{C_{cf}}\right)$ and $C_{cf} = 5\ \text{GHz}$ as well as an effective light to sensor travel distance of 0.5m, we can approximate a $T_{lag} \approx 0.2\ \text{ns}\ (N_{Layers} + 2) + 2\ \text{ns}$ which for a useable but relatively shallow neural net such as a 10 layer system this would be approximately 4ns. This is a short enough time scale that hints at the possibility of using AI control even in very fast nuclear systems.

The final point is a bit more onerous for nuclear systems, how much of a change can be caused in the system to counter any error function to keep the system within an acceptable bounds. The difficulty of analytically calculating the response of a real nuclear system already necessitates the use of approximation techniques such as Monte Carlo for current computers to accurately model. However, the purpose of using an NN as a controller is that after an initial learning stage this computational difficulty can be ignored as the NN follows a weight mapping to find output values [21,22,25] within the necessary error bounds to respond rather than solving the modeling equations. This weighting can additionally be continuously updated at the rate of the much longer isotopic evolution time scales and through reinforced learning to continually increase accuracy or allow control through system states not previously simulated.

A full short time scale simulation is beyond the current scope of this paper and so we will focus on looking at the feasibility. We would mainly be looking at the system in terms of energy deposition and neutron population. These can be simplified in terms of the effective criticality (k_{eff}) and equated with thermal deposition. The isotopic concentration are considered static here as they would drift on a time scale that is much larger than the control time scale and can be considered to be a steady state initial condition within certain bounds. However, these isotopic concentrations would be a factor on choosing the weighing of the NN and would be done prior to any real time control through the use of repetitive modeling to build a NN learning library. These weightings may also possibly be updated in parallel with a corrective feedback to the NN. As criticality would be one of the main criteria, the input source can be expressed in terms of a reactivity insertion and the output as a resultant change in criticality.

NN methods are data driven approaches where their performance is highly dependent on the quantity and quality of training data. Unfortunately, there is very limited experimental data with the desired conditions that restricts the use of purely data-driven approaches. To partially circumvent this we produce the learning data through realistic Monte Carlo simulation. These simulations, while not necessary to be initially optimized are still partially bounded by optimization goals so as to produce sufficient relevant information in a short enough time frame as to be useful for this research. To that end a hybrid heuristic [20] using an Evolutionary Algorithm with Simulated Annealing, is applied to find testable pathways and test solutions for proposed models. An Evolutionary Algorithm / Genetic Algorithm (EA/GA) is used for diversification through a population model. This also uses a breeding/cross-over model after the scoring against the objective function in choosing the next set of solutions in the iterative set.

Simulated Annealing is used by increasing intensification through the use of an annealing/cooling schedule on the randomness of how much an EA mutation can change the solution and on the size of the solution set (population) from breeding (EA terms). A "Tabu" search [38] style memory is also used to prevent loops. This allows for simulation approximation from a Monte Carlo based "non-exact" or "noisy" objective functions as is often done to good results.

3 Physics of Subcritical Systems

To develop a toy model to test our assertion, we need an understanding of the basic physics of a nuclear system. This is a quick overview of the physics of the system and it's toy model with a focus on transmutation and the principals being used for control. Neutron interaction is the primary driver for transmutation, additionally the use of a series of chain reaction produced neutrons allows for the goal of highly efficient systems. For this reason, the neutron flux spectra and its importance to fission and the overall balancing of the neutrons economy through the addition and removal neutrons must be understood in context of the new neutron source.

3.1 Neutron Flux Energy Spectra and Neutron Economy

The instantaneous balance of neutron production and neutron removal at each point of the phase space (energy E, space r, and angle Ω) for the neutron flux ϕ_0 can be found by solving the Boltzmann equation,

$$\begin{aligned} &-\Omega\nabla\phi_0(r,E) - \Sigma_0(r,E)\phi_0(r,E) \\ &+\int\int\Sigma_0(r,E')f_0(r;\ \Omega',E'\to\Omega,E) \\ &\times\ \phi_0(r,\Omega',E')d\Omega'dE' + \frac{\chi(E)}{4\pi} \\ &\times\int\int\nu\Sigma_{0,f}(E')\phi_0(r,\Omega',E')d\Omega'dE' \\ &= 0, \end{aligned} \tag{2}$$

where the overall flux change is found through adding losses and additions of neutron flux by each process. Neutron losses to capture, use in fission, and loss due to energy transfer in scattering are found by combining neutron flux acting on the total macroscopic cross section Σ_0. The changes from neutron energy transfer to a different part of the flux would be found from convolving f_0 the probability of neutron scattering transfer from $\Omega', E' to\ \Omega, E$. While additional flux from fission would be found by the convolution of the excitation function represented by the fission macroscopic cross-sections Σ_0 and ϕ_0.

This is usually solved by first assuming a time invariant case. The isotopic evolution being negligible in instantaneous effect is typically applicable for large ranges of changes but will be important later in considering time evolution. Alternatively, to include the flux from the source it may be easier to represent the Boltzmann in a matrix from for use in discrete system representation as is done in [39].

$$A\phi_{in} + P\phi_{in} + s = 0, \tag{3}$$

where ϕ_{in} represents the solution of the multigroup inhomogeneous equation using the multigroup neutron flux vector ϕ_0, A is a matrix operator accounting for neutron leakage, absorption and scattering transfer, P is a matrix operator accounting for fission neutron production, while s is the representation of the external source $s(E,r,\Omega)$ in a stationary state. This will result in an energy discrete steady state neutron flux similar to that seen in [**Fig. 3**].

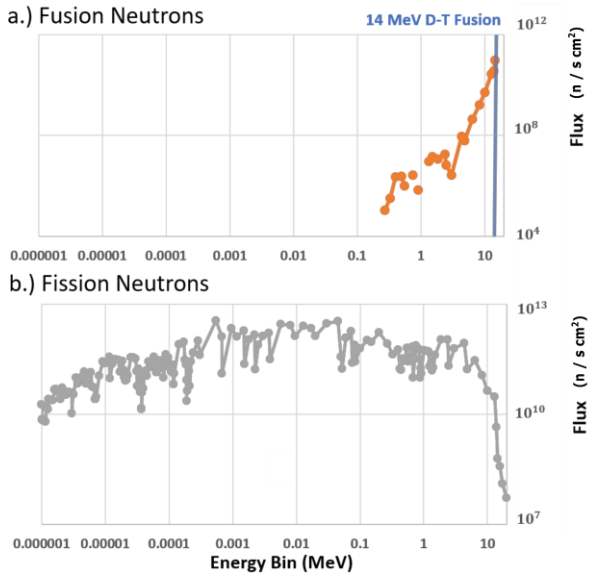

Fig. 3 The initial steady state neutron flux at interior wall boundary of a minor actinide only fuel. (a) The flux of neutrons from a singular D-T fusion source that has not yet fissioned accounting for slow down (lethargy) due to scattering. (b) The flux of all resulting fission chain reactions

Neutron generation through neutron-induced fission that can result in further fission via chain reaction provides a neutron multiplicative effect. Transmutation by fission resulting from this self-continuing process would greatly affect the input cost of neutrons to resulting transmutations and should be shown as a separable factor. This can be found through observables such as power and its relationship to the 'neutron economy' which can be found by the criticality (k) of a system. The criticality is the generation averaged neutron multiplication rate, or the probability that an incoming neutron will create another neutron by acting as a new neutron source. In a finite medium where neutrons can escape the system the effective criticality (k_{eff}) can be found by finding the eigenvalue solution that excludes the source by $A\phi + \frac{1}{k_{eff}} P\phi = 0$, which is a solution to $A\phi_{in} + P\phi_{\text{in}} = 0$ of a critical or super critical system ($k_{eff} \geq 1$), where $\phi_0 \sim \phi_{in}$ are approximately equal.

In a subcritical system ($k_{eff} < 1$) the criticality, referred to as k_{sub}, can be considered by the total number of neutron sources, or the sum of the external neutron sources (S) and internal or secondary fission neutron sources (F). This would be written as

$$S + F = S + Sk_{sub} + Sk_{sub}^2 + Sk_{sub}^3 + \ldots, \quad (4)$$

which can then be rewritten as

$$k_{sub} = \frac{F}{S + F}. \quad (5)$$

To find this we use angle space integration, denoted here by bracket $<.>$ of the source s and the fissions that would occur in an extraneous source free system using P, where F is $< n_0^*,\ P\phi_{in} >$ and S is $< n_0^*,\ s >$, and n_0^* is the importance function representing the importance of the source neutrons to the secondary fission neutrons to observables such as power

$$k_{sub} = \frac{< n_0^*,\ P\phi_{in} >}{< n_0^*,\ s > + < n_0^*,\ P\phi_{in} >}. \quad (6)$$

This is the generational average and represents a physical observables meaning to k_{sub} but is not the representation of each generation or how variations in flux for the source differ from the similar eigenvalue solution. A single value φ^* importance parameter for n_0^*, as the system approaches criticality, can also be found through simulations and comparisons of observables experimentally, where

$$\varphi^* = \frac{\left(\frac{1}{k_{eff}} - 1\right)}{\left(\frac{1}{k_{sub}} - 1\right)}. \quad (7)$$

This can then be used to break out the sensitivity to various parameters and the difference between the source neutrons and the secondary fission neutrons similar to as done in [39,40]. This can be done

through the use of generalized perturbation methods.

Critical or supercritical systems are important to separate as in such a system where a single neutron would continuously produce more and more neutrons not stopping until the system has changed through transmutation, expansion, or artificial changes in k_{eff} to bring it below unity. However, due to the very high speeds of fission and the speed of the relevant neutrons this can be difficult to control, particularly for the fast spectra and prompt heavy to delayed neutron counts involved here, having a neutron generation time on the order of 10^{-7} seconds. This can result in dangerous levels of neutron multiplication in very short times. Alternatively, a steady state arises from a sub-critical system ($k_{eff} < 1$) as the same neutron generation time and a decreasing neutron return converges to 0, resulting in a multiplication factor (M) for time independent neutron economy of

$$M = \frac{1}{1 - k_{eff}}. \tag{8}$$

Driven as this subcritical chain reaction is, the efficiency can still be propelled well past unity despite a driving cost from producing the fusion neutrons. However, such a system needs to keep close watch on what happens in time.

3.2 Transmutation by Atomic Ladder

By definition transmutation is not a steady state, as the excitation function represented by the group cross section is changed by the isotopic evolution. This includes changes from natural decay, a loss of nucleon counts induced fissionability from MA waste fission, as they split into lower nucleon counted FPs, and a change by the production of more TRUs from capture. Even though the newly capture transmuted TRUs are generally more fissionable, if there is a shell stability, the next generation may balance downwards by bringing down the criticality (and thus the efficiency, which is the original source of the MA waste). The natural decay channels, with the exception of self-fission, are also typically much slower, as they have to work through increasingly stable isotopes to drop below TRU status and are a less sure means to quicker waste disposal.

To see this isotopic time evolution, represented by the neutron flux from above, the Bateman [29,39] equation should be solved. Here the change in quantity of an isotope is found through adding two sets of terms representing losses and additions from transmutations between isotopes *i* and *j*. The first set through radioactive decay, and the next set being reactions such as fission, capture, n2n, etc.:

$$\frac{\delta N_i}{\delta t} = -\lambda_i N_i + \sum_j \lambda^{j \to i} N_j + \sum_j N_j \sigma^{j \to i} \langle \phi \rangle - N_i \sum_{\forall r} \sigma_i^{(r)} \langle \phi \rangle . \tag{9}$$

This is basically working backwards to solve what rate of change of the isotopes matches the earlier solved for flux to produce an instantaneous picture for the rate of change, this is opposed to using a full analytical form, which would be highly coupled.

To move between two different steps some assumption of invariance has to be made to decouple the times. This assumption holds typically that the time derivative of the flux from the Boltzmann is negligible, or changes only in scale not shape, or that a change in flux itself is instantaneous as a result of a large differences of time scales between neutron propagation and second order changes to the flux from isotopic changes or other means of changes such as artificial addition or removal of material. This infers that an initial state is found from the Boltzmann equation, and the time is evolved by solving the Bateman equation for a steady state flux independently and the isotopic evolution is used to find the next state. Any non-negligible flux changes are either avoided by reducing the time step till the change is

negligible, or by artificially changing the rates or flux by adding additional terms to the Bateman, or changes from modification to the flux used prior to its' solving. These changes can include; chemically induced changes such as demonstrated by Doligez in the Bateman variation in [29], and changes to the flux to account for changes due to thermal effects that can sometimes change much faster than solved for time steps but allow isotopic changes to remain negligible.

This assumed that time invariance of the flux in a changing system results in a perturbative view of nuclear systems, which looks mainly at departures from the invariance case and quasi-linear results by time stepping. If there is no departure, each end of the flux → evolution → flux step series represents a return to equilibrium, and the system is once again represented by its neutron economy's connection to observables such as power. The criticality changes between steps by approximately following the stepwise flux changes resulting from isotopic evolution represented by way of the Bateman equation. If the criticality is changed by a perturbation but it returns to equilibrium, then this change, known as the reactivity or ΔK insertion, can be simplified as just the relative power distance between the two steps over the time that the insertion took place. This is a useful view for temporarily bypassing kinetics as the overall system and local system changes can then usually be looked at by way of linear changes to the neutron economy between two equilibrium states and the time it take change between them by "reactivity insertion". A neutron economy following this view that has doubled in an hour, for example, has then doubled its integral flux or power output, while approximately following a linear path between. It must also be noted that while flux may return to the invariant case, realistically it may do so following a peak path that, being connected to the power, is not acceptable in terms of safety.

Departure from the invariance case is found by perturbing variables on the time scales they are likely to change in. We can then find the appropriate time scales that would allow equilibrium as denoted by the criticality to be achieved. This is done by representing criticality (k) in terms of reactivity (ρ) as $\rho = \frac{k-1}{k}$, so that feedbacks that result in further changes in reactivity resulting in changes in criticality can be easier to see. This is useful because the derivative of the change in terms of the factor being changed will result in a logarithmic relation that can then be shown as sum of terms known as the reactivity coefficients. This can then be done over the time of the change or insertion to find the time based change in criticality and power through the point kinetic equation. If the cases follow very large differences in allowable time steps then they can be treated as independent from each other (decoupled). If not they can be treated only within the scope of shortest non- independent time scales to minimize or linearize the coupling, then solved for the simpler case using just the coupled variables. This could be for example the Doppler shifting effects applied to a cross section where a flux can be seen as different at different temperatures. The temperature can be linked to the above neutron economy so a shift here could potentially result in the next step shifted to a greater change in temperature in the same time resulting in a feedback loop. Solving for this reactivity change by integrating all these changes over time can show how the system will evolve on this shorter time scale. If the integral is 0, then the system will again return to the invariant case path, if it diverges then it will similarly diverge in the same time scale as the time step since this has been reduced to lowest order time scales and follows the invariance assumption. This can be examined at progressing longer time scales as each term becomes independent of each other and treating their instantaneous case linearly and additively $k_f = k_i + \Delta K_0 + \Delta K_1 + \cdots$. If the coefficients for the perturbations add to a positive value then the tendency is reinforced by positive feedback to the reactivity and similarly is negatively reinforced for negative coefficients.

The ladder, or fertile breeding, process is possible due to an increase fissionability by nucleon count

[**Fig. 2**], but is made more useful by utilizing the relatively slow rate it climbs the ladder. This slower rate is due to single nucleon adding captures being slower at creating more waste by having non FPs or non-waste TRUs advance to MA status waste status than becoming too unstable to be sustained. This is simply a result of the pathway back to waste status combined with its cross section and quantity being much longer than the pathway to most probable point of fission. As a result having significant quantitates of a non-waste TRU such as uranium, plutonium, or thorium that can be transmuted into a waste MA and covers significant portions of the overall cross section, is likely not helpful to getting the most advantage of a continuous waste burning ladder process. Alternatively, if little or no additional TRU waste is created these transmutations would eventually move all transuranic elements out of the transuranic group and into the less radiotoxic natural elements. This becomes the primary component to a pathway to removal of most or all the most dangerous long-lived parts of nuclear waste.

3.3 Differences in Reactivity and Reactivity Coefficients for Control Purposes

The differing rates of the change in criticality from capture to fission, or chance of fission before decay also offer new forms of possible control based around the source [15]. If split the effects of the reactivity and their coefficients, by way of linearization through short time step, into those caused by source neutrons (ΔK_s) and the proportional secondary fissioning (ΔK_f) neutron it could then be possible to create a system that has controllability using the speed at which the source can be controlled if it is on the order of the fastest levels of reactivity insertion. This could be done by first finding system properties where ΔK_s and ΔK_f have large differences or opposing directions and then using them to balance the steady state conditions. Possible opposing reactivity insertions could be found as suggested here by way of sensitivities in time, power, spatial, and operational scheduling of processes and source inputs. An area that is likely to have a strong split in reactivity directions is the isotopic evolution around a high fission to high capture ratio split between the two spectra. Another is in balance of a rate of fast decaying nuclides and use of the intensity of fast source neutrons to make their interaction more important or less important to the point of negligible by source intensities. Even the resulting isotopic evolutions revolving around promoting different FP distributions could be used to promote desired results. This would make pulsed, feedback variable power level, or opposition to secondary fissions source operations possibilities. Each of these control operations may even be combined taking full advantage of control through the source.

A pulsed source could take advantage of large or narrow reactivity insertions. This would create an immediate increase in fission due to the fast spectra's heavy fission to capture ratio. This could take advantage of:

1. Power based reactivity coefficients that provide positive or negative feedback but within a narrow range soon to decay way.
2. Quick localized depletion of the most fissionable isotopes in bursts of outputs that require replenishment before they dominate the higher capture ratio nuclides that remain. Possibly to counter slower moving thermal reactivity insertions.
3. Alternatively, the quick addition of FPs from fission that have greater capture ratios than a removal process is removing them, used to dampen feedback in slower thermal feedbacks.
4. Changeable focus to and from short lived isotope with a greater desired cross section too short lived to build sufficient quantities for lower fluxes or by alternating passes of capture favorable

and fission favorable reactivity insertions.

This overall addition to control would be boosted by the ability to place multiple small tunable sources that allow faster coordination in a more equal measure throughout the system. Reactivity insertions by sources can be large for single source systems but suffer from gradients in relevant fast neutrons that form locally around the source. This diminishes pulsed effects away from the source and limits the use of strong but narrow effects near the source to only what is acceptable to this source entry limited area leaving capacity further away untouched. This would differ from high energy ADS systems which could suffer from its lack of controllable gradient in spatial manner for the input beam time scales due to large target requirements that require either high densities or long target path lengths. This brings another possible spatial gradient effect that may be of use from multiple sources, interference effects.

The ability to provide a near and relatively even fast spectra distribution system wide means that the control can be shaped to very narrow pulses dependent mainly on the sources temporal resolution. This would decrease the overall time for the reactivity insertion represented by changes in the reactor power (P_N) to be added or removed nearly instantaneously as shown by solving for the subcritical point kinetic equation in [39]

$$I_{eff}\frac{dP_N}{dt} = \left(\rho_{gen} - \alpha\beta\right)P_N + \alpha\sum_{i}^{I}\lambda_i\xi_i + \zeta(1-P_N) + \rho_{source}, \quad (10)$$

$$\frac{d\xi_i}{dt} = \beta_i P_N - \lambda_i\xi_i, \quad (11)$$

where I_{eff} is the effective prompt neutron lifetime, ρ_{gen} is the generalized reactivity relevant to perturbation at $t = 0; A \to A + \delta A,\ P \to P + \delta P$, α is a coefficient for the delayed neutron distribution, ζ is the subcriticality index, β is the effective delayed neutron fractions, ξ is the i-th effective precursor density.

The changeable gradients can also be utilized more readily by the method of waves, by thinking of the reactivity insertion as a traveling interfering wave. In this way time effects can be further amplified by resonance, beating frequencies, and other wave effects that can follow differing diffusion rates by neutron energies. Boosted power effects can be made to amplify or diminish by shortening times beyond the sources directly controllable input using where sufficient positive feedback effects constructively interfere with another wave only to be driven back down by second sources wave or negative power or thermal feedback effects of the slower secondary fission energy neutrons.

This would likely involve utilizing reactivity insertions or coefficients that are opposite in directions for each type of reaction or that can vary primarily by the intensity of the source. This would most easily be utilized by having one or more of any two conditions (source higher intensity, source lower intensity, and secondary fission) straddle a changeover condition for their feedback or insertion effect. Additionally longer term effects such as isotopic evolution end-points, and longer-term thermal effects can be manipulated by these choices. Isotopic evolution can be done through balancing burn rates of different isotopes based on this ratio difference. Thermal effects can also be enhanced by increased control of a thermal gradient which influence bulk transport.

Transmutation driven in this way by fusion neutrons has long offered tantalizing hints that it could be a solution, and as new fusion based neutron sources become possible [15,41,42], transmutation needs a more thorough examination within the scope of these new sources to determine the best possible setup to allow for efficient large scale real world solutions. For this reason, a transmutator using a fusion based neutron source driving a subcritical reactor core is being developed and simulations to test these assertions performed. This is a multidisciplinary effort that already has

many efforts to explore all aspects of the transmutation process in order to ensure its place as a safe and feasible technical solution.

4 Processing Control Simulations of Transuranic Transmutation

In this section we will discuss possible operational modes of the transmutation process of the transuranic (TRU) elements based on a fusion neutron subcritical condition in the molten salt [15]. This will be done with an eye towards overall trends in steady state isotopic evolutions and trends rather than a specific case real world operation. Additionally reactivity coefficients and time scale information are gathered for later use in creating testing points for short time scale operations.

The molten salt chosen for the initial tests is FLiBe ($LiF.BeF_2$: 66-34 mol%), with other salts such as FLiNaK (LiF-NaF-KF: 46.5-11.5-42 mol%) also in future consideration [14,15,34,35]. FLiBe and FLiNaK were developed starting in the late forties, specifically for their ability to dissolve actinides for use as a fuel carrier in liquid homogeneous reactors. However, it is not yet well understood what the solubility limits for many isotopes are [34]. As a generalization for most of the actinides it has been suggested that 2% mol [14] should be used. While 1% mol should be used as a limit for plutonium in FLiBe. Therefore, as an additional precaution a limit was put on the molar ratio of added nuclear waste to molten salt at no more than 1% mol. This is below the likely real limits for many mixtures that can be considered. To avoid cooling design issues, power density limits contained here do not exceed a steady state 64 W/ cm^3, which is below even the lower end of other FLiBe TRU carrying liquid designs such as MOSART [30] at 140 W/ cm^3.

The walls used in the simulation are carbon based modeled using pure graphite, for their high tolerance to radiation (low DPA), low chemical reactivity, and neutron reflectivity. This is an incomplete picture of the wall materials as laid out in [15], nevertheless being another allotrope of carbon, it is the best match for the behavior in the transmutator. This material was chosen to simplify the simulation but retain the dominant neutronic effecting characteristics of moderation and reflectiveness. However further studies about the radiation damage to the walls, chemical interaction, and the thermal behavior will still need to be performed.

The initial dimensions chosen simulate a cylindrical tank 2 m in length and 1 m in interior diameter [**Fig. 4**]. The wall thickness is 30.5 cm, chosen for its relation to 3 times the mean free path of the 14 MeV fusion neutrons, which results in negligible further changes in criticality based on wall thickness. Thus this model has a total diameter of 1.61 m in diameter, and includes a top and bottom cap of equal thickness, resulting in a total length of 2.61 m. The dimension and geometry have a noticeable effect on criticality, as will be expanded on later. Additionally, there were several optimization runs performed with 2 cm thick carbon walls at various radii, and creating annular ring or sectional divisions in the tank.

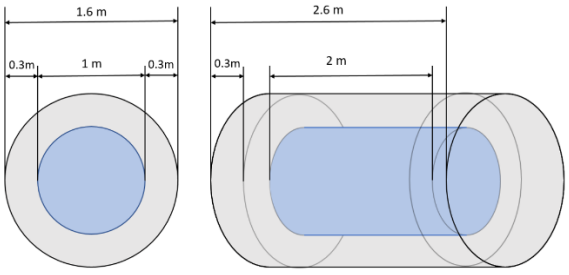

Fig. 4 *Example of geometry and dimensions of our transmutator*

Finally, it is important to note that nuclear waste often does not come in the desired form for use as either as a target mix or even as separate components to allow for mixtures and optimization. Thus some form of reprocessing is needed to prepare the target mixture and separation of end

products, even if the molten salt allows for minimal other fuel preparations to be made due to its ability to act as a solvent to the actinides [14,35,37]. There is currently only one non-weapons specific, already industrial scale, process that separates nuclear waste into aqueous mixtures that would be useful for the transmutation process known to us. This is Plutonium Uranium Redox Extraction or PUREX [37,43]. PUREX also has several closely related or derivative methods that allow for different useable fuels. PUREX originally separated both plutonium and uranium allowing for fuel reprocessing, and later was modified to extract only uranium (UREX) leaving plutonium for use as a fuel in possible Fast Reactors (FR) designs. For minor actinides separating processes there are derivatives such as Energy Solutions' NUREX and the French CEAs' (COEX + DIAMEX-SANEX). Choice of these process allows for separable groups of: U, Pu, Am + Cm + Np, Pu + Np, Am + Cm as possibly needed. These can currently reach purity levels above 90-99% which is more than enough for this purpose and for now avoid the issue of the utility of the 99% to 99.9% that is currently subject to debate for need vs. cost [37].

4.1 Source Input Sensitivity for the Transmutation of Minor Actinides

Our simulation of the transmutation of a minor actinides fuel at 10 MW constant output thermal fission power for 1 year is shown in [**Fig. 5**]. The starting mixture is 92 wt% FLiBe with an additional 8 wt% fuel which contains 7% neptunium, 1.1% americium, and 91.9% curium. This is a fuel derived from concentrating the results of a spent nuclear fuel (SNF) run. Due to the increased curium levels and decreased americium levels this is one of many possible secondary starting cases. However, this example was chosen for its more straightforward observation of ladder climbing's ability to increase fissionability. A derivative from an earlier stage of a SNF transmutation may be more appropriate as not so much curium is typically needed. This would be done by combining of separate transmutation tanks and adding fresh SNF, the end result would then only need a typical FP removal schedule and a single Pu removal (PUREX) to obtain.

The content of FLiBe while not completely unchanged, remains virtually constant due to its low interaction cross section [14,34,35] at 92% by weight (wt%), and thus the evolution of the system can be quickly observed by only looking at non FLiBe components. As shown in [**Fig. 5**] the TRUs are depleted at a linear rate until almost all TRU's are gone. This shows a consistency in output thermal power to the rate of TRU depletion.

After 1 year the amount of transuranic elements initially at 360 kg is decreased by 3.8 kg to 356 kg. Curium is decreased 4.4% from 328.6 kg to 314.2 kg, americium is decreased 10.6% from 6.6 kg to 5.9 kg, and neptunium is decreased 5.5% from 24.9 kg to 23.5 kg. Additionally 12.65 kg of non-weapons usable plutonium is produced along with less than 10 grams of other TRUs.

This operation uses a preprocessed waste such that the criticality allows the desired output power level. As mentioned, and can be seen in [**Fig. 6**], using minor actinides as taken directly from typical reprocessing methods as the primary driver for transmutation would yield a neutron multiplication effect that would be very close to unity. The $k_{eff} = 0.4$ or $M = \frac{1}{1-k_{eff}} = 1.6$ at a high 1.6 mol% (~10 wt%) concentration would be well below what is achievable with a $M = 50$ or more of typically considered criticality limits for sub critical systems. Transmutation of waste directly would thus relies strongly on the efficiency of the neutron source to result in significant amounts of transmutations having lost the advantage of the neutron rich environment from neutron multiplication. As an additional note for [**Fig. 6**] there is a nonlinear part where the concentration of MA drops enough to allow the beryllium in the FLiBe to receive a significant portion of the incoming neutrons, this

would not itself fission but would undergo n2n reactions. This would release an increased number of neutrons of sufficient energy to fission causing a boost in criticality. This boost would eventually be lost as the amount of MA to fission drops below what even the additional neutron production can fission.

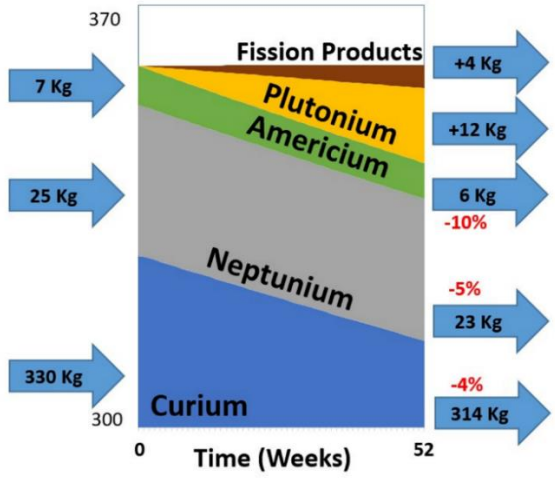

Fig. 5 Minor actinide transmutation. Inventory changes in our subcritical operation at the level of 10 MW thermal fission energy production over 1 year. (Note: the inventory mass is displayed with an expanded scale between 300 and 370 kg)

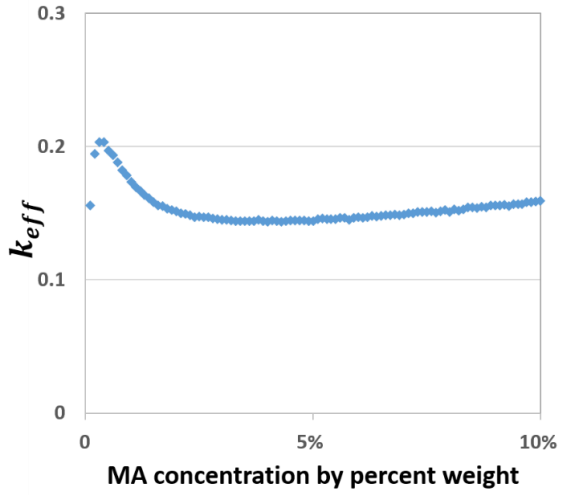

Fig. 6 Instantaneous criticality (k_{eff}) with different concentration in the molten salt. Our transmutator with FLiBe and MA taken from PUREX processed spent nuclear fuel from a PWR reactor.

Due to the continued increasing criticality based on the burn power as shown in [**Fig. 6**] this system is ran at 10 MW_{th} output thermal power as opposed to 100 MW_{th} or more. This is an important characteristic that allows for the continuous addition of criticality dampening minor actinides without significant losses in efficiencies or the power feedback controls as mention in [Sec 1.5]. For the MA burner this occurs due to the ladder climbing production of isotopes through the capture process in more often thermally moderated neutrons. This creates isotopes with more favorable fission cross sections than the original isotopes increasing criticality. Particularly: Neutron capture $^{241}\mathrm{Am} + \mathrm{n} \rightarrow {}^{242}\mathrm{Am}$ followed by β^- decay $^{241}\mathrm{Am} \rightarrow {}^{242}\mathrm{Cm} + \beta^-$ and α decay $^{242}\mathrm{Cm} \rightarrow {}^{238}\mathrm{Pu} + \alpha$, which can also alternatively branch after neutron capture to an electron capture $^{241}\mathrm{Am} + \mathrm{ec} \rightarrow {}^{242}\mathrm{Pu} + \nu$ and another neutron capture $^{242}\mathrm{Pu} + \mathrm{n} \rightarrow {}^{243}\mathrm{Am}$ another capture $^{243}\mathrm{Am} + \mathrm{n} \rightarrow {}^{244}\mathrm{Am}$ and β^- decay $^{244}\mathrm{Am} + \beta^- \rightarrow {}^{244}\mathrm{Cm}$. These decay chains have a combined half-life of less than a year and a high chance of occurring due to the large capture cross section of americinum-241. This counteracts the downward pull on the criticality from the fission products and the overall loss of TRUs to fission. Notably the highest burn ratio fuel is americium, which due to large thermal energy generation through alpha decay is the primary concern in most waste treatment plans. It also has an unfavorable cross section ratio of fission to radiative capture for a majority of the fission neutron energies [Fig. 1]. This ratio typically makes it the primary reason traditional nuclear reactors fuels becomes unusable after its accumulation, even when most of the fuel remains unused. This loss vs. gain ratio in Am-241 can allow for a desired distinction between the fast and slower spectra.

Additionally the isotopes of plutonium that are produced (Pu-238, additional to larger quantities of Pu-240) are not easily weaponizable and are useful as a possible power source thorough use as an radioisotope thermoelectric generator (RTG) [44]

especially in non-orbital satellite and space based settings. It may then possibly be considered a fulfillment of its primary goal of transmutating minor actinides into either useful or more benign product this could even be considered part of a successful transmutation if continuously removed.

The overall process is an example of a desired non-waste producing and waste driven transmutation. This type of initial loading, resulting in changes to the time evolution of the criticality, is of particular interest to meeting the long-term goal of burning disproportionately hard to transmute elements such as americium. Once the criticality tends upwards additional americium heavy mixes including the original separated with curium and neptunium from the original SNF could be added without dampening the system below practical efficiencies. They would be continuously added to level off the increasing criticality by initially acting through capture as a neutron poisons. While later, as some of the nuclides capture neutrons they will likely become the fission driver due to increased fissionability

New elements as represented by the FP created as a result of fissions over the course of the year are the direct indicators of transmutation. Removing them [34,45] will further help prevent the criticality from decreasing and possibly impairing operations as FPs as a whole if left in the transmutator insert a negative reactivity. This can be accomplished through partitioning and separation methods with various levels of difficulties and rates. This part will require a more through chemistry review, although some of the process considered are already well known and used. FP's such as some of the gases (such as Kr and Xe which are major neutron poisons) can be quickly and easily removed on-line through sparging, or gas bubbling, helium [46]. Most other FP's can be removed off-line through other various processes such as UREX or UREX derivative chemical processes. Alternatively, many FPs can also be removed by electrometallurgical processing techniques ('electro winnowing' or 'pyro processing'). These can all be implemented to various degrees in situ. Additionally, some elements such as the lower level wastes and long lived fission products (LLFP) radiotoxic isotopes of iodine, cesium, and strontium can possibly be left in the tank or in a nearby blanket to have them further transmute through neutron capture. This will not recover the entire amount of lost criticality, however it does represent a good lower bound for the criticality in its most passive undisturbed state and thus bounds the efficiency of the system by leaving them in for the simulation.

4.2 Operational Sensitivity for Transmutation of Spent Nuclear Fuel

Nuclear waste mainly comes in the form of Spent Nuclear Fuel (SNF). Our simulation of the transmutator cannot start with the MA burner initially and is thus started with SNF as would be taken from a Pressurized Water Reactor (PWR) after a short cooling period and subjected to UREX [37,43] to remove the uranium and fission products to simulate the most direct waste to initial transmutation burn of a common nuclear power system. While this is not a physical run, there are important observations in the near linear response that help in understanding how a physical run can be built. The initial simulation is started and left to run as if the transmutator was uninterrupted with no refueling or fission product removal yet considered. While the content of FLiBe is at 96% by weight (wt%). The reactor power was also considered as a constant thermal power outputted as is traditionally done [33,47–54]. In this case shown in [**Fig. 7**] the power used is 100 Megawatts thermal (MW_{th}). This would require an adjustment of the source's intensity and is done only for clarity.

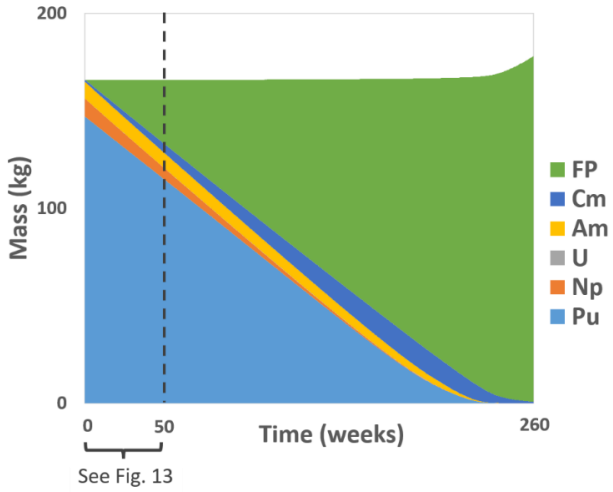

Fig. 7 Reduction of SNF by our transmutation process in time. The operation at a constant burn rate of 100MW thermal fission energy production over 5 years. (Results by linkage code M.O.P.)

Plutonium is decreased at a similar rate at 37.0 kg / year (25.1% by mass), to the overall depletion of TRUs overall at 37.2kg / year (22.4% by mass). While the minor actinide contents does so unevenly including some additional production to match the difference in overall rate of total TRU depletion. Neptunium which starts as the next most abundant element in the fuel decreases the quickest at 3.9 kg / year (42.8% by mass), faster even than Plutonium. Americium also decreases but at a relatively slow rate of 0.9 kg / year (10% by mass). Curium, however, increases at a significant rate considering its negligible starting mass at 3.4 kg per year. While seemingly at odds with the goal of burning minor actinides, this may be advantageous as use for starting fuels in other setups similarly as shown in Sec. 3.1. The relatively low transmutation rate occurs primarily due to processes normally seen in power based fission reactors. Plutonium-241 has a half-life of 14 years and β decays into americium 241. This results in a rate of about 5% a year [55] of the plutonium mass becoming americium. The initial inventory of plutonium 241 is 22 kg and thus would create about 1.1 kg in a year, which counteracts the depletion. Americium would thus not greatly benefit in transmutation while in the presence of large amounts of plutonium-241. Removing plutonium as the dominant driver of fissions would prevent the accumulation of new waste.

The primary goal of the transmutator is the transmutation or burning of nuclear waste, so determining how much waste is burnt is an important aspect of determining success. How much waste was burned can be seen in the mass of FPs produced, as the loss of mass converted to energy from fission is small compared to the overall mass. The mass here is 37.2 kg of fission products produced per year. With the proposed offline laser based spectroscopy this is possible to determine directly. However typically the mass of many different elements would be difficult to determine actively and a more observational method may also be desired for confirmation. The amount that has been transmuted (T) can also be found as a function of the measured amount of thermal energy (E) that is released into the molten salt. The transmuted mass is given by the average amount of energy released per fission ($\bar{\epsilon}$) substituted in for the number of fissions that occur (N) by combining with the molar atomic mass ($\bar{m}_a$) of what is fissioned

$$T = \frac{\bar{m}_a E}{\bar{\epsilon}}. \qquad (12)$$

While (11) does not include a breakdown of the materials fissioned nor does it include other possible heat contributors, such as beam deposits, radioactive decays and chemical processes, but does still allows for the quick determination of the expected amounts of TRU's that are burnt in any operating process through traditionally measured means. Which is still useful to see the efficiency through relative measure of waste production through captures to TRUs total burning in terms of scale. This is useable because the average values for the TRU's have a sufficiently narrow range and is not likely to consist of significant amounts of non TRU fission. This TRU specific burn is because the TRU's are also the largest cross sections in this neutron energy range by a significant amount for most operable cases and thus dominate the averages. Additionally, the other contributions to the thermal energy deposited are in general several orders of magnitude smaller than the thermal

energy produced by fission. Thus for an expected average thermal energy released by TRU fission between 180 and 220 MeV per fission, and a molar atomic mass average between 238 and 248 u. A generalization of 40.5 ± 4.8 kg per 100 Megawatt year (MWyr) of output thermal power can be estimated as demonstrated below. This can further have narrowed by using expected transmutation targets and excluding ratios that would not be in any likely operating range. SNF would have an expected transmuted mass of 37.4 ± 1.3 kg per 100 MW yr.

This continuous process along with a good approximation of the amount of transmuted waste as a function of burn power will also allow for the estimations of the amount of power needed to burn waste being produced and the waste stockpiled. Or at least put a lower bound based on efficiency by assuming an eventual paring down of each element in accordance with the stockpile level, an important key to determining how effective the transmutation could be.

In addition to starting waste and burn power a large number of factors for each operation are needed to determine how some of the key components such as criticality and isotopic inventory develops over time. These are often, but not always, optimized ahead of time and held relatively constant. This is because each of these factors can often be coupled in complex ways and deemed to be hard to change in time. Once implemented, however, time evolutions can follow the more stable linearization used above. These factors do allow for a significant level of adjustability if the wide array of inter linked effects can be handled or flexibly calculated. The major neutronic factors used for the transmutator that fall in these groups include: source characteristics, tank geometry, the removal and addition schedules, rates of various FPs, and waste.

Shown in [**Fig. 8**] are some generalized ranges of the various disparate physics (and chemistry) and their timescales related to transmutation. These include prompt nuclear fission, delayed neutron fission, neutron absorption, deuteron transport and its fusion, fast and slow neutron transport, neutron slowdown, expansion of the (local) reactive domain, chemical reactivity (s. a. FP, fueling, etc.), rep rates of lasers and other control operations, hydrodynamics, and thermal processes. We recognize that the first categories of timescales are very short or relatively short, while the latter half are much longer or very long compared with the first. We also understand that in our concept of liquid transmutator with distributed laser-driven neutron sources can make these two categories of timescales approach each other much more than the conventional solid state operation. For example, the concentrations of the components of SNF and FP can be distributed in the liquid sparsely so that their reactivity can be changed and made longer if they wished to be, rather than stuck to the precondition of the solid fuel cell values.

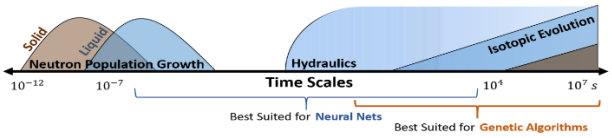

Fig. 8 Timescales related to transmutation. The brown colors refer to solid reactors, while blue colors to our liquid transmutator. Our controlling AI time scales for neural net and genetic algorithm indicated.

The convergence of the timescales gives us an opportunity to combine the monitoring and control of various processes. Since the converged timescales, toward the ball park μs regime, are called for to monitor and control the processes that influence these processes. This encourages us to introduce the AI (artificial intelligence) control and real time monitoring, as the time scales to control are beyond the human perception time scales. In the below we also outline the broad operational modes sensitively dependent on some of the crucial parameters. This sensitivity requires us to carefully implement enough monitoring and feedback control systems in our simulation. Such monitoring and real time control should not only significantly increase the safety operation, but also the efficiency of the transmutator and waste management.

Source characteristics are an important component in subcritical systems, as they provide the directly correlated resulting burn power both system-wide and locally and they couple strongly with individual systems geometry and source arrangement. Local power densities are especially important around the sources entry as too high of a local power density can cause state changes resulting in possible void bubbling which will cause sudden reactivity insertions. This would be necessary to account for. This is the case even if the parts of the system are not near a source, or reflector, or if the localized heterogeneity is well below safer power densities, resulting in lower overall average power densities and limits. Placement also has effects on losses due to the walls or containing structure from absorption, escape and changes to the flux profile from either reflection or pass through to possibly adjacent fissionable materials such as a blanket or separate tanks.

Nevertheless the key to making the transmutator sustainable is the adjustment of the concentrations of waste and FP's in the tank. By setting this process to have isotopic concentrations loop over a usable range when more waste is added then something equivalent to a fuel replacement cycle could be created. This can be a steady state operation that allows for the continuous processing of waste. This cycle is not generally straightforward to find as the concentration of isotopes in the transmutator tanks are one of the most significant factors in operation. Molten salt system do allow for many adjustments in-situ, but they are also all separately limited by chemistry and focus on individual or particular groups of elements, the waste also continues to come in the same limited isotopic ratios as initially started.

As can be seen below in [**Fig. 9**], the weekly, complete FP removal or replacement with additional SNF waste for SNF simulations can result in significant changes. This is true even if changes are limited only to the FPs produced during operation as shown here. Large possible branching pathways in natural decay and a broad range of possible isotopes from fission makes the changes sensitive to minute difference in schedules and the degree of change may sometimes not be immediately apparent upon processing. This means that planning must be done to schedule or account for these changes, which are critical to setting course of the end results and the criticality needed for efficient operation. A range of operating conditions can be planned for prior to operation, but this would require a great deal of computational power to perform many simulation to be able to account for all useable possibilities prior to operation. This may require limits on the range of other operating parameters that may have coupled effects.

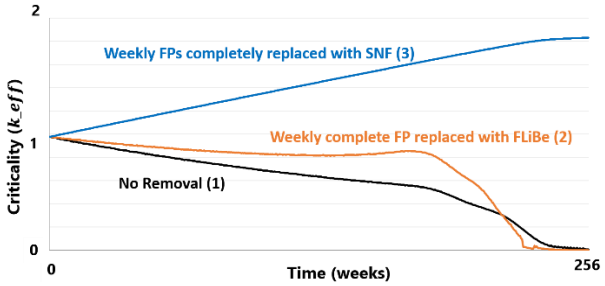

Fig. 9 Comparison of the criticality depending on the operational modes of transmutator: (1) no removal of FP nor input of SNF; (2) weekly removal of all FP replaced with FLiBe; (3) weekly FP removal of all FP replaced by fresh SNF. SNF Loading Fuel is at the 100 MW Thermal Burn Power

The intensity of the source for the MA burner can, as shown in [**Fig. 10**], below play a significant role in how quickly important variables such as the criticality can be changed. This case is opposite in direction to the SNF fuel which has further negative reactivity insertions as the input source intensity increases. Neutron source placement as compared to tank and especially multi tank geometries can also play a significant role. Simulations completed with distributions of sources at different radial placement distances show different levels of criticality for equal neutron intensity. Similarly for overall tank size, particularly when close to the collisional mean free path lengths. Nearby tanks that can receive neutrons that would have escaped will also cross contribute between tanks. These are

just some of the large number of factors that can play a role in systems operation. Which while often simple as a single variable problem can lead to large areas of seemingly chaotic parameter space due to their coupling and strong individual effects on operation.

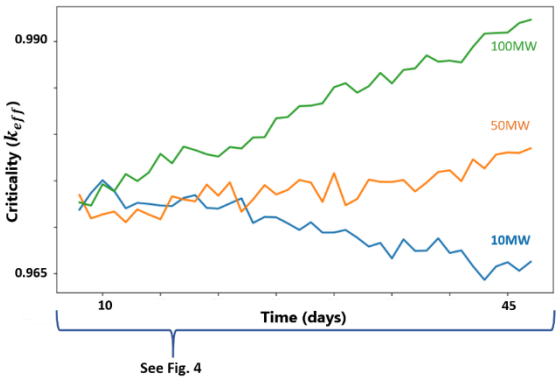

Fig. 10 Criticality time history comparison as a function of the burn power 10, 50, 100 MW thermal power production in MA burner operation as seen in [**Fig. 5**].

5 Active Control by Artificial Intelligence in a Molten Salt Transmutator

Molten salts are optically clear and accessible and can allow for homogenous or near homogenous off-loading in each tank and active and passive monitoring that can be done in real time. This would provide active feedback opportunities for a control system as well as safety monitoring for divergence from predictions. Time scales are typically considered to be the main technical hurdle for artificial intelligence (AI) implementation in solid fueled nuclear systems. There are two primary reasons for this; difficulty in real time monitoring, and time scales often shorter than computers can operate at. Solid fuels typically require removal and intensive processing before they can to be analyzed for isotope concentrations. Solid fuels also lack sufficient bulk transport to equilibrate as liquid solvents would. This would make measuring local isotope concentration both necessary and difficult in solid fuels. The shortness of the time scale for neutron growth is also an issue that molten salts acting as fuel carriers help overcome. The TRUs that are the primary driver of fission can be suspended in the low collisional cross sectioned salts resulting in lower effective densities and larger mean neutron collisional times or neutron generation times. This increase in collisional time is further enhanced with a thermal reactivity effect through Doppler broadening that increases the capture to fission ratio in the resonance regions of the nuclear cross sections allow for a negative temperature coefficient for non-source neutrons. This negative feedback coefficient provides a major component of the inherent safety of aqueous homogenous systems such as molten salts. As the thermal effects are less important for the faster neutrons of the source it can also act as separation point between fast neutron effects produced by the controllable source and slower neutron effects of the bulk secondary reactions.

Time scales of control would then be the major obstacle to feedback control if real time monitoring due to the molten salt can be done. Traditional time scales for nuclear control are still many orders of magnitude longer as most forms of control rely on thermal neutron effects and the moderation required to get there. These include almost all considered reactivity coefficients thermally capturing neutron poisons, to thermal expansion, thermally driven Doppler broadening, and moderating and void effects which are typically seen as spectrum hardening or softening due to changes in capture or elastic scattering through collisions. The primary effect that would stand out in fast spectra is the isotopic evolution. Fortunately, the lower effective density increases the rate of transmutation in proportion to the original contents. This is another extreme in time scales, typically considered too long to be an active part of considerations and left for external optimization and not used for control. Nevertheless, there may be significant advantages in control from isotope based criticality monitoring,

particular in relations to feed forward criticality predictions.

AI has already been commonly used for neutronics modeling, such as in computational optimization. Often it is used to greatly improve the speed of long-term fuel cycle simulation. However, they have not yet found use in control. This is mainly due to the lack of perceived advantage informed by how traditional systems operate more so than technical or feasibility concerns of alternative systems. A large part of this has to do with the choice of solid fuels rather than fluids, and the difference of time scales this confers. The other part has to do with the limitations in sub critical systems typically lack a range of methods for quick control, from source limitations on input locations to number of sources with separate intensities. The options for adjusting these methods are typically perceived as singular or static to slow moving that have limited active opportunities for control or optimization. This does not usually apply to molten salt fuel carriers, and for sources that are miniscule in size, have tunable repetition rate based intensities and are easily placeable in many otherwise unusable locations. This allows a form of control arising from the input source's distinction of energy from that of the typical released energies of fission as seen in [Sec 1.5]. Often because of the large time scale of differences, which typically results in the neutron flux spectra being considered spatially invariant over a cell region. This leads to solutions to the neutron growth rates being treated as instantly uniform and applied over the entire system requiring system sized but slower spreading reactivity insertions. While this is effectively true in traditional case when considering the difference between neutron diffusion rates for the fast spectra compared to the changes that more often effect the thermal spectra dependent fission range of the major actinides such as U^{235}. It is an after effect of treating neutrons at higher energies as unimportant until after they are moderated.

If there are multiple sources disbursed throughout the system, and they overlap in such a way that the higher energy source neutrons play an import role in the entirety of the system, then their adjustment can be also treated just as effectively instantaneous. An example of this is shown in [**Fig. 11**] highlighting the difference between a large single source and smaller distributed sources. This difference may be important as both a near instant global reactivity insertion or by taking advantage of sharp differences between the importance of the faster source neutrons and the fission neutrons. A possible pulsed system could also treat both the reactivity changing effects and the diffusion of spectra by neutron energy can be treated as a series of coupled waves spreading throughout the system. These can be coupled in such a way that effects can be amplified and diminished by their interference or overlaying effect this would allow temporary boosts that diminish in turn as two waves pass through or an opposing effect arrives. In this way feedback effects can be made transient by the same time scale as their traveling effect, allowing for controlled boosts of transient near critical or even super critical effect that would diminish without feedback while keeping feedback causing reactivity changes from aligning the feedback reinforcing effects.

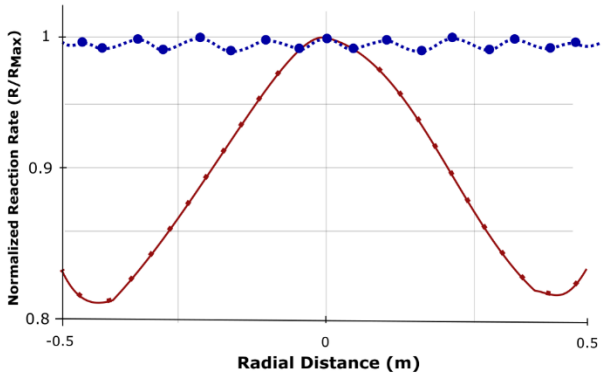

Fig. 11 Relative reaction rate based distance from center of example cylindrical core with reflective wall. Single source vs. multiple sources

5.1 .Possible Advantages of Artificial Intelligence Control

Overall AI has many possible uses and serious advantages in the areas of model selection and control that can be explored. With the aid of AI it would be possible to explore multiple tanks each with different concentrations of TRUs arranged in such a way that their secondary neutron production can be different than closer tanks. These tanks can also have their emphasis changed to focus on tanks that are operating in different stages of an overall transmutation process. This is advantageous as operation in individual stages can be less readily transparent as beneficial, such as through the accumulation of a curium as demonstrated in the minor actinide burner, but be used in longer terms such as possibly allowing increased transmutation rates for harder to transmute isotopes. Other areas include source placement. Traditionally sub critical sources are placed furthest from all walls due to wall absorptions resulting in efficiency losses. However, small localized sources can allow power densities to be more efficiently spread over the entire transmutator for more efficient feedback and thermal control. This may also allow for sectional shutoffs and maintenance without impacting total operability, or mixed operating modes where parts of the transmutator can be working on different stages of the transmutation process and changing them can be done by changing additive and removal schedules or source intensities rather than physically moving molten salt contents. These can be added in addition to thermal hydraulics, adjusting for regional densities, heat flow with forced convection, and localized input neutron source intensities to greatly enhance safety and efficiency of the transmutator. Finally different FP removal and waste addition schedules can be actively tailored to keep waste transmutation in ratio with the stockpile of waste needed to be disposed of, while keeping in mind the many different factors in removal for each isotope. This would prevent the buildup of a new category of waste and allow for the eventual complete transmutation of all waste. Thus implementing this kind of scheme will help introduce a methodology to explore repeatable loops for steady state operations.

Monitoring and feedback applied to the change in the criticality (reactivity insertion) can be through monitoring of the output thermal power level and modifying the mass flux of FPs out with additional waste added to replace it. This process as shown in [**Fig. 12**]. If applied on a timed schedule can result in the desired steady state operation loop where MA waste is continually added to be burned and the FP's from a similar process burned earlier is removed in equal measures.

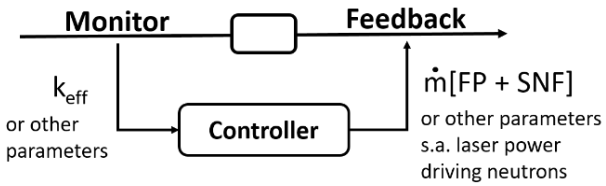

Fig. 12 Monitor various operational parameters such as k_{eff}, concentrations of SNF, FPs, etc., which is processed by the controller. Feedback the changes to the operation parameters s.a. the input or removal of SNF and FP and the laser power level, etc.

Overall it is shown that transmutation can be made a viable solution, by taking advantage of the increasing fissionability that comes through increasing the atomic mass. A possible implementation of this can be seen in [**Fig. 13**]. Utilizing this increased fissionability only speeds up what must eventually happen by nature, which is the complete removal through transmutation of all the minor actinides. Optimization through choosing methods to ensure pathways to get there will be the primary goal of future studies. Focus on the in-process cycles such as removal and waste additive rates will also be explored. This will be used to create an equivalent to a fuel replacement cycle that burns the desired type of waste using only waste as an additive. This may be initially inefficient, but has results that more than make up for this lost efficiency. These would be the primary focus of the future directions. However, to get there the completion of some aspects of the AI control system will need to be completed and verified.

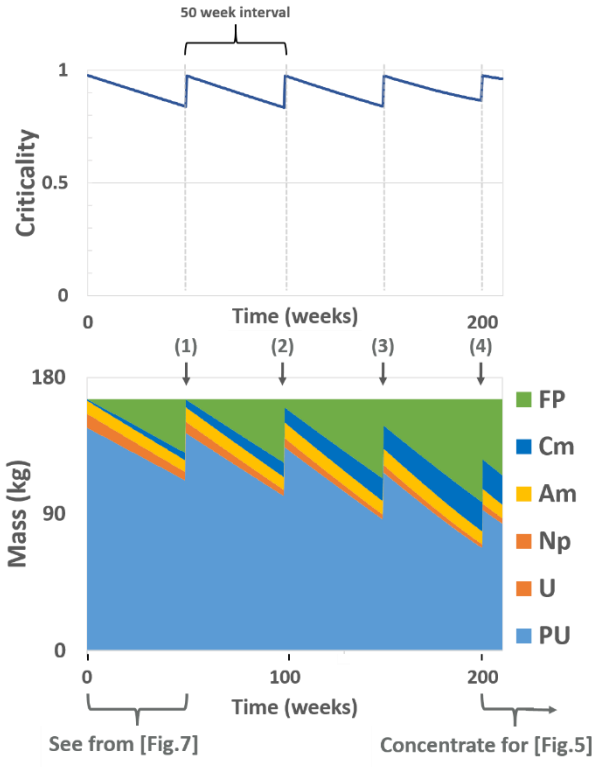

Fig. 13 An example of feedback control scenario, showing (a) criticality change and (b) isotopic evolution with operational scheduling of FP replacement with fresh SNF waste at 50 week intervals. Where to obtain a repeating cycle the percentage of FP replaced with SNF were: (1) 99%, (2) 86%, (3) 66% and, (4) 41%

While thermal hydraulic simulations are not required for early stages of simulation, they will also need to be done to ensure that the transmutator can safely sustain the reactions taking place. This is a result of how the accumulation of thermal energy from fusion and its removal limits is the main ceiling to efficiency that we expect to encounter. Additionally, the homogeneity effects should be validated due to the significant role the isotope density plays in this transmutation process. In-process material changes as outlined in [15] through refueling and FP removal will also need to be accounted for in greater detail, including their schedules and inclusion of process limitations. This would be tuned by an AI that monitors the neutronics conditions in the system and makes an adjustable feedback loop for performance. In instituting such control elements in our code, we should benefit from our experience in working closely with the AI scientists (such as those of Google under the ongoing collaboration established between TAE and Google), as seen in ref [56].

6 Summary

In principle the elimination of minor actinide waste through transmutation has been known to be possible, here it is shown that there are controllable sensitivities in reactivity that could be used to realize transmutation as a practical solution. These sensitivities in addition to normal nuclear process would be used in a systemic optimization process that improves the effectiveness of the waste elimination. An example of this sensitivity and possible process step can be seen best by the minor actinide burning example where it is shown there are configurations that can, with additional operational control, transmute waste without the creation of more waste. This example also demonstrates how it may be possible to overcome local minima efficiencies that are sometimes focused on by trying to find power producing processes before considering the waste treatment processes. Typically this minima comes about as a focus on one of the lower hanging fruits such as the use of more readily available fissionable fuels lower than the waste meant to be treated on the atomic ladder. This process would still allow for an opportunity for the type of further optimizations and exploration of controls that can result in the power multiplier effects envisioned by Rubbia [11] as a future extension. However, power production can now be left as something to be found after treatment of the currently long unsolved problem has already begun and to more completely meet the true objective of waste treatment processes. This would be as a similar technology to the deep waste repository which looks to the management of waste as a true back end solution, with according attention paid to the safety, and efficiency in terms of

manageable costs and operation conditions and benefits for its expected time scales.

Through the simplified simulations shown here we can see that by adjusting previous assumptions about limits, by pairing the complementary properties of a liquid system and taking advantage of new cheaper smaller and distributed fusion neutron sources currently being developed, we can take advantage from multiple less explored sensitivities and capabilities. These new capabilities rely on the ability to take advantage of the sensitivities to source power, multi-source placement, source intensity tuning, as well as homogenizing and bulk transport effects in a safer molten salt system that allow for better control in the sensitives to operational scheduling and isotopic evolution in a controlled system. Shown directly is part of a possible initial burn pathways from SNF currently process able from nuclear power plants as in [**Fig. 7**] that are controlled to reach desired isotopic ratios to then allow for steady state burns in the style of [**Fig. 5**]. Steady state can be done through adjustments to the overall operational scheduling that cycle the applied power levels but keep in tack the near linear progression of their isotopic evolutions. This is possible due to SNF's relative insensitivity to the FP and FLiBe carrier ratios at long time scales except as an overall decrease to their operating power due to moderating effects that are less important to the fissioning of the SNF's initial primary fission driver plutonium.

The sensitivities could be taken advantage of by the use of AI to adjust operational sensitivities in the style of [**Fig. 13**] to allow a near linear progression in the isotopic evolution while keeping the overall neutron flux up through the use of breeding to climb the atomic ladder. Breeding would provide compensation in fissionability to compensate for the inability or rate limited removing of all or some FP's. This removal would keep the system from going so deeply subcritical that neutron multiplying advantages in efficiency are lost. Through the concentration of a resultant mixture, and removing non waste TRUs to leave behind just the MA waste transmutation without creating new waste from TRU capture is also possible. This could be done through use of the sensitivities which could allow for greater source based control. In addition this method would be primarily targeting Americium which is the hardest waste to get rid of. This may also allow excess neutrons to be used to transmute possible blankets of LLFP waste in the future.

Control of the neutron flux would primarily be through the spatial spreading by homogenizing or by temporal manipulation of the power density while keeping the fissionability strongly dependent on the source through isotopic evolution, which could allow for fast non-thermal speed controls. This control could be accomplished by finding points of opposing or directionally turning points of reactivity insertion sensitivities as done here, and through power sensitivities, where faster burnups can be triggered to a feedback that shortly reverses. Such as what could be found around a peak similar to the weekly FP removal [**Fig. 9**] (but not shown here). Or that would allow compensation for increased FP production to evenly balance with atomic ladder.

This can all be monitored by an AI that follows tailoring made possible by the convergence in time scales of its ability to effect change in a system and the large range of multiple spatial controls. This could take advantage of all the sensitivities here and add more through the differences and sensitivities in short lived isotopes in the isotopic evolution, or periodic thermal gradients changing densities in thermal hydraulics through differential heating, or even spatially separate and directional pulsing through reactivity feedback effecting waves with fast spectra dependent fission. This could be affected to adjust power densities, and isotopic evolution pathways, while keeping the system in a state unable to maintain super critical for dangerous lengths of time on its own.

Combined these updated avenues for exploration in reactivity sensitivities and their use in a transmutation system could allow for efficient and through waste burning tailored to fit any TRU waste in question without creating additional MA waste.

Which with further work on the transmutation process in the areas of chemistry, and material science may then finally begin to reduce the long standing nuclear waste problem.

In conclusion we have devised and created a numerical simulation tool utilizing the Monte Carlo based neutronics of MCNP along with the isotopic evolution tracking of ORIGEN and built using Python to demonstrate how atomic ladder climbing, by breeding, can fix power production focused nuclear waste disposal issues by using heavier fuel to ensure a diminished secondary waste producing disposal process. This code accomplishes this by considering the isotopic monitoring made possible by a molten salt core and using the sensitivities of reactivities in nuclear processes, based on properties of a distributed tunable neutron source, to allow for the use of AI based operational feedback control by converging the time scales of nuclear and control processes. This code may also be used for broader nuclear energy problem settings.

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