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IRVINE

# Neutronics Study for AI Controlled Fusion Driven Transmutator

DISSERTATION

submitted in partial satisfaction of the requirements

for the degree of

DOCTOR OF PHILOSOPHY

in Physics

by

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Dissertation Committee:

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# DEDICATION

This work is dedicated to my wife, Yuqi, who has been a constant source of support and encouragement during the challenges of graduate school and life. I am truly thankful for having you in my life.

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## REFEREED JOURNAL PUBLICATIONS

Electron Dynamics in the High-Density Laser-Wakefield Acceleration Regime, B.S. Nicks, S. Hakimi, E. Barraza-Valdez, K.D. Chesnut, G.H. DeGrandchamp, K.R. Gage, D. B. Housley, G. Huxtable, G. Lawler, D.J. Lin, P. Manwani, E.C. Nelson, G.M. Player, M.W.L. Seggebruch, J.Sweeney, J.E. Tanner, K. A. Thompson, and T. Tajima, Photonics 8(6), 216 (2021).

Collisions of low energy electrons with isopropanol, M. H. F. Bettega, C. Winstead, V. McKoy, A. Jo, A. Gauf, J. Tanner, L. R. Hargreaves, and M. A. Khakoo, Phys. Rev. A 84, 042702 (2011).

High accuracy optical inverse square law experiment using inexpensive light to frequency converters. Keith H Wanser, Steve Mahrley, and Joshua Tanner, IOP Phys. Edu. 47, 174 (2012).

Low-energy electron scattering by tetrahydrofuran, A. Gauf, L. R. Hargreaves, A. Jo, J. Tanner, M. A. Khakoo, T. Walls, C. Winstead and V. McKoy, Phys. Rev A. 85, 052717 (2012).

# ABSTRACT OF DISSERTATION

Neutronics Study for AI Controlled Fusion Driven Transmutator

By

Joshua Tanner

Doctor of Philosophy in Physics

University of California, Irvine, 2021

Professor Zhihong Lin, Chair

Neutronics study to determine the validity and real world extendibility through Artificial Intelligence (AI) optimization of a transmutation concept with the aim of significantly reducing the radiotoxicity of Spent Nuclear Fuel (SNF) together with its required storage duration and volume. In this transmutation concept (the transmutator) utilizes a novel source providing laser-generated neutrons to transmute transuranic elements separated from SNF and dissolved in a molten salt within a subcritical core. The source neutrons are generated via beam-target fusion whereas the beam is created by laser irradiation of nanometric foils through the Coherent Acceleration of Ions by Laser (CAIL) process. This relatively low deuteron energy is catapulted by fusion and eventually by secondary fission processes. A significant factor in this study is that this can be accomplished using relatively cheap fiber lasers terminating onto small scale targets. Consequently, this makes possible the use of multiple tunable and distributable neutron sources. Such a source has not previously been considered and encourages an investigation with the aid of AI into new spatial arrangement and temporal control operation strategies as done here. This source is combined with a molten salt core whose liquid state allows and facilitates homogeneity by mixing, safety, laser irradiation, in-situ processing, and monitoring of chemical and physical properties. The combined use of molten salt and laser also allows for the introduction of rapid feedback or feedforward control of the system's operation. This further extends the transmutator concept as this work demonstrates here by neutronics simulations

showing: Efficient and continuous Minor Actinide (MA) waste only burning through AI optimized and balanced processing scheduling without the use of isotopic separation. As well as shaped thermal reactivity insertion for increased tank usage efficiency and active thermal management.

# Chapter 1

## 1 Introduction

### *1.1 Climate Impact*

The world is facing a major global crisis at issue is nature's response to human activities, rather than human confrontation, i.e. the human-created global issue of climate change and related global energy issues. It is apt for us to consider, study, and act in global scales on this issue, as our human impacts have become so permeating and global.

Since the Industrial Revolution, human societies have developed and enjoyed the convenience of fossil fuel energy. Further, since the nuclear revolution of the 20th century, human societies have entered, and been indulged in, by the power and convenience of nuclear energy. Meanwhile, humanity has conveniently forgotten or postponed the treatment of its exhaust (backend) side of these powers, i.e. $CO_2$ and radioactive waste production associated with energy production. All the while the consumption of fossil fuels have been dangerously accumulating $CO_2$ in our Earth environment. Scientific literature is in good agreement confirming that the climate changes have been incurred and are accelerating.

Climate change and its majority energy issues derive from an imbalance in priorities between production and waste management. We can't let it pass on to the same with nuclear power as with fossil fuels. Though nuclear energy production is in principle insulated from the $CO_2$ production of fossil fuels, we have not realized the treatment of radioactive waste production associated with nuclear energy production, which could leave its impact as long as millions of years on Earth. Transmutation is a possible solution and would help with this matching of priorities. Consequently it is worth studying.

## *1.2 Origin and Components of Nuclear Waste*

Nuclear waste can be the result of many activities, including nuclear energy, nuclear medicine, nuclear research, rare-earth mining, and nuclear weapons reprocessing or development. However, nuclear waste from energy production makes up the vast majority of the unique long lived danger that compel this research and so the characteristics of this subset of nuclear waste of concern, specifically parts of the Spent Nuclear Fuel (SNF), are highlighted here.

### 1.2.1 Energy and Waste Production

The climate crises is the largest and most urgent problem we face today. The climate crisis is also fundamentally a problem of priorities. This problem stems from humanities' over focus on the means of production at the neglect of how to process the waste created from that production. The brunt of the climate issue also revolves around energy production, typically through their emission of greenhouse gases. However, while within nuclear energy we already have and use a well-developed source of power that seems to avoid the problems of an enormous carbon footprint. It unfortunately has other issues that of: Weapons proliferation, Safety, and Nuclear Waste. These issues typically make it an often debated subject on whether it really is the solution that we need, even while it is already in significant use.

Nuclear power currently generates more than 10% of the world's electricity through about 440 reactors operating in nearly 30 countries [1]. Furthermore, in the countries where nuclear power is employed, it can be a major source of the overall power generation as shown with France generating roughly 70% of electricity from nuclear. This results in an ongoing and significant amount of waste being continuously generated in an area of high dependence and already significant structural investment. Nuclear power generation currently produces a stream of radioactive Spent Nuclear Fuel (SNF), with about 10,000 tons created each year according to International Atomic Energy Agency (IAEA) reports [1].

While how the world approaches power generation in the future is not the focus of this work, it is clear that if nuclear power does play a part the generation of waste would need to be addressed. Looking at just the reduction of the production of waste, as well as increasing safety, and addressing proliferation issues through the continued use of nuclear power, there are already many suggestions and innovative ideas. This is exemplified with the many proposed generation IV (gen IV) reactors which are, with an international effort, currently in development. However, gen IV reactors still place the priority on the production of power and often does little to examine the issue of a backend for what has already been created. Yet even if nuclear power is not a significant part of our power generating means in the future, there is already a large quantity of waste that should not be ignored. Worldwide SNF inventory is approximately 300,000

metric tons which has developed from almost 70 years of past power generation [1]. Waste management sufficient to handle at least this preexisting waste burden in concert, or even outside the scope of energy production, should then become a necessity.

To highlight the two main hazards of nuclear waste, and provide a quick assessment of the degree of radiotoxicity and the time scales they involve, the IAEA created a five level classification for radioactive waste based on radioactivity and heat output. This is further simplified into the designation of "High Level Waste" (HLW) and "Low Level Waste" (LLW) where HLW includes categories I (high-level of radioactivity, long-lived) and II (intermediate-level, long-lived) and LLW encompasses the others. The HLW in particular is problematic to deal with and their storage or elimination is the primary focus of the research here and elsewhere. The definition of nuclear waste is also sometimes a controversial one, since what may be considered waste in some countries may be considered valuable fuel or too susceptible to diversion for weapons development in others. The most important example is the plutonium from spent fuel, considered waste in, for example, Spain and the US and then used as part of mixed oxide fuel (MOX) in others such as in France and Japan. The problems radioactive waste presents however are universal and are a major drawback to the production of energy through nuclear power.

The origin of waste within SNF is a result of less fissile or reaction dampening materials accumulating in the solid pins used as nuclear fuels until the point that they become no longer effective as a fuel. Because this waste is trapped inside the solid this occurs before a significant amount of the fuel has been used. As shown in Table 1, which shows the typical components of 1 ton of SNF from the common operation of a Pressurized Water Reactor (PWR) operated until no longer useable by undergoing a $50\,\frac{\mathrm{GWd}}{\mathrm{t}}$ burnup before being placed in a cooling storage pool for 6 years. The solid pins with an actinide fuel such as uranium oxide metals ($UO_2$) usually enclosed in a Zirconium Cladding would be used, but about 93% of the original fuel will remain and the rest has become various levels of waste. The two main HLW components are the transuranic (TRU) elements and some Long Lived Fission Products (LLFP such as Tc-99 and I-129). The

TRU's in particular, which are produced through consecutive neutron captures and their subsequent decay chains and where half-lives of some isotopes reaches millions of years, are responsible for most of the long term radiotoxicity from SNF. For this reason, we will consider the waste to consist of these groups:

- Unused Fuel: Such as uranium oxide
- Possible Fuel/Possible Waste: Plutonium
- Minor Actinides (MA) waste: TRUs (except plutonium)
- Fission Products (FP): All remaining fission byproducts.

**Table 1** Composition of 1 ton of SNF from 4.2% enriched UOX fuel in PWR with 50 GWd/tM burnup and 6 year cool down [2]

| **Composition of 1 ton of SNF** | | |
|---|---|---|
| **Element** | **Isotope** | **Mass (kg)** |
| U | 235 | 7.6 |
| | 236 | 5.46 |
| | 238 | 922 |
| Np | 237 | 0.7 |
| Pu | 238 | 0.339 |
| | 239 | 6.09 |
| | 240 | 2.84 |
| | 241 | 1.33 |
| | 242 | 0.85 |
| Am | 241 | 0.502 |
| | 242m | 0.000902 |
| | 243 | 0.205 |
| Cm | 242 | 0.000005 |
| | 243 | 0.000656 |
| | 244 | 0.0715 |
| | 245 | 0.00611 |
| | 246 | 0.00762 |
| FP | | 51.997 |

### 1.2.2 Radiotoxicity

Radiotoxicity is defined as the activity of a certain quantity of radionuclides weighted by the intake dose factors. This indicates the degree of effect from a "biokinetic" perspective for any who has ingested or inhaled any of these substances. A measure of the hazards from waste elements can then be provided by the radiotoxicity arising from their radioactive nature rather than their chemical form. The rationale for a focus on the TRU parts of the SNF is thus exhibited in **Fig. 1** in terms of radiotoxicity. A reference point is the radiotoxicity associated with the raw material (~7.83 tons of uranium) used to fabricate 1 ton of enriched uranium, including not only the uranium isotopes, but also all their radioactive progenies. As shown the radiotoxicity of the FPs dominates the total radiotoxicity during the first 100 years. Long-term radiotoxicity is dominated solely by actinides, mainly plutonium and americium isotopes.

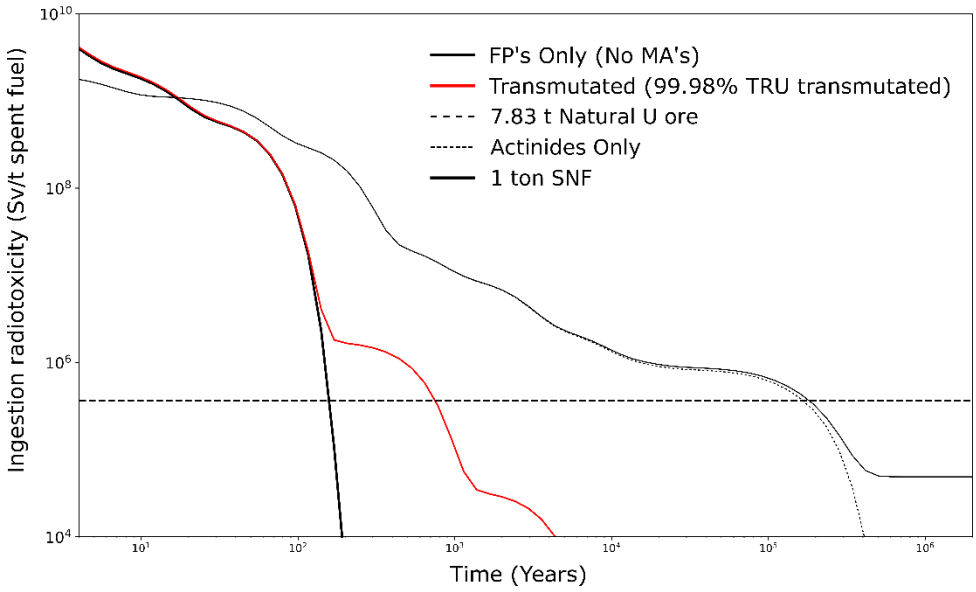


**Fig. 1** The ingestion radiotoxicity for 1 ton of the following: Spent Nuclear Fuel, Fission Products of SNF, Actinides of SNF, and scenario of 99.98% of all TRU's Transmutated. Reference line given for 7.83 tons of natural Uranium ore which would be needed to process into 1 ton of Uranium fuel.

The reference radiotoxicity level is reached by SNF after periods of more than 100,000 years. In more detail, the radiotoxicity of FPs dominates the first 100 years after discharge and decreases to the natural reference level in about 300 years. However, in the longer term, the radiotoxicity is mainly dominated by TRUs, particularly plutonium isotopes and decay products of Pu-241. Approximately 100–1000 years after fuel discharge, the radiotoxicity is dominated by Am-241, the radioactive daughter of Pu-241, with a level of about $3x10^7 \frac{\text{Sv}}{\text{ton}}$ U, i.e., about 300 times as large as the natural reference. Between 1000 and 10,000 years, radiotoxicity is dominated by Pu-240, with a value of about $4x10^6 \frac{\text{Sv}}{\text{ton}}$ U. Thereafter, Pu-239 is the main contributor to radiotoxicity with a value of $2x10^6 \frac{\text{Sv}}{\text{ton}}$ U. Beyond 100,000 years, the total radiotoxicity decays to the level of $10^5 \frac{\text{Sv}}{\text{ton}}$ U. After that, the main sources of radiotoxicity come from the descendants of Am-241.

### 1.2.3 Limited Options for Waste Disposal

To date, other than a deep earth burial of the SNF, there are no well-established long-term approaches that are available to dispense these radioactive materials. Deep geological storage, while considered safe, has several issues that has led to many started and canceled projects but only 1 existing deep geological storage that still does not yet store HLW at Onkalo (in Finland) after decades of attempts. The intrinsic radiotoxicity of some of the elements present in SNF often justifies the great concern of the safety authorities in the control of these substances, as the possible leakage of some of these elements into the biosphere could have unpredictable consequences for public health and for the environment in general. This is also no idle concerns as many difficult to control threats such as minor container defects, or penetration of water into the repository could occur. Water penetration could set the conditions for a recriticalization of the waste due to the reflection and thermalization of the radiation, which would otherwise escape from the waste

canisters. A recriticalization of the waste elements could have fatal consequences not only for the containing units but for the whole repository as well. Moreover, the persistence of these elements makes changes to the geological conditions of their storage possible and thus these changes facilitating the escape of radiotoxic waste centuries after the closure of the repository also possible.

Most importantly deep geological storage is not a means of reduction only a means of storage away from likeliest danger. Transmutation is the only means of reducing waste. Additionally, even in the case of only partial transmutation it can be combined deep geological storage with smaller hurdles for greater effect.

### 1.2.4 Nuclear Fuel Cycle

The goal of this research is to suggest a method to transmute radioactive transuranic elements and thereby to reduce the overall radiotoxicity of the SNF. This is the backend of the nuclear fuel cycle, in contrast to the frontend (mining, enrichment, fuel fabrication) which is the "energy production" part of the nuclear fuel cycle. As human activities globally grow, so does the importance on the backend science to dispense the SNF waste associated to this increased energy consumption and accumulated waste.

The backend management of SNF can follow three main paths:

(1) The once-thru cycle (OTC) where the SNF is sent to storage without reprocessing apart from packaging to isolate from biosphere;
(2) The reprocessing fuel cycle (RFC) with recycling of plutonium and uranium using PUREX;
(3) Partitioning and Transmutation (P&T) [3].

OTC is where the fuel goes through the reactor with no post processing and after the spent nuclear fuel leaves the reactor the whole pin is considered waste including the unused excess uranium. This is the process currently employed by the majority of the world and in particular the US.

The RFC method, which removes and reuses the unused fuel components of SNF, reduces the longevity and overall volume of the waste that would need to be stored. However, RFC does not reduce any of the MA or lower level wastes present in the FPs.

Alternatively, the P&T method as would be used in this and most transmutation schemes is an additional step after RFC post processing that takes the minor actinide, and optionally the plutonium and possibly even select fission product, recovered from post processing and seeks to reduce them or completely transmute them. However, while the OTC and RFC method reached industrial scale maturity, P&T is currently in the research stage.

Even though the class of highly toxic long-lived radioactive components of SNF (transuranic elements) is a minority, their toxicity is particularly high. By following the P&T management we greatly reduce the radiotoxicity of SNF. This motivation for transmutation may be seen in **Fig. 1**. Overall, the P&T of the SNF allows 100 times volumetric reduction and 1000 times duration reduction of the storage facility of the remaining waste (mainly fission products) [4].

## *1.3 Transmutation*

Neutronic transmutation is transmutation caused by an incident neutron resulting in either fission or capture. Transmutation is this manner is the only means of reduction of the nuclear waste. The spent fuel contains several elements which are resistant to further elimination from the slower thermal neutrons in thermal

reactors. Fortunately, a vast majority of the long lasting radiotoxicity (> 500 years) are found in a few of the separable TRU elements making up only about 1% of the spent fuel. In order to eliminate these resilient nuclides (TRUs and LLFPs) in an efficient way, fast neutrons (> 1 MeV) are necessary. Fast neutrons are particularly important for TRUs which can generally only be eliminated by fission, while LLFPs can be primarily transmuted by neutron capture.

These fast neutrons can be produced for transmutation in several ways such as by Molten Salt Fast Reactors (MSFR), which include Russia's MOSART [5,6] and Europe's eponymous MSFR [5,7], Accelerator-Driven Systems (ADS), such as MYRRHA [8,9], and fusion-fission hybrid models. The concepts presented here would fall into both ADS and fusion-fission hybrid. These collectively form the partition and transmutation waste management scenario and have the goal to eliminate 99.9% of the TRUs. These goals if technically feasible and economically viable present a realistic and safer alternative to the geological repositories in order to eliminate HLW.

### 1.3.1 Transmutator

This neutronics study was specifically for an examination of the core principals of the transmutator as presented in [10]. In this transmutation concept (the transmutator) utilizes a novel source providing laser-generated neutrons to transmute transuranic elements separated from SNF and dissolved in a molten salt within a subcritical core.

The overall transmutator system is based on distributed neutron sources. Each neutron source is driven by a unit of fiber laser system that drives the production of fusion neutrons. The fusion neutron energy (such as 14 MeV neutrons from the D-T fusion reaction) has an advantage in transmutation. As shown in **Fig. 2**, 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.

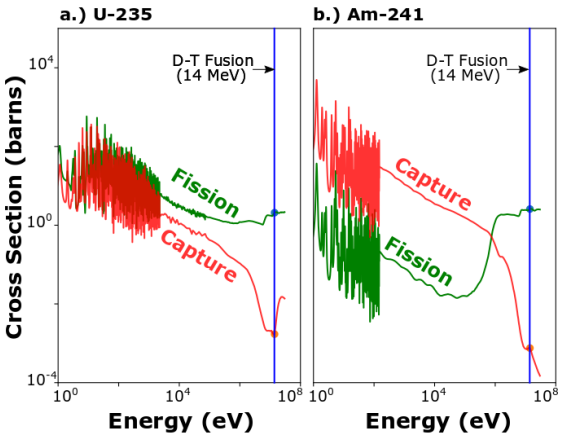


**Fig. 2** Comparison of cross sections of the fission induced by a neutron and the neutron capture by $^{235}$U and $^{241}$Am

This helps to overcome small dips in the fissionability that would otherwise be steadily increasing from an increasing nucleon count. 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.

To develop the transmutator as presented in [10] 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 [6,11–13] 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).

# Chapter 2

## 2 Neutronics

To develop a model to test our assertion, we need an understanding of the basic physics of a nuclear system. Additionally, this should be the context of the method of best approach to be solved. As such a quick overview of the physics of the system with a focus on transmutation and the principals being used is presented here. 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 neutron source.

For this system for the sake of generalization and possible optimization we will have open parameters that are not known at the start as to which is the best direction to go, particularly in geometry. This changes the focus from the more specific and limited cases solvable by things such as exact analytical solutions, diffusion theory, and even the generalized Neutron Transport Equation to a more a broadly useable numerical methods.

## *2.1 Physics of Subcritical Systems*

A critical system entails a system where the inherent properties of the system, such as the composition of the fuel, allows for the system to continue to maintain itself through chain reactions long after any initial driver is no longer present. Alternatively, a subcritical system requires a neutron source as the primary driver of further reactions. As one of the primary focus of this research is the control of a nuclear system through control of the neutron source, and because the source neutron source becomes increasingly less important the closer to critical a system is, I will focus on subcritical systems here. This is more than a matter of preference as there are also limitations to the use of minor actinide based systems in critical systems as will be shown. These limitations are also key to the differences of this type of system, with a high importance placed on the source, from the existing "Fast Reactor" concept.

### 2.1.1 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 $\phi_0$ can be found by solving the Boltzmann equation,

$$-\Omega\nabla\phi_0(r,E) - \Sigma_0(r,E)\phi_0(r,E) + \int\int \Sigma_0(r,E')f_0(r;\ \Omega',E' \rightarrow \Omega,E) \times \phi_0(r,\Omega',E')d\Omega'dE' + \frac{\chi(E)}{4\pi} \times \phi_0(r,\Omega',E')d\Omega'dE' + \frac{\chi(E)}{4\pi} = 0, \quad (2\text{-}1)$$

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 $\Sigma_0$. 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 $\Sigma_0$ and $\phi_0$. This is usually solved by first assuming a time invariant case. This can be done for a large range of cases because the isotopic evolution is negligible for instantaneous effect. More broadly this means the flux is slow to change in shape even if it can change in magnitude quickly, this 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 [14].

$$A\phi_{in} + P\phi_{in} + s = 0, \tag{2-2}$$

where $\phi_{in}$ represents the solution of the multigroup inhomogeneous equation using the multigroup neutron flux vector $\phi_0$, $A$ 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**.

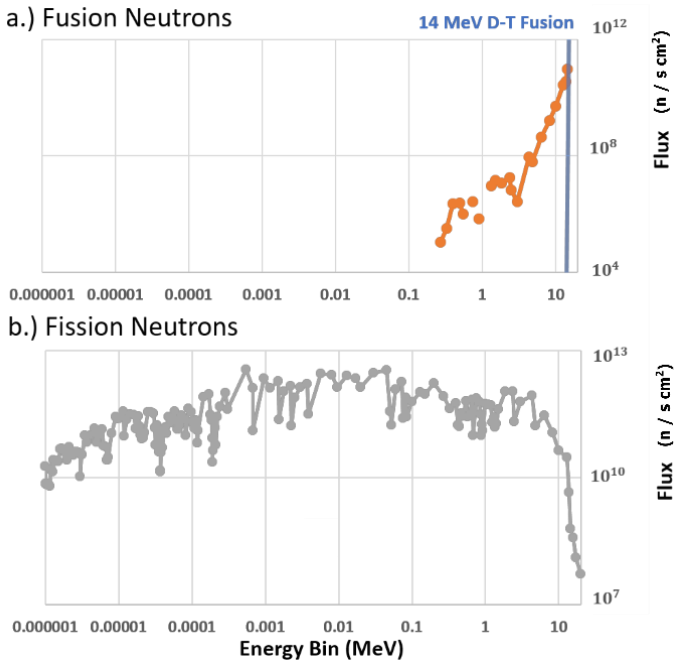


**Fig. 3** The initial steady state neutron flux at interior wall boundary of a minor actinide only fuel. (a) The flux of neutrons from a singular D-T fusion source that has not yet fissioned accounting for slow down (lethargy) due to scattering. (b) The flux of all resulting fission chain reactions

Neutron generation through neutron-induced fission that can result in further fission via chain reaction provides a neutron multiplicative effect that is the shining characteristic of the efficiency of nuclear systems. 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

$$S + F = S + Sk_{sub} + Sk_{sub}^2 + Sk_{sub}^3 + \cdots, \tag{2-3}$$

which can then be rewritten as

$$k_{sub} = \frac{F}{S + F}. \tag{2-4}$$

Important to note here is that the value of $k_{sub}$ cannot exceed "1" here, as a source neutron does not reproduce the source only starts a fission chain reaction. To find $k_{sub}$ 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

$$k_{sub} = \frac{< n_0^*,\ P\phi_{in} >}{< n_0^*,\ s > + < n_0^*,\ P\phi_{in} >}. \tag{2-5}$$

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 $\varphi^*$ importance parameter for $n_0^*$, as the system approaches criticality, can also be found through simulations and comparisons of observables experimentally, where

$$\varphi^* = \frac{\left(\frac{1}{k_{eff}} - 1\right)}{\left(\frac{1}{k_{sub}} - 1\right)}. \tag{2-6}$$

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 [14,15]. This can be done through the use of generalized perturbation methods.

Critical or supercritical systems are important to separate, as in such a system a single neutron would continuously produce more and more neutrons not stopping, and no longer bound by the source for control, 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 with the mechanical means normally available outside of the use of the source, particularly for the fast spectra and prompt heavy to delayed neutron counts involved here, having a neutron generation time on the order of $10^{-9}$ to $10^{-7}$ seconds. This can result in dangerous levels of neutron multiplication in very short times. Alternatively, a steady state can arise from a continuously driven 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

$$M = \frac{1}{1 - k_{eff}}. \tag{2-7}$$

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

### 2.1.2 Depletion and Isotopic Evolution

To see the isotopic time evolution, represented by the neutron flux from above, the Bateman [13,14] equation (2-8) should be solved. Here the rate of change in quantity of an isotope is found through adding

two sets of terms representing nuclide sources and sinks from transmutations between isotopes *i* and *j*. The first set through radioactive decay, and the next set being reactions such as fission, capture, (n,2n), etc.:

$$\frac{\delta N_i}{\delta t} = -\lambda_i N_i + \sum_j \lambda^{j\to i} N_j + \sum_j N_j \sigma^{j\to i}\langle\phi\rangle - N_i \sum_{\forall r} \sigma_i^{(r)}\langle\phi\rangle . \tag{2-8}$$

This is solvable through exponential matrix methods and 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. A steady state assumption of the flux is done instead of using a full analytical solution, as it would otherwise be highly coupled. Differences can be accounted for and corrected through simulation steps that will discussed later.

To move between two different steps the assumption of flux 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 size until 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 [13], 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.

### 2.1.3 Reactivity and Reactivity Coefficients

An 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 $\Delta K$ 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 linearized path between. However, 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 ($\rho$) 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 scale 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 progressively 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.

### 2.1.4 Kinetics of a subcritical system

Neutrons originating from fission appear on two different time scales: (1) Prompt neutrons appear instantaneously following fission event. (2) Delayed neutrons appear milliseconds to minutes following fission event. It is the presence of the delayed neutrons that allows for the critical operation of most nuclear reactors, such as a light water reactor (LWR). This is because their postponed appearance allows for a region of hybrid time scales where the total system is critical but only while including the delayed neutrons. This mixed time scale is long enough it enables mechanical adjustments to maintain steady-state neutron

flux and control. However, the fraction of delayed neutrons, which defines the width of this region and thus the ease of staying within this region, depends on the fuel; fission of $^{235}$U or $^{239}$Pu produces 0.7% and 0.2% of neutrons as delayed respectively, with MA having fractions < 0.1 %. Therefore, a critical reactor based on minor actinides as fuel suffers from decreased delayed neutrons and largely cannot be maintained stable within this desired time scale region. As a consequence, to incinerate large quantities of minor actinides subcritical operation is required and externally injected neutrons are needed to offset losses.

For subcritical systems changes in the reactor power ($P_N$) can be shown by solving for the subcritical point kinetic equation as demonstrated in more detail in [14]

$$I_{eff}\frac{dP_N}{dt} = (\rho_{gen} - \alpha\beta)P_N + \alpha\sum_{i}^{I}\lambda_i\xi_i + \zeta(1 - P_N) + \rho_{source}, \quad (2\text{-}9)$$

$$\frac{d\xi_i}{dt} = \beta_i P_N - \lambda_i\xi_i, \quad (2\text{-}10)$$

where $I_{eff}$ is the effective prompt neutron lifetime, $\rho_{gen}$ is the generalized reactivity relevant to perturbation at $t = 0; A \to A + \delta A,\ P \to P + \delta P$, $\alpha$ is a coefficient for the delayed neutron distribution, $\zeta$ is the subcriticality index or $\frac{1-k_{sub}}{k_{sub}}$, $\beta$ is the effective delayed neutron fractions, $\xi$ is the *i*-th effective precursor density. This can be used if the kinetic parameters or reactor power constants ($I_{eff}$, $\beta$, etc.) are determined along with a measure of reactivity insertion and their coefficients to determine safety and control factors.

## *2.2 Application to Transmutator Concept*

The concepts from above need just a few additions to best determine suitability and explain the more unique aspects of this system. In particular the philosophical principal used to justify the search for a minor actinide only driven system, as well as the factors that should be considered for controllability.

### 2.2.1 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 transmuted by capture 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 and already part of the consideration for the overall longevity of the waste in the first place.

The ladder, or fertile breeding by relying on higher nucleon counts, process is possible due to an increase fissionability by nucleon count **Fig. 4**. Additionally, there is an overall increase in the number of neutrons that are produced from each fission. This does not mean that it is inevitable that all neutrons either transmute or produce a more transmutation ready target for subsequent neutrons, but it is close particularly if a few factors are considered. These factors include: How many neutrons it costs compared to neutrons gained, factor with change in cross section size, and slowdown of neutrons (lethargy) and thermal fissioning ability.

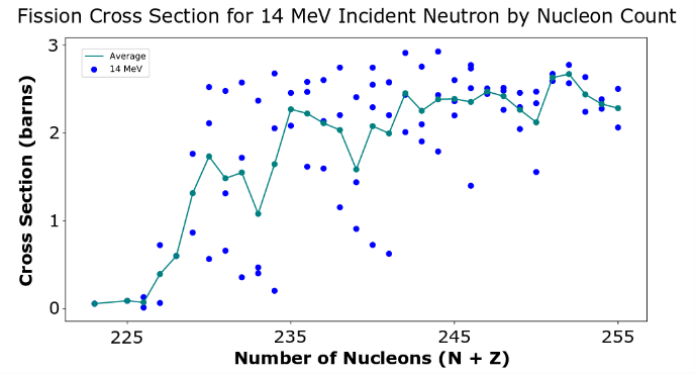


**Fig. 4** Increasing fission cross section showing ease of fission following nucleon count. Uses nuclides with greater than 224 nucleons from ENDF/B-VIII.0 library with 900K temperature profiles.

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 could 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.

### 2.2.2 Macroscopic Efficiency

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. 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

$$T = \frac{\bar{m}_a E}{\bar{\epsilon}}. \tag{2-11}$$

While (2-11) 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.

To see the relations of input power to output power for determining efficiency. First start working from energy of the output ($E_{out}$) and finding the needed energy of the input ($E_{in}$). Is to take the average energy released by fission ($\bar{E}_f$) divide by the average neutron energy (from fission) ($\bar{E}_n$) and adjust for number of neutrons using normal neutron economy rules (i.e. 1 neutron in = 1/(1-$k_{eff}$) neutrons out except fission is

"one step" behind so multiply by another $k_{eff}$). This is done as a substitution for number of fissions so divide by number of neutrons per fission ($\bar{\nu}_f$)

$$E_{in} = \frac{E_{out}\,\bar{E}_n\left(1-k_{eff}\right)\bar{\nu}_f}{\bar{E}_f\,k_{eff}}. \tag{2-12}$$

This can/should be adjusted for actual input in by using adjustment of source to "input" neutrons: Average energy of source neutrons ($\bar{E}_s$), average neutrons per secondary fission ($\bar{\nu}_f$), and average neutrons per source neutron fission ($\bar{\nu}_s$):

$$E_{in} = \frac{E_{out}\,\bar{E}_s\left(1-k_{eff}\right)\bar{\nu}_f\bar{\nu}_s}{\bar{E}_f\,k_{eff}k_{src}} = \frac{E_{out}\,\bar{E}_s\left(\frac{1}{k_{eff}}-1\right)\bar{\nu}_f\bar{\nu}_s}{\bar{E}_f\,k_{eff}k_{src}} \tag{2-13}$$

Similarly, with power (E→P) in place of energy and net multiplication (M):

$$P_{in} = \frac{P_{out}\bar{E}_s\bar{\nu}_f\bar{\nu}_s}{k_{eff}k_{src}M\bar{E}_f} \tag{2-14}$$

## *2.3 Monte Carlo*

Because a change in incoming flux and can shift the entire chain reaction pathway by promoting new self-sustaining reactions or push otherwise self-sustaining reactions below sustainability small change in incoming flux can have large changes in resultant reactions and thus the change the outgoing flux of any neutronics system. As a result, every boundary in a neutronics problem acts as an additional nonlinear coupled term in a neutronics problem. Consequently, direct solutions are not typical possible except for the simplest of geometric parameters. Even inhomogeneous conditions that would quickly arise from depletion

in any system not actively correcting for this such as with solid fuels would constitute as a boundary quickly limiting the usefulness of directly solving a real world neutronics problem.

The best means of solving real world geometries then becomes Monte Carlo. This is because Monte Carlo can ignore vastly improbable yet computationally expensive branching probability ratios by treating them as the small probability they are. Monte Carlo uses randomness to solve for systems that have underlying deterministic principles. Statistical systems with well-behaved underlying function and large number of samples will strongly converge on a solutions of a mapping of its probability density or its peak by averaging these samples [16]. This allows for solving for a number of the simpler classical dynamics problem of single random neutrons to give a greater picture of a much larger number of neutrons in the highly coupled system.

Monte Carlo is especially well suited for neutronics problem as it was privately developed for this purpose in the 1930's by Enrico Fermi, and later secretly expanded for real world use in the Manhattan Project and finally codified in computer science for this still same purpose by Metropolis [16,17] in the 1950's. The programs used to simulate neutronics through Monte Carlo methods have become a fundamental aspect of any neutronics study and were used here in conjunction with other solvers, linkages, and self-developed tools to determine the neutronics information fundamental to this research.

### 2.3.1 MCNP / ORIGEN

Monte Carlo N-Particle Transport code also known as MCNP is the oldest and most well-known of the neutronics solvers and was developed by Los Alamos National Lab as a result of the Manhattan Project. To evaluate neutronics characteristics and "burnup" of the transmutators subcritical molten salt transmutation

process as a function of time the MCNP and SCALE software packages are used through an internally developed code that prepares and parses information from each in a ”linkage” code.

Software such as MCNP and SCALE are already thoroughly validated & verified for accuracy for their particular scope. However, the needed scope for all parts of the setup described in is not fully covered by any individual software available. Fortunately, many individual aspects are well covered by existing software, and by using a linkage code to connect these separate codes, a complete and coherent picture can be obtained. Additionally, as the code would be linked and controlled by a central platform that is editable in process, this allows for optimization to be performed and feedback controls simulated by artificial intelligence.

Neutron transport which can solve for neutron flux is the primary focus of the Monte Carlo N-Particle code suite MCNP 6.2 and is at the heart of many neutronic linkage codes. However, as subcritical operations are envisioned, special care is taken to include the source neutrons and keep them separable from fission produced neutrons. This is also necessary because they are not typically tracked for critical systems, that are the primary focus of MCNP, as they become greatly outnumbered the closer to critical a system becomes. As source neuron energies are significantly different than neutron energies from fission, there are important differences in cross sections as described by that play a significant role in the transmutation process. This lack of tracking or inclusion of source neutron is the reason the ORIGEN-S module in the SCALE software package is used instead of CINDER, MCNP’s included depletion solver.

### 2.3.2 OpenMC

OpenMC is a relatively new, open-source, and community-developed Monte Carlo neutron and photon transport simulation code originally developed by members of the Computational Reactor Physics Group

at the Massachusetts Institute of Technology starting in 2011. It is capable of performing fixed source, k-eigenvalue, and subcritical multiplication calculations on models built using either a constructive solid geometry (CSG) or CAD representation. The code supports both continuous-energy and multigroup transport. Since this code is open source, its use is not subject to licensing, with no restrictions on modifications, developments and addition of new capabilities.

# Chapter 3

## 3 Laser Driven Fusion Neutron Source

### *3.1 Coherent Acceleration of Ions by Laser*

Recent research shows that ultrafast intense laser pulse interacting with an appropriately designed thin target can accelerate ions to designed energies [18]. This physical interaction arises from the Coherent Acceleration of Ions by Laser (CAIL) process [19,20] when a nanometrically thin target is properly conditioned. Unlike the earliest laser ion acceleration regime of Target Normal Sheath Acceleration (TNSA) [21], CAIL directly uses nonlinear ponderomotive laser forces (i.e., electromagnetic forces) on accelerating electrons coherently, which in turn accelerate ions coherently by the subsequent electrostatic force, thus yielding higher efficiency. In order for this combination between the laser ponderomotive acceleration of electrons and subsequent ion acceleration by electrons' electrostatic pull to be matched, the laser intensity and the ionic inertia have to satisfy an optimized condition (among other conditions). This condition is expressed [18,22,23] as the normalized target thickness equal to the normalized laser strength:

$$\sigma = a_0, \tag{3-1}$$

where the normalized target thickness $\sigma = \frac{n_e}{n_c}\frac{D}{\lambda_L}$ and the normalized laser field $a_0 = \frac{e\,E}{m_e c\,\omega_L} = 0.85\lambda_{L,\mu m}\left(\frac{I_L}{10^{18}}\right)^{1/2}$ with $n_e$the electron density, $n_c$ = the critical density, $D$ the foil thickness, $\lambda_L$the laser wavelength, and $I_L$ the laser intensity. The critical density, $n_c$, is determined from the condition, $\omega_p(n_c) = \omega_L$, where $\omega_L$is the laser frequency and $\omega_p$ is the plasma frequency. Under this optimal condition the maximal ion energy accelerated by laser may be given as

$$\varepsilon_{max} = (2\alpha + 1)Q\Phi, \tag{3-2}$$

where

$$\Phi = m_e c^2\left(\sqrt{a_0^2 + 1} - 1\right) \tag{3-3}$$

is the ponderomotive potential of the laser. Here, Q is the ion charge, and the parameter $\alpha$ is the coherence parameter [24] and indicates the nature of the coherence between the driver (laser) and the electrons and ions.

## *3.2 Laser Driven Fusion*

In this coherence of electron dynamics of the CAIL process and its' predictive and relatively high efficiency of laser coupling to ions [18], we can design the deuteron accelerators for our system based on ultrafast intense lasers. Because each accelerator and each target of deuterons is small, we can make the neutron generation section small. This makes our neutron generation unit small, local, and controlled in real time through laser control.

The conceptual suggestion of the laser-driven neutron source for transmutation is shown in **Fig. 5a**, where neutrons are generated by a large array of femtosecond pulsed fiber lasers with high-repetition and high efficiency coherently added CAN fiber lasers [25]. By taking advantage of the CAN high-repetition rate and good efficiency, high-fluence laser pulses are generated, which irradiates a nanometric foil ejecting deuterons onto a solid or gaseous catcher target in order to generate deuterium-deuterium (D-D) or D-T fusion neutrons. **Fig. 5b** shows our simulated deuteron energy spectrum. Our simulation performed with the two-dimensional (2-D) EPOCH particle-in-cell code [26] uses a linearly polarized laser of intensity $I = 1x10^{18}$ W/cm$^2$, wavelength $\lambda = 1$ μm (or $a_0 = 0.8$) and pulse length 60 fs and has been focused onto a 3-μm beam footprint. In this simulation the foil is composed of deuterium with density of $1.0x10^{23}$ cm$^{-3}$ and thickness of $D = 10$ nm: Such cases satisfy $\sigma = a_0$; thus, the foil thickness is on the order of the skin depth $c = \omega p$ permitting a fraction of the incident laser to penetrate to fit the CAIL optimal condition Eq. (3-1). Our computational domain of 13λ × 10λ and 20 000 × 2000 grids used 100 particles per cell.

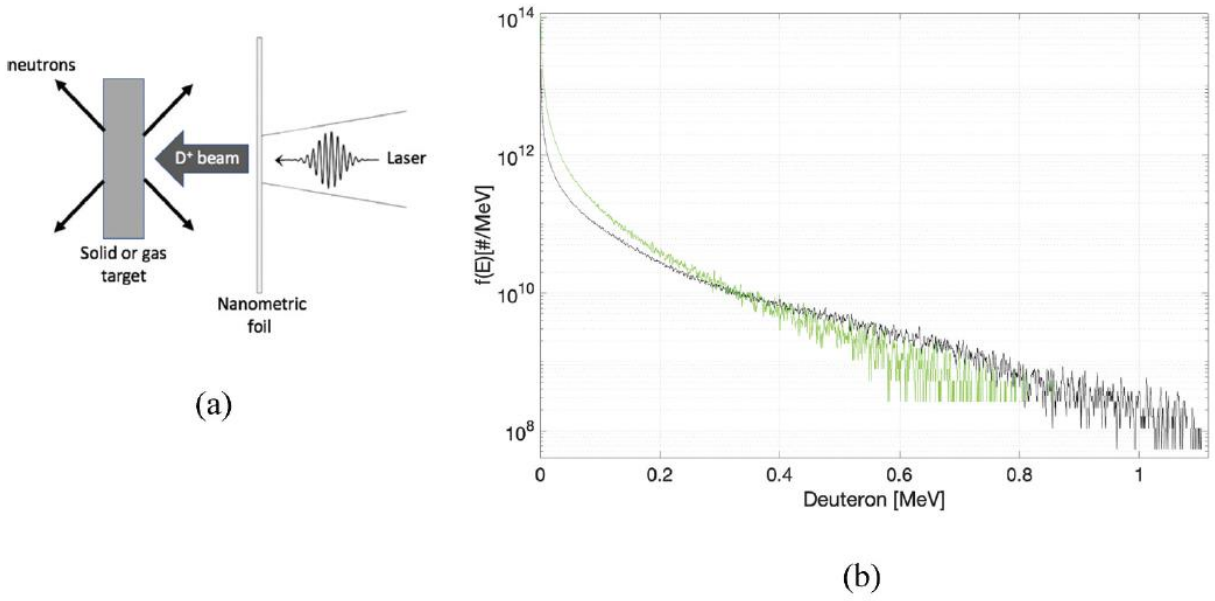


**Fig. 5** Neutrons are generated by the laser irradiation of a nanometric deuteron foil, deuteron acceleration and interaction with titrated solid or gas target. (A) Schematic of neutron generation using laser. (B) D+ energy spectrum.

Once the deuteron beam is produced by the laser and neutrons are generated, we design beam-target fusion (D and T, respectively), and a secondary target of tritium is placed behind the primary thin target of the deuterium beam with a neutron per deuteron yield, $Y_{N/D}$:

$$Y_{N/D}(E) = \int \sigma_{DT}(E')n_T dx = \int_0^E \frac{\sigma_{DT}(E')}{\frac{dE_s}{dx}(E')} n_T \, dE', \tag{3-4}$$

where $\sigma_{DT}$ is the deuterium-tritium cross section, $dE_s/dx$ is the energy loss per distance, $n_T$ the target density of tritium.

We note that such a beam-target fusion device is compact and devoid of any magnets (a condition favorable for an environment of high neutron flux). The neutron per deuteron yield $Y_{N/D}$ is shown in **Fig. 6** for D-D and D-T cross sections. Integrating Eq. (3-4) over the deuteron energy spectra in **Fig. 5b**, we can obtain the neutron yield per laser pulse. The $\sigma = a_0$ (green) gives a total neutron yield of $10^6$ n/laser shot; this regime is optimized to attain maximum deuteron energy. The deuteron energy spectrum for $\sigma = 4a_0$ (black) is optimized to obtain maximum average energy with a neutron yield of $10^7$ n/laser shot. Assuming a repetition rate of 100 kHz, the neutron rate per single laser is $10^{12}$ n/s. The compactness and scalability of the CAN laser allow us to deploy a large number of units to meet the overall need of the total number of neutrons. Thus, a neutron rate of mid $10^{15}$ can potentially incinerate 10 kg of TRUs a year.

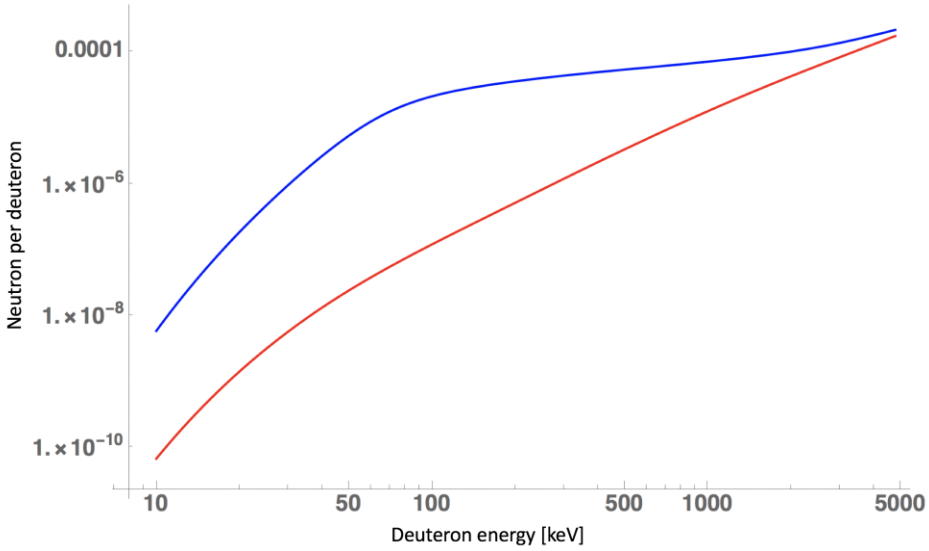


**Fig. 6** Neutron per deuteron yield for a thick target of deuterium (red) or tritium (blue) calculated using Eq. (3-4).

# Chapter 4

## 4 Molten Salts

Another central theme of our transmutator is to operate in the liquid state by dissolving TRUs in a molten salt (e.g. FLiBe, FLiNaK). Such operation allows for a real-time passive and active monitoring and feedback and consequently control of the TRU refueling, reprocessing and criticality. This is then coupled with the monitoring of the distributed neutron sources. Monitoring in real-time allows for a flexible and an optimized operation.

The liquid operation enhances safety and efficiency as it allows to employ active and passive laser monitoring. Additional passive safety could be provided by using a frozen plug on the bottom of the transmutator and allowing it to melt in an event of undesired core temperature increase and discharge the transmutator content into a tank containing neutron absorber, e.g. boron, and simultaneously freezing and encasing the content of the transmutator core thus mitigating dispersal of radiotoxic materials. As a consequence of using molten salt the pressure in the vessel is close to atmospheric thus in an unlikely event of a catastrophic rapture, an explosion and dispersal has low probability. Furthermore, due to the molten salts high melting temperature (~300 ℃) the content would quickly freeze if no longer actively heated.

A further advantage of the liquid Transmutator is the removal of the fission products (FP) and refueling while the Transmutator is operating without requiring to shut down. The molten salt and TRU refueling can be done jointly as a mixture or separately whereas the TRU fuel is injected as a pellet or a gas. The removal of FP depends on their chemistry. The noble gases (e.g. xenon, krypton) are removed online by *in-situ* by helium injection [27], the remainder composition of the molten salt, FP, and actinides is partitioned to the pyrochemical reprocessing performed off-line whereas FP are removed and actinides and molten salt are returned for further transmutation.

To extract energy from the molten salt, we pump a coolant (such as $CO_2$ or non-alkaline-liquid) through the transmutator core in lieu of pumping out the mixture of molten salt, TRU and FP into an external heat exchanger. This helps to minimize the presence molten salt and fuel in the external circuitry.

We note that there exist efforts in transmutation R&D is based on possible burning of TRU in the Next generation of Fast Breeder Reactor [28], MOSART project [29] or in the ADS system (Accelerator Driven System) which consists of 100s MeV class proton superconducting Linear accelerator (600 MeV, 2-4 mA) coupled to subcritical core reactor loaded with TRU as fuel elements [30]. Other approaches employ fusion-fission hybrid technology [31]. We have introduced the rationales and a set of new operations and technologies that accompany the present approach.

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 [32] 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 [5–7,33] 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 [34] 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 [5–7,32,33,35] 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. 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.

## *4.1 FLiBe and FLiNaK*

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 [6,10,32,33]. 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 [32]. As a generalization for most of the actinides it has been suggested that 2% mol [6] 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.

## *4.2 Laser Monitoring*

The liquid core is also suited for active and passive spectroscopy since electromagnetic signals pass through the liquid in contrast to solid fuel rods that are opaque and transmissive of no spectral information. The spectroscopy and monitoring may be done real-time and in-situ (or in a chamber adjacent the main vessel and possibly shielded against background noise). Active gamma spectroscopy may be adopted relying on the nuclear resonance fluorescence (NRF) to collect data pertaining to the isotopic composition (e.g. the NRF is a well-known technique [36] to detect $^{240}$Pu). Passive gamma spectroscopy collects gamma rays emitted by the decay of radioactive isotopes. This may be used to verify spent nuclear fuel content [37]. Meanwhile, the laser-induced fluorescence (LIF) provides information regarding the elemental composition by atomic excitation. The LIF has been used for actinides and lanthanides analysis in the nuclear fuel cycle [38]. For this purpose, either a broad-band laser or a scanning laser might be used. While velocimetry style measurements to gather thermal data are important for the shorter time scales. These can all work together to provide the needed measurement for the appropriate time scale inputs. Finally soluble fluorides such as $UF_4$ and $NiF_2$, can dramatically change the salt's color in both solid and liquid state, making spectrophotometry a viable monitoring tool as has been done for MSRE operations. The information from optical monitoring as well as thermocouples, neutron detectors etc. can then be fed to the AI to adjust fuel and molten salt concentration, laser power, etc. in order to assist real-time operation (in μs and ms timescales).

## *4.3 PUREX and Other Process*

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 additional fuel preparations to be made due to its ability to act as a solvent to the actinides [6,33,35]. 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 [35,39]. 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 [35].

## *4.4 Carbon Based Wall Materials*

The first wall is a wall component immediately adjacent to and in direct contact with the core, which is a mixture of molten salt, dissolved actinides and fission products. The severe physical conditions which the first wall of the transmutator must withstand include:

- High dose of neutrons that can transmute, ionize, elevate temperature and degrade physical properties (displacement per atom) of the wall
- High dose of other radiations such as X-rays and other electromagnetic radiations
- Lattice embrittlement by the diffusion of helium atoms, fission products or TRUs
- Creep and fatigue by the high pressure and high heat load
- Corrosion by reduction-oxidation (REDUX) reactions [40,41].

To address these issues, a proposed multilayer structure with a carbon-based material can be incorporated within the first wall as shown in **Fig. 7**. The carbon allotrope proposed for the first wall is a Chemically Vapor Deposited (CVD) diamond. CVD diamond sheets are supported by struts. The combination of struts and carbon-based first wall takes advantage of the tensile strength of the thin diamond wall. Neutrons could be moderated and stopped in a surrounding water tank or by a material such as a graphite filler for an outer shielding.

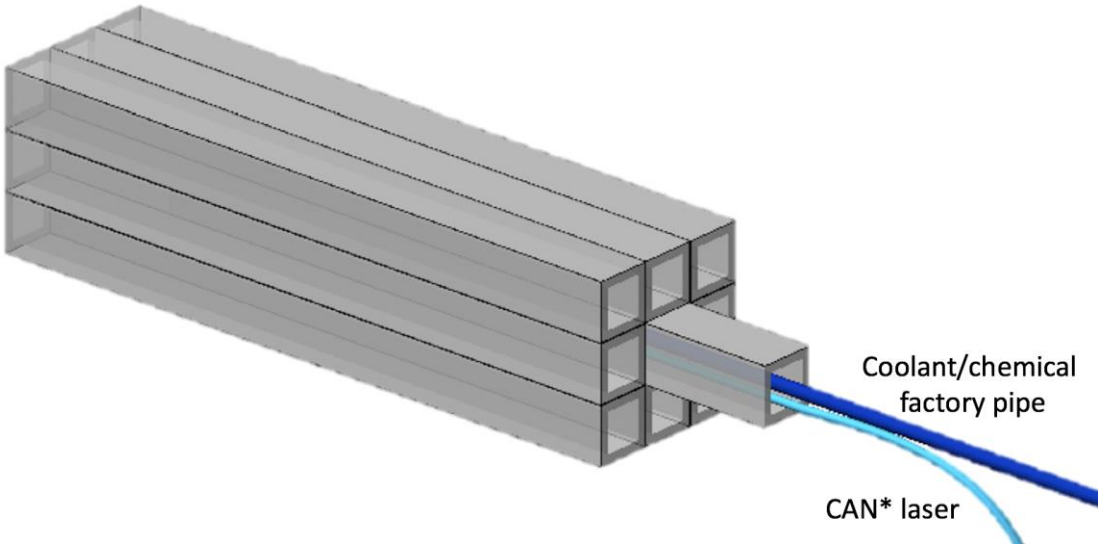


**Fig. 7** Schematics of a laser driven liquid transmutator. In this embodiment the tanks have a square cross section. The wall is made of thin diamonds with struts. The center of each chamber is used to deliver the

Coherent Amplification Network (CAN) fiber laser for neutron generation as well as pipes delivering coolant and chemicals.

Carbon is not easy to transmute by neutrons (and other nuclear processes). While a thin first wall allows for electromagnetic radiation and neutrons to pass through, therefore minimizing damage and heating. Meanwhile, some carbon allotropes show extraordinary strong tensile strength (e.g. graphene [42]), while others such as diamond show a very high 3D rigidity due to its tetrahedral structure [43]. Furthermore, in terms of helium embrittlement, it has been shown that by coating a metal such as tungsten with a single layer of graphene, it is possible to stop damage to the tungsten surface [44] when exposed to ionized high energy helium. The effect can decrease the mass loss of the material by as much as a factor of 10. Having strong covalent bonds, diamond may also have a resistance against such an effect.

Unlike metals, carbon chemical bonds (four of them) may be all saturated by the covalent bonds, such as in graphene and diamond. These bonds introduce a strong and stable chemical state of these materials against the highly reactive chemicals of the molten salt and its solvents. These also provide the foundation for the high electric and thermal conductivities if these materials are under certain conditions or treatments. Thus, carbon-based materials may serve for a chemically robust first wall, as already demonstrated by graphite which has already been used extensively in the MSRE project at ORNL exhibiting little corrosion [45]. However, carbon may be reactive to oxidation at high temperature in the presence of oxygen, in which case, we should avoid high temperature for burning and/or the presence of oxygen.

The past 15 years has seen a progress in new carbon-based materials. Graphene, which is basically a carbon monolayer, was discovered in 2004 [46] and since then, it is already available commercially on multiple types of substrates [46]. Opposite to the 2D structure, diamond has been known for much longer. However, only recently did it become an available off the shelf engineering material. The development of plasma-assisted technology of CVD diamond growth made it possible to engineer the diamond material for specific applications. Different grades of diamond are available for different applications: polycrystalline diamond

(freestanding or on substrate, up to several mm thick), single-crystalline, nanocrystalline and smooth ultra-nanocrystalline diamond [43].

Diamond is known for its multiple outstanding properties, especially high thermal conductivity and hardness. In addition, diamond can be a wide bandgap semiconductor. It can be doped, for example with boron, yielding "metallic" electric conductivity at doping level of $4 \times 1020$ $cm^3$ (0.2 at%) and higher [47]. Due to its large bandgap (~5.5 eV) in comparison with free electrons in metal and high displacement energy, diamond is an intrinsically radiation resistant material. In the past the combination of its fast charge collection, low dielectric constant and low thermally generated leakage current, made diamond a material of choice for detectors of high energy particles in radiation harsh environments [48].

Both diamond and graphene are chemically stable materials in comparison with other traditional wall materials, s.a. metals, and are inert to most chemical reagents at room temperature. However, there is lack of data on corrosion studies on these carbon allotropes in molten salt (i.e. FLiBe / FLiNaK) and their resilience needs to be demonstrated.

# Chapter 5

## 5 Artificial Intelligence

Because of the large parameter space that the features of this transmutator allows but were left open, much of this study revolved around the integration of AI as a tool to optimize and search for solutions. In essence there are two types of AI's involved here that will be covered. The primary one that I currently use is the Evolutionary Algorithm (EA). This is a class of heuristics used to explore large parameter spaces to find optimized results. The other type is the Neural Net (NN) which takes a trove of previously explored parameter space to "learn" by building up a "weight mapping". The Neural Net can then, with enough learning, quickly (and accurately) "solve" the very hard problems needed without actually "solving" the model equations behind them by mapping a series of inputs onto predicted outcome. Done by following through a much simpler set of functions adjusted by their "weight mapping". NNs are important here because of this speed from not having to solve these complex equations. However, because they require a system to already have a previously explored parameter space for their learning library, which does not exist for many aspects of this system, luckily an EA can and are often used to generate this data and train neural nets.

## *5.1 Meta Heuristics*

Heuristics are algorithms or techniques designed to search for a solutions when direct methods may be too slow, or where a direct method to solve is unknown but an approximation solution is sufficient. Likewise, a meta-heuristic, from the same named field created by Glover (1986) [49], as defined in [50] is a "high-level problem-independent algorithmic frame-work that provides a set of guidelines or strategies to develop heuristic optimization algorithms". Or more broadly an examination of possible heuristics independently of any problem to develop guidelines in choices of appropriate heuristics for possible optimization problems.

Neutronics already encompass a very large parameter space that leads to highly nonlinear branching pathways that requires a non deterministic heuristics such as Monte Carlos, overlaying another such heuristic would seem to only further complicates this. According to the "No Free Lunch" theorem in metaheuristics this would leave determining the best or most efficient heuristic to solve this problem prior to any attempt a likely unfruitful endeavor. As such overall a trial and error approach to the development of an AI is needed. However, there are still aspects that were considered in the selection and creation of the AI's from a metaheuristic standpoint as indicated by quotation marks below without in-depth explanation.

An EA is used for "simulation approximation" from a Monte Carlo based "non-exact" or "noisy" "objective functions" as is often done to the best results (Fu 2002). The EA provides "diversification" through a population model rather than a single point, and uses a breeding/cross-over model after the scoring against the "objective function" in choosing the next set of solutions in the iterative set. Additionally, Tabu Search "memory usage" keeps a hashed history of previous attempts to prevent loops, and Simulated Annealing is used for increasing "intensification" by the reduction of free energy, or randomness through mutation and population size, using an annealing/cooling schedule.

The Neural Net was also approached in terms of suitability for use in a further developed Transmutator but is not directly used in this study due to the undefined and generalized nature of the parameters and extreme time scales most important to validating their use and making the accumulation of a useable learning library difficult.

## *5.2 Evolutionary Algorithms*

There is some issue with calling the algorithm (heuristic) I developed to determine the operational problem any single specific name. The method names such as: Evolutionary Algorithm (EA), Evolutionary Computing (EC), Genetic Algorithm (GA), Simulated Annealing (SA), and Tabu Search (TS) etc. are not in themselves specific "recipes" but rather frameworks that can overlap. This would be a "hybrid method". An EA could be considered the foundational algorithm used in this hybrid method and is used as the overall description of the AI used for parameter space searching with the GA as its near equivalent as the subpart.

In a broader view Evolutionary Computing (Dortmond 1991) [51] is used as an umbrella term for all algorithms, collectively the "Evolutionary Algorithms", that are inspired by biological evolution specifically meant to cover and merge the independently developed lines of Evolutionary Programming (Fogel 1962), Evolutionary Strategies (Rechenberg 1973), and Genetic Algorithms (Holland 1975) which were all preceded by less defined strategies of Artificial Life (Barricelli 1953), Automatic Programming (Friedberg 1958), and Artificial Selection (Fraser 1957) and even earlier less referenced work with many additional sub branches (most notably Genetic Programming) later. EA's would be more general of a term than GA, although John Holland made GA a very popular term that is still often used [51].

In EA's (or ECs or EC Algorithms etc.), an initial set of candidate solutions is generated and iteratively updated. Each new generation is produced by stochastically removing less desired solutions and introducing

small random changes. In biological terminology, a population of solutions is subjected to natural selection (or artificial selection) and mutation. As a result, the population will gradually evolve to increase in fitness, in this case the chosen fitness function of the algorithm.

### 5.2.1 Genetic Algorithms

Genetic algorithms (GA) are the foundational AI in the evolutionary algorithm family. They are a metaphor based heuristic defined by their attempt to model choice selections of fitness in a similar manner as genetics. They are as real world genetic trends evolved by a simulated natural selection. This is accomplished by creating "individuals" which are defined by the genomes made of discretized parameter options in the manner of base pairs in DNA. Many individuals are first created in random manner as part of a population of an initial generation which are then measured for fitness. These initially separate individuals then work to build to the most successful possible parameters by matching up and combining the most successful individuals as determined by fitness scoring. New children based on these combined parameters then repeat the process until an optimal parameter setting is converged on.

This process allows for solutions that incorporate possibly innovative solutions by allowing many different parameter combinations to be tested without presumption to their importance. The importance will naturally emerge as result of the iterative process. This is important for problems that have large parameter spaces with difficult to measure or distinguish effects. Neutronics can have an endlessly branching set of possibilities that make many parameters that are innocuous in one set of problems be quite important in another and so it is naturally fitting to use here.

Genetic Algorithms like their metaphorical inspiration must take into account several factors to allow for the optimization. These include:

- Fitness Measurement
- Mating Selection
- Combination/Reproduction
- Mutations

The measurement of fitness is one of the most important aspects of genetic algorithms as it defines the quantitative goal of the heuristic and allows for progression by determining which set of individuals best allow for it. This is also an often underestimated criteria for more complex systems such as this one. The main reason being that there are often conflicting criteria's of good. A complete and quick transmutation of all the waste in the tank in a single time step with minimal input of energy would by the most energy efficient and best score on one measure and is additionally marginally possible due to self-sustaining chain reactions, but the amount of energy released all at once would be an utter failure on the measure of fitness for safety. As such a careful consideration that ultimately and quantifiably transforms all these consideration into a single measurement must face some scrutiny for its meaning.

Selecting from a pool of individuals for use in creating the next generation is also a factor that must be considered. An instance where only the top two individuals are chosen would lead to a rapid decline in efficiency of this heuristic for a variety of reasons the most prominent being a loss of diversity. The loss of diversity immediately narrows parameter space search to only combinations of initial parents. This will also bias results only towards whatever initial guesses randomly hit as the most important factors found which is especially troublesome for small generation sizes and large parameter spaces. As a result of this typically methods that introduce some level of randomness that biases towards the most successful sets of pairing is involved. This biases the next generation to improve upon each prior generation but does not make too many early presumptions that were the reason for choosing to do a GA in the first place. This can take the form of such methods such as using "Elites" by taking something such as the top 20% of fitness scorers and then randomly pairing this pool. Other methods such as tournament select random individuals and then

compare only the subset of these individuals. These choices are also a factor in such things as the number of offspring from each mating set or the question of using the same parent in multiple different sets or restricting each mating set to a one time choice.

The combination of each mating set to create the next generation is another factor that is key to the optimization abilities of a GA. This is in essence a smaller sub optimization problem. To see this one can look at the simplest method of generating children which is by a crossover where the genes are chosen from one parent and at a specified cross over point (such as halfway or at a random point etc.) is switched and completed by the other parent. This has the effect of ordering the parameters and immediately limiting the number of children that come from each mating set. So, this must also be devised in accordance to parameter space size and the number of children and possible parent combinations while also keeping the hereditary aspects of each parameter sequence at the heart of this iterative solution but allowing for a consistently broad possible exploration of the encompassed parameter combinations.

Even with a very large number of possible combinations from any possible mating set while covering disjointed areas of a parameter space, any choice immediately narrows the solution space tremendously. This could allow local maxima and local minima's to dominate even if much better solutions were to be had. To combat this there must be a small amount of randomness inserted to skip the rails constraining the explored space. This randomness must also come in limited measure so as not to lose the evolutionary effect from building on prior results. This is accomplished by inserting limited random mutations, such as by randomly scrambling a parameter gene for a small percentage of individuals during reproduction.

### 5.2.2 Simulated Annealing

Simulated Annealing was introduced by Kirkpatrick in 1983 and is based on [16] is considered a "metaphor-based" metaheuristic (although the term "metaheuristic" did not exist at that time). Simulated Annealing is most commonly used as an analog to increasing "intensification" by the reduction of free energy, or randomness, using an annealing/cooling schedule.

An SA aspect was used here, as while not the primary goal of this research it was still necessary to implement some computational efficiency improving methods. The most computationally intensive aspect that is faced by this AI is the cost function. With greater precisions and reduction of statistical uncertainty a single cost function simulation can be a task that is typically its own intensive project. To keep the computational load at a reasonable level reducing the number of total cost function runs is the low hanging fruit for efficiency. This was done by reducing the number of individuals in addition to the degree of change allowed through the mutation aspect of the GA as the parameter search progressed.

### 5.2.3 Tabu Search

A pre simulation check to prevent rerunning an exact copy of prior attempted individuals to prevent loops known as a Tabu search (Glover 1977) was implemented [49]. This was accomplished by creating a history of fingerprint based hash sequence for each genome, which was aided by the already discretized parameters. Each generation then has the list filtered by a list from a changeable number of prior generations. This is particularly important for children creation as the much narrower parameter space deriving only from those allowed by the parents and possibly repeated if more than one child is needed from each mating set.

There is additional overhead for this check especially if the list covers a large number of generations but if the number of parameters is not so much greater than the number of children from each pair then this catches a great number of duplicates that saves a greater amount of computation time in fitness scoring. This could

alternatively be used in conjunction with a prior score lookup to bypass simulation step but allow the repeat if loss of diversity or loops are not an issue.

### 5.2.4 Hybrid AI in Transmutation Optimization

A self developed object based method written in Python was used to allow a mix and match approach of some features of multiple heuristics. Which is often done and considered named by "personal taste" as per [52]. The EA's process as shown in **Fig. 8** is:

1. Create a population representing individuals of random guesses of parameter settings
2. Run individuals through a simulator and score them through a cost function. Where the cost functions setup will determine the optimization goals.
3. After scoring the population select and match the most fit parameters sets by these scores.
4. Prior guesses are then combined by to create a completely new population of parameter settings.
    a. Occasionally some level of mutation is also added to randomly adjust some individuals. This helps prevent results from falling into local score minimas
5. This new population will be biased towards improvement by taking parts from the most successful of prior guesses
6. This process is then repeated for a set number of times or until some criteria is met such as no significant improvement is being made.

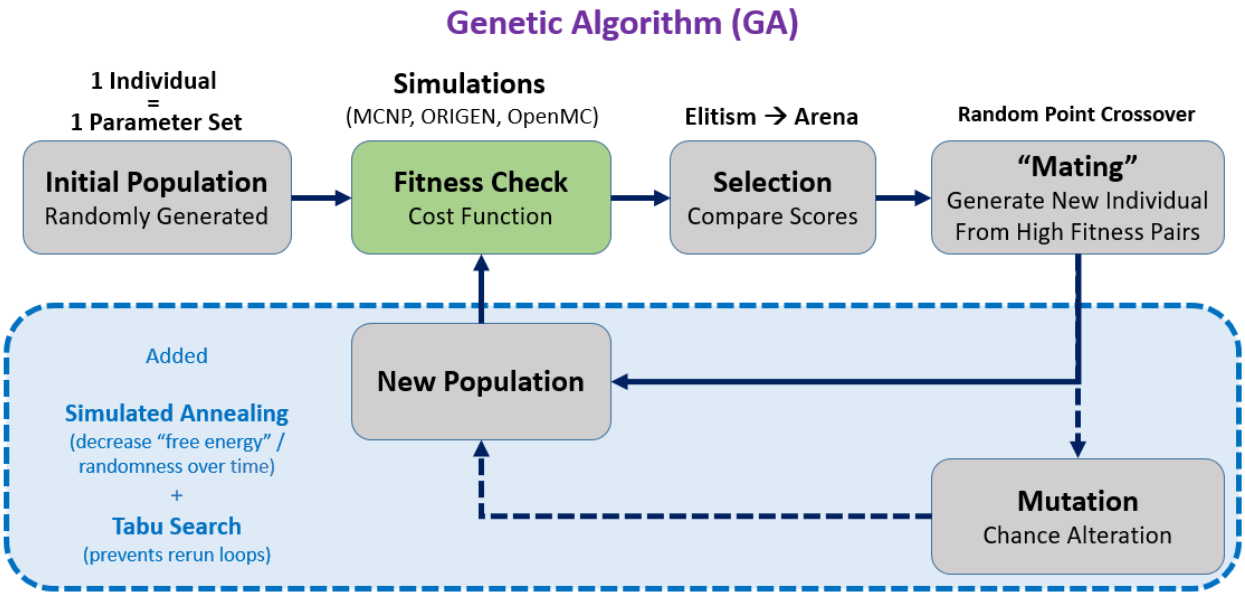


**Fig. 8** Process diagram of hybrid EA AI used for optimization

## *5.3 Neural Nets*

Neural nets are especially intriguing for their potential to leverage control in the short time scales of nuclear systems as they have already been demonstrated to be useful in similar very fast but very complex systems. Such as predicting disruptions in a tokamak [53] as far back as 1996, and more recently used for detections of beam driven modes in FRC plasmas by TAE [54]. This is now possible because of the added forms of monitoring and the ability to enact feedback or feed forward control at optical speeds. This was further enhanced by molten salt shifting the short time scales slightly by allowing for a lower density of targets through the dispersal of the waste in a low interacting medium.

Feedback or feedforward control of a nuclear system is not otherwise 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 ($l_{Prompt}$) 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 research is to our knowledge the first attempt to suggest the use of a NN for control in the prompt time scales of a fission system.

Feedback or Feedforward control may be possible if a controller can reactively adjust the system's operations in a short enough time and to a degree that the systems output stays within an acceptable bound. To see the effectiveness of the adjustment we need to know how fast the system can react (latency) and what level of change this response can cause (local or total reactivity insertion). There are five main parts to this:

1. The time for a system state to be relayed to a controller by sensor medium.
2. The sensor response time.
3. The controller response time
4. The time to enact this correction. (controller 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 ($N_{layers}$) [55] 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 or minimize 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.
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.

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 input source can be expressed in terms of a reactivity insertion and the output as a resultant change in criticality.

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 [55–57] within the necessary error bounds to respond rather than solving the modeling equations.

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 the learning data can be produced through realistic Monte Carlo simulation. To that end a hybrid heuristic [51] using an Evolutionary Algorithm can be applied in parallel to optimize operation and to build a NN learning library and through reinforced learning to continually increase accuracy

.

# Chapter 6

## 6 Transmutator

The Transmutator that prompted this work as shown in [10] was envisioned as an optimized result of the exploration of several concepts related to the impact of a novel source combined with the use of molten salts and the other concepts listed above. This work is the exploration on a generalized transmutator with many parameters considered open to allow the best general understanding applicable to many offshoots rather than a specific design. A generalization was also best served by the simplification of some parameters, when possible, to allow the least amount of interference by factors that could easily change by design. This also allows for the application of similar concepts to other designs beyond a particular device. However, these parameters were often still set to best demonstrate as close to a real case of an entire transmutator with multiple concepts working together and not just the theoretical framework. The tools and methods such as the use of Monte Carlos were left generalized to allow for quick adaption to more specific cases as needed or for use in future works.

## *6.1 Spatial*

### Geometry

Initial tests were done to find the effect of geometry to establish a reasonable baseline to be used throughout for comparison purposes. An overly symmetric design such as a sphere or slab was not desired as it may obscure possible shape effects while some symmetry and simplicity were desired for computational purposes. As a result, the baseline use of a cylindrical tank was chosen. Additionally, this baseline had to be large enough that the molten salt was the primary interaction medium, and the overall size was not a primary factor, while not so large that it was not in a realistic scope for multiple possible designs. As the D-T fusion neutrons had a mean free path much smaller than spallation neutrons and on the order of ten centimeters depending on the fuel concentrations a tank that was 1 meter in diameter and 2 meters in length for the interior core as seen in **Fig. 9** was used to possibly accommodate multiple sources.

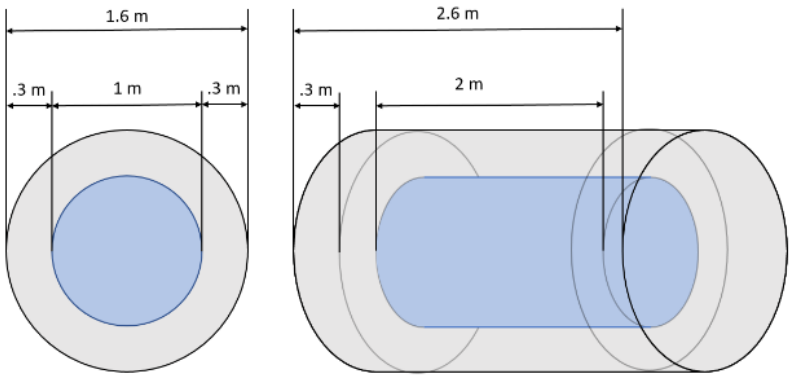


**Fig. 9** Schematic of the transmutation tank for the neutronics study.

### Outer Wall

A graphite outer wall 30.5 cm thick was chosen as a quick set of simulations showed negligible change in reactions once the wall was more than 2 or 3 times the mean free collision length. Graphite is not required

to be the outer wall material and would not be useful for a structural material but was chosen for its high tolerance to radiation (low DPA), low chemical reactivity, and neutron reflectivity that could be used a filler for the bulk shielding. Graphite was also chosen over alternatives such as water because it is an allotrope matching in composition to the diamond skin wall material proposed in [10]. The outer wall material was also confirmed to not represent a significant effect on the overall concept by tests with purely reflective and void boundaries although an observable effect is still present which would require an in-depth study for a completed design.

## Homogeneity

Homogeneity achievable by mixing of a molten salt is the most important spatial difference as compared to solid fuel systems. Artificial heterogeneity allowing for advantages as control through selective neutron energy fissioning/capture ratios can be accomplished using multiple coupled tanks with differing fuel concentrations such as a series of annular tanks or distributed modular cells, as shown in **Fig. 7** and ***Fig. 10***. Multiple coupled tanks could extend controllability, efficiency and even change overall size significantly. Additionally, a bi-stable approach can be used where a breed burn zones are created by a heterogeneous core arranged either mechanically or through depletion development as in a traveling or standing wave reactor. A multiple tank design could also negate the concept of a 'blanket' because of the ability to distribute multiple sources no longer needing a central point would make the blanket simply non source activated tanks on outer edges of activated source regions. Alternatively, a monolithic single tank design could still be desired for simplicity and reliability. The single tank could use of such concepts as using the lower activity a distance from active source being cooler to keep the molten salt as a solid for less chemical and structural stress at the outer wall. These possibilities are certainly open and more easily allowed by the nature of the source and the molten salt so are worth mentioning but are diverging and beyond the scope of a generalized study.

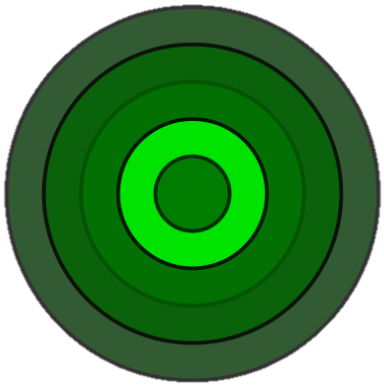

**Fig. 10** Example alternate design of transmutator with coupled tanks arranged as annular rings

## Multiple Source Distribution

The small size of the source, combined with its relatively cheap construction end point of a fiber make the use multiple sources and their distribution the most important difference spatially between the laser driven source and the single source single input site traditional ADS system. Input site or distribution is important as any non-infinite medium will have a loss of efficiency due to neutron escape or absorption through a systems outer wall. If a system is subcritical, and thus source driven, this can be minimized by either centralizing the source or making the core large in comparison to the fissioning neutrons mean free path. For this I used a good approximation of an evenly distributed group of sources through layers of phyllotaxis arrangements with even spacing in accordance with the total number of sources. This was done as leaving the individual placement of sources as open to the AI tended to increase the parameter space enough to make the required number of simulation runs computationally prohibitive. The AI also had a discretized control on the intensity level of each source for similar reasons.

Many additional spatial parameters could also be considered and optimized by AI but these would fit to more specific conditions rather than general more in the scope of engineering. These optimizable parameters include the use of coupled tanks, exact geometry, and interior design features such as heat exchangers. Thermal Hydraulics could also be considered an important spatial factor, but since anomalous

transport would be minimized by the use of a fast spectrum because of the time scale difference between bulk transport and from reaction lifetime from an initial source neutron. As a result, thermal hydraulic were ignored beyond the time scale for mixing for the desired homogeneity as this is a machine specific design focused consideration. An emphasis was instead placed on the relations of the source to transmutator concept particularly the use of multiple spatially distributed sources. This factor is best demonstrated by a spatially shaped reactivity insertion.

### 6.1.1 Source Controlled Spatially Shaped Thermal Insertion

The biggest danger and limitation for any fission based system comes from thermal deposition. This is because unplanned material vaporization, be it in a structural components, coolant, or fuel, needs to be avoided and become the primary limit to any safe operation. This also extends to secondary effects such as thermal based over pressurization that result in structural breach. Thermal management is especially important to most nuclear systems as overall thermal deposition is usually correlated with the overall power and efficiency.

The operational limits for transmutation is based on the capacity to cool any part of the system. A molten lead (LBE) system typically has a $500\ \mathrm{W_{th}/cm^3}$ limit which by equation 2-11 would be approximately $190\frac{kg}{m^3\ yr}$ for the volume cooled this way. This is where a major limitation for Aqueous Homogenous Reactors (AHR) utilizing water as its core medium as the water will break down at very low activity levels. It is also not a straightforward matter to utilize this upper limit through the entire tank for a subcritical system. Local power densities become especially important around the sources entry as too high of a local power density can cause state changes resulting in possible issues such as void bubbling which can cause sudden reactivity insertions.

Traditional ADS systems are especially limited by thermal deposition as most accelerators are large and expensive and do not have the option to change input location. While technical improvements can lead to the possibility of higher beam currents, there is an upper limit for single input site efficiency based on thermal management that concentrates on this peak input site. This results in a loss of transmutation efficiency in terms of transmuting the peak amount of material throughout the tank following activity drop off away from this peak input site. Proton spallation alleviates this problem somewhat by greatly increasing the overall initial disbursement area because of the long mean collisional distance from much higher proton energies. Targets made of materials such as tungsten and lead on the scale of a meter long are often used to make spread out distribution of secondary neutrons, that have much smaller mean collisional distances. This disbursement can be further flattened by varying target density or material but is still limited by target design. This contributes to a major efficiency loss from evaporation, and the use of secondary neutrons rather than primary source particles. This can result in losses on the order of a single GeV proton on average producing only 2 or 3 useable ~20 MeV neutrons for an approximately 4-6% efficiency. Compared to D-T fusion produced neutrons this is balanced by obtainable Q factors and importantly here the greater initial spread of neutrons utilizing a higher single source limit in a larger tank.

Sources placed by the wall would indeed suffer more edge-based neutron losses but they allow for a more efficient use of the tank that would otherwise be bounded by the centers highest rate of energy deposition and the limits of cooling at that location. The overall efficiency represented by $k_{eff}$ can also be increased by a higher concentration of fissionable material as fuel to counter this loss somewhat. A placement of multiple sources that balances these efficiencies can yield a higher overall efficiency for each Transmutator.

An effectively random source placement, as shown in **Fig. 11a**, will often produce a typical centered distribution where there is little benefit from putting sources closer to a wall where efficiency is lowered by neutron escape losses. However, a more even energy deposition allows for more distributed cooling and greater overall power density can be found through iterative AI design as seen in **Fig. 11b**. Here a

Phyllotaxis arrangement of 1000 neutron sources with parameters setting the 10 levels of normalized power is used.

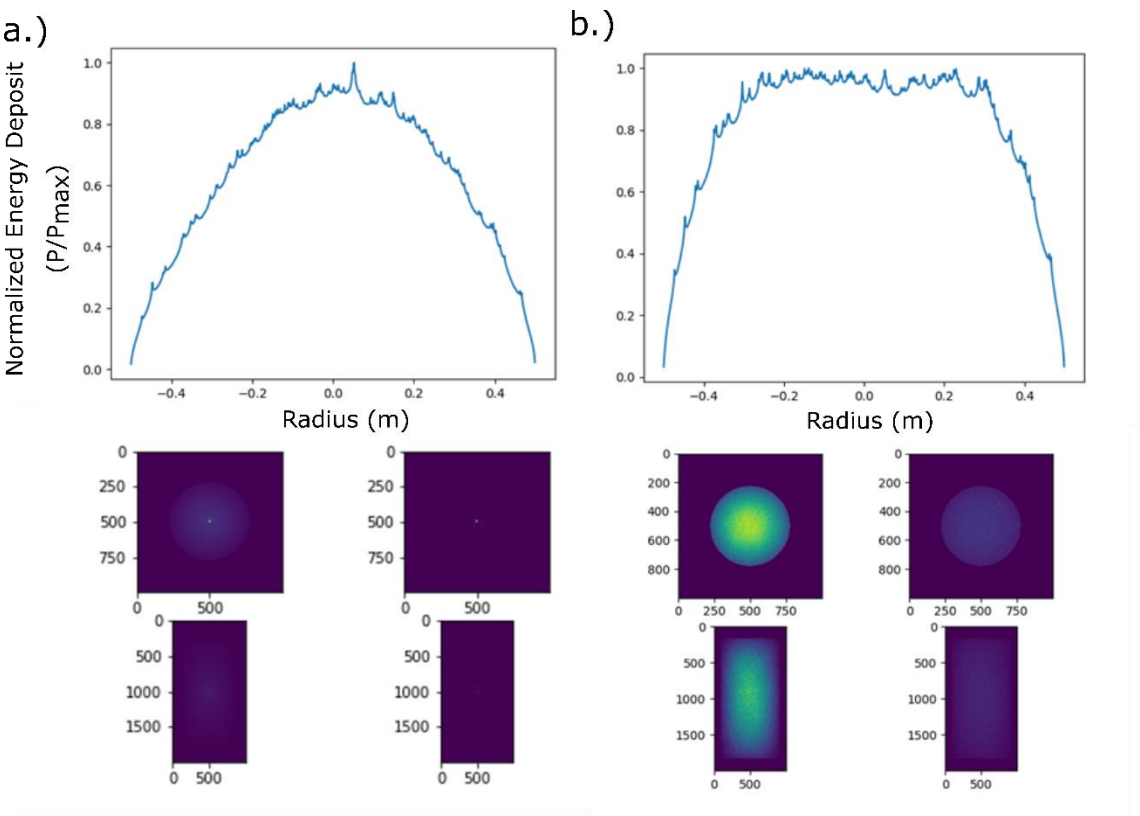


**Fig. 11** Thermal deposition profiles for early (a) and late (b) AI optimization results.

This was found through an EA with a population of 30 at start decreasing by one till a population is acheived for a total of 100 generation steps. Scoring was done by combining the criticality and the difference from maxium for the energery deposition for each part of the tank. In this way losses in efficency from increased escape is directly measured with the increases in efficiency from a more even power density.

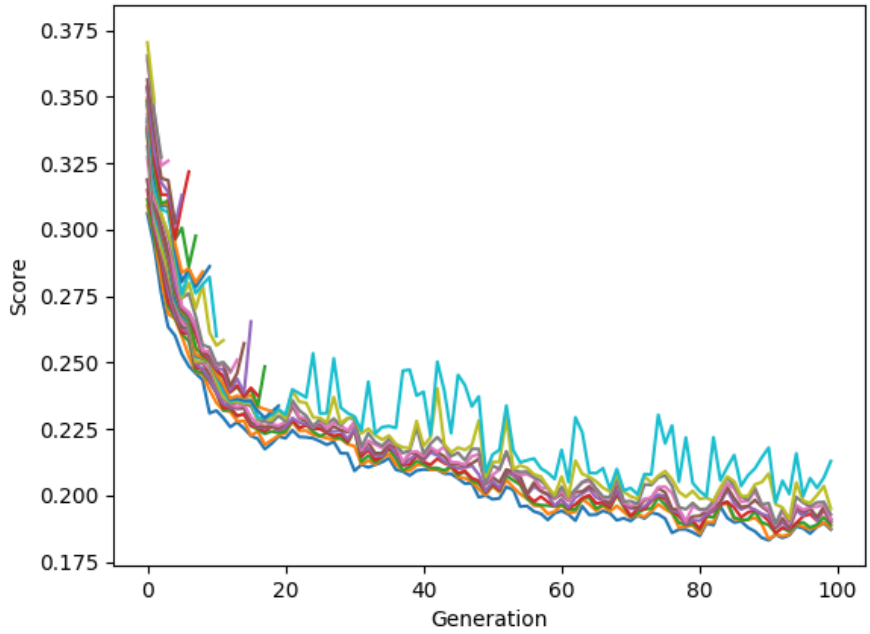


**Fig. 12** Scores for all individuals in EA optimization AI population by generation for Fig. 11 a-b.

Additional efficiency can be found by expanding the parameters to include fuel mix and concentration as well as geometric design for separate tanks such as the annular ring style shown below. This would for example allow for a boost to the outer regions of the tank by decreasing the neutron production efficiency (not necessarily the burn efficiency if more desirable but less efficient burn targets such as Am is used) in the most commonly overlapped regions of the center. This thermal insertion shaping can also be extended to safety and operational conditions through active monitoring, allowed by the molten salt, by actively shaping thermal input to bring a current measured system towards an ideal shape to match local thermal dissipation limits.

## *6.2 Temporal*

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.

### 6.2.1 Operational FP Replacement Process Control by AI

A major goal for this study is verifying pathways for a no isotope separation waste burner. This is an important factor in the advantage of using a molten salt system in a plausible real-world solution for nuclear waste. Finding such pathways can be accomplished by using AI to show how long-time scale in-situ operational control such as quantity and timing for waste adding and removal allowed by the liquid nature of the molten salt can operate as an efficient waste burner.

For this to be a good candidate as a real-world solution for nuclear waste that can be quickly developed this study was constrained to already existing process such as PUREX to determine what could be added or removed. This is an issue though, since as can be seen in **Fig. 13**, using MA as taken directly from typical reprocessing methods as the primary driver for transmutation would be very inefficient. Source neutrons 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 concentration of 1.6 mol% (~10 wt%) would be well below what is achievable with a $M = 50$ or more of typically considered sub-critical systems. Transmutation of waste directly would then

rely 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. 13** 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 and undergo (n, 2n) 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.

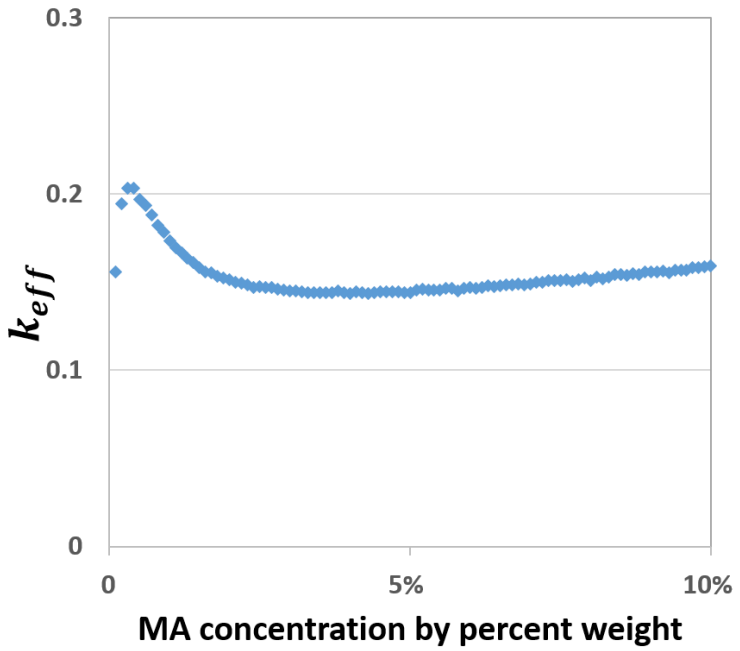


**Fig. 13** Criticality of system by fuel to molten salt medium weight with only MA processed from SNF as fuel.

An inefficient MA fuel can be countered by an initial quantity of actinide (normally fissile) fuel, particularly plutonium being initially left mixed with the minor actinides from PUREX. However, our goal is a minor actinide burner and while efficiently using neutrons is good this is still not an efficient MA burner.

Minor actinides are very inefficient because even if the first set of nuclides easily fission from fusion neutrons the resultant fission neutrons are not good at maintaining the chain reaction as their energies are more likely to capture than fission again. However, waste is still fertile and if it captures becomes more and more efficient, as shown in **Fig. 4** because of the atomic ladder discussed in section 2.2.1. This improves the current mixtures efficiency even if after starting only additional MA waste with no Pu is added.

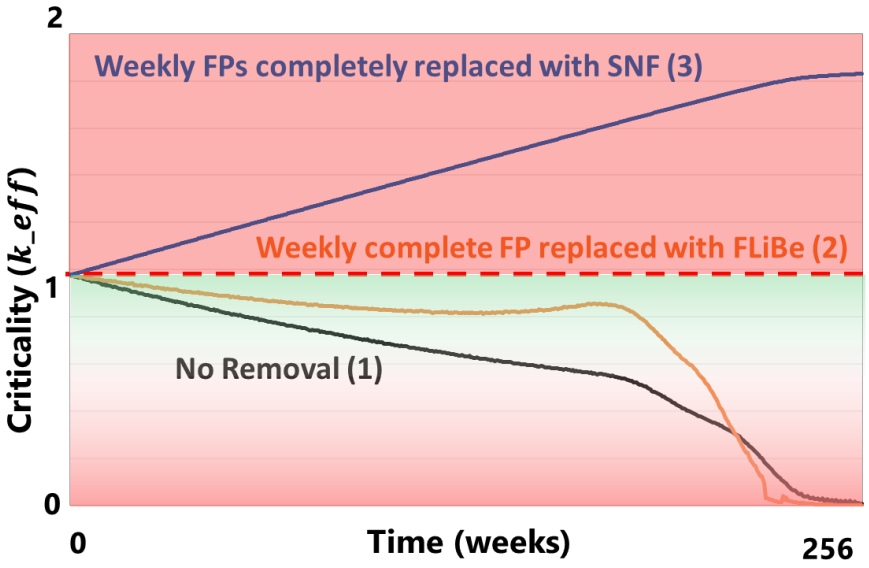


**Fig. 14** Comparison of the criticality depending on the operational modes of transmutator: (1) no removal of FP nor input of SNF; (2) weekly removal of all FP replaced with FLiBe; (3) weekly FP removal of all FP replaced by fresh SNF. SNF Loading Fuel is at the 100 MW Thermal Burn Power

This would normally be hindered by the accumulation of fission products or the reduced overall quantity and thus density of fissile materials, but this can be balanced process wise during operation because of the molten salt. As long as a target trend remains between the trends represented by maximum processable and replaceable with waste or additional molten salt and the trend represented by no processing the fuel can be developed, this would extend to other waste outputs from differing types of plants and inverted trend lines as well. Our goal then becomes allowing for the remaining waste to develop and become a fuel so efficient that the initial Plutonium is no longer needed. This would then turn into a generalized 3-stage process **Fig.**

**15**, the initial being this ‘development’ phase with a target goal being developing increasingly efficient fuel until the focus can change to efficiently burning waste. After the development phase the transmutator can then be ran in a ‘static’ phase whereby balancing the in-operation fission product removal and the addition of more minor actinide waste the transmutator runs continuously burning all the waste that is inputted. A ‘shutdown’ phase could also be considered as the final burn with no additional waste added and TRU quantity reduced until minimal. The ‘shutdown’ phase while included as markedly different focus will not be covered as the overall importance and effect would be negligible in real world operation as tank contents could be continually combined or operation in the static phase continued for an indefinite time.


**Fig. 15** 3 stage transmutation operational plan

This is started with the development phase where, while minor actinide waste burning still occurs, initially a majority of neutrons go to depleting the plutonium. However, enough capture occurs that then develops an increasingly efficient fuel. This primarily derives from Am-241, the normally most problematic waste, capturing a lot of these neutrons resulting in an easier to fission fuel mostly consisting of curium. This is continued until it can be self supporting, as can be seen in*Fig. **16*** An example of a 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 for the development phase.**Fig. 16a**, by the increasing amount of curium. In a solid fuel this would quickly become too inefficient to be worth running or the curium too heterogeneously developed and concentrated and possibly dangerous. However, molten salt can be mixed to continuously homogenize the fuel and can also be processed either continuously during operation or at scheduled stop then start steps as shown in*Fig. **16*** An example of a 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 for the development phase.**Fig. 16b** in an excessively large period for demonstration purposes.

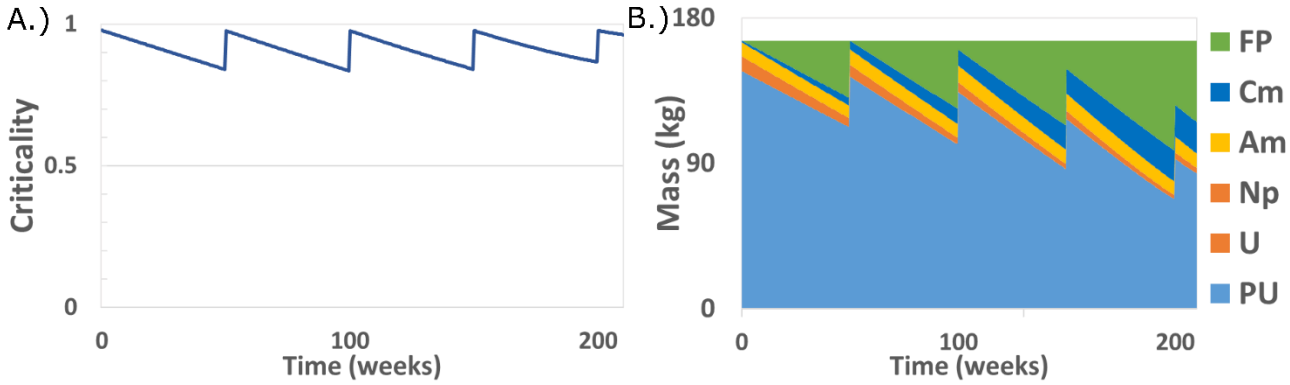


**Fig. 16** An example of a feedback control scenario, showing (a) criticality change and (b) isotopic evolution with operational scheduling of FP replacement with fresh SNF waste at 50 week intervals for the development phase.

This would also normally be hindered by the steady drop in criticality which is due to fission products capturing neutrons that could otherwise be causing fissions or even being captured and breeding the curium fuel. However, the AI here is determining the ideal amount of processing to perform at these steps to continue operating efficiently. The scoring also keeps in mind other aspects of the system such as power reactivity insertion coefficients for safety purposes. This example is just an extreme on process limiting and step length allowing an exceptionally large power efficiency loss, but in-process removals and more frequent small partition processing can retain better overall power efficiencies as have also done. This is not as important though as this operation type will typically converge on a better or sufficient fuel mixture for reasons such as the higher neutron production per fission of curium than plutonium if other possible neutron losses are controlled, and a fast spectrum is being maintained. Once this mixture is developed it can then be used for the next stage, although it may need to be a combined result from more than one tank to obtain sufficient quantities as in this case where 9 tanks are combined if all plutonium is then removed.

A developed mixture is then used in the ‘static’ phase where a similar processing schedule is found by an AI to maintain a consistent balance of creating more of its own fuel and burning all the purely MA waste that is added as shown in **Fig. 17**. This allows for burning of an arbitrary amount of waste by continuing the process as long as needed. It should also be noted that the balance is not only just restoring criticality but keeping the integral ‘effective’ breeding or conversion ratio equal at 1.

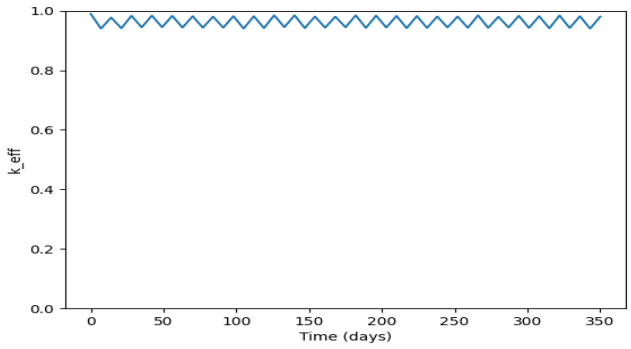


**Fig. 17** An example of a feedback control scenario, showing criticality change with operational scheduling of FP replacement with fresh SNF waste at 1 week intervals for the static phase. Transmuted: ~7kg from operation at.98 keff and 1.416 MWth power output.

# Chapter 7

## 7 Final Conclusion

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 steady state 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 [58] 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 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 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. 16** that are controlled to reach desired isotopic ratios to then allow for steady state burns in the style of **Fig. 17**. 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. Combined these updated avenues in transmutation could allow for efficient and thorough 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 humanity may finally begin to reduce the long standing nuclear waste problem.

# REFERENCES


[1] IAEA, *ESTIMATION OF GLOBAL INVENTORIES OF RADIOACTIVE WASTE AND OTHER RADIOACTIVE MATERIALS*, 2008.

[2] J. Magill, V. Berthou, D. Haas, J. Galy, R. Schenkel, H. W. Wiese, G. Heusener, J. Tommasi, and G. Youinou, Nucl. Energy **42**, 263 (2003).

[3] INTERNATIONAL ATOMIC ENERGY AGENCY, *Implications of Partitioning and Transmutation in Radioactive Waste Management* (INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna, 2004).

[4] S. David, H. Nifenecker, and O. Meplan, *Accelerator Driven Subcritical Reactors*, Vol. 20031722 (Institute of Physics Publishing, Bristol and Philidelphia, 2003).

[5] V. Ignatiev, in *Gen IV International Forum* (GIF, Moscow, 2018).

[6] J. Serp, M. Allibert, O. Beneš, S. Delpech, O. Feynberg, V. Ghetta, D. Heuer, D. Holcomb, V. Ignatiev, J. L. Kloosterman, L. Luzzi, E. Merle-Lucotte, J. Uhlíř, R. Yoshioka, D. Zhimin, J. Leen, L. Luzzi, E. Merle-Lucotte, J. Uhlí, and D. Zhimin, Prog. Nucl. Energy **77**, 308 (2014).

[7] J. L. Kloosterman, in *Proc. TU Delft* (TUDelft, Delft, 2017).

[8] M. Vazquez and F. Martin-Fuertes, in *International Conference on the Physics of Reactors 2012*, Vol. 3 (American Nuclear Society, La Grange Park, 2012).

[9] A. C. Mueller, in *Journal of Physics: Conference Series*, Vol. 420 (CNRS, Paris, 2013), p. 012059.

[10] T. Tajima, A. Necas, G. Mourou, S. Gales, and M. Leroy, Fusion Sci. Technol. **77**, 251 (2021).

[11] R. C. Briant and A. M. Weinberg, Nucl. Sci. Eng. **2**, 797 (1957).

[12] Rosenthal M.W., Kasten P.R., and Briggs R.B., Nucl Appl Technol **8**, 107 (1970).

[13] X. Doligez, D. Heuer, E. Merle-Lucotte, M. Allibert, and V. Ghetta, Ann. Nucl. Energy **64**, 430 (2014).

[14] A. Gandini and M. Salvatores, J. Nucl. Sci. Technol. **39**, 673 (2002).

[15] G. Chiba, K. Nishihara, and T. Endo, in *International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering* (American Nuclear Society, Rio de Janeiro, 2011).

[16] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. **21**, 1087 (1953).

[17] N. Metropolis, *THE BEGINNING of the MONTE CARLO METHOD*, 1987.

[18] A. Necas, T. Tajima, G. Mourou, K. Osvay, C. Kamperidis, G. Korn, S. V. Bulanov, J. Wheeler, M. Matys, and P. Valenta, Submitt. to Phys. Rev. Accel. Beams (2019).

[19] S. Steinke, A. Henig, M. Schnürer, T. Sokollik, P. V Nickles, D. Jung, D. Kiefer, R. Hörlein, J. Schreiber, T. Tajima, X. Q. Yan, M. Hegelich, J. Meyer-Ter-Vehn, W. Sandner, and D. Habs, Laser Part. Beams **28**, 215 (2010).

[20] T. Tajima, D. Habs, and X. Yan, Rev. Accel. Sci. Technol. **02**, 201 (2009).

[21] R. A. Snavely, M. H. Key, S. P. Hatchett, I. E. Cowan, M. Roth, T. W. Phillips, M. A. Stoyer, E. A. Henry, T. C. Sangster, M. S. Singh, S. C. Wilks, A. MacKinnon, A. Offenberger, D. M. Pennington, K. Yasuike, A. B. Langdon, B. F. Lasinski, J. Johnson, M. D. Perry, and E. M. Campbell, Phys. Rev. Lett. **85**, 2945 (2000).

[22] D. Habs, T. Tajima, J. Schreiber, C. P. J. Barty, M. Fujiwara, and P. G. Thirolf, Eur. Phys. J. D **55**, 279 (2009).

[23] X. Q. Yan, T. Tajima, M. Hegelich, L. Yin, and D. Habs, Appl. Phys. B Lasers Opt. **98**, 711 (2010).

[24] F. Mako and T. Tajima, Phys. Fluids **27**, 1815 (1984).

[25] G. Mourou, S. Mironov, E. Khazanov, and A. Sergeev, Eur. Phys. J. Spec. Top. **223**, 1181 (2014).

[26] C. S. Brady and T. D. Arber, Plasma Phys. Control. Fusion **53**, 015001 (2011).

[27] T. J. Dolan, Molten Salt Reactors and Thorium Energy.

[28] S. C. Chetal, V. Balasubramaniyan, P. Chellapandi, P. Mohanakrishnan, P. Puthiyavinayagam, C. P. Pillai, S. Raghupathy, T. K. Shanmugham, and C. S. Pillai, Nucl. Eng. Des. **236**, 852 (2006).

[29] R. J. Sheu, C. H. Chang, C. C. Chao, and Y. W. H. Liu, Ann. Nucl. Energy **53**, 1 (2013).

[30] C. Rubbia, C. Roche, J. A. Rubio, F. Carminati, Y. Kadi, P. Mandrillon, J. P. C. Revol, S. Buono, R. Klapisch, N. Fiétier, and Others, *Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier*, 1995.

[31] A. V. Anikeev, in *AIP Conference Proceedings*, Vol. 1442 (2012), pp. 153–158.

[32] R. Serrano-López, J. Fraderaa, and S. Cuesta-Lópeza, Chem. Eng. Process. Process Intensif. **73**, 87 (2013).

[33] C. N. A. C. Z. Bahri, W. M. Al-Areqi, M. I. F. M. Ruf, and A. A. Majid, in *AIP Conference Proceedings*, Vol. 1799 (AIP, Selangor, 2017), p. 40008.

[34] S. Gales, D. L. Balabanski, F. Negoita, O. Tesileanu, C. A. Ur, D. Ursescu, and N. V Zamfir, Phys. Scr. **91**, 093004 (2016).

[35] http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/processing-of-used-nuclear-fuel.aspx.

[36] B. J. Quiter, T. Laplace, B. A. Ludewigt, S. D. Ambers, B. L. Goldblum, S. Korbly, C. Hicks, and C. Wilson, Phys. Rev. C **86**, 34307 (2012).

[37] I. Gauld and M. Francis, in *Proceedings of INMM 51st Annual Meeting* (2010), pp. 1–8.

[38] C. Moulin, P. Decambox, and P. Mauchien, Le J. Phys. IV **1**, C7 (1991).

[39] R. G. Geier, *Purex Process Solvent: Literature Review*, 1979.

[40] J. R. Keiser, *Compatibility Studies of Potential Molten-Salt Breeder Reactor Materials in Molten Fluoride Salts*, 1977.

[41] L. C. Olson, J. W. Ambrosek, K. Sridharan, M. H. Anderson, and T. R. Allen, J. Fluor. Chem. **130**, 67 (2009).

[42] Y. Gao and P. Hao, Phys. E Low-Dimensional Syst. Nanostructures **41**, 1561 (2009).

[43] O. A. Williams, M. Daenen, J. D'Haen, K. Haenen, J. Maes, V. V. Moshchalkov, M. Nesládek, and D. M. Gruen, Diam. Relat. Mater. **15**, 654 (2006).

[44] M. X. Navarro, R. R. Delgado, M. G. Lagally, G. L. Kulcinski, and J. F. Santarius, Fusion Sci. Technol. **72**, 713 (2017).

[45] P. N. Haubenreich and J. R. Engel, Nucl. Appl. Technol. **8**, 118 (1970).

[46] A. K. Geim and K. S. Novoselov, Nat. Mater. **6**, 183 (2007).

[47] T. Klein, P. Achatz, J. Kacmarcik, C. Marcenat, F. Gustafsson, J. Marcus, E. Bustarret, J. Pernot, F. Omnes, B. E. Sernelius, C. Persson, A. Ferreira da Silva, and C. Cytermann, Phys. Rev. B **75**, 165313 (2007).

[48] D. R. Kania, M. I. Landstrass, M. A. Plano, L. S. Pan, and S. Han, Diam. Relat. Mater. **2**, 1012 (1993).

[49] F. Glover, Comput. Oper. Res. **13**, 533 (1986).

[50] K. Sörensen, M. Sevaux, and F. Glover, *A History of Metaheuristics* *, 2017.

[51] K. De Jong, D. B. Fogel, and H.-P. Schwefel, in *Handbook of Evolutionary Computation*, edited by T. Baeck, D. . Fogel, and Z. Michalewicz (IOP Publishing Ltd and Oxford University, 1997).

[52] K. Sörensen, Int. Trans. Oper. Res. **22**, 3 (2015).

[53] J. V. Hernandez, A. Vannucci, T. Tajima, Z. Lin, W. Horton, and S. C. Mccool, Nucl. Fusion **36**, 1009 (1996).

[54] C. Scott, S. Dettrick, T. Tajima, R. Magee, and E. Mjolsness, Nucl. Fusion **60**, 126025 (2020).

[55] J. J. Hopfield, Proc. Natl. Acad. Sci. U. S. A. **79**, 2554 (1982).

[56] T. Tajima, *Computational Plasma Physics: With Applications to Fusion and Astrophysics* (Addison Wesley Publishing Company, Inc., Redwood City, California, 1989).

[57] W. A. Little and G. L. Shaw, Math. Biosci. **39**, 281 (1978).

[58] F. Carminati, C. Roche, J. A. Rubio, C. Rubbia, J. P. C. Revol, and R. Klapisch, **47**, (1993).