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ADVANCED FUEL REPROCESSING AND MULTI-PHYSICS ANALYSIS OF
THE MOLTEN SALT BREEDER REACTOR

BY

ANDREI RYKHLEVSKII

THESIS

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Master's Committee:

Assistant Professor Kathryn Huff, Chair
Associate Professor Tomasz Kozlowski

ABSTRACT

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INTRODUCTION

1.1 Background and motivation

Nowadays humankind has only few ways to generate reliable, nonintermittent base load power, namely, in no particular order: fossil fuel, hydroelectric, biomass, geothermal, and nuclear energy. Because of increasing global warming and climate change concerns, sources that have negligible CO₂ footprints represent crucial measures for control global temperature change. From environmental viewpoint these observations allow conclude that hydro and nuclear power are preferable ways to generate nonintermittent power¹. Nevertheless, potential for hydro power is strictly limited by local geographical conditions and have significant impact on environment, affecting land use, homes, and natural habitats in the dam area, consequently, one could argue that nuclear power should find wide usage around the world.

In reality, nuclear power plants currently generate 4.9% of the global energy production [2], a figure which is projected to stay constant up to 2040 while electricity demand predicted to expand by 30% [3]. In contrast, contribution of coal in 2040 projected to be about 30%. This visible fear towards large-scale switch from fossil to nuclear power based on a number factors such as concerns regarding safety, nuclear weapon proliferation, radioactive waste treatment, and competitiveness with other sources of energy (i.e. renewables). Unfortunately, because of all these fears, a negative public attitude to nuclear has formed in many developed countries which makes challenging to advocate its zero emissions benefits.

Molten Salt Reactors (MSRs) is one effort to overcoming the problems related to conventional Nuclear Power Plants (NPPs). As one of six advanced

¹Biomass could be classified as renewable energy source but recent research has proved that it also produce considerable amount of CO₂ and not necessarily beneficial for the environment over time scales that are relevant when considering global climate change [1].

reactor concepts that have been chosen by the Generation IV International Forum (GIF) for further research and development, MSRs offering significant improvements “in the four broad areas of sustainability, economics, safety and reliability, and proliferation resistance and physical protection” [4]. To achieve the goals formulated by the GIF, MSRs attempt to simplify the reactor core and improve inherent safety by using liquid coolant which is also a fuel². In the thermal spectrum MSR, fluorides of fissile and/or fertile materials (i.e. UF_4 , PuF_3 and/or ThF_4) are mixed with carrier salts to form a liquid fuel which is circulated in a loop-type primary circuit [5]. This innovation leads to immediate advantages over traditional, solid-fueled, reactors. These include near-atmospheric pressure in the primary loop, relatively high coolant temperature, outstanding neutron economy, a high level of inherent safety, reduced fuel preprocessing, and the ability to continuously remove fission products and add fissile and/or fertile elements [6].

The thermal spectrum Molten Salt Breeder Reactor (MSBR) was designed to realize the promise of the thorium fuel cycle, which uses of natural thorium instead of enriched uranium. Thorium breeds fissile ^{233}U and avoids uranium enrichment. The mixture of $\text{LiF}-\text{BeF}_2-\text{ThF}_4-\text{UF}_4-\text{PuF}_3$ has a melting point of 499°C, a low vapor pressure at operating temperatures, and good flow and heat transfer properties [7].

The MSBR complex geometry is challenging to describe in software input and usually require major geometric approximations [8].

1.2 Methods

1.3 Objective and outline

The thermal spectrum MSR is an advanced type of reactor that consists of constantly circulating liquid fuel (i.e., mixture of $\text{LiF}-\text{BeF}_2-\text{ThF}_4-\text{UF}_4$ or $\text{LiF}-\text{BeF}_2-\text{ZrF}_4-\text{UF}_4$) and solid graphite moderator structures. This liquid fuel form leads to immediate advantages over traditional, solid-fueled, reactors. The molten-salt carrier with dissolved fissile and/or fertile material allows

²Molten-salt-cooled reactors with solid fuel are usually also referred to MSRs. However, in this thesis, MSRs are assumed to be reactors with liquid fuel which simultaneously serves as coolant.

for online refueling and reprocessing. Thus, MSRs can potentially operate years without shutdown, achieving maximum fuel utilization and outstanding neutron economy [6]. Moreover, this type of fuel does not need complex fabrication since the fissile material could be transported from any enrichment plant to the NPP in the form of uranium hexafluoride (UF_6). Additionally, MSRs have a high level of inherent safety due to their characteristically the strong negative temperature coefficient of reactivity, near-atmospheric pressure in the primary loop, stable coolant, passive decay heat cooling, and small excess reactivity [9].

The thorium-fueled MSBR was developed in the early 1970s by Oak Ridge National Laboratory (ORNL) specifically to realize the promise of the thorium fuel cycle which allows the use of natural thorium instead of enriched uranium as the fertile element. Thorium breeds the fissile ^{233}U and avoids uranium enrichment [7]. In the matter of nuclear fuel cycle, the thorium cycle produces a reduced quantity of plutonium and minor actinides (MAs) compared to the traditional uranium fuel cycle. Consequently, it may significantly increase proliferation resistance when the MSR operates in the breeder regime. The MSRs also could be employed as a converter reactor for transmutation of spent fuel from current Light Water Reactors (LWRs).

Recently, interest in MSRs has resurged, with multiple new companies pursuing commercialization of MSR designs³. To further develop these MSR concepts, particularly with respect to their strategies for online reprocessing and refueling, computational analysis methods capturing their unique reactor physics and process chemistry are needed. However, most contemporary nuclear reactor physics software is unable to perform depletion calculations in an online reprocessing regime. Powers *et al.* suggested a novel method for conducting depletion simulations for MSR. This suggested method takes into account fuel salt composition changes due to online reprocessing and refueling based on the deterministic computer code NEWT in SCALE [10]. This approach was later used by Jeong *et al.* to find an equilibrium fuel composition for the MSBR and was validated with a Monte Carlo N-Particle code (MCNP)/CINDER90 model [11].

The current paper presents a single-cell model developed using the continuous-energy Serpent 2 Monte Carlo reactor physics software. It was employed to

³Examples include both liquid-fueled molten salt designs from Transatomic, Terrapower, Terrestrial, and Thorcon.

establish a Serpent-based method for finding the equilibrium core composition and core depletion of the MSBR. All calculations presented in this paper were performed using the Serpent 2 code version 2.1.29 with ENDF/B-VII.0 nuclear data [?, ?]. Serpent 2 is an improvement upon Serpent 1, and contains a complete redesign of memory management using hybrid OpenMP + MPI parallelization. This hybrid parallelization is important in depletion calculations using computer clusters with multiple cores [12]. This work used the built-in Serpent 2 depletion capabilities and its built-in online reprocessing subroutine. Another feature of the MSBR, its circulating liquid fuel and corresponding delayed neutron precursor drift, is not treated here.

MOLTEN SALT REACTORS

2.1 History

MSR development started in the late 1940's as part of the United States' program to design a nuclear powered airplane [13]. Particularly, liquid fuel appeared to offer a number of advantages, so experiments to demonstrate the feasibility of molten salt fuels were begun in 1947. "At the enthusiastic urging of Bettis and on the recommendation of W.R. Grimes, R.C. Briant adopted molten fluoride salts in 1950 as the main line effort of the Oak Ridge National Laboratory (ORNL)'s Aircraft Nuclear Propulsion program." The fluorides appeared exceptionally suitable because they have high solubility for uranium, are among the most stable of chemical compounds, have low vapor pressure even at temperature more than 1300°C, have fairly good hydraulic and thermal properties, do not react furiously with air or water, are not damaged by high neutron fluxes, and are inert to some common structural materials [14].

A small test reactor, the Aircraft Reactor Experiment (ARE), was built at Oak Ridge site to probe the use of molten fluoride fuels for aircraft propulsion reactors and to study the nuclear stability of the circulating fuel system. The fuel salt for the ARE was a mixture of NaF, ZrF₄, and UF₄. BeO served as moderator, and all the piping was nickel-chromium alloy Inconel. The experiment was successful: in 1954 the ARE was operated for 9 days at steady-state outlet temperatures up to 860°C and at powers up to 2.5 MW_(th). No mechanical or chemical problems were observed, and the reactor was found to be stable and self-regulating [13].

The great potential of MSRs for civilian power application was recognized from the beginning of Aircraft Nuclear Propulsion program, and in 1956 H.G. MacPherson founded a group to study the technical characteristics, nuclear

performance, and economics of molten salt converting and breeding reactors. After few years of research with number of concepts, MacPherson and his colleagues concluded that graphite-moderated thermal reactors operating on a thorium fuel cycle would be the best choice for applying molten salt systems for producing economic energy [14]. Breeding ^{233}U from ^{232}Th was found to give better performance in a molten salt thermal reactor neutron energy spectrum than a uranium fuel cycle in which depleted uranium (^{238}U) is the fertile material and fissile ^{239}Pu is produced and recycled. Homogeneous reactor designs that have an entire core consisting liquid salt were rejected because the moderation by the salt was limited compared to a reactor moderated by graphite. Furthermore, intermediate spectrum reactors did not appear to have high enough breeding ratios to compensate their higher inventory of fuel [14]. Later studies of fast spectrum molten salt reactors have shown that effective breeding could be obtained with extremely high power densities that needed to avoid excessive fissile inventories [15]. Acceptable power densities appeared challenging to achieve without using novel and untested heat transfer technologies [14].

Two types of graphite-moderated reactors were selected by MacPherson's group for further research: single-fluid reactors in which thorium and uranium are dissolved in the same carrier salt, and two-fluid design in which a fertile salt accommodated ^{232}Th is separated from the fissile salt which contains ^{233}U and/or ^{239}Pu as initial fissile load for reactor startup. The two-fluid reactor could operate as breeder except that construction materials for flows separation would significantly deteriorate neutron economy and, consequently, breeding ratio. The single-fluid design is much simpler, easier to build and offers lower power costs, even for that time technology which could only achieve breeding ratio slightly below 1.0. The chemical reprocessing method, namely the fluoride volatility process [16], which separates uranium from fluoride salts, had been already demonstrated during ARE for recovery uranium from ARE fuel salt and might be used for partial reprocessing of salts from another type of reactor.

The U.S. Atomic Energy Commission Task Force has considered results of the ORNL research and made a comparative evaluation of liquid-fueled reactors early in 1959. One conclusion of the Task Force was that the MSR even limited in potential breeding gain, had "the highest probability of achieving technical feasibility" [17].

In the 1960s more complete conceptual MSR designs have been developed. ORNL concluded that both single-fluid and two-fluid concepts would lead to reactors with low cost of power generation, and that moving to the breeder either directly or using the converter would create reactors with good fuel utilization characteristics [14]. Because many of the features of commercial power reactors would differ from those for the ARE, and the ARE had been operated only a short period of time, new reactor experiment with molten salt was necessary to investigate some of the technology for civilian power reactors.

The developing of the Molten Salt Reactor Experiment (MSRE) was started in 1960. Creators selected a single-fluid design because it is similar to a converter, but the fuel salt did not contain thorium, and, consequently, was similar to the fuel salt composition for a two-fluid breeder. The MSRE fuel salt is a mixture of uranium, ^7Li , beryllium, and zirconium fluorides. Bare graphite serves as the moderator because the salt cannot penetrate into its pores if the pore sizes are small. Specially developed in the aircraft program, nickel-based alloy INOR-8 (also called Hastelloy-N) for use with molten fluorides was employed as a main construction material for piping and system components. The maximum power is about 8MW_{th} , and the heat is dissipated to the atmosphere [5].

Construction of the MSRE began in 1962, and the reactor first became critical in 1965. Figure 2.1 shows assembling of a graphite reactor core. Continuous operation at full power began in December 1966. Successful completion of a six-month test campaign in March of 1968 closed the first phase of operation, all initial objectives were achieved. The molten fluoride fuel salt was used in the reactor core for many months at temperatures $\geq 649^\circ\text{C}$ without corrosive damaging of the metal and graphite elements of the system. All reactor equipment worked reliably, radioactive liquids and gases were retained safely, the fuel salt was absolutely stable. Xenon was removed continuously from the salt. Radioactive equipment was repaired or replaced in acceptable time without overexposing maintenance personnel.

The second stage of MSRE started in August 1968 when a small chemical processing facility connected to the reactor was used to remove the original uranium from the fuel salt using fluorine gas. ^{233}U fuel was added to the same carrier salt, and on October 2 the MSRE began operation using ^{233}U . Six days later the power 100 kW was achieved by Glenn T. Seaborg, Chairman

of the U.S. Atomic Energy Commission, bringing to power the first reactor in the world to operate using ^{233}U [5].



Figure 2.1: The MSRE core, shown while being assembled, contains about 1.95 m^3 of reactor graphite. The 1'140 fuel channels contain about 0.57m^3 of fuel salt.

After MSRE was built and brought into operation, most of the research and development work on MSR's was in support of the MSRE. However, molten fluoride salts chemistry continued developing during this period. One discovery during this time was that the lithium fluoride and beryllium fluoride can be separated from rare earths by vacuum distillation at temperature about 1000°C [18]. This method provided an inexpensive, on-site way for recovering valuable rare materials, and following this, the efforts for future reactors changed focus to a two-fluid breeder. In this reactor, the fuel salt should be fluorinated to recover the uranium and distilled to separate carrier

salt from fission products. The blanket salt must be processed by fluorination alone, since few fission products would be generated in the blanket if the uranium concentration were kept low [14]. Graphite tubes in the core were designed to prevent the fuel and fertile streams from mixing.

Two-fluid system analyses have shown that breeding ratio could be in the range of 1.07 to 1.08, which with low fissile inventory would lead to relatively good fuel utilization. Consequently, the development effort for future molten salt reactors by ORNL was aimed mainly at the features of two-fluid breeders [19]. The main drawback of those reactors was identified as graphite pipes damaging by very high neutron fluxes.

Later, in 1967, new experimental information obtained from MSRE and an advance in core design lead to shift the ORNL molten salt program R&D focus from the two-fluid to a single-fluid breeder. This switch was based on concerns about graphite behavior at higher radiation exposures that had been achieved previously, graphite changes dimensions more rapidly than had been anticipated. To use in MSBR reactor graphite type which was tested during MSRE, lower core power densities enabled acceptable graphite lifetime but, even still, these components required frequent replacement. Furthermore, due to core assembly complexity, the entire core and reactor vessel required replacement when any graphite element reached its irradiation limit or developed a leak [14].

To achieve acceptable breeding ratio in single-fluid reactor, ^{233}Pa (27.4-day half-life) must be separated from the fuel salt and held outside the core until it decays to ^{233}U . Laboratory experiments demonstrated liquid-liquid extraction process for removing protactinium and uranium from molten fluoride salts. The method is to exchange thorium and lithium dissolved in molten bismuth for the components to be removed from the salt. Additional data have confirmed that the uranium can be selectively separated from the salt, the protactinium can be trapped in the salt in a decay tank, and the uranium can be returned back to the fuel salt by electrolysis for subsequent transfer to the core. Analysis indicated that the extraction and electrolysis could be carried out rapidly and continuously.

The fertile “blanket” in the single-fluid breeder is obtained by increasing the volume fraction of fuel salt and reducing the volume fraction of graphite in the outer part of the reactor. This advanced core design makes the outer region undermoderated and increases neutron capture there by the thorium.

Moreover, most of neutrons are born in the inner region, at some distance from the reactor boundary, and captures in the outer region which reduce the neutron leakage. Further studies indicated that the fuel utilization in single-fluid, two-region MSR can be as good as in two-fluid prototype, and even with the limitation on graphite lifetime the economics might be better [14]. Thus, in 1968 ORNL MSR Program was oriented toward the development of single-fluid breeder reactor.

Despite the success of ARE and MSRE, the MSR program closed down in the early 1970s in favor of the liquid metal fast-breeder reactor (LMFBR),[20] after which molten salt reactor research stagnated in the United States. As of 2018, the ARE and MSRE remained the only MSRs ever operated in the world.

Recently, interest in MSRs has resurged, with multiple new companies pursuing commercialization of MSR designs (e.g. liquid-fueled molten salt designs from Transatomic, Terrapower, Terrestrial, and Thorcon). China initiated a thorium molten salt reactor research project, and demonstrations of the liquid fuel version (TMSR-LF) are targeted for 2024. European Union funds the Safety Assessment of the Molten Salt Fast Reactor (SAMOFAR) project, in which several European research institutes and universities are developing various molten salt reactor prototypes such as the Molten Salt Fast Reactor (MSFR), the Molten Salt Actinide Recycler and Transmuter (MOSART), the Fluoride-Salt-Cooled High-Temperature Reactor (FHR). To further development of these MSR concepts, particularly with respect to their strategies for online reprocessing and refueling, computational analysis methods capturing their unique reactor physics and process chemistry are needed.

2.2 Thorium fuel cycle overview

In the early days of nuclear energy industry, in the United States as a follow-up of the Manhattan Project (1945-1960), leading U.S. national laboratories studied thorium as a possible substitute for uranium and the possibility of using ^{233}U in a nuclear weapon. In the Atoms for Peace Program, with its great variety of developments (1955-1975), thorium appeared to be an interesting resource for supplementing limited uranium availability in the

context of a fast-growing nuclear industry because thorium is at least 4-5 times more abundant than uranium in Earth's crust and preparation of thorium fuel does not require difficult and expensive enrichment processes. International Fuel Cycle Evaluation Conference (INFCE) of 1978 predicted thorium would someday be almost equal in importance to uranium. It stated that in case of the optimistic nuclear energy development scenario, thorium would be called upon massively in the future. These predictions were too optimistic but in a long-term, the use of thorium along with uranium could significantly improve the potential of nuclear energy [21].

During this pioneering period, thorium fuel cycle research and development for prototype demonstration reactors were initiated, first in the United States under cooperation between the United States Atomic Energy Commission (USAEC) and U.S. industry, then in Europe. About 1500 kg of ^{233}U have been bred in the United States from 900 metric tons of thorium. Many reactor prototypes as well as thorium extraction plants were built and operated in many countries. The U.S. and France have each separated from the ore about 2000 metric tons of thorium, part of which is still available [21]. However, for most countries uranium was relatively abundant and research in thorium fuel cycles diminished from late 1970s to 2000s. A notable exception was India's three-stage nuclear power program [22]. In the twenty-first century thorium's potential for improving proliferation resistance and waste characteristics is generating renewed interest in the thorium fuel cycle [23].

Compared to natural uranium which contains 99.284% ^{238}U , thorium almost exclusively composed of ^{232}Th . It can be seen from figure 2.2 that the fertile isotopes are ^{238}U and ^{232}Th for uranium-plutonium and thorium fuel cycle, respectively. Accordingly, the fissile isotopes are ^{235}U , 0.711% of natural uranium present in nature, and the artificial fissile isotopes are ^{239}Pu and ^{233}U for U-Pu and thorium cycles, respectively.

In the Uranium-Plutonium cycle, production of fissile material (^{239}Pu) in a fast-spectrum reactor occurs by neutron irradiation of fertile material (^{238}U), while in the thorium fuel cycle ^{232}Th absorbs a neutron in either a fast or thermal reactor. Next, the ^{232}Th emits an electron and an anti-neutrino by β^- decay to become ^{233}Pa . The protactinium then emits another electron and anti-neutrino by a second β^- decay to become ^{233}U , which in turn is used as fuel. In MSR designs, the ^{233}Pa is extracted and protected from neutrons (to prevent the core's poisoning via the ^{233}Pa transmutation into

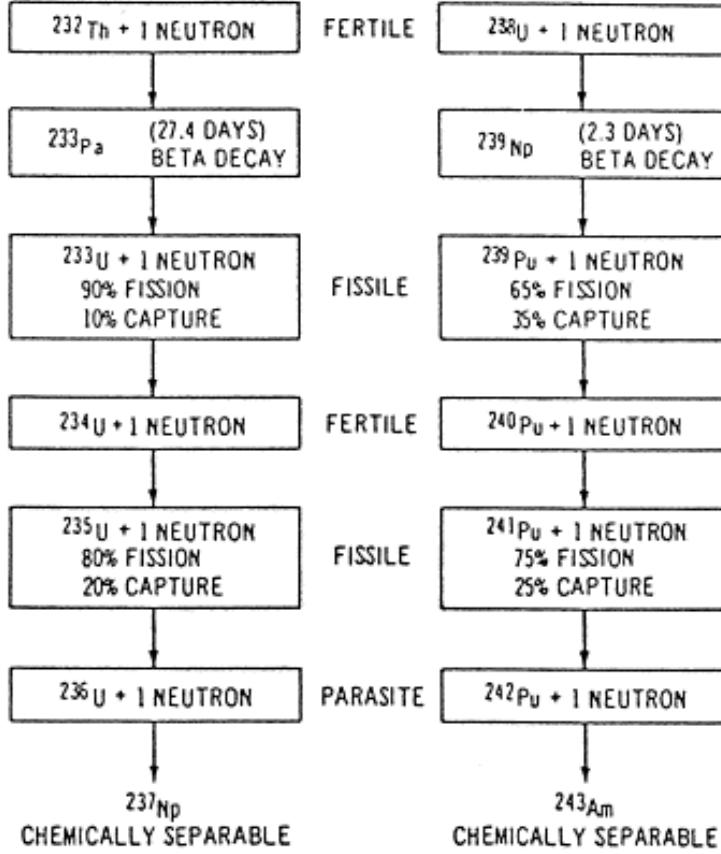


Figure 2.2: Isotopic build-up in ^{232}Th and ^{238}U breeding systems [24].

^{234}Pa and then to ^{234}U), until it has decayed to ^{233}U . Figure 2.2 demonstrates transmutations in the thorium and U-Pu fuel cycles. This is done in order to improve the breeding ratio which is low compared to fast reactors.

Although the thermal neutron fission cross section (σ_f) of the resulting ^{233}U is comparable to ^{235}U and ^{239}Pu , it has a much lower capture cross section (σ_c) than other two fissile isotopes, providing fewer non-fissile neutron absorptions and improving neutron economy. Figure 2.3 shows thermal utilization factor (η) which in ^{233}U is greater than other two over a wide range of energies, including the thermal spectrum. Consequently, thorium fuels can be the basis for a thermal breeder reactor [25], while a breeding reactor in the U-Pu cycle requires a fast neutron spectrum, because, in the thermal spectrum, one neutron absorbed by ^{239}Pu in average produces less than two neutrons.

Another advantage of the thorium fuel cycle is inherent proliferation resistance due to contamination of fissile ^{233}U with ^{232}U in proposed power reactor designs. ^{232}U cannot be chemically separated from ^{233}U and emits high-energy gamma radiation. These high-energy γ -rays are a radiological

hazard, thus, remote handling is necessary for separated uranium and such materials could be passively detected.

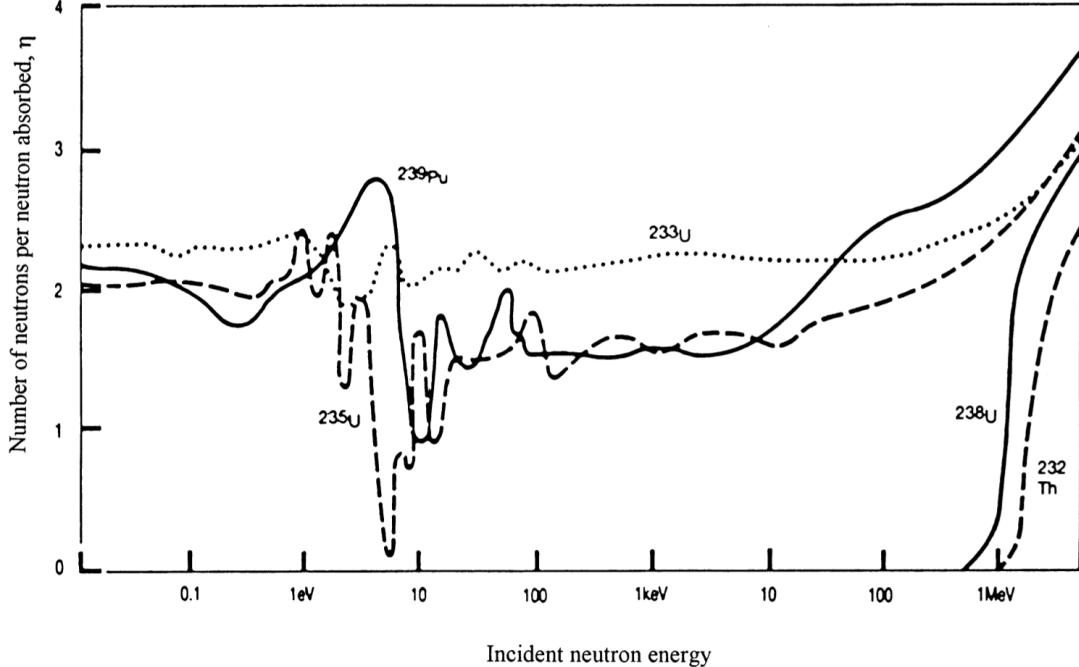


Figure 2.3: Neutron yield per neutron absorbed [26].

Moreover, from the respective positions of uranium and thorium in the periodic table, the long-lived minor actinides resulting from fission are in much lower quantity in the thorium cycle, especially compared with the uranium-plutonium cycle. Because of this, thorium is a potentially attractive alternative to uranium in mixed oxide (MOX) fuels to minimize the generation of long-lived transuranic elements and maximize the destruction of plutonium.

For the many reasons explained above, the thorium fuel cycle has not so far been able to compete on par with uranium, which currently dominates nuclear energy. The time has come to have another hard look at what was perhaps too quickly set aside forty years ago and restart with new advanced computational methods.

2.3 Literature review

While most contemporary nuclear reactor physics software is unable to perform depletion calculations in an online reprocessing regime. Furthermore, no established liquid-fueled MSR tool for neutronics and fuel cycle evaluation

exist, there are codes from universities, research institutions, and internally developed tools for online refueling approximation [27]. The foundation for these tools was based on early MSR programs methods at ORNL, which integrated neutronic and fuel cycle codes [28] into operational plant tools [29] for MSR and reprocessing system designing. More recent research efforts in Europe and Asia mainly focused on fast spectrum reactors fuel cycle analysis and use some external tools to couple neutron transport and depletion codes to take into account continuous feeds and removals in MSRs. Four of these efforts listed in table 2.1.

Table 2.1: Tools and methods for fast spectrum system fuel cycle analysis.

#	Neutronic code	Depletion code	Authors
1	MCNP [30]	REM [31]	Doligez <i>et al.</i> , 2014; Heuer <i>et al.</i> , 2014 [32, 33]
2	ERANOS [34]	ERANOS	Fiorina <i>et al.</i> , 2013 [35]
3	KENO-IV [36]	ORIGEN [37]	Sheu <i>et al.</i> , 2013 [38]
4	SERPENT 2 [12]	SERPENT 2	Aufiero <i>et al.</i> , 2013 [39]

Most of these methods are applicable to thermal spectrum reactors, moreover, additional tools developed specifically for thermal MSR applications listed in table 2.2.

Table 2.2: Tools and approaches for thermal spectrum system fuel cycle analysis.

#	Neutronic code	Depletion code	Authors
5	MICODE [40]	ORIGEN2 [41]	Ahmad <i>et al.</i> , 2015 [42]
6	MCNP6	CINDER90 [43]	Park <i>et al.</i> , 2015; Jeong <i>et al.</i> , 2016 [8, 11]
7	SCALE [44]	SCALE/ ChemTriton [10]	Powers <i>et al.</i> , 2014; Betzler <i>et al.</i> , 2017 [10, 45, 46]
8	SERPENT 2	SERPENT 2	Rykhlevskii <i>et al.</i> , 2017 [47]
9	MCNP	REM	Nuttin <i>et al.</i> [48]

Methods (1,3,4) provide some form of reactivity control, and methods (1,4,5,6,8,9) use inventory of all nuclides in depletion calculations.

Liquid-fueled MSR have online separations and/or feeds, where material is moved to or from the core at all times (continuous) or at specific time steps (batch). To account for batch discards, a depletion tool should have capability to remove fraction or all material at specified interval. This requires the burn-up simulation to stop at a given time and restart with a new liquid fuel composition (after removal of discarded materials and addition fissile/fertile materials). Accounting for a continuous removal or addition is more difficult because it required adding a term to the Bateman equations. In SCALE [44], ORIGEN [37] solves a set of Bateman equations using spectrum-averaged fluxes and cross sections generated from transport calculation. Methods (1,4,8) provide opportunity to work with true continuous feeds and removals, while other methods employed batch-wise approach. ORNL researchers have developed ChemTriton, Python-based script for SCALE/TRITON which uses a semi-continuous batch process to simulate a continuous reprocessing. This tool models salt treatment, separations, discards, and refill using an unit-cell MSR SCALE/TRITON model over small time steps to simulate continuous reprocessing and deplete the fuel salt [10].

Thorium-fueled MSBR-like reactors similar to one in this thesis described in (6,7,8,9). Nevertheless, most of these efforts considered only simplified unit-cell geometry because depletion computations for few year cycle are very computationally expensive even for simple models.

Nuttin *et al.* broke up reactor core geometry into tree MCNP cells: one for salt channels, one for two salt plena above and below the core and the last cell for the annulus, consequently, two-region reactor core was approximated by one region with averaged fuel/moderator ratio [48]. Similar approach was used by Powers *et al.*, Betzler *et al.*, Jeong *et al.* [10, 45, 49, 46, 50, 11] and clearly misrepresent two-region breeder reactor concept. The unit-cell or one-region models may produce reliable results for homogeneous reactor cores (i.e. MSFR, MOSART) or for one-region single-fluid reactor designs (i.e. MSRE). Two-region MSBR must be simulated using whole-core model to represent different neutron transport in inner and outer region of the core, because most of fissions happens in inner region while breeding occurs in outer zone.

Aufiero *et al.* extended Monte Carlo burn-up code SERPENT 2 and employed it to study the material isotopic evolution of the MSFR. The developed extension directly takes into account the effects of online fuel reprocessing

on depletion calculations and features a reactivity control algorithm. The extended version of SERPENT 2 was assessed against a dedicated version of the deterministic ERANOS-based EQL3D procedure [34] and adopted to analyze the MSFR fuel salt isotopic evolution. Rykhlevskii *et al.* employed this extended SERPENT 2 for simplified unit-cell geometry of thermal spectrum thorium-fueled MSBR and obtain contradictory results with existing MSBR depletion simulations [11].

Chapter 2 and 3 of current study most similar to the works described in (6,7,9), but focus on developing new external open-source tool for online reprocessing simulation named Saltproc. The tool works with Monte Carlo code SERPENT 2, has reactivity control module which allows adjust reactivity by changing feed material flow and avoid control rod movement. Moreover, recent research efforts are being extended by using for online reprocessing simulation high-fidelity full-core 3-D model without any approximations in the core geometry.

Another challenge presented by liquid-fueled systems is the fuel materials movement. Fuel flow is important because of delayed neutron emission. In reactor with solid fuel, the fission product delayed neutron precursors remain very close to the location where fission happened, later emitting delayed neutrons at that location. Delayed neutrons has softer energy spectrum than prompt neutrons. In case of liquid-fueled reactors the precursors drifting, consequently, the fission and delayed neutron emmition locations are different. The reactor design determines the effect of the precursor drift on the core physics. The flow parameters (e.g., flow rate, pipe diameter, primary loop length) affect on the effective delayed neutron fraction β_{eff} . This quantity has significant impact on reactor safety because delayed neutron production occurs on a relatively large time frame to allow control a reactor. Hence, to take into account tightly coupled MSR neutronics, thermal-hydraulics and the precursors drift multi-physics code required.

There are number of multi-physics tools developed by many authors which successfully describe steady-state and transient behavior of various MSR concepts. Krepel *et al.* extended the LWR diffusion code DYN3D to consider drift of delayed neutron precursors alongside the reactor temperature profile, re-introducing the extended code as DYN3D-MSR [51]. That work compared DYN3D-MSR against experimental MSRE data to simulate local fuel channel blockage accidents as well as local temperature perturbations.

Similarly, Kophazi *et al.* used iterative coupling between three-dimensional neutronic and one-dimensional heat conduction models DALTON and THERM to analyze normal MSRE operation as well as channel-blocking-incident transients [52]. The Kophazi model added entrance effects of heat transfer coefficients as well as thermal coupling between fuel channels through moderator heat conduction. Later, Cammi *et al.* performed a 2D-axisymmetric single-channel analysis of the MSBR using the commercial finite element package COMSOL Multiphysics [53]. That work directly solved the fuel salt velocity field, used heterogeneous group constants in fuel and moderator regions.

More recently, Aufiero *et al.* [54] have approach to transient simulations in the MSFR within the finite volume OpenFOAM multiphysics toolkit [55]. This approach benefits from pre-implemented turbulence models available in the OpenFOAM library and captures the full-core three-dimensional geometry of the reactor primary circuit. OpenFOAM Computational Fluid Dynamics (CFD) has additionally been shown by Laureau *et al.* [56] to couple well with Transient Fission Matrix neutronics within the MSFR.

Concurrently, Lindsay *et al.* have introduced Moltres, a physics application for multiphysics modeling of liquid-fueled MSRs [57]. It couples equations for neutron diffusion, thermal-hydraulics, and delayed neutron precursor transport. Moltres solves arbitrary-group neutron diffusion, temperature, and precursor governing equations in anywhere from one to three dimensions and can be deployed on an arbitrary number of processing units. That work compared 2D-axisymmetric many-channel analysis of the MSRE in Moltres against experimental MSRE data in steady-state mode.

On the whole, all these research efforts used initial fuel salt composition, consequently, considered reactor core for at the moment of startup. Chapter 4 of the present thesis introduces the steady-state multi-physics analysis of MSBR using Moltres code for both initial fuel composition and for equilibrium fuel composition.

STEADY-STATE FULL-CORE MSBR BENCHMARK

3.1 SERPENT 2 code overview

SERPENT is a continuous-energy Monte Carlo neutronics code capable of solving the neutron transport problem by tracking individual neutrons within the problem geometry and using stochastic method to determine chain of events for each neutron [12]. SERPENT has been under active development at the VTT Technical Research Centre of Finland from 2004, where it was initially conceived as a tool to simplify group constant generation in a high-fidelity Monte Carlo environment. During this period, SERPENT has seen as widely used transport code and number of users grows steadily. Now SERPENT used by more than 500 registered individuals in 155 organisations located in 37 countries around the world. This success is not only a result of the simple and naive cross section generation procedures, but also its high-performance parallelization and user-friendly usage. The burnup calculation capability in SERPENT was established early on, and is fully based on built-in calculation routines, without using any external solvers. A restart features allows performing fuel shuffling or applying any modifications in the input by dividing the calculation into several parts which is crucial for online reprocessing simulations.

Latest version, SERPENT 2, supports advanced geometry types and has advanced burnup capabilities, including online refueling capabilities which are necessarily for neutronic computations of pebble-bed reactors and liquid-fueled MSRs [39]. Furthermore, recently was demonstrated multi-physics simulations using SERPENT 2, i.e. coupled calculations with thermal hydraulics, CFD and fuel performance codes [58]. Two-way coupling to thermal hydraulics, CFD and fuel performance codes has been a major topic in Serpent development for the past several years and operate on two levels:

internal coupling to built-in solvers for fuel behavior and thermal hydraulics and external coupling via a universal multi-physics interface.

SERPENT 2 can be effectively run in parallel on computer clusters and multi-core workstations. Parallelization at core level is handled by thread-based OpenMP, which has the advantage that all processors use shared memory space. Calculations can be divided into several nodes by distributed-memory MPI parallelization. SERPENT 2 is an improvement upon SERPENT 1, and contains a complete redesign of memory management using hybrid OpenMP [59] + Message Passing Interface (MPI) parallelization. This hybrid parallelization is important in depletion calculations using computer clusters with multiple cores, and allows to achieve significant speed-up in depletion calculations on computer clusters with more than 4'000 cores [60]. In addition to the particle transport simulation, parallelization in the burnup calculation mode divides also the preprocessing and depletion routines between several cores.

All calculations presented in this thesis were performed using SERPENT 2 version 2.1.28 on Blue Waters XK7 nodes. For cross section generation, JEFF-3.2 was employed [61].

3.2 Molten Salt Breeder Reactor description

The MSBR vessel has diameter of 680 cm and a height of 610 cm. It contains a molten fluoride fuel-salt mixture that generates heat in the active core region and transports that heat to the primary heat exchanger by way of the primary salt pump. In the active core region, the salt flows through channels in moderating and reflecting graphite blocks. Salt at about 565°C enters the central manifold at the bottom via four 40.64-cm-diameter nozzles and flows through the lower plenum and upward via the channels in the graphite to exit at the top at about 704°C through four equally spaced nozzles which connect to the salt-suction pipes leading to primary circulation pumps. The fuel salt drain lines connects to the bottom of the reactor vessel inlet manifold.

Reactor graphite experiences significant dimensional changes due to neutron irradiation, consequently, the reactor core was designed for periodic replacement. The reference MSBR design has an average core power density of about 6.666 W/g, which, based in the irradiation behavior of materials

obtained from MSRE, allows to achieve useful core graphite life of about 4 years and reflector graphite life during 30-year lifetime of plant [7].

Moreover, it was decided to remove and install the core graphite as an assembly rather than by individual blocks, because it relatively quickly, easier for maintenance personnel and has lower probability of radioactive elements escape. In addition, handling the core as an assembly also allows the replacement core to be carefully preassembled and tested under factory conditions.

The core has two radial zones bounded by a solid cylindrical graphite reflector and the vessel wall. The MSBR core consists of two different zones. The central zone, zone I, in which 13% of the volume is fuel salt and 87% graphite. zone I composed of 1320 graphite cells, 2 graphite control rods, and 2 safety rods consisting of boron carbide clad. The under-moderated zone, zone II, with 37% fuel salt, and radial reflector, surrounds the zone I core region and serves to diminish neutron leakage. zones I and II are surrounded radially and axially by fuel salt. This space for fuel is necessary for injection and flow of molten salt. Fig. 3.1 and 3.2 demonstrate MSBR vessel, core configuration, “fission” (zone I) and “breeding” (zone II) regions position inside the vessel.

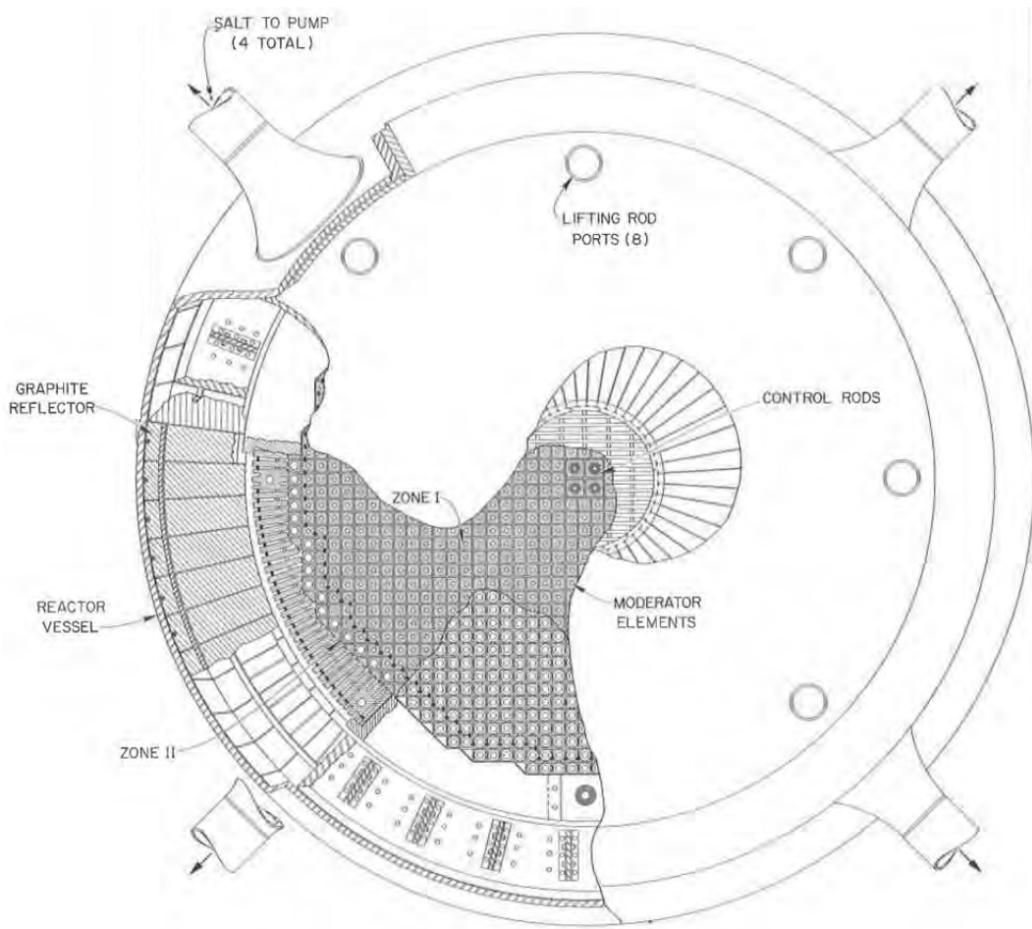


Figure 3.1: Plan view of MSBR vessel [7].

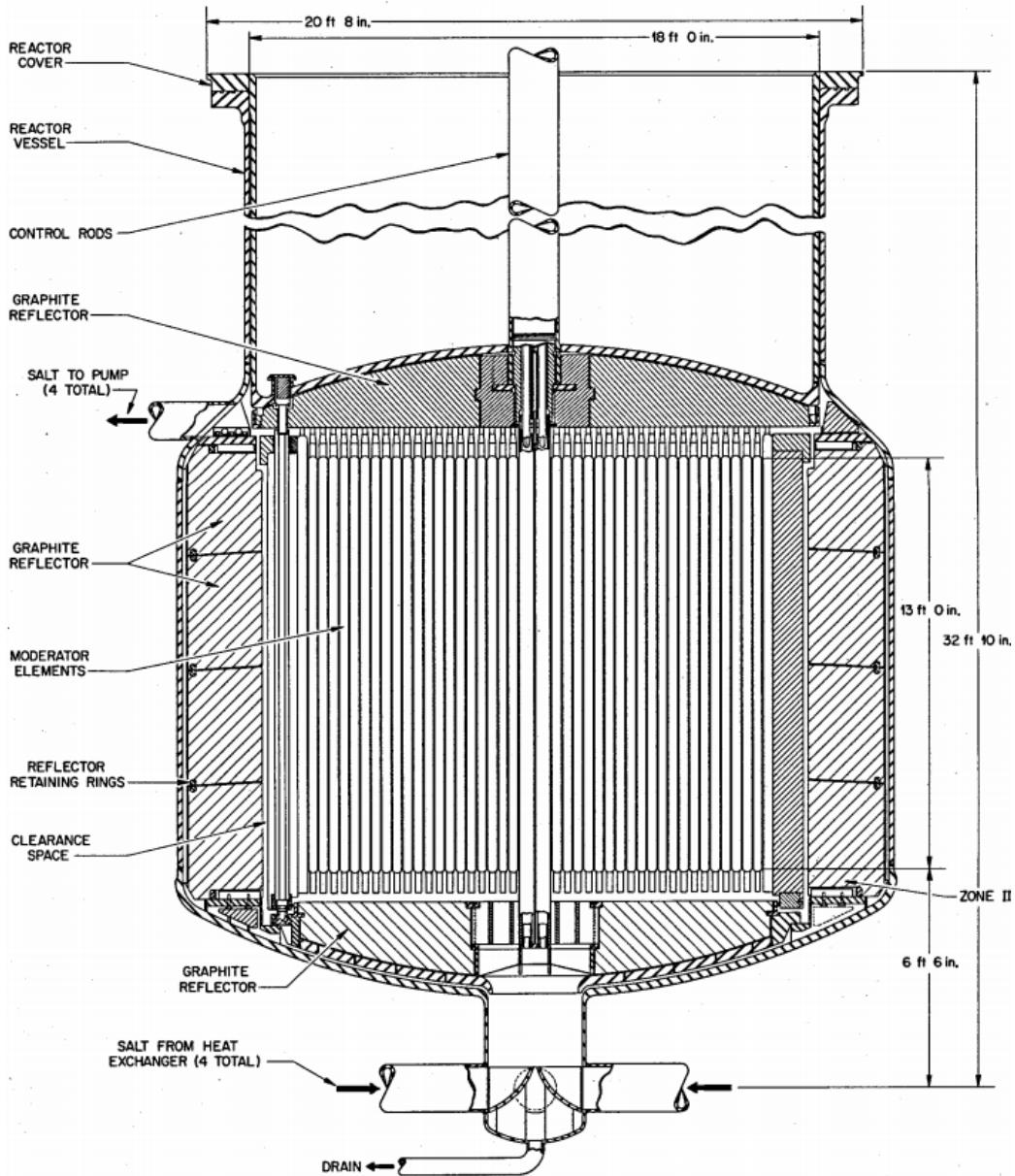


Figure 3.2: Sectional elevation of MSBR vessel [7].

There are eight graphite slabs with a width of 15.24 cm in zone II one to each other, one of which is illustrated in Fig. 3.3. The holes in the centers are for the core lifting rods used during the core replacement operations. These holes also allow a portion of the fuel salt to flow to the top of the vessel for cooling the top head and axial reflector. Fig. 3.3 also demonstrates the 5.08-cm-wide annular space between the removable core graphite in zone II-B and the permanently mounted reflector graphite. This annulus, 100% consists of fuel salt, provides space for moving the core assembly, helps compensate the out-of-roundness dimensions of the reactor vessel, and serves to reduce

the damage flux at the surface of the graphite reflector blocks.

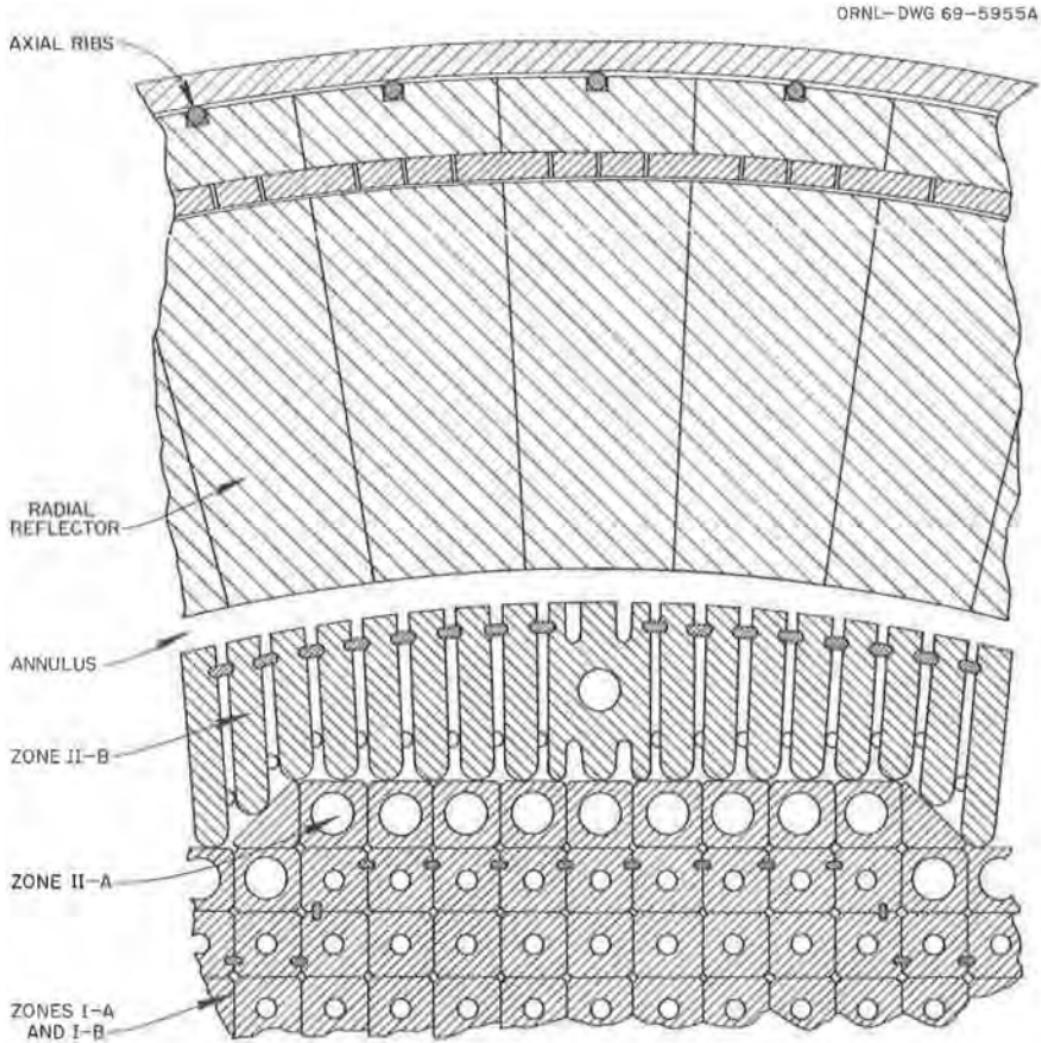


Figure 3.3: Detailed plan view of graphite reflector and moderator elements [7].

3.2.1 Core zone I

The central region of the core, called zone I, is made up of graphite elements, each $10.16\text{cm} \times 10.16\text{cm} \times 396.24\text{cm}$. In zone I, 13% of the volume is fuel salt and 87% is graphite. Zone I is composed of 1'320 graphite cells and 4 channels for control rods: two for graphite rods which both regulate and shim during normal operation, and two for backup safety rods to assure sufficient negative reactivity for emergency situations.

These graphite elements have a mostly rectangular shape with lengthwise ridges at each corner that leave space for salt flow elements. Various element sizes reduce the peak damage flux and power density in the center of the core prevent local graphite damage. Zone I is well-moderated which is necessarily to achieve desired fission power density. Figure 3.4 demonstrates the elevation and sectional views of graphite elements of zone I [7] and these elements SERPENT model [62].

3.2.2 Core zone II

The undermoderated zone, zone II, surrounds zone I. Combined with the bounding radial reflector, zone II serves to diminish neutron leakage. This zone is formed of two kinds of elements: elements like those in zone I with a larger channel diameter (zone II-A), and radial graphite slats (zone II-B).

Zone II has 37% salt by volume and each element has a fuel channel diameter of 6.604cm. It is divided into two different zones: zone II-A and zone II-B. The graphite elements for zone II-A are prismatic and have elliptical-shaped dowels running axially between the prisms and needed to isolate the fuel salt flow in zone I from that in zone II. Fig. 3.5 shows shape and dimensions of these graphite elements and SERPENT model. Zone II-B elements are rectangular slats spaced far enough apart to provide the 0.37 fuel salt volume fraction. The reactor zone II-B graphite 5.08cm-thick slats vary in the radial dimension (average width is 26.67cm) as shown in figure 3.3. Zone II serves as “blanket” to achieve the best “performance” associated with a high breeding ratio and a low fissile inventory. The neutron energy spectrum in zone II is made harder, to enhance the rate of thorium resonance capture relative to the fission rate, thus limiting the neutron flux in the outer core zone and reducing the neutron leakage [7].

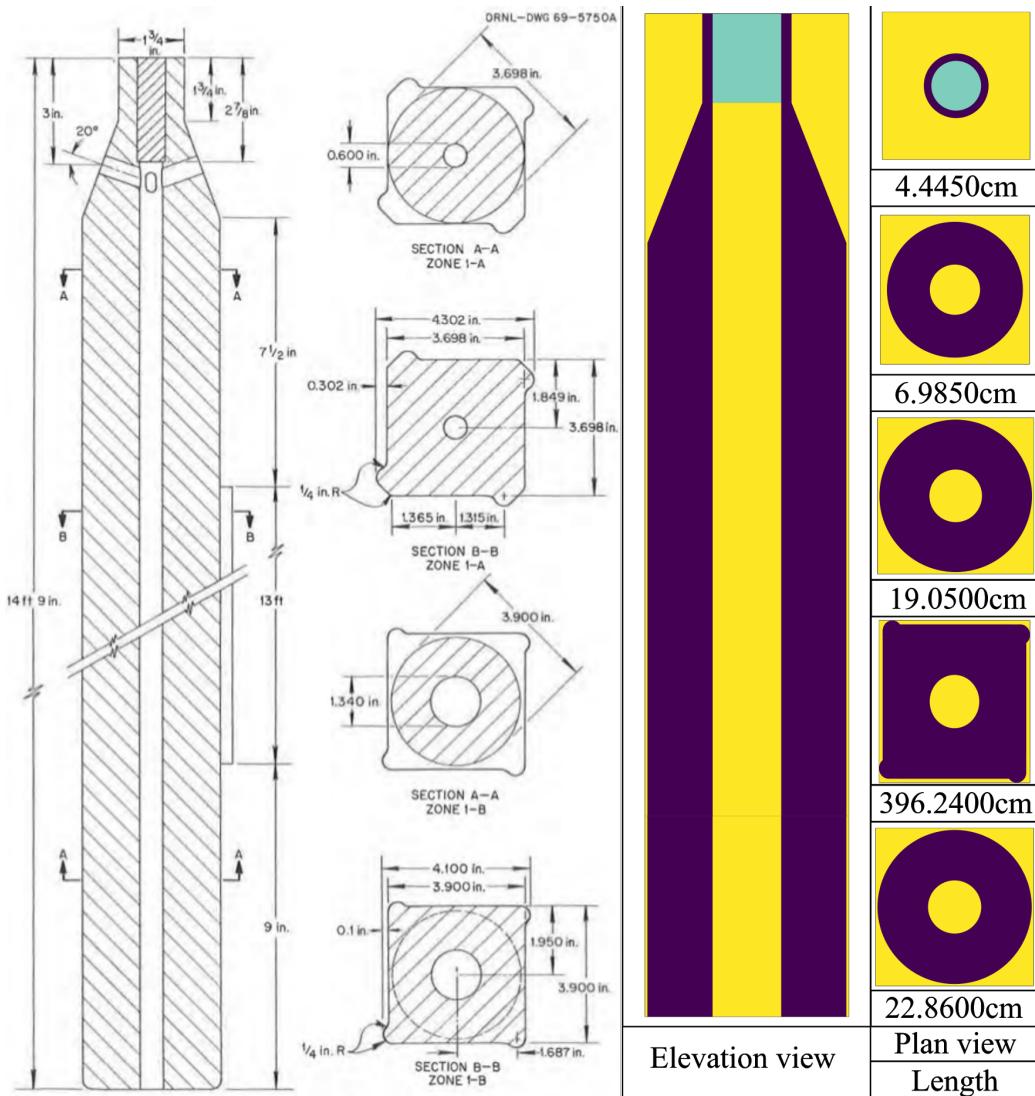


Figure 3.4: Graphite moderator elements for zone I [7, 62].

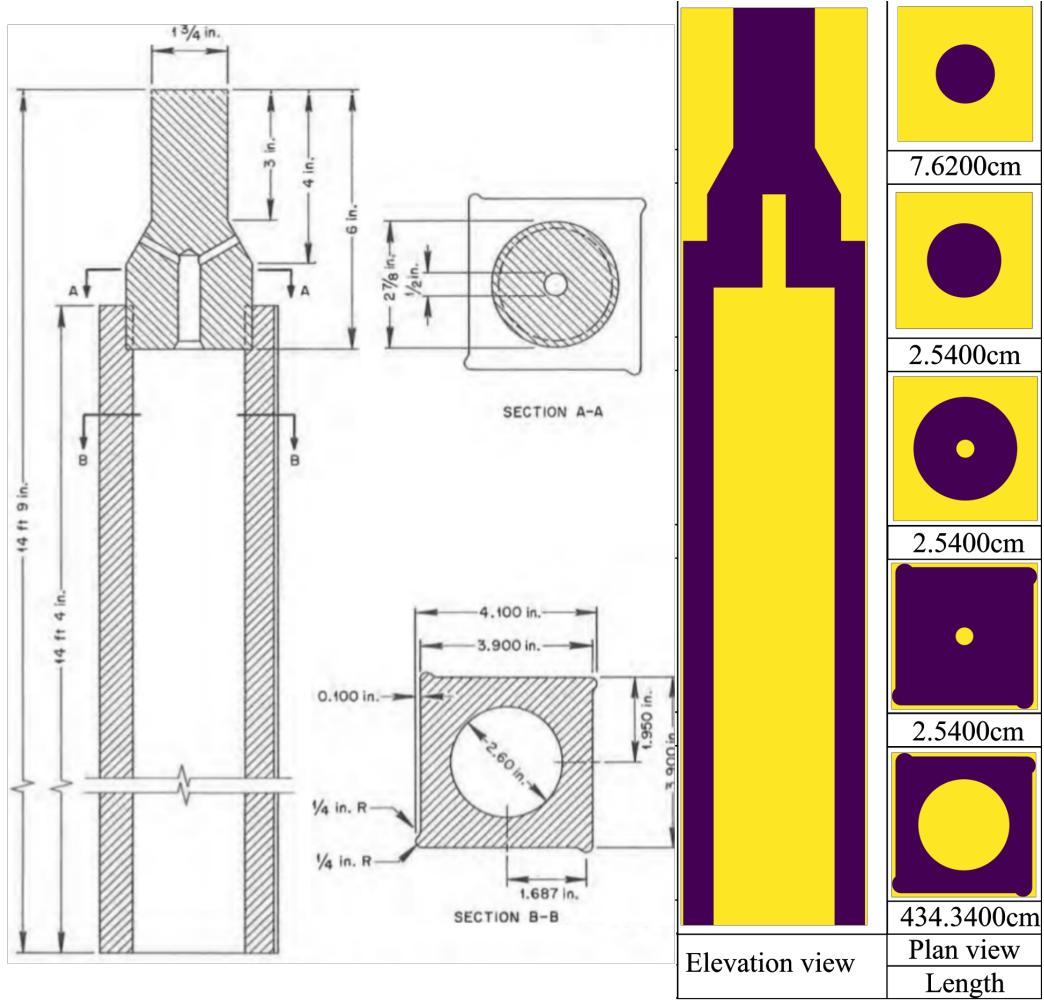


Figure 3.5: Graphite moderator elements for zone II-A [7, 62].

3.3 SERPENT 2 model

To represent complex irregular MSBR core geometry advanced geometry surfaces in SERPENT was employed. Fig. 3.6 shows the plan view of the whole-core configuration at the expected reactor operational level when both graphite control rods are fully inserted, and the safety rods are fully withdrawn. The safety rods only get inserted during an accident and was not considered in steady state simulation.

Fig. 3.7 shows the longitudinal section of the reactor. The violet color represents bare graphite, and the yellow represents fuel salt. The blue color shows Hastelloy-N, a material used for the plenum and vessel wall, and the

white color is a void space. The model contains about 2000 geometry surfaces and 2066 calculation zones. In this thesis, all figures of the core were generated using the built-in SERPENT plotter.

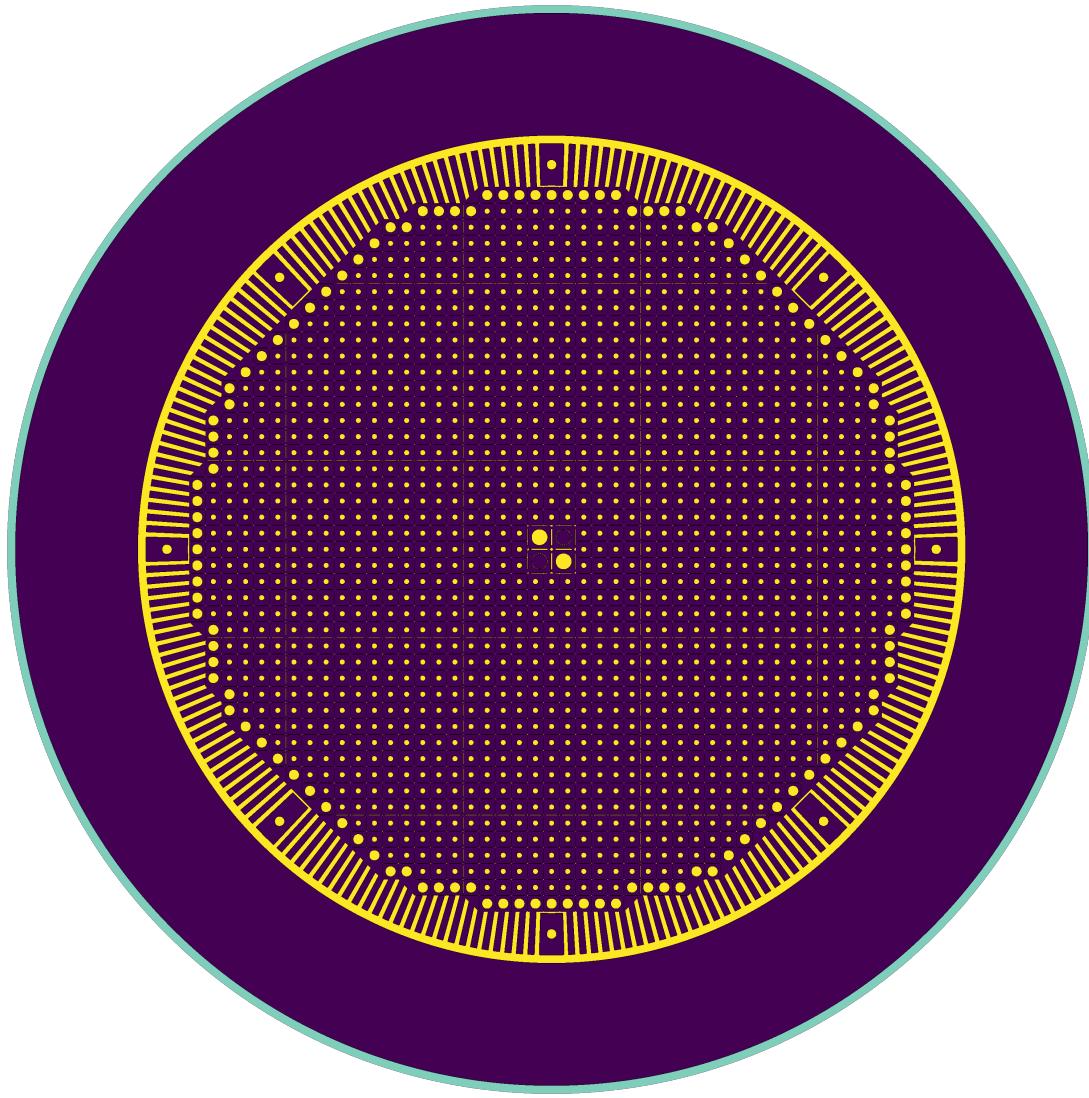


Figure 3.6: Plan view of MSBR model.

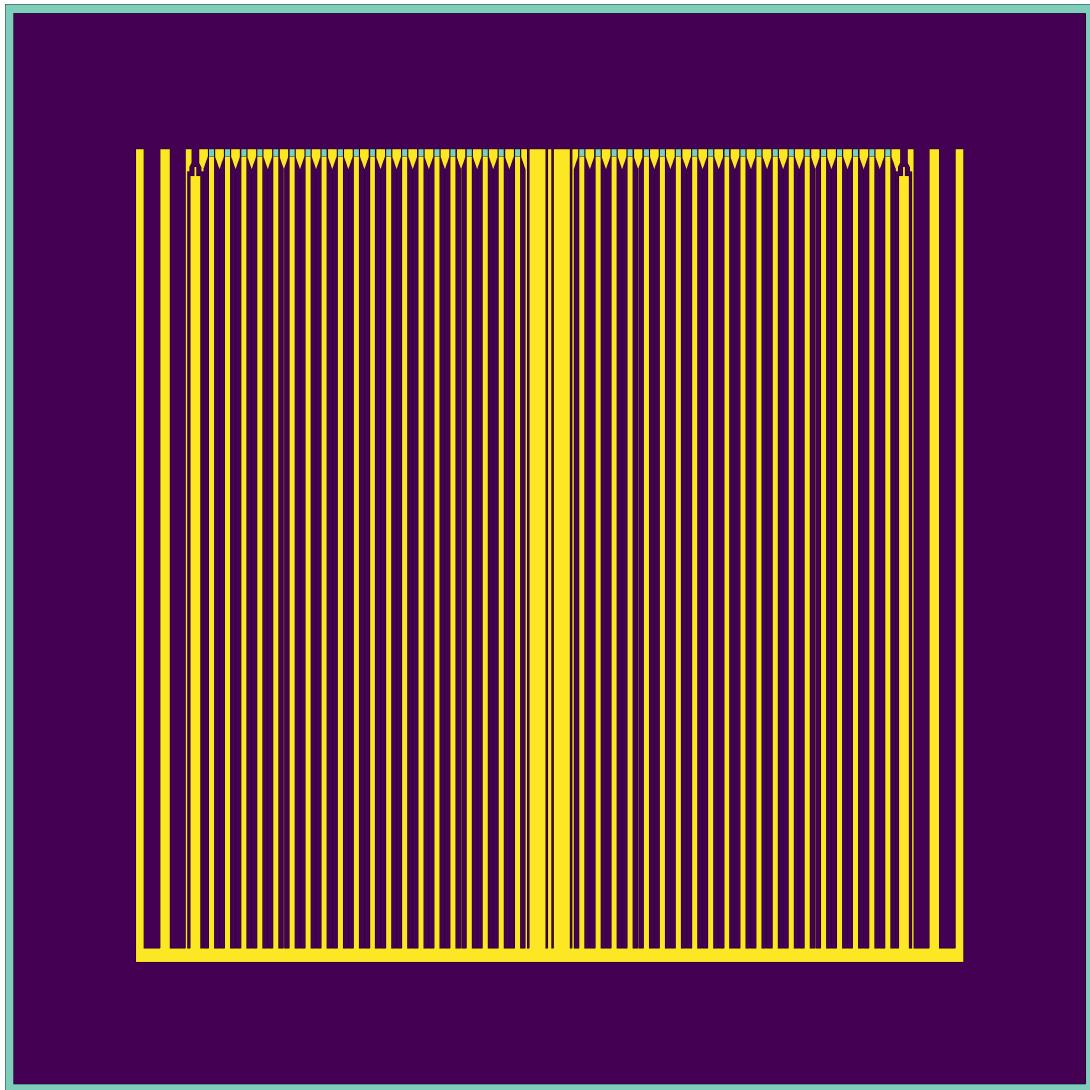


Figure 3.7: Elevation view of MSBR model.

In the model, zone I, zone II-A graphite blocks was described using circular cylinder and square cylinder surface types, lengthwise ridges at each corner mentioned earlier was specified using dodecagonal cylinder surfaces and general planes. Zone I of the core was described using square lattice inscribed in the octagonal cylinder surfaces to accurately represent geometry of that region.

The main challenge was accurately represent zone II-B because it has irregular elements with sophisticated shape. From the ORNL report [7], the suggested design of zone II-B has 8 irregularly-shaped graphite elements every 45° as well as salt channels (figure 3.3). These graphite elements were simplified into right-circular cylindrical shapes with central channels. Fig. 3.8 illustrates this core region in SERPENT model. Volume of fuel salt in zone II kept exactly 37%, consequently, this simplification did not considerably change neutronics of the core. This is the only simplification made to the MSBR conceptual geometry in this work.

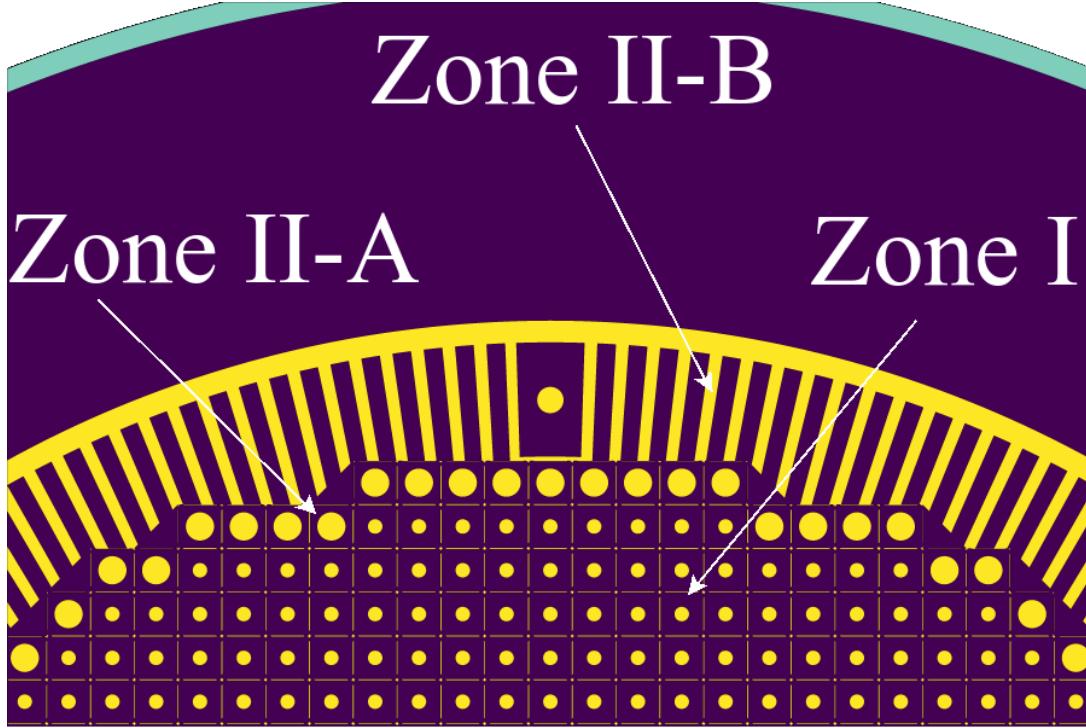


Figure 3.8: Detailed view of MSBR zone II model.

3.3.1 Material composition and normalization parameters

The fuel salt, the reactor graphite, and the modified Hastelloy-N are materials unique of the MSBR and were created at ORNL. The initial fuel salt used the same density (3.35g/cm^3) and composition $\text{LiF-BeF}_2\text{-ThF}_4\text{-}^{233}\text{UF}_4$ (71.8-16-12-0.2 mole %) as the MSBR design[7]. The lithium in the molten salt fuel is a fully enriched ^7Li because ^6Li is a very strong neutron poison and becomes tritium upon neutron capture.

For cross section generation, JEFF-3.2 was employed [61]. The specific temperature was fixed for each material to correctly model the Doppler-broadening of resonance peaks when Serpent generate problem-oriented nuclear data library. The isotope composition of each material at the initial state was described in detail in the MSBR conceptual design study [7] and has been applied to Serpent model without any modification. Table 3.1 is the summary of the major MSBR parameters used by this model [7].

Table 3.1: Summary of principal data for MSBR.

Thermal capacity of reactor	2250 MW(t)
Net electrical output	1000 MW(e)
Net thermal efficiency	44.4%
Salt volume fraction in central core zone	0.13
Salt volume fraction in outer core zone	0.37
Fuel salt inventory (Zone I)	8.2 m^3
Fuel salt inventory (Zone II)	10.8 m^3
Fuel salt inventory (annulus)	3.8 m^3
Total fuel salt inventory	48.7 m^3
Fissile mass in fuel salt	1303.7 kg
Fuel salt components	$\text{LiF-BeF}_2\text{-ThF}_4\text{-}^{233}\text{UF}_4$
Fuel salt composition	71.9-16-12-0.2 mole%

4

ONLINE REPROCESSING SIMULATION

4.1 Online reprocessing method

4.2 Python code description

4.3 Results and equilibrium state analysis

RESULTS AND DISCUSSION

5.1 Results

This section presents calculation results, such as the effective multiplication factor for whole core, neutron flux spectrum, temperature reactivity coefficients, and compares the results against those obtained by Park *et al.* using Monte Carlo MCNP6 [8].

Figure 5.1 illustrates Park *et al.* model which ignores lengthwise graphite ridges at each corner of each graphite element in zone I, II-A, and significantly simplifies in zone II-B and annulus. The normalized neutron flux distribution is calculated for the whole core using continuous-energy nuclear data. The temperature coefficients for both fuel salt solution and reactor graphite are computed by comparing effective multiplication factors for two temperatures in the working range.

5.1.1 Neutron spectrum

Fig. 5.2 demonstrates the normalized neutron flux spectrum for the whole core in the energy range from 10^{-9} to 10 MeV. The results show close fit with the MCNP simulation [8], especially in thermal energy range. It is important to obtain the epithermal and thermal spectrum to produce ^{233}U from ^{232}Th because radiative capture cross section of thorium monotonically decreases from 10^{-10} MeV to 10^{-5} MeV. Hardening the spectrum tends to significantly increase resonance absorption in thorium and decrease the absorptions in fissile and construction materials. Thus, a significant amount fissile material will be needed to make the reactor critical.

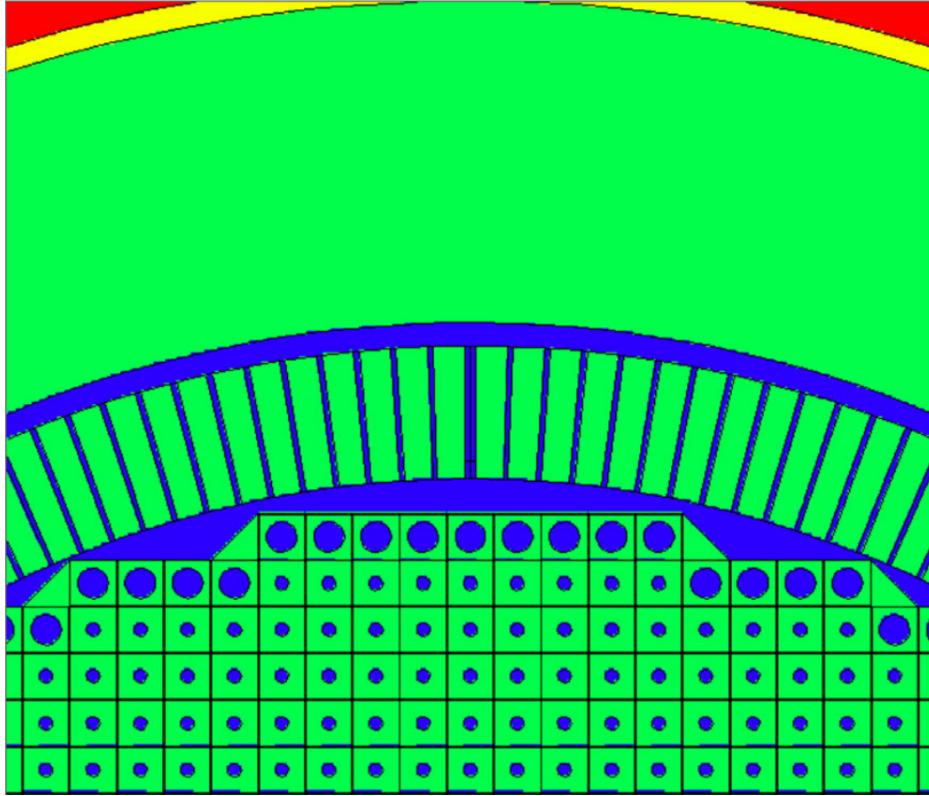


Figure 5.1: Detailed plan view of Park (MCNP6) model [8].

5.1.2 Effective multiplication factor

Table 5.1 shows the effective multiplication factor for both MCNP6 and Serpent 2 whole core models. The factor obtained using Serpent 2 is 300 pcm lower than that obtained by Park *et al.* using MCNP6 [8]. Standard deviations are 5 and 9 pcm, respectively. The discrepancy is likely due to simplifications to the Zone II geometry model used in Park *et al.*

TABLE 5.1: Effective multiplication factor of whole core model.

	Serpent2	MCNP6 [8]
K_{eff}	1.00389 ± 0.00005	1.00736 ± 0.00009

5.1.3 Temperature effect of reactivity

Table 5.2 shows temperature effects on reactivity calculated in this work as compared to both [8] and [7]. Uncertainty for each temperature coefficient also appears in Table 5.2. The main physical principle underlying the reactor

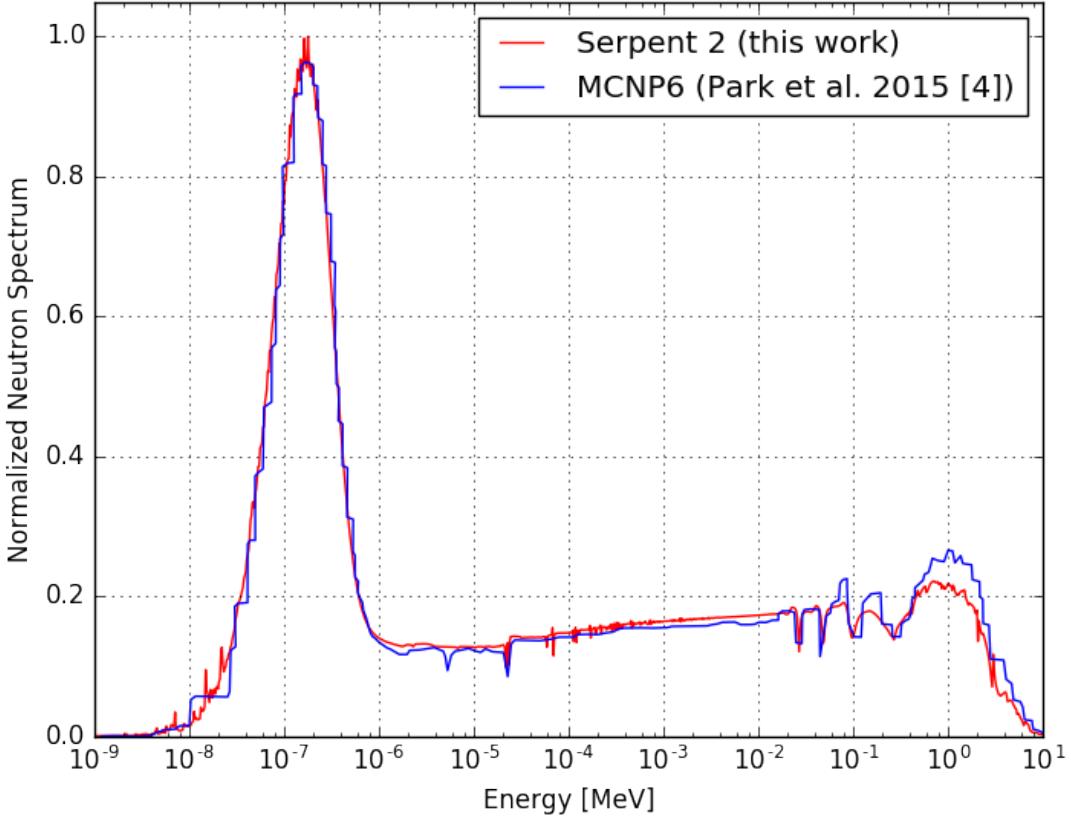


Figure 5.2: Neutron flux spectrum of MSBR for MCNP6 and SERPENT model.

temperature feedback is an expansion of matter when it is heated. When the fuel salt temperature increases, the density of the salt decreases, but at the same time, the total volume of fuel salt in the core remains constant because it is bounded by the graphite. When the reactor graphite temperature grows, the density of graphite declines creating additional space for fuel salt. To determine temperature coefficients, the cross-section temperatures for fuel and moderator were changed from 900K to 1200K. Three different cases were considered:

1. Temperature of fuel salt rising from 900K to 1200K.
2. Temperature of graphite rising from 900K to 1200K.
3. Whole reactor temperature rising from 900K to 1200K.

In the first case, changes in the fuel temperature only impact fuel density. In this case, the geometry is unchanged because fuel is a liquid. However,

TABLE 5.2: Temperature coefficients of reactivity.

Reactivity coefficient [pcm/K]	Serpent2	MCNP6 [8]	Reference [7]
Fuel salt	-3.70 ± 0.016	-3.20 ± 0.05	-3.22
Moderator	$+2.33 \pm 0.027$	-0.11 ± 0.05	+2.35
Total	-1.57 ± 0.033	-3.21 ± 0.04	-0.87

when the moderator heats up, both the density and the geometry change due to thermal expansion of the solid graphite blocks and reflector. Accordingly, the new graphite density was calculated using a linear temperature expansion coefficient of $1.3 \times 10^{-6} \text{ }^{\circ}\text{C}/\text{K}$ [7]. A new geometry input was created based on this information.

The fuel temperature coefficient (FTC) is negative due to thermal Doppler broadening of the resonance capture cross sections in the thorium and is in a good agreement with early research [7, 8]. The moderator temperature coefficient is positive due to changing density and would increase during reactor operation because of spectrum hardening along with fuel depletion [8]. Finally, the total temperature coefficient of reactivity is relatively large and negative, despite graphite components, and affords excellent reactor stability and controllability.

DISCUSSION AND CONCLUSION

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