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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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# MOLTEN SALT REACTORS

## 1.1 History

Molten Salt Reactor (MSR) development started in the late 1940's as part of the United States' program to design a nuclear powered airplane [1]. 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 [2].

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, ZrF<sub>4</sub>, and UF<sub>4</sub>. 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<sub>(th)</sub>. No mechanical or chemical problems were observed, and the reactor was found to be stable and self-regulating [1].

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 [2]. Breeding  $^{233}\text{U}$  from  $^{232}\text{Th}$  was found to give better performance in a molten salt thermal reactor neutron energy spectrum than a uranium fuel cycle in which depleted uranium ( $^{238}\text{U}$ ) is the fertile material and fissile  $^{239}\text{Pu}$  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 [2]. 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 [3]. Acceptable power densities appeared challenging to achieve without using novel and untested heat transfer technologies [2].

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  $^{232}\text{Th}$  is separated from the fissile salt which contains  $^{233}\text{U}$  and/or  $^{239}\text{Pu}$  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 [4], 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 Oak Ridge National Laboratory (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" [5].

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 [2]. 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,  $^7\text{Li}$ , 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  $8\text{MW}_{th}$ , and the heat is dissipated to the atmosphere [6].

Construction of the MSRE began in 1962, and the reactor first became critical in 1965. Figure 1.1 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.  $^{233}\text{U}$  fuel was added to the same carrier salt, and on October 2 the MSRE began operation using  $^{233}\text{U}$ . 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  $^{233}\text{U}$  [6].



Figure 1.1: The MSRE core, shown while being assembled, contains about  $1.95 \text{ m}^3$  of reactor graphite. The 1'140 fuel channels contain about  $0.57\text{m}^3$  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  $1000^\circ\text{C}$  [7]. 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 [2]. 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 [8]. The main drawback of those reactors was identified as graphite pipes damaging by very high neutron fluxes. Figure 1.2 demonstrates design of two-fluid Molten Salt Breeder Reactor (MSBR) single cell.

Later, in 1967, new experimental information obtained from MSRE and an advance in core design led 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 [2].

To achieve acceptable breeding ratio in single-fluid reactor,  $^{233}\text{Pa}$  (27.4-day half-life) must be separated from the fuel salt and held outside the core until it decays to  $^{233}\text{U}$ . 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.

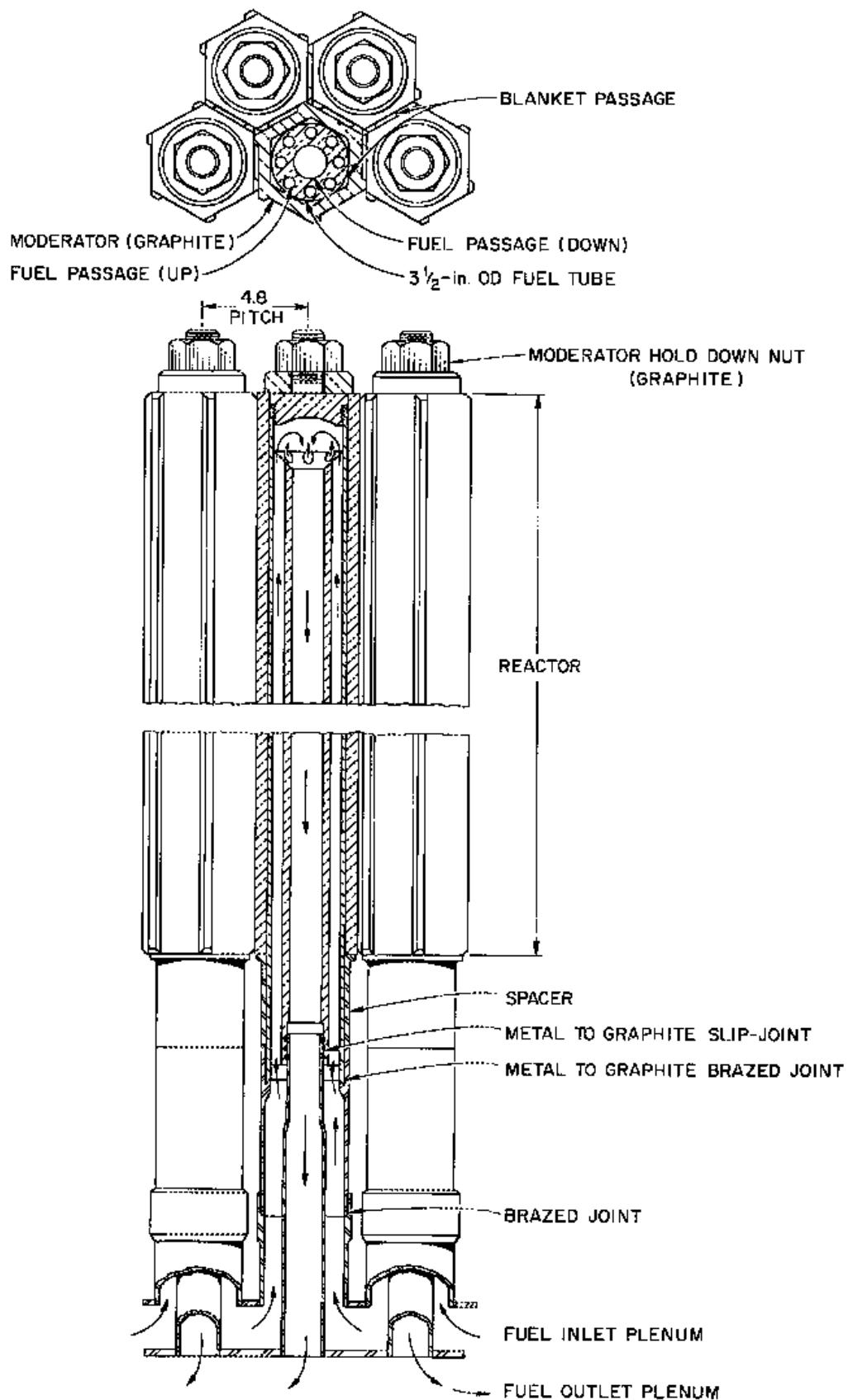


Figure 1.2: A single graphite “fuel cell” for a molten-salt breeder reactor. Fuel salt flowed upward from the entrance plenum through eight channels at 45-degree angles to one another, then downward through the central channel to the exit plenum [9].

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 [2]. 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),[10] 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.

## 1.2 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  $^{233}\text{U}$  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 [11].

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  $^{233}\text{U}$  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 [11]. 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 [12]. In the twenty-first century thorium's potential for improving proliferation resistance and waste characteristics is generating renewed interest in the thorium fuel cycle [13].

Compared to natural uranium which contains 99.284%  $^{238}\text{U}$ , thorium almost exclusively composed of  $^{232}\text{Th}$ . It can be seen from figure 1.3 that the fertile isotopes are  $^{238}\text{U}$  and  $^{232}\text{Th}$  for uranium-plutonium and thorium fuel cycle, respectively. Accordingly, the fissile isotopes are  $^{235}\text{U}$ , 0.711% of natural uranium present in nature, and the artificial fissile isotopes are  $^{239}\text{Pu}$  and  $^{233}\text{U}$  for U-Pu and thorium cycles, respectively.

In the Uranium-Plutonium cycle, production of fissile material ( $^{239}\text{Pu}$ ) in a fast-spectrum reactor occurs by neutron irradiation of fertile material ( $^{238}\text{U}$ ), while in the thorium fuel cycle  $^{232}\text{Th}$  absorbs a neutron in either a fast or thermal reactor. Next, the  $^{233}\text{Th}$  emits an electron and an anti-neutrino by

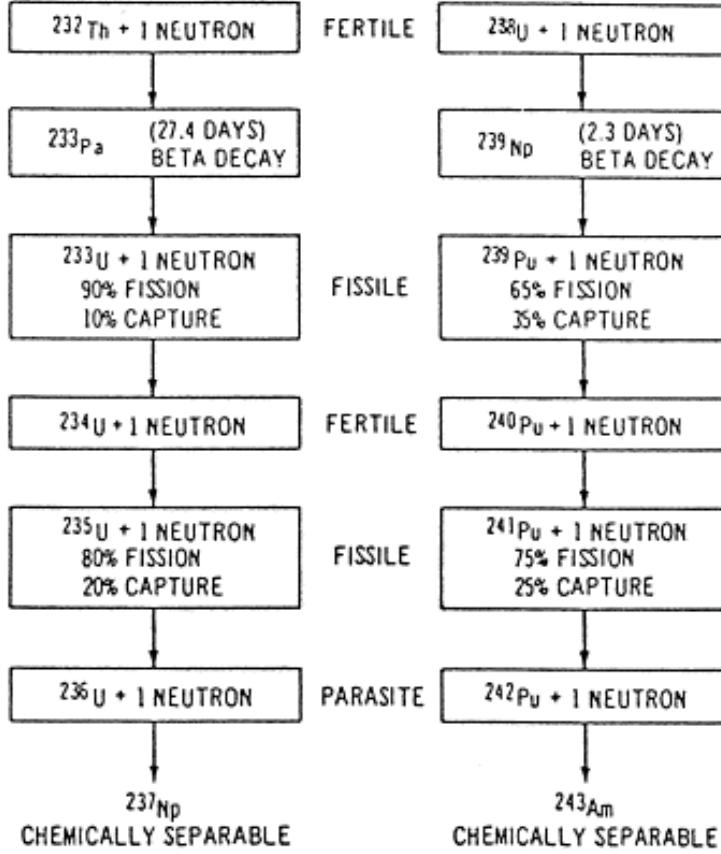


Figure 1.3: Isotopic build-up in  $^{232}\text{Th}$  and  $^{238}\text{U}$  breeding systems [14].

$\beta^-$  decay to become  $^{233}\text{Pa}$ . The protactinium then emits another electron and anti-neutrino by a second  $\beta^-$  decay to become  $^{233}\text{U}$ , which in turn is used as fuel. In MSR designs, the  $^{233}\text{Pa}$  is extracted and protected from neutrons (to prevent the core's poisoning via the  $^{233}\text{Pa}$  transmutation into  $^{234}\text{Pa}$  and then to  $^{234}\text{U}$ ), until it has decayed to  $^{233}\text{U}$ . Figure 1.3 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 ( $\sigma_f$ ) of the resulting  $^{233}\text{U}$  is comparable to  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , it has a much lower capture cross section ( $\sigma_c$ ) than other two fissile isotopes, providing fewer non-fissile neutron absorptions and improving neutron economy. Figure 1.4 shows thermal utilization factor ( $\eta$ ) which in  $^{233}\text{U}$  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 [15], while a breeding reactor in the U-Pu cycle requires a fast neutron spectrum, because, in the thermal spectrum, one neutron absorbed by  $^{239}\text{Pu}$  in average produces less than two neutrons.

Another advantage of the thorium fuel cycle is inherent proliferation resistance due to contamination of fissile  $^{233}\text{U}$  with  $^{232}\text{U}$  in proposed power reactor designs.  $^{232}\text{U}$  cannot be chemically separated from  $^{233}\text{U}$  and emits high-energy gamma radiation. These high-energy  $\gamma$ -rays are a radiological hazard, thus, remote handling is necessary for separated uranium and such materials could be passively detected.

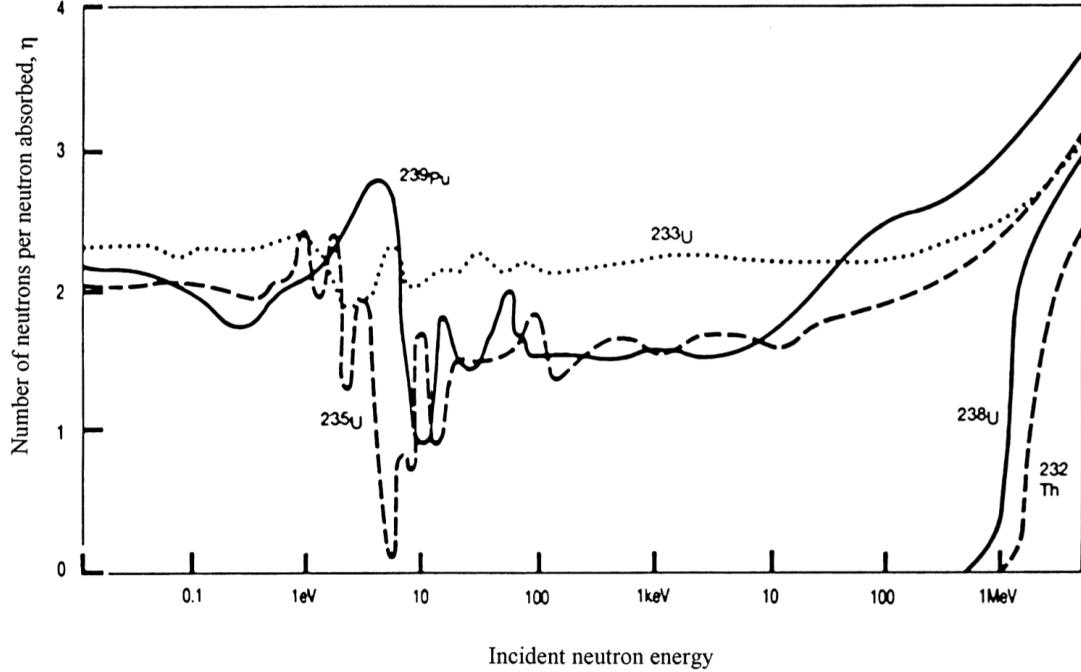


Figure 1.4: Neutron yield per neutron absorbed [16].

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.

### 1.3 Literature review

While there is most contemporary nuclear reactor physics software is unable to perform depletion calculations in an online reprocessing regime. Furthermore, no established tool for liquid-fueled MSR neutronics and fuel cycle evaluation exist, there are internally developed tools from universities, research institutions for online refueling approximation [17]. The foundation for these tools was based on early MSR simulation methods at ORNL, which integrated neutronic and fuel cycle codes [18] into operational plant tools [19] for MSR and reprocessing system design. More recent research efforts in Europe and Asia mainly focus 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 are listed in table 1.1.

Table 1.1: Tools and methods for fast spectrum system fuel cycle analysis.

#	Neutronic code	Depletion code	Authors
1	Monte Carlo N-Particle code (MCNP) [20]	REM [21]	Doligez <i>et al.</i> , 2014; Heuer <i>et al.</i> , 2014 [22, 23]
2	ERANOS [24]	ERANOS	Fiorina <i>et al.</i> , 2013 [25]
3	KENO-IV [26]	ORIGEN [27]	Sheu <i>et al.</i> , 2013 [28]
4	SERPENT 2 [29]	SERPENT 2	Aufiero <i>et al.</i> , 2013 [30]

Most of these methods are applicable to thermal spectrum reactors, and, additional tools developed specifically for thermal MSR applications are listed in table 1.2.

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

Liquid-fueled MSR designs 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 discharge, a depletion tool should have the capability to remove some 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 of fissile/fertile materials). Accounting for a continuous removal or addition

Table 1.2: Tools and approaches for thermal spectrum system fuel cycle analysis.

#	Neutronic code	Depletion code	Authors
5	MICODE [31]	ORIGEN2 [32]	Ahmad <i>et al.</i> , 2015 [33]
6	MCNP6	CINDER90 [34]	Park <i>et al.</i> , 2015; Jeong <i>et al.</i> , 2016 [35, 36]
7	SCALE [37]	SCALE/ ChemTriton [38]	Powers <i>et al.</i> , 2014; Betzler <i>et al.</i> , 2017 [38, 39, 40]
8	SERPENT 2	SERPENT 2	Rykhlevskii <i>et al.</i> , 2017 [41]
9	MCNP	REM	Nuttin <i>et al.</i> [42]

is more difficult because it requires adding a term to the Bateman equations. In SCALE [37], ORIGEN [27] solves a set of Bateman equations using spectrum-averaged fluxes and cross sections generated from a deterministic 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, discharge, and refill using an unit-cell MSR SCALE/TRITON model over small time steps to simulate continuous reprocessing and deplete the fuel salt [38].

Thorium-fueled MSBR-like reactors similar to the one in this thesis are 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 [42]. A similar approach was used by Powers *et al.*, Betzler *et al.*, and Jeong *et al.* [38, 39, 43, 40, 44, 36] and clearly misrepresent the 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). A two-region MSBR must be simulated using a whole-

core model to represent different neutron transport in the inner and outer regions of the core, because most fissions happens in the 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 [24] and adopted to analyze the MSFR fuel salt isotopic evolution. We employed this extended SERPENT 2 for a simplified unit-cell geometry of thermal spectrum thorium-fueled MSBR and obtained results which contradict existing MSBR depletion simulations [36].

Chapter 2 and 3 of current study are mostly similar to the works described in (6,7,9), but the focus of this work is on developing new external open-source tool for online reprocessing simulation named Saltproc. The tool works with Monte Carlo code SERPENT 2, and has a reactivity control module which allows reactivity adjustment by changing feed material flow to avoid control rod movement. Moreover, this work extends recent research efforts 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 material movement. Fuel flow is important because of delayed neutron emission. In a reactor with solid fuel, the delayed neutron precursor fission products remain very close to the location where fission happened, later emitting delayed neutrons at that location. Delayed neutrons have softer energy spectrum than prompt neutrons [40]. In case of liquid-fueled reactors the precursors drifting, consequently, the fission and delayed neutron emission 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  $\beta_{eff}$ . This quantity has significant impact on reactor safety because delayed neutron production occurs on a relatively long time frame and enables control of the reactor. Hence, to take into account tightly coupled MSR neutronics, thermal-hydraulics, and precursors drift a multi-physics code is required.

There are number of multi-physics tools which successfully describe steady-

state and transient behavior of various MSR concepts. Krepel *et al.* extended the Light Water Reactor (LWR) diffusion code DYN3D to consider drift of delayed neutron precursors alongside the reactor temperature profile, re-introducing the extended code as DYN3D-MSR [45]. 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 [46]. 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 [47]. That work directly solved the fuel salt velocity field, and used heterogeneous group constants in fuel and moderator regions.

More recently, Aufiero *et al.* [48] approached transient simulations in the MSFR with the finite volume OpenFOAM multiphysics toolkit [49]. 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.* [50] 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 [51]. 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, these research efforts used initial fuel salt composition, thus, considering the reactor core 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

## 2.1 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 [29]. 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 organizations 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 [30]. Unfortunately, build-in online refueling features still under active development and do not available for ordinary users. Furthermore, recently was demonstrated multi-physics simulations using SERPENT 2, i.e. coupled calculations with thermal hydraulics, CFD and fuel performance codes [52]. Two-way coupling to thermal hydraulics, CFD and fuel perfor-

mance 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 Message Passing Interface (MPI) parallelization. SERPENT 2 is an improvement upon SERPENT 1, and contains a complete redesign of memory management using hybrid OpenMP [53] + MPI parallelization. This hybrid parallelization is important in depletion calculations using computer clusters with multiple nodes, and allows to achieve significant speed-up in depletion calculations on computer clusters with more than 4'000 cores [54].

All calculations presented in this thesis were performed using SERPENT 2 version 2.1.30 on Blue Waters XE6 nodes. For cross section generation, JEFF-3.1.2 was employed [55].

## 2.2 Molten Salt Breeder Reactor description

The MSBR vessel has a 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 [56].

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 1'320 graphite cells, 2 graphite control rods, and 2 safety rods. 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. 2.1 and 2.2 demonstrate MSBR vessel, core configuration, “fission” (zone I) and “breeding” (zone II) regions position inside the vessel.

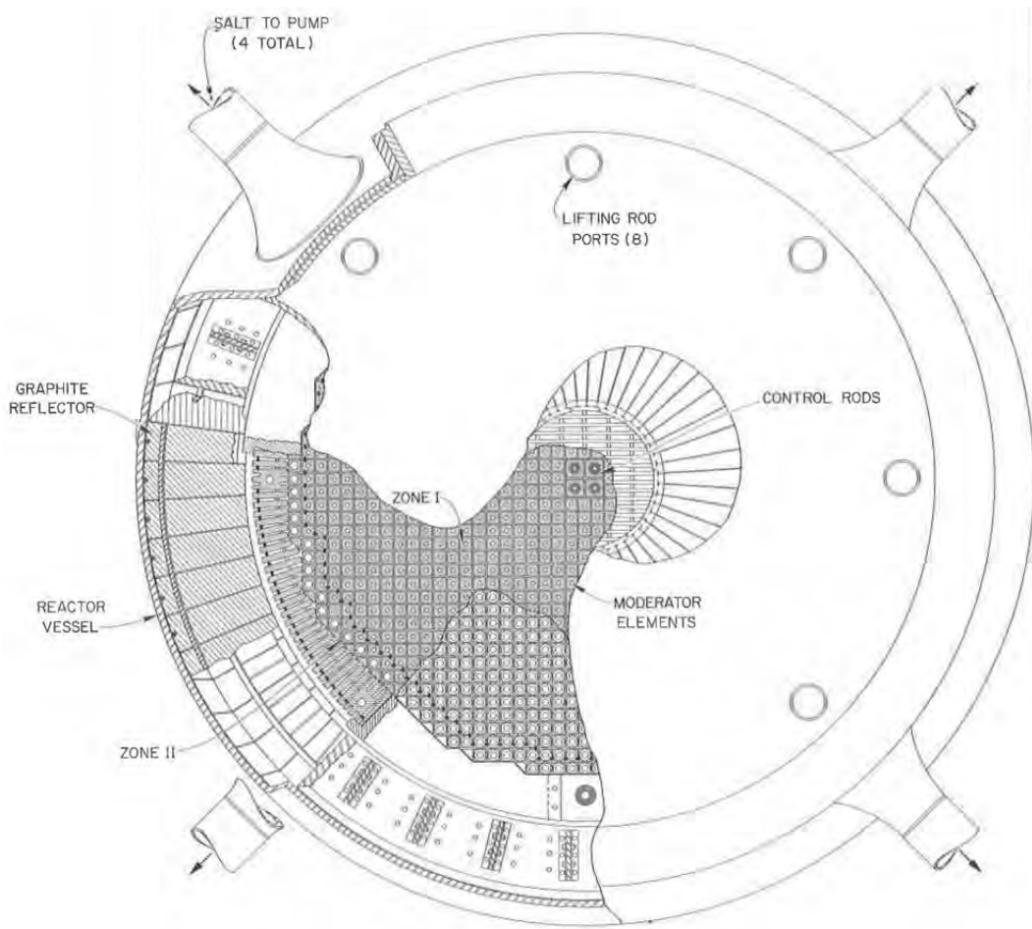


Figure 2.1: Plan view of MSBR vessel [56].

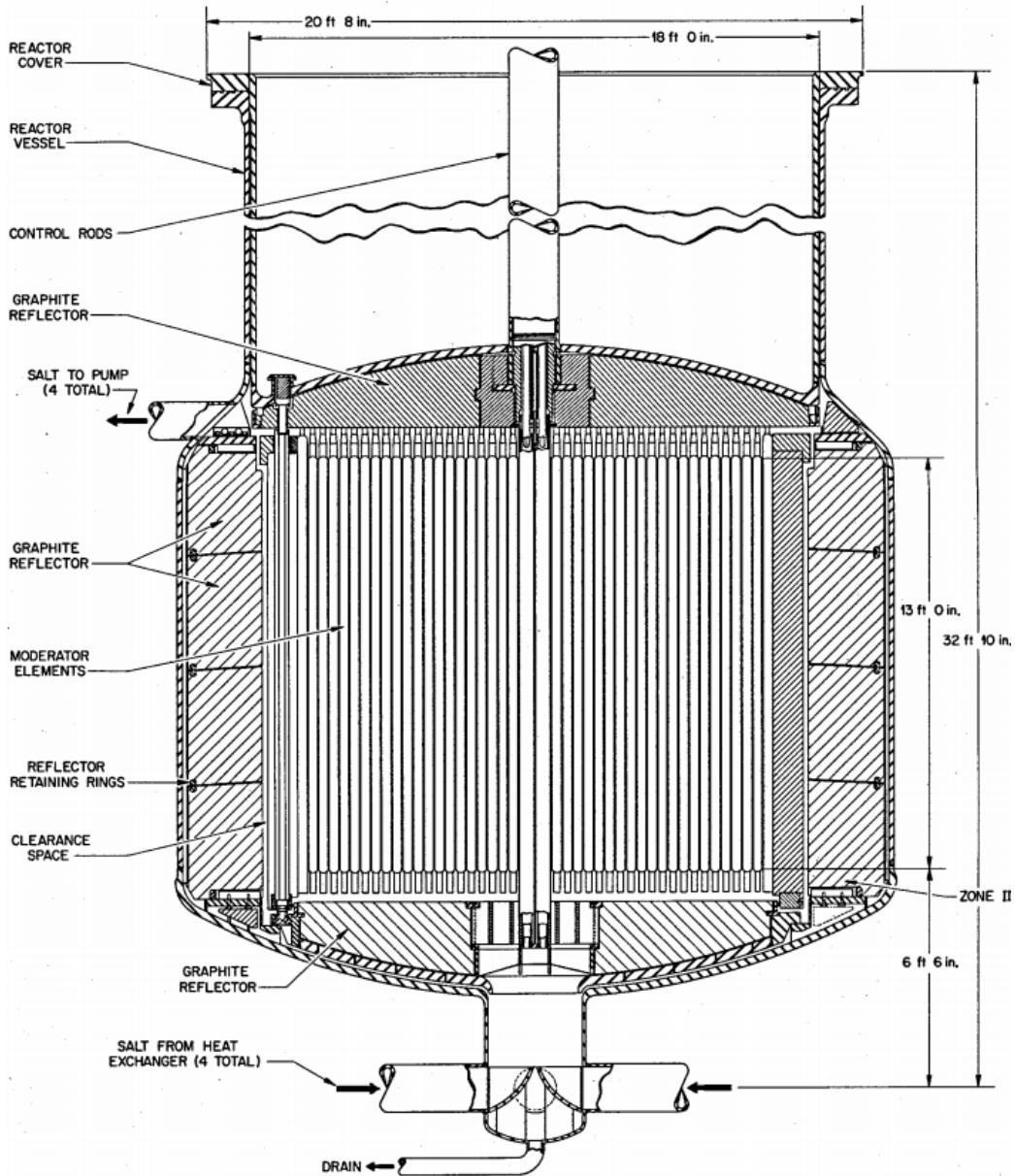


Figure 2.2: Sectional elevation of MSBR vessel [56].

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. 2.3. 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. 2.3 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.

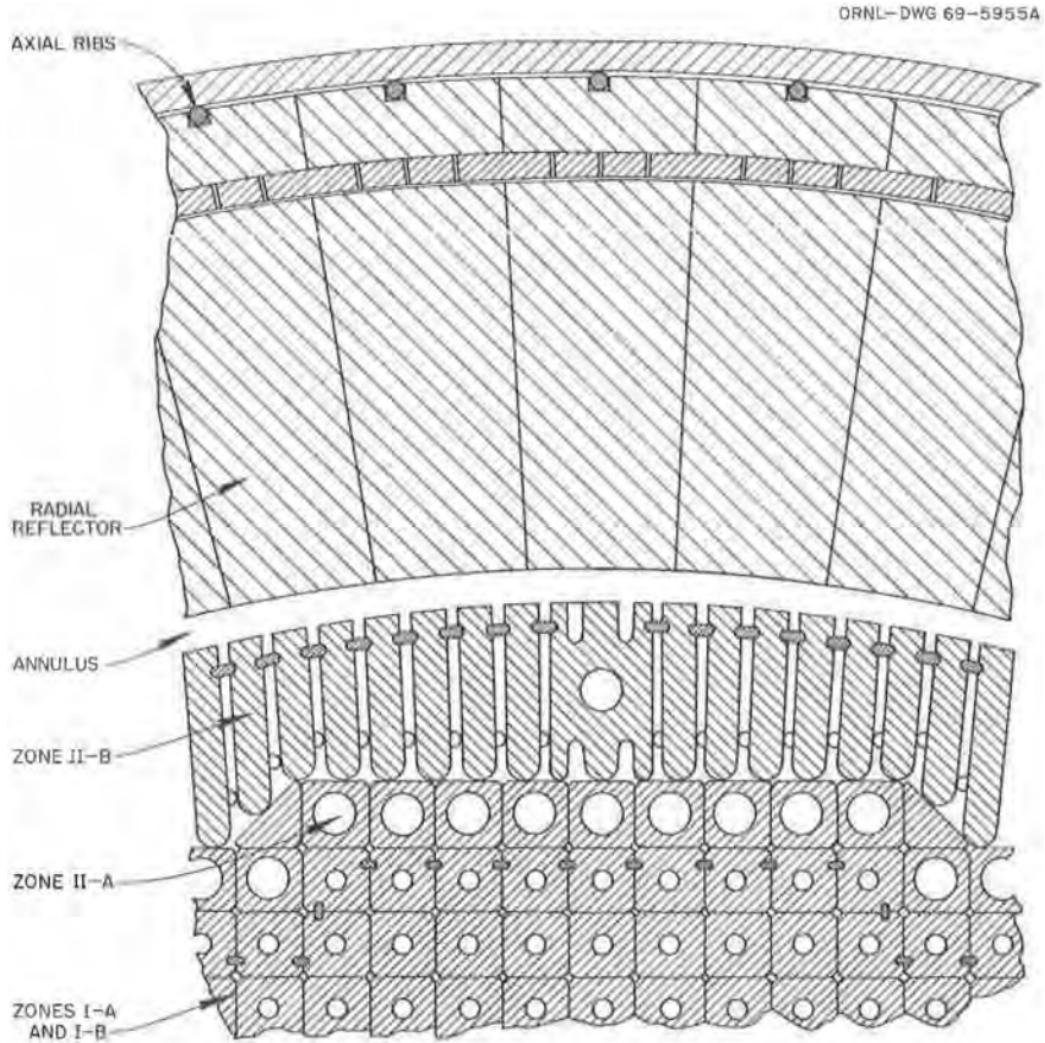


Figure 2.3: Detailed plan view of graphite reflector and moderator elements [56].

### 2.2.1 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 consisting of boron carbide clad 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 2.4 demonstrates the elevation and sectional views of graphite elements of zone I [56] and these elements SERPENT model [57].

### 2.2.2 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% fuel 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. 2.5 shows shape and dimensions of these graphite elements and their 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 2.3. 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 [56].

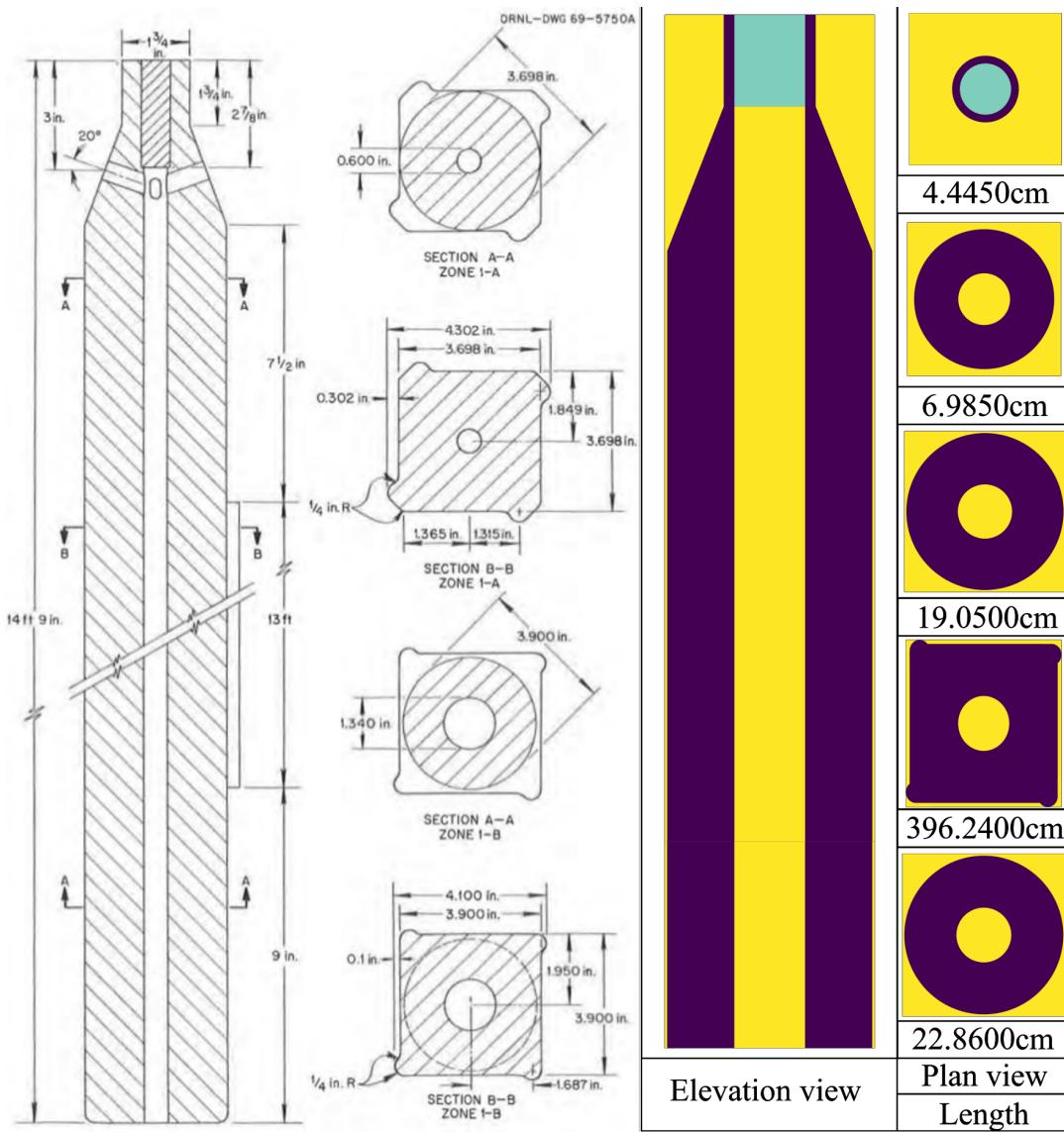


Figure 2.4: Graphite moderator elements for zone I [56, 57].

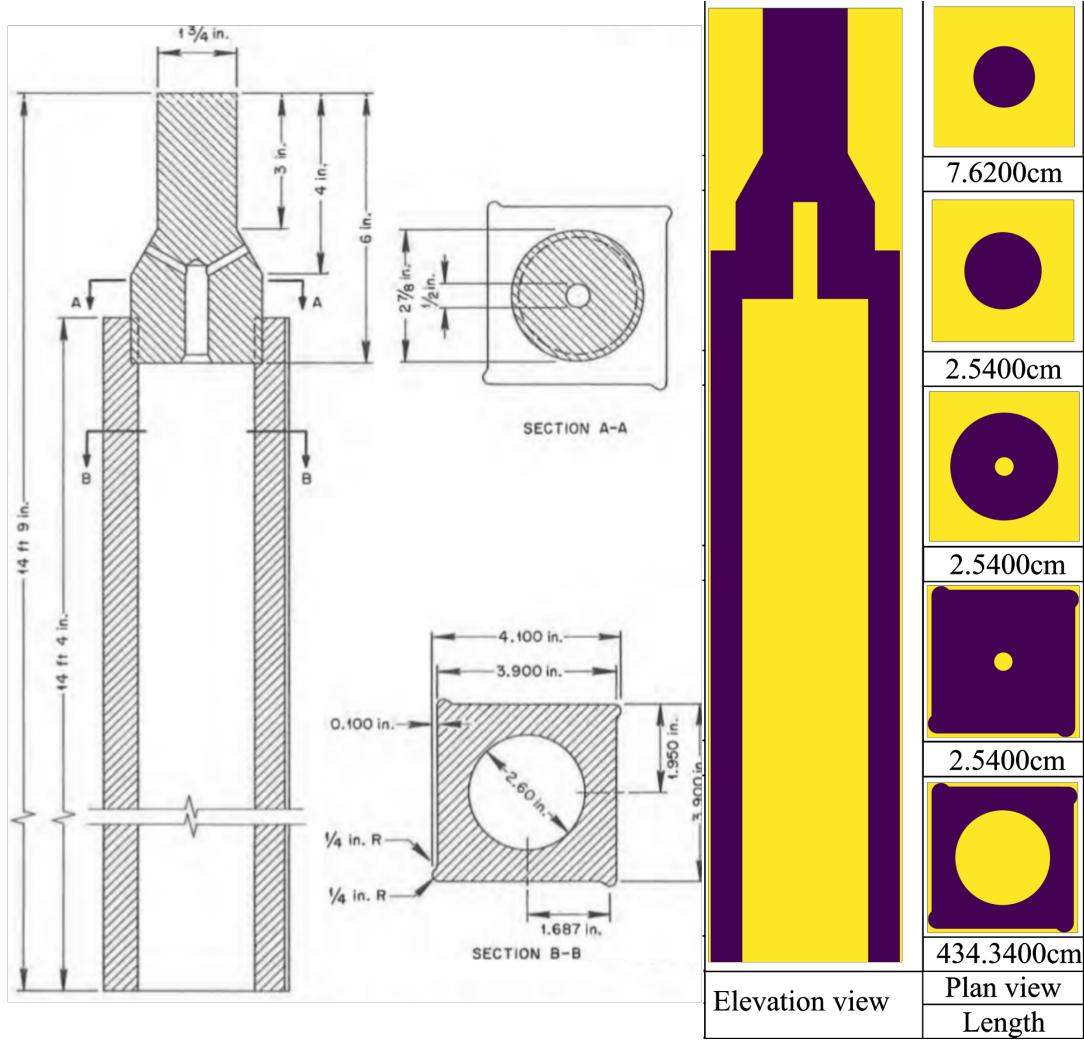


Figure 2.5: Graphite moderator elements for zone II-A [56, 57].

## 2.3 Existing full-core MSBR models

There are few recent studies presented full-core MSBR models for neutronics analysis. First, MCNP6 model developed by Park *et al.* for burn-up computations and safety parameters analysis [35]. This model has significant simplifications in zone II-B graphite elements geometry, and completely ignore lengthwise ridges at each corner of cell. Figure 2.6 shows simplifications in geometry of the model. More recently Skirpan *et al.* built a model of the core using Shift [58] to compare fidelity of one-cell, two-cell and full-core models of MSBR [59]. In this model complex cell geometry in zone I and zone II-A were approximated to slightly rotated square cylinder (figure 2.7).

Moreover, as can be seen from figure 2.8, zone II-B described using horizontal, vertical and  $45^\circ$ -degree graphite elements, that might significantly distort neutron flux and reaction rates in that region, and, consequently, misrepresent breeding parameters of the whole reactor.

In sum: full-core Monte Carlo model with sufficient fidelity necessarily for online reprocessing and refueling simulation. Moreover, high-fidelity model essential for problem-oriented homogenized nuclear data (multi-group cross sections and diffusion constants) generation for deterministic reactor codes, and for coupled simulations.

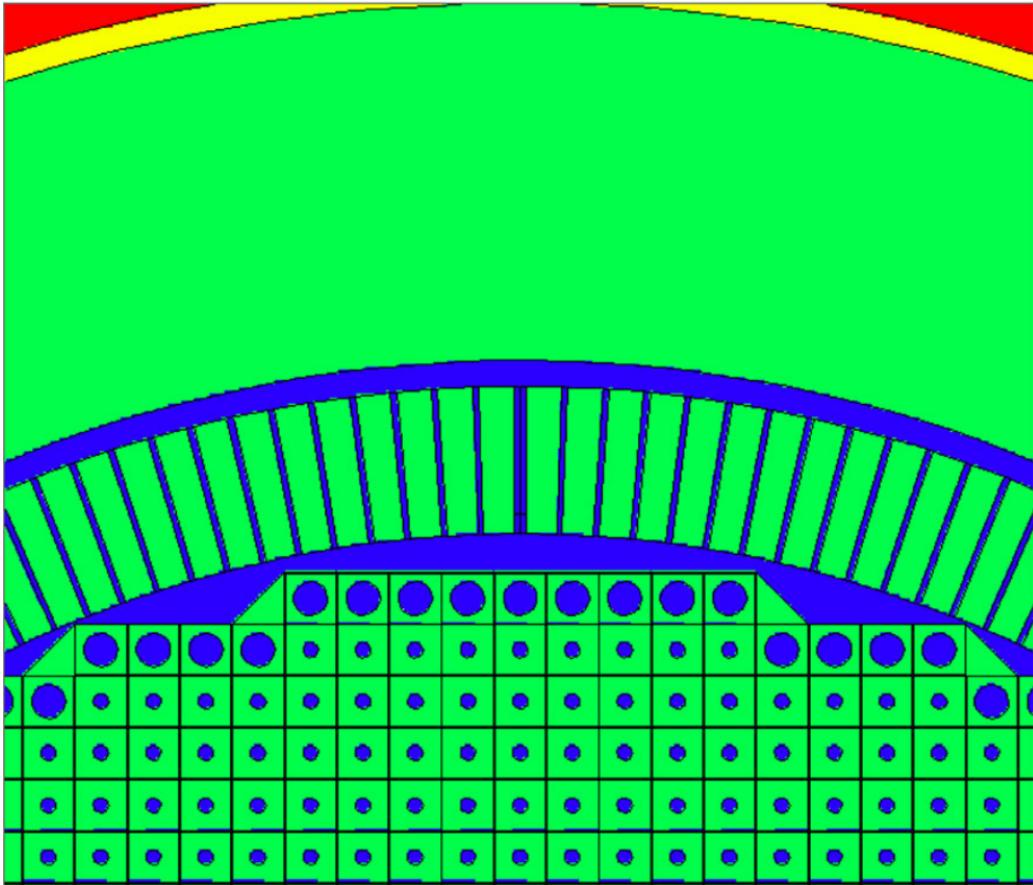


Figure 2.6: Graphite moderator elements for zone II and reflector from Park MSBR model (MCNP6) [35].

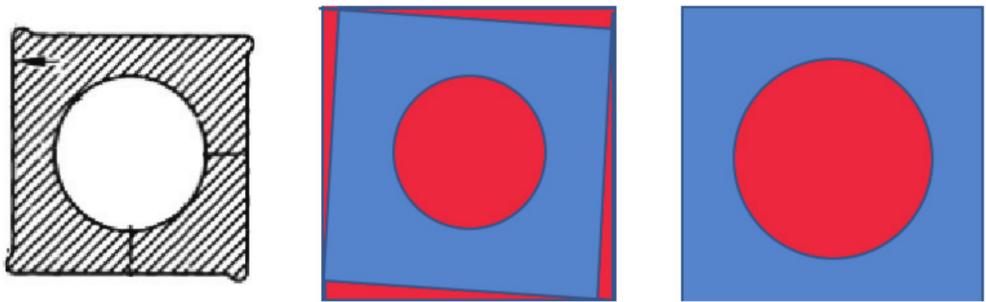


Figure 2.7: Geometry of an MSBR fuel channel (left) approximated with a simple geometric model (center) to calculate appropriate volumes to reduce to a two-region model (right) [59].

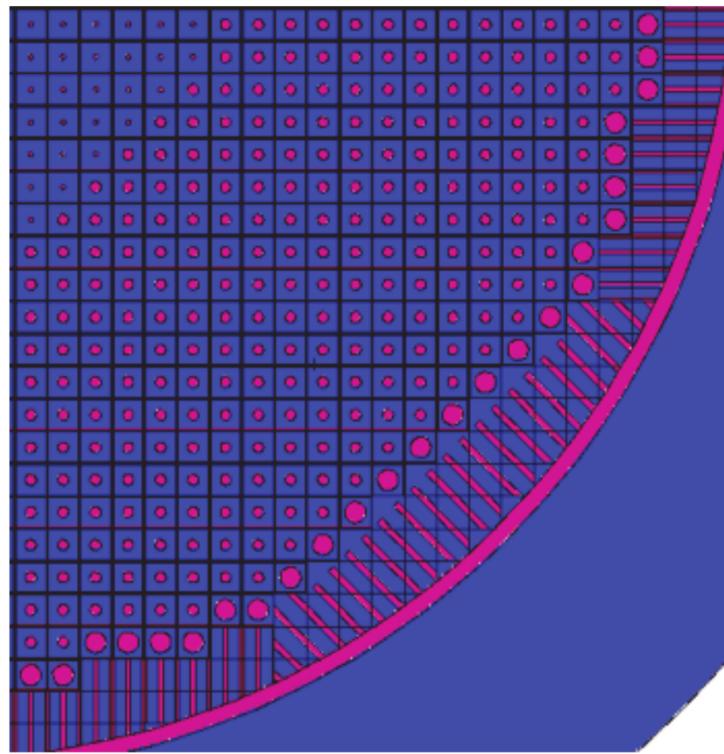


Figure 2.8: Plan view of the MSBR full-core transport model at core horizontal midplane [59].

## 2.4 SERPENT 2 model

To represent complex irregular MSBR core geometry advanced geometry surfaces in SERPENT was employed. Fig. 2.9 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 were not considered in this model. Another feature of the MSBR, its circulating liquid fuel and corresponding delayed neutron precursor drift, is not treated here also.

Fig. 2.10 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.

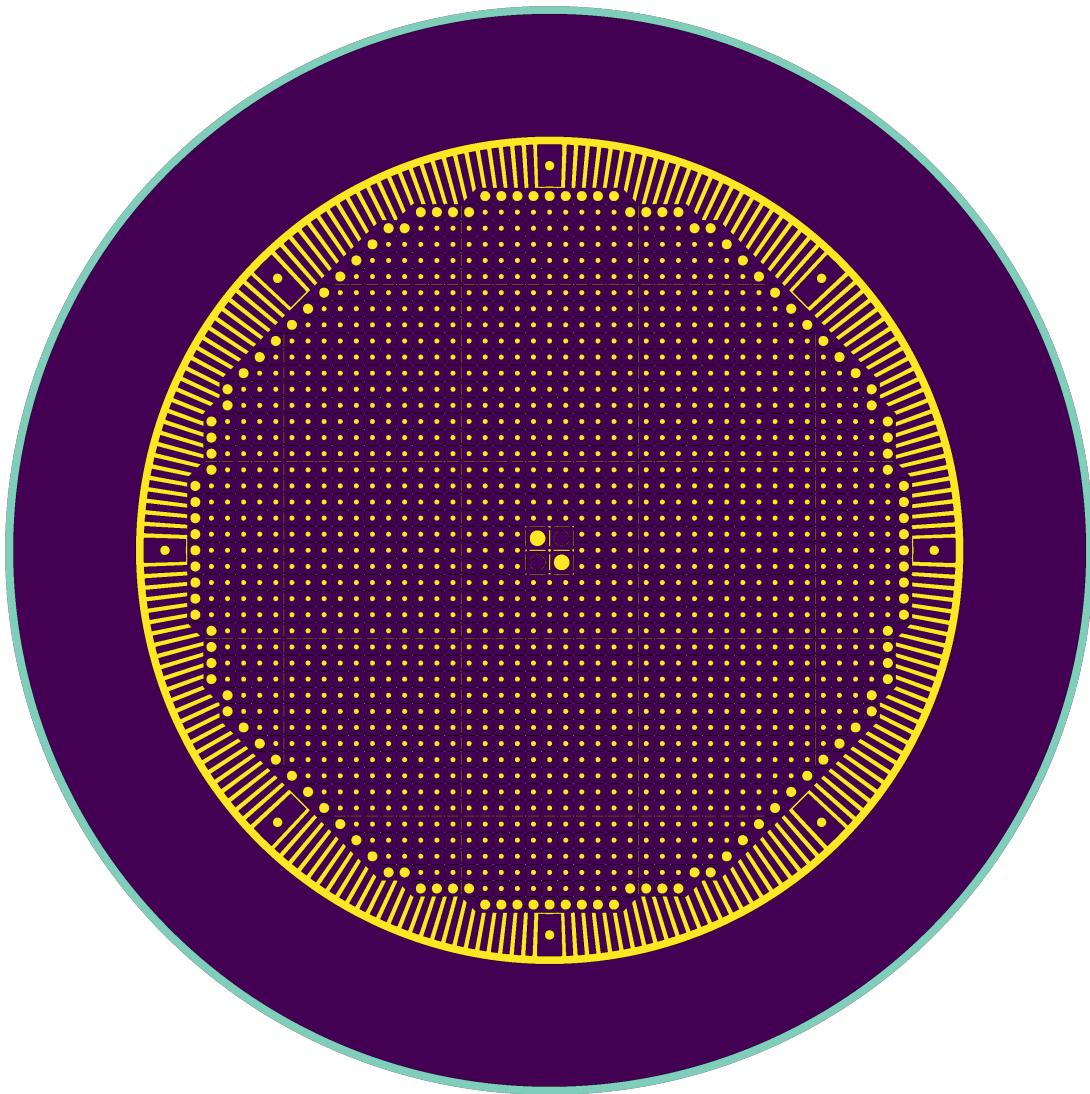


Figure 2.9: Plan view of MSBR model.

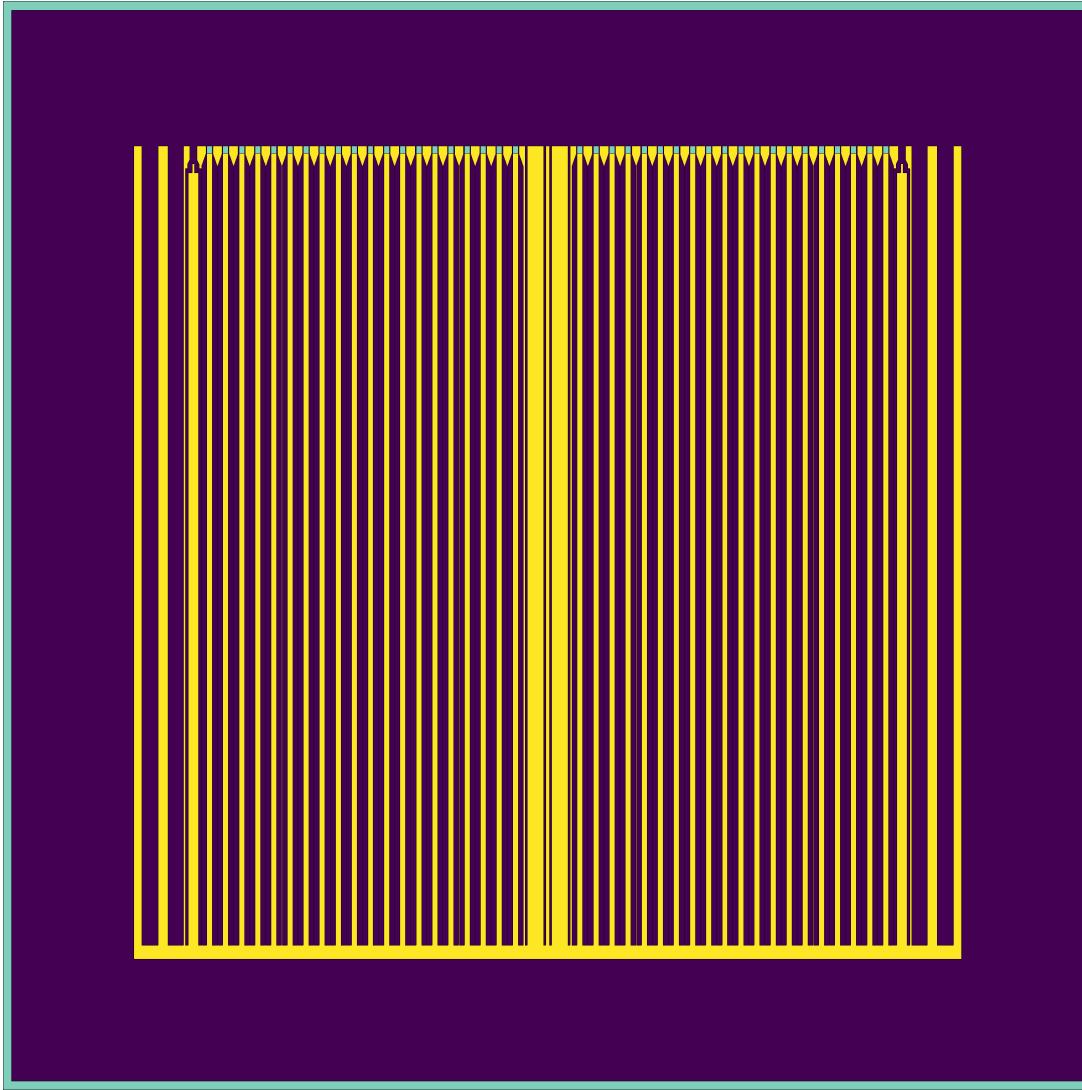


Figure 2.10: Elevation view of MSBR model.

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 (figure 2.4, 2.5). 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 [56], the suggested design of zone II-B has 8 irregularly-shaped graphite elements every  $45^\circ$  as well as salt channels (figure 2.3). These graphite elements were simplified into right-circular cylindrical shapes with central channels. Fig. 2.11

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.

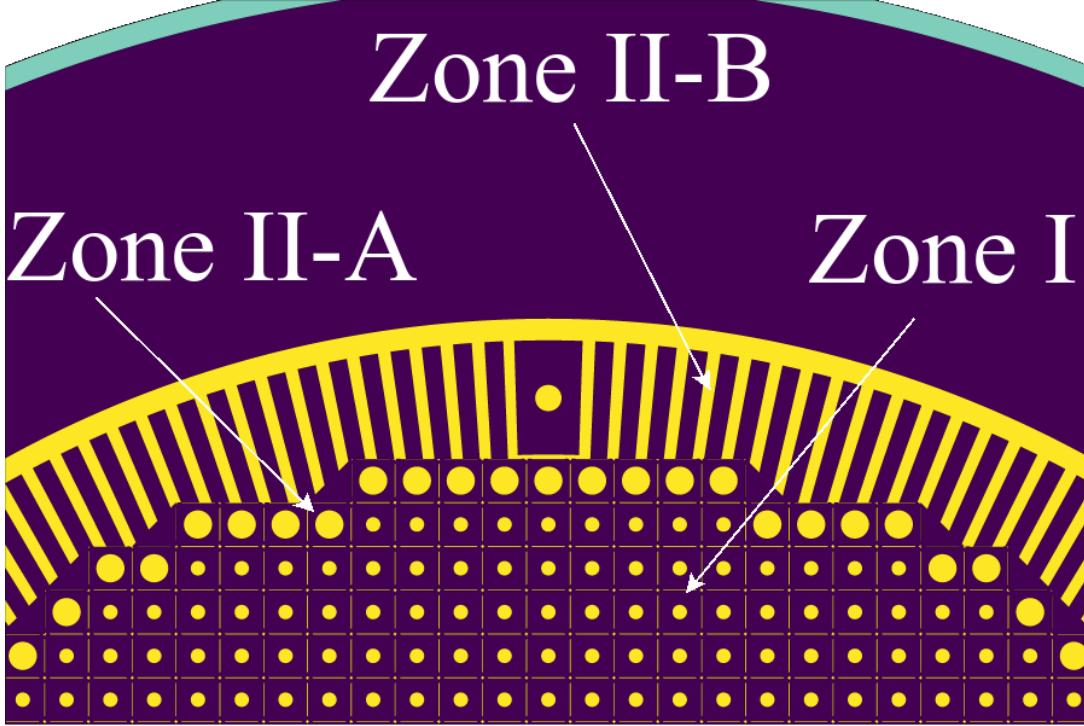


Figure 2.11: Detailed view of MSBR zone II model.

#### 2.4.1 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 ( $3.35 \text{ g/cm}^3$ ) 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[56]. The lithium in the molten salt fuel is a fully enriched  ${}^7\text{Li}$  because  ${}^6\text{Li}$  is a very strong neutron poison and becomes tritium upon neutron capture.

For cross section generation, JEFF-3.1.2 was employed [55]. 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 [56] and

has been applied to Serpent model without any modification. Table 2.1 is the summary of the major MSBR parameters used by this model [56].

Table 2.1: Summary of principal data for MSBR [56].

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 <sup>3</sup>
Fuel salt inventory (Zone II)	10.8 m <sup>3</sup>
Fuel salt inventory (annulus)	3.8 m <sup>3</sup>
Total fuel salt inventory	48.7 m <sup>3</sup>
Fissile mass in fuel salt	1303.7 kg
Fuel salt components	LiF-BeF <sub>2</sub> -ThF <sub>4</sub> - <sup>233</sup> UF <sub>4</sub>
Fuel salt composition	71.9-16-12-0.2 mole%
Fuel salt density	3.35 g/cm <sup>3</sup>

# ONLINE REPROCESSING SIMULATION

## 3.1 Fuel salt processing systems

### 3.1.1 Fuel salt chemical processing facility

All liquid-fueled MSR designs involve varying levels of online fuel processing. Minimally, volatile gaseous fission products (e.g. Kr, Xe) escape from the fuel salt during routine reactor operation and must be captured. Additional systems might be used to intensify those elements removal rate. Most designs also call for the removal of rare earth metals from the core since these metals act as neutron poisons. Some designs suggest more complex processing elements list (figure 3.1), including the temporary removal of protactinium from the salt or other regulations of the actinide inventory in the fuel salt [33].

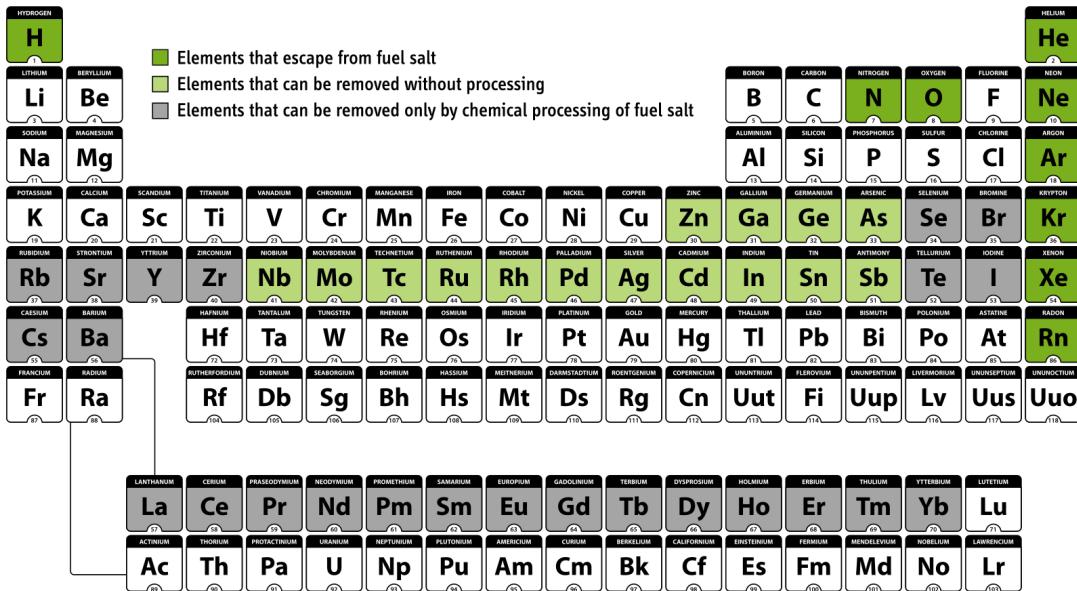


Figure 3.1: Processing options for MSR fuels [33].

In the one-fluid MSBR, considered in this work, thorium, uranium, protactinium, and fission products are all mixed together in a single fluoride salt (FLiBe). Separation of thorium from lanthanide (atomic numbers 57 through 71) fission products is rather challenging because of their chemical similarities.

Principal scheme of MSBR reprocessing facility has shown on Figure 3.2. Fuel salt was first held up for cooling and decay of the shortest lived fission products, then directed to the primary fluorinator, where most of the uranium was removed by fluorination to  $\text{UF}_6$  using gaseous molecular form of fluorine ( $\text{F}_2$ ) as the fluorination agent. After that the salt was routed to an extraction column where mixture containing metallic bismuth, lithium and thorium were contacted with the salt. The remaining uranium and protactinium were reductively extracted to the bismuth, leaving a salt that only contained fission products dissolved in carrier salt (base composition of  $\text{LiF}-\text{BeF}_2-\text{ThF}_4$ ). This salt entered another reductive extraction column where bismuth containing lithium separated from the salt lanthanide fission products and some thorium. The salt then went through a reduction column where  $\text{UF}_6$  was reduced to  $\text{UF}_4$  in the salt, refueling it and preparing it for return to the reactor. Refill  $\text{BeF}_2$  and  $\text{ThF}_4$  were also added and all residual bismuth was removed from the salt. After a final cleanup step and valence adjustment the purified salt was returned to the reactor [60],[61].

The bismuth accommodating some uranium and protactinium was routed to a hydrofluorination column where the metallic solutes in the bismuth were oxidized into their fluoride forms in the presence of a decay salt. The decay salt, containing  $\text{UF}_4$ ,  $\text{PaF}_4$ , and  $\text{ThF}_4$  passed into a decay tank where  $^{233}\text{Pa}$  was decaying to  $^{233}\text{U}$ . This uranium generated by protactinium decay was removed through fluorination to  $\text{UF}_6$  and directed to the reduction column to refuel the purified fuel salt. A hydrofluorinator and a fluorinator have capability to remove about **95%** of the uranium from the stream.

The fully processed salt, on its way back to the reactor, has uranium added from the protactinium decay tank at the rate required to maintain or adjust the uranium concentration in the reactor (and, consequently, control the reactivity). This is performed by contacting the salt with  $\text{UF}_6$  and hydrogen to produce  $\text{UF}_4$  in the salt and HF gas [56].

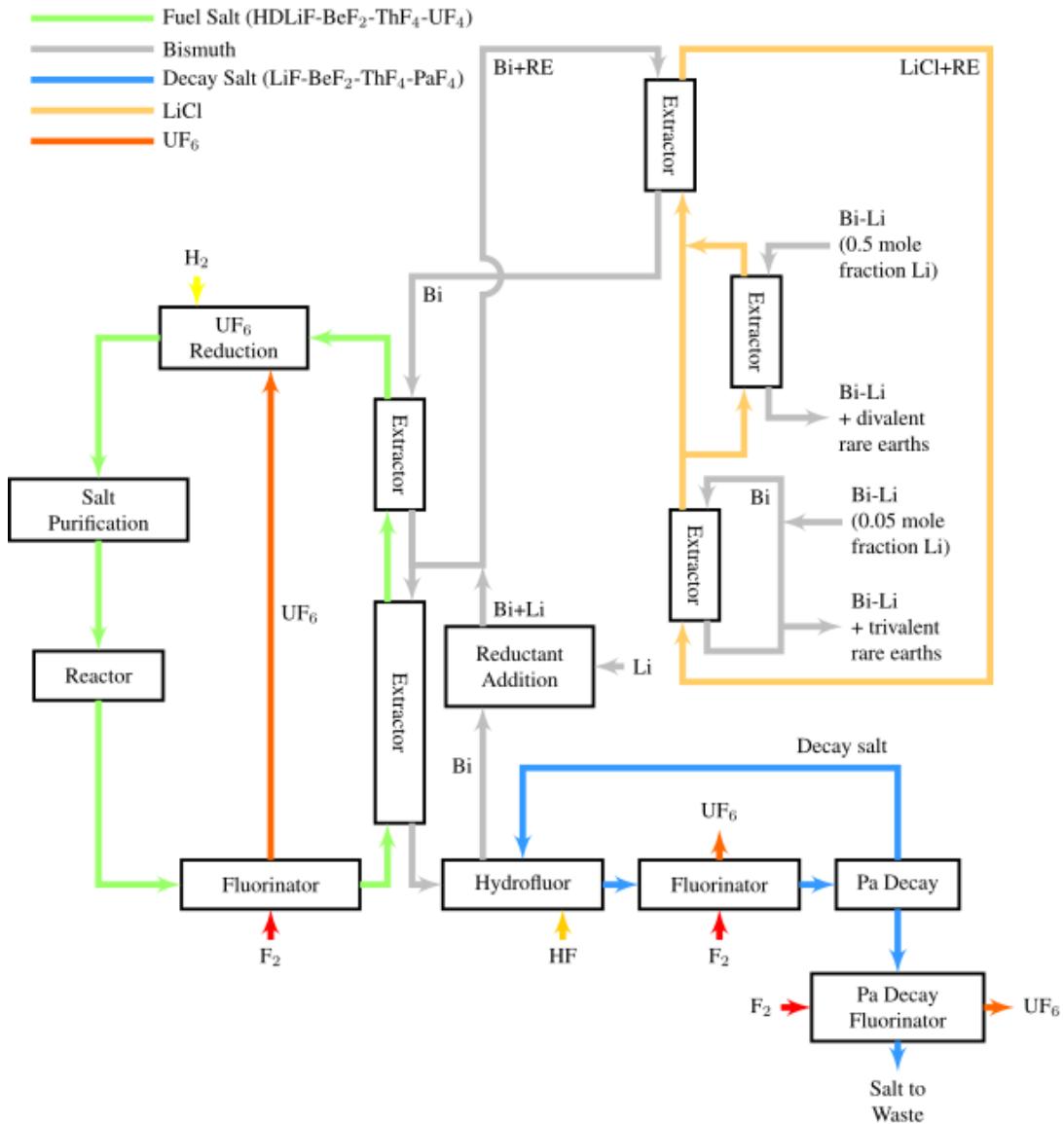


Figure 3.2: Detailed block diagram of chemical processing scheme for one-fluid MSBR [56],[61].

### 3.1.2 Gas separation system

Volatile gaseous fission products (e.g. Kr, Xe) must be removed from the fuel salt to avoid reactor poisoning especially during startup and maneuvering. This is particularly true for  $^{135}\text{Xe}$ , with its very large absorption cross section. Tritium, xenon, and krypton are sparged from the fuel salt by helium introduced in a bypass stream by a bubble generator and subsequently removed by a gas separator. Indeed, noble gases, because of their exceptional insolubility in the salt, will migrate promptly to any gaseous interface available. Because they form a true solution in salt, they will migrate in accordance with the conventional laws of mass transfer. If tiny helium bubbles are circulated with the fuel salt, they will absorb xenon and krypton fission products. The fission-product-rich bubbles of helium may then be separated from the salt and discharged to the off-gas system. Xenon migration to the circulating bubbles is in competition with xenon migration to the porous moderator graphite. The graphite is especially of concern because it absorbs xenon and holds it in the core which leads to parasitic neutron absorption. In ORNL report [56] in Appendix A it is concluded that, with moderate success of the coated-graphite program, the 0.5% target value for  $^{135}\text{Xe}$  poison fraction can be achieved when circulating helium bubbles 0.508mm in diameter. This is accomplished by bypassing 10% of the fuel salt from the pump discharge through a bubble separator to remove the xenon bubbles, then through a clean helium bubble generator for replenishment of helium bubbles, and back into the pump suction, as shown in Figure 3.3. The average residence time of a bubble in the fuel loop would be 10 full cycles.

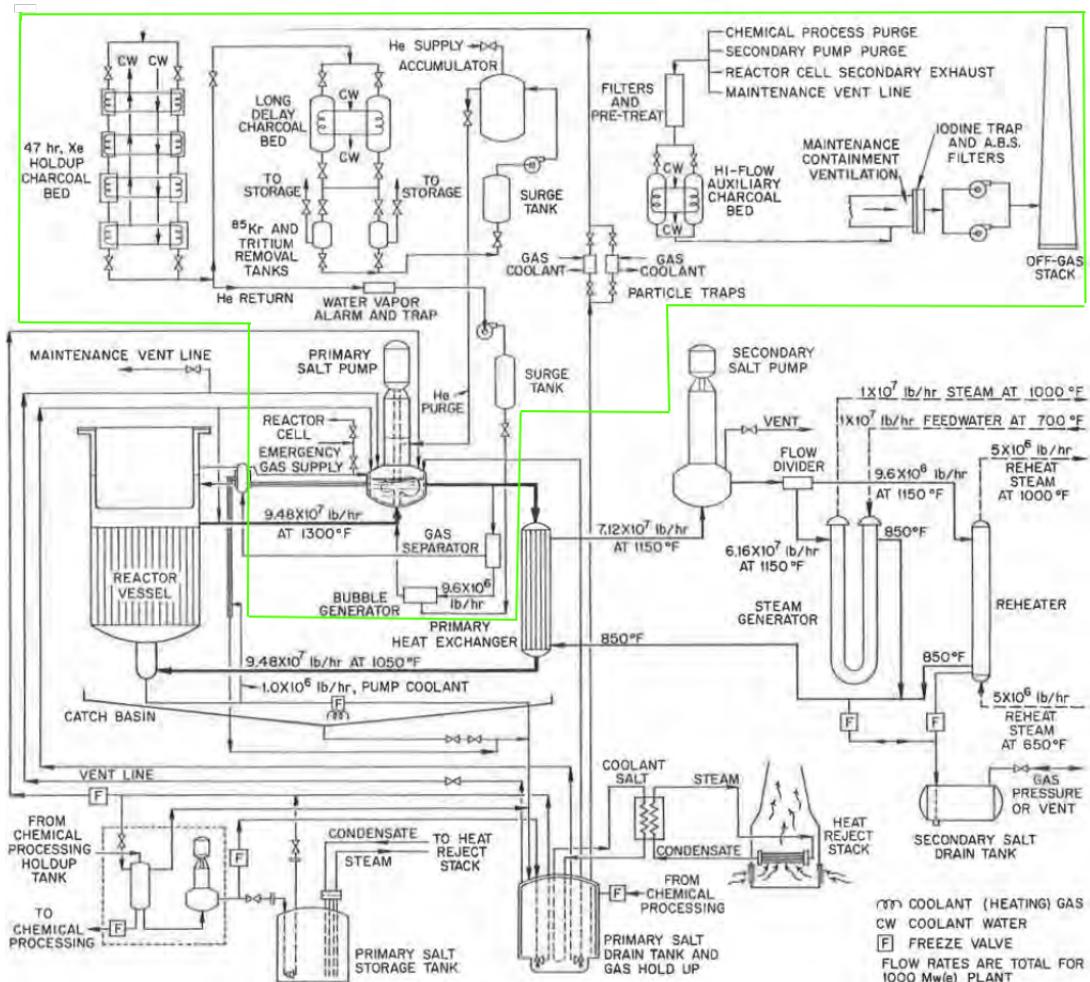


Figure 3.3: Flow diagram for MSBR plant. Green line indicates gas separation and off-gas system [56].

## 3.2 Online reprocessing method

### 3.2.1 Online separations and feeds

The ability to perform online reprocessing of fuel salt improves the potential neutronic performance of liquied-fueled reactors. Firstly, it is unnecessary for liquid-fueled reactors to operate with excess reactivity because fissile material is continuously adding into the core. Secondly, ability to continuously removing fission products that includes strong absorbers (poisons) might significantly improve fuel utilization and decrease neutron parasitic absorption. Finally, neutronic parameters could be adjusted “on-the-fly” without operational cycle interruption. Nevertheless, removal of each element from the liquid fuel salt presents a exclusive problem in terms of storage and disposal of the separated materials.

To take into account online reprocessing two principal approaches might be implemented. One is a batch-wise approach where material is moved into or from the core at specific time intervals (batch). This approach based on assumption that specific material accumulation in the core during the time between separations or feeds does not affect on reactor physics. This method requires the simulation to stop at a given moment in time and restart it with a new fuel salt composition (after removal discharged materials and addition fresh fissile and/or fertile materials). This approach was implemented in a ChemTriton script [38] which has been developed by T.J.Harrison, ORNL, and actively using for online reprocessing simulation with SCALE/TRITON [37] and Shift [58].

Another approach is continuous reprocessing where material is separating from (or added into) the core at all times and exactly simulate a true continuous online reprocessing. This method is more difficult because it requires adding a term to the Bateman equations. In SCALE/TRITON, ORIGEN [27] solves a set of Bateman equations using one-group averaged fluxes and cross-sections obtained from a transport calculation. Bateman equations that describe the rate of change of the isotopes due to neutron induced reactions and decay processes could be written in this form [30]:

$$\frac{dN_i}{dt} = \bar{\Phi} \sum_j N_j \sigma_{j \rightarrow i} - \bar{\Phi} \sum_j N_i \sigma_{i \rightarrow j} + \sum_j N_j \lambda_j b_{j \rightarrow i} - N_i \lambda_i$$

where  $N_i, N_j$  are the number densities of isotopes  $i$  and  $j$ ;

$\bar{\Phi}$  is averaged in the space and energy spectrum neutron flux;

$\sigma_{j \rightarrow i}$  is the microscopic one-group transmutation cross section of nuclide  $j$  to nuclide  $i$ ;

$\lambda_i$  and  $\lambda_j$  are the decay constants of nuclides  $i$  and  $j$ ;

$b_{j \rightarrow i}$  is the branching fraction for neutron absorption by isotope  $j$  that lead to the formation of isotope  $i$ .

The four terms on the right-hand side of the equation represent (1) the production rate of nuclide  $i$  from irradiation, (2) the loss rate of nuclide  $i$  due to irradiation, (3) the decay rate of nuclide  $i$  into nuclide  $j$ , and (4) the loss rate of nuclide  $i$  due to decay. Mentioned earlier deterministic code SCALE/TRITON and Monte Carlo codes MCNP, Shift, KENO-VI do not support non-zero removal or feeds rates for depletion simulations.

Online fuel reprocessing can be explicitly introduced in the system of equations by adding effective decay and transmutation terms for the different nuclides. During fuel composition evolution calculations, the total mass fraction of thorium fluoride is kept constant at 12%. For this purpose, transmuted into  $^{233}\text{Th}$  isotope are replaced with the fresh  $^{232}\text{Th}$  feed material. It could be achieved by modifying the Bateman equation adding the following additional gain term to the right-hand side:

$$\bar{\Phi} \sum_{k=^{232}\text{Th}} N_k \sigma_{k,c}$$

where  $\sigma_{k,c}$  is the one-group capture cross section of thorium-232.

The removal of fission products and protactinium is achieved by adding an explicit decay term to the Bateman equations. For the generic fission product  $l$ , following additonal loss term might be added:

$$-N_l \lambda_{l,reproc}$$

where  $\lambda_{l,reproc}$  is the effective removal time constant of the particular chemical specie. This approach was recently implemented as a purpose-made exten-

sion of the continuous-energy Monte Carlo reactor physics and burn-up code SERPENT [30] but it is not properly tested and unavailable for ordinary users so far.

To validate and verify in the nearest future the SERPENT 2 online reprocessing capability, I have been developed Python-based script, SaltProc, implementing batch-wise approach on the top of SERPENT 2 burnup routine. High-fidelity full-core MSBR model serves as a benchmark in online reprocessing simulation described in this thesis. Assessment against the SERPENT 2 continuous online reprocessing procedure based on the benchmark is not treated here.

### 3.2.2 Fuel material flows

The MSBR has the capability to remove all poisons (e.g.  $^{135}\text{Xe}$ ), noble metals, and gases (e.g.  $^{75}\text{Se}$ ,  $^{85}\text{Kr}$ ) every 20 seconds. The  $^{232}\text{Th}$  in the fuel absorbs thermal neutrons and produces  $^{233}\text{Pa}$  which then decays into the fissile  $^{233}\text{U}$ . Protactinium presents a challenge, since it has a large absorption cross section in the thermal energy spectrum. Accordingly,  $^{233}\text{Pa}$  is continuously removed from the fuel salt into a protactinium decay tank and allowing  $^{233}\text{Pa}$  to decay to  $^{233}\text{U}$  without poisoning the reactor. The reactor reprocessing system is designed to separate  $^{233}\text{Pa}$  from the molten-salt fuel over 3 days, hold it while  $^{233}\text{Pa}$  decays into  $^{233}\text{U}$ , and return it back to the primary loop. This feature allows the reactor to avoid neutron losses to protactinium, keeps fission products to a very low level, and increases the efficiency of  $^{233}\text{U}$  breeding. Table 3.1 summarizes full list of nuclides and the cycle times used for modeling salt treatment and separations [56].

Since removal rates vary among nuclides in this reactor concept, the SERPENT 2 build-in reprocessing subroutine is unable to capture the desired reprocessing strategy. The removal rates also dictate the necessary resolution of depletion calculations. If the depletion time intervals are very short an enormous number of depletion steps are required to obtain the equilibrium composition. On the other hand, if the depletion calculation time interval is too long, serious impacts of short lived fission products are not captured in a manner that is faithful MSBR conceptual design. To compromise, the time interval for depletion calculations in this model was selected as 3 days

Table 3.1: The effective cycle times for protactinium and fission product removal [56].

Processing group	Nuclides	Cycle time (at full power)
Rare earths	Y, La, Ce, Pr, Nd, Pm, Sm, Gd	50 days
	Eu	500 days
Noble metals	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te	20 sec
Seminoble metals	Zr, Cd, In, Sn	200 days
Gases	Kr, Xe	20 sec
Volatile fluorides	Br, I	60 days
Discard	Rb, Sr, Cs, Ba	3435 days
Salt discard	Th, Li, Be, F	3435 days
Protactinium	$^{233}\text{Pa}$	3 days
Higher nuclides	$^{237}\text{Np}$ , $^{242}\text{Pu}$	16 years

to correlate with the removal interval of  $^{233}\text{Pa}$  and thorium was continuously added to maintain the initial mass fraction of  $^{232}\text{ThF}_4$ .

### 3.2.3 Simplifying assumptions

The main goal of present study is to identify the effects of moving materials from and to the reactor core, and find equilibrium performance of a thorium fuel cycle using MSBR. To highlight these effects and simplify the analyses, several assumptions have been made.

First of all, thorium loading during operation was held constant and equal initial thorium loading (i.e.  $m_{Th}(t) = m_0$ ) with variable feed rate (in kg/day) of fresh thorium. Because thorium is a fertile material with relatively high absorption cross section, this has few important impacts on reactor physics, including negatively impacting reactivity and distorting the fuel-to-moderator ratio which makes neutron energy spectrum harder. While a reduction in the thorium loading reduces the amount of initial fissile material required to achieve criticality, the breeding rate of  $^{233}\text{U}$  should be sufficient to maintain the core critical during operation.

The solubility of heavy metals is a known problem for MSRs but it is

fundamentally dependent from the type of carrier salt. For this work, solubility limits for uranium were deprioritized because the molar fraction of  $\text{UF}_4$  was less than percent which considered to be reasonable to first-order for this study. In addition, it was assumed that addition or removal of soluble material (e.g.  $\text{UF}_4$ ) has a small influence on the fuel salt volume, this volume change is not treated here.

Figure 1.3 from Chapter 1 demonstrates that transformation  $^{232}\text{Th}$  to  $^{233}\text{U}$  takes about 30 days because  $^{233}\text{Pa}$   $\beta$ -decay has half-life 27.4 days. Therefore, if protactinium decay tank is empty at the moment of reactor startup than expected fissile material stream would appears only after few weeks of reactor operation at full power. To avoid time-dependent feed rate for  $^{233}\text{UF}_4$  it is assumed that protactinium decay tank contain some amount of  $^{233}\text{UF}_4$ , and the rate of fissile material flow from the tank to the core is set equal to  $^{233}\text{Pa}$  removal rate. Moreover, simulated cycle time at full power in this study is limited by 3435 days ( $\approx 9.41$  years), consequently, Rb, Sr, Cs, Ba separation, salt discard and high nuclides ( $^{237}\text{Np}$ ,  $^{242}\text{Pu}$ ) removal not considered here because those nuclides have cycle time longer than 3435 days (see table 3.1). Finally, 100% reprocessing separation efficiency was assumed.

The thermal fission of a  $^{233}\text{U}$  in fluoride salts can be shown to be an overall oxidizing process to the salt. This happens because the uranium nucleus balances the charge of four fluorine ions in the salt (e.g.  $^{233}\text{UF}_4$ ), but fission products tend to not bind to all the four fluorines released after the uranium fissions. Figure 3.4 demonstrates an example of an oxidative fission reaction. This excess of fluorine must be compensated, otherwise chemical reactions harmful to reactor components would occur [62]. In this study, fissions oxidizing effects are ignored.

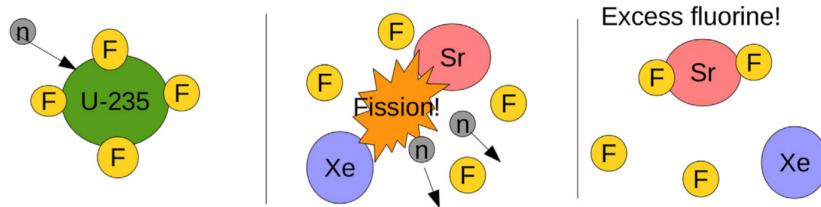


Figure 3.4: Process of production an excess of fluorine due to fission of a  $^{233}\text{U}$  in fluoride salts [62].

Finally, for this study equilibrium is defined as when  $k_{\text{eff}}$  of the full-core model with black boundary conditions and the  $^{233}\text{U}$  concentration in the fuel

salt are both significantly invariant in time (i.e., vary less than percent over several months). The fissile material was loaded into the system (core and protactinium decay tank) only at startup, therefore, no additional external feed or removal of fissile material assumed during reactor operation.

### 3.3 Python code description

The objectives for the SaltProc tool were to expand SERPENT 2 burn-up capabilities for modeling liquid-fueled MSR and provide an open-source tool for the simulation of reactors where material is removing or adding at any time during fuel irradiation. The code written in Python 2.7, uses HDF5 [63] to store data and the Nuclear Engineering Toolkit - PyNE [64] for SERPENT output files parsing. As was discussed earlier, SaltProc maintains the iterative semi-continuous approach to simulate a continuous feeds and removals.

The tool structure and capabilities are close to ChemTriton tool for SCALE developed in ORNL [38]. The primary function of SaltProc is to manage material mixtures while SERPENT 2 performs most of the computationally heavy work namely neutron transport and burnup calculations. Each material stream represents a fluid in the core design and has specific parameters (e.g. isotopic composition, reprocessing interval, mass rate, removal efficacy, etc). In addition, there is a set of available functions for each stream: read and write isotopic data in/from database, separate out specific isotopes from stream with defined efficiency, feed in specific isotopes to stream, maintain number density of specific nuclide in the core constant. These attributes and functions are crucial to simulate the operation of a complex, multi-zone, multi-fluid MSR and are universal enough to apply it for various types of systems.

Figure 3.5 demonstrates the algorithm of online reprocessing simulation with SaltProc and SERPENT 2. To perform depletion step, SaltProc reads a external SERPENT 2 template file which must be defined by user. This file contain input cards with all required for burnup calculations data such as geometry, moderator and construction materials isotopic composition, neutron population, criticality cycles, total heating power, boundary conditions. After depletion calculation completes, SaltProc reads the burned fuel com-

position file into memory and store it into HDF5 database. SaltProc only knows the number density and isotopic composition of a given fuel stream which provides the tool with the flexibility to model any geometry: an infinite medium, a unit cell, a multi-zone simplified assembly, or a full-core. While in some applications the simple single-cell is sufficient to get an accurate results within the fuel for depletion calculations with reduced simulation time, this flexibility provides high-fidelity full-core geometry capabilities for comparisons to more scrupulous models.

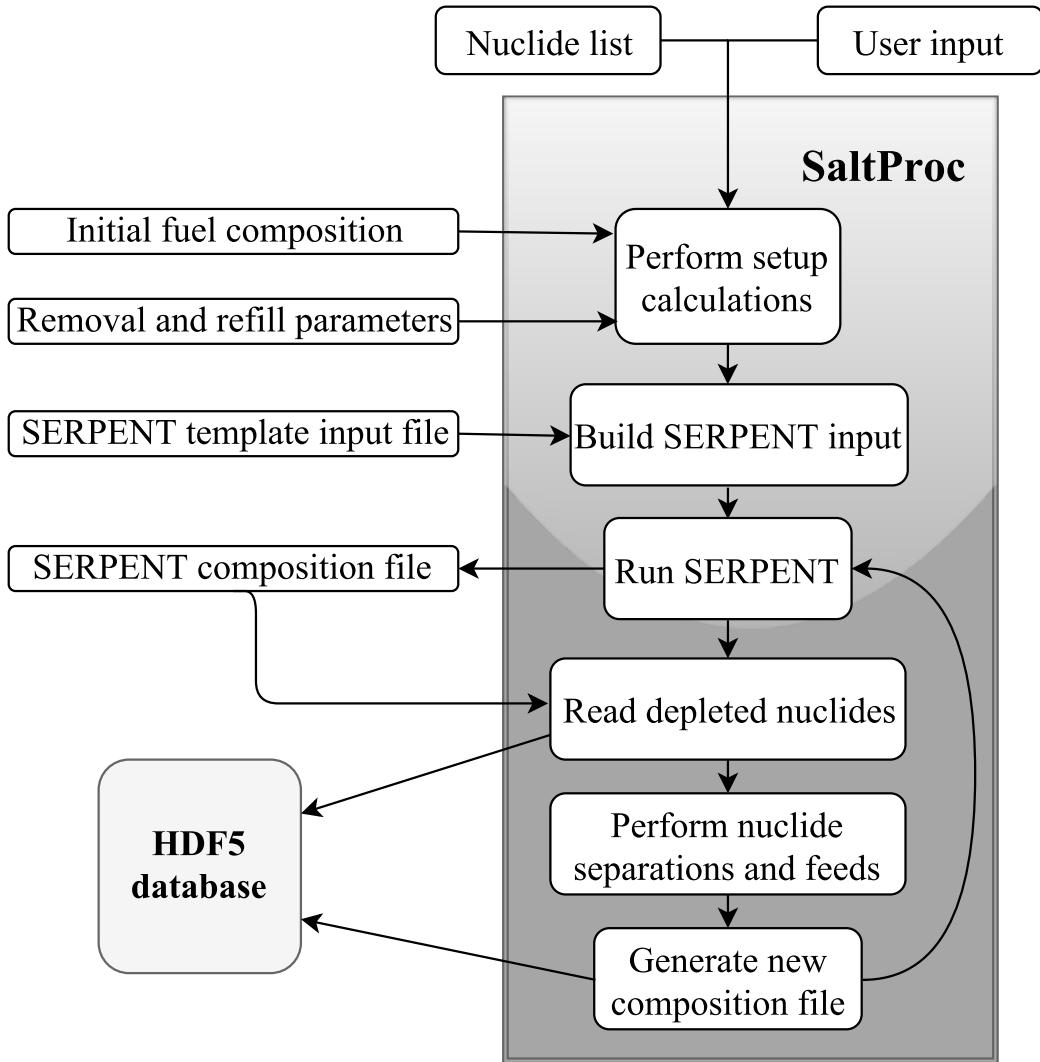


Figure 3.5: Flow chart for the Saltproc python-based tools.

SaltProc could manage as many fuel streams as desired. It also may work with multiple depletion materials. At the end of each depletion step, SaltProc reads the separate depleted compositions and tracks each material

stream individually. Following this, chemical reprocessing functions applying to fuel stream vectors. These vectors then combining in matrix, storing in database and printing into SERPENT composition file for the next step calculations.

Liquid-fueled MSBR design focuses on the state of the core at an equilibrium condition, after fission products have built up in the fuel salt during years of operation. Changes in isotopic composition of the fuel salt continues encounter small changes even after decades of operation, but the dominant nuclides that have significant impact to the neutronics behavior tend to reach an equilibrium concentration (e.g., vary about  $10^{-5}$  over several years). In contrast, from the startup of an MSBR until equilibrium condition, the fuel salt composition undergo significant changes (e.g., fission products, minor actinides, and fissile materials number density). During this period, the material feeds and removals should be optimized for the fastest MSBR transition to an equilibrium state, where the material streams are more constant in time. A faster transition simplifies the reactor operation because in an equilibrium state the fissile and fertile feed rates, safety parameters, and fission product removal rates are more constant in time.

In addition, SaltProc is able to define time-dependent material feed and removal rates to investigate the effect of different nuclide separations and/or feeds. The time dependence of the streams could be defined as piecewise functions and could be dependent on certain conditions being met. For instance, the tool could increase fissile material feeding rate if effective multiplication factor,  $k_{eff}$ , below a specific limit. Moreover, it could be useful to keep fissile material number density in the core approximately constant to accumulated excess of  $^{233}\text{U}$  into protactinium decay tank. These capabilities allow SaltProc to investigate the effect of lower concentrations of fissile and fertile startup loads. In sum, the development approach of SaltProc focused on producing a generic and flexible tool to give SERPENT 2 Monte Carlo code the ability to conduct advanced fuel cycle analysis as well as simulate a myriad of online refueled systems.

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