Master of Science Thesis

M/S Thunder - Design and analysis of a conceptual thorium fueled heavy water moderated molten salt reactor



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1 Abstract

In this thesis the basic design of a conceptual molten salt reactor, THUNDER, is introduced. THUNDER is a heavy water moderated molten salt reactor, running on a closed thorium fuel cycle. The design is loosely based on the CANDU reactor with individual fuel channels and unpressurized, cold moderator. The purpose of THUNDER is to combine the attractive properties associated with:

- The superior neutron economy of having heavy water as moderator
- The low production of long lived minor actinides in radioactive waste and the ability to obtain breeding in a thermal neutron spectrum, wich are attractive features of the thorium fuel cycle
- The unique safety characteristics of molten salt reactors along with the ability for online fuel reprocessing

The fundamental physical characteristics of the THUNDER reactor is examined with regards to basic reactor parameters like, fuel channel radius, fuel channel pitch, reprocessing time and fuel composition. Particular attention is given to how these parameters affect temperature feedbacks, rectivity and conversion of fertile material. Furthermore heavy water moderation is compared to the the standard case of using graphite as moderator and an unmoderated design, with regards to safety characteristics and efficiency of fertil to fissile conversion.

The results of the study are encouraging but not conclusive. By selecting an optimal set of design parameters it seems feasible to achieve a critical reactor with acceptable safety characteristics, and that a self breeding equilibrium state can be obtained, in which the reactor is fuelled only with fertile thorium. THUNDER is shown to have a slight advantage over graphite through a higher conversion ratio, but it is not clear if THUNDER has any significant overall advantage over graphite moderated molten salt reactors.

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3 Introduction

After a long slump in activity, nuclear energy is once again on the table as an option for energy production. Hardly no new reactors has been built in the west during the last twenty years, and while asian countries has continued to build reactors nuclear power have not come close to the optimistic projections of the 60's and 70's. The revival in interest for nuclear energy can to a large extent be attributed to a growing awareness of the beneficial environmental characteristics of nuclear power and increased concern about the effects fossil fuels have on the climate. A valid question might be raised if nuclear power have indeed overcome the obstacles it faced that put a halt to its first expansion? It would not be out of line to claim that the main reason for the end of the first nuclear era is long lived waste, public perception of nuclear safety, high capital costs and concerns about weapons proliferation. One additional issue has arisen that might impact public perception of nuclear power, the sustainability of uranium and the environmental effects of uranium mining.

It does not take much effort to realize that many of those issues are tied to the present use of once through fuel cycles in light water reactors, and not intrinsic to all possible nuclear energy generation. Light water reactors can utilize less then a percent of the available energy in natural uranium and in the process they produce significant quantities of transuranic elements that require storage for tens of thousands of years. For the nuclear renaissance to be long lasting, a gradual shift away from light water reactors and towards closed fuel cycles seems advisable. The Generation IV International Forum ¹ has been established to examine new nuclear systems that might overcome the issues with current nuclear reactors. One of the most promising gen-4 reactor types in the long term is the molten salt reactors(MSR).

The focus of this work is THUNDER, a conceptual heavy water moderated MSR. THUNDER rests on three legs, molten salt fuel, heavy water moderator and a thorium fuel cycle. MSRs are characterized by their fuel being in a liquid state and simultaneously acting as the coolant for the reactor system while graphite generally serves as moderating material. Liquid fuel gives the MSR unique advantages in terms of reactor safety, waste management and resource utilization. Heavy water have some distinct advantages over graphite as a neutron moderating material, it absorbs less neutrons and moderates more efficiently yielding better neutron economy. In addition, unlike graphite, heavy water does not get damaged by neutron irradiation and the issue of replacing graphite moderator periodically is avoided completely. Thorium is 3-5 times as abundant as uranium in the earth's crust and its the only nuclear fuel that allows thermal breeding.

Combining the excellent properties of molten salt fuel, heavy water moderator and a thorium fuel cycle gives THUNDER.

This thesis is the MSc thesis of the author and the research for it has been conducted at the nuclear engineering department of Chalmers university of technology during spring and summer 2008.

4 Background

4.1 Thorium

Thorium is a weakly radioactive heavy metal that is fairly abundant in the earth crust. The only naturally occuring isotope is Thorium-232. Its estimated to be 2-4 times as abundant as uranium², this abundance potentially makes thorium a sustainable energy resource for the very long term if utilized properly. The total world production of thorium today is a few hundred tons, mostly as a by-product when extracting rare earth elements. Its primarily used in special alloys and glasses. Brazil, Turkey and India has the largest known thorium deposits. Norway and the US also has significant deposits.

There are no fissile isotopes of thorium, but thorium is a fertile material that can transform into fissile uranium-233 by the following reaction.

$$^{232}Th + n \rightarrow ^{233}Th \xrightarrow{\beta - 22,3 \ min \ 233}Pa \xrightarrow{\beta - 27 \ d \ 233}U$$

Like plutonium-239, uranium-233 doesn't exist in nature. hence another fissile nuclide has to be used to start the process. Thorium was early recognized as a possible fuel for nuclear reactors but interest dropped when uranium was found to be more abundant than first thought. Uranium-233 has one significant advantage over other fissile isotopes, in the thermal spectrum the number of neutrons emitted by fission per neutron absorbed is larger than for any other fissile nuclide. Uranium-235 or plutonium-239 produce to few neutrons per absorbed thermal neutron to breed in a thermal spectrum but uranium-233's higher neutron yield makes thermal breeding plausible. In figure 1 the neutron yield for different isotopes is shown.

Breeding with thorium has in fact been demonstrated in the Shippingport light water reactor ⁴ hinting that other reactor types with more beneficial neutron economies can achieve breeding more easily. One factor that makes thermal breeding with thorium more problematic than fast breeding with uranium-238 is the relatively long half life of the uranium-233 precursor Protactinium-233. Protactinium-233 has a non-negligible neutron capture cross section in the thermal region, and to avoid unnecessary losses of Pa the power density has to be lowered, or the fuel continuously reprocesses to remove Protactinium-233, allowing it to decay into uranium-233 outside of the core. Despite this complication a reactor running on a thorium fuel cycle has the potential to combined the effective fuel utilization of fast breeder reactors with the prominent safety characteristics and much lower fissile inventory of thermal reactors.

There is relatively much experience with thorium as a nuclear fuel. Besides the aforementioned Shippingport reactor many other reactors has been fuelled with thorium⁵. The German pebble bed reactor program was centered around using thorium and highely enrichened uranium(HEU) in a closed fuel cycle. Both the experimental reactor, the AVR, and the industrial demonstration reactor, the THRT(thorium high temperature reactor) was fuelled with thorium in periods. Similary the the American Fort Saint Vrain prismatic block type high temperature



Figure 1: Number of neutrons emitted in fission per neutron absorbed ³

reactor was fuelled with HEU/Thorium fuel. In Russia experiments are running with thorium fuel rods in pressurised water reactors. India is the country most heavily investing in thorium, India has scarce uranium resources but vast thorium deposits. The long term goal of India is to have its entire nuclear sector fuelled by U233 breed from indigenous thorium in fast reactors. Thorium has recently started to receive attention in Norway due to Norway's large thorium deposits and declining oilfields. Thorium is also a proposed fuel for the Pebble bed reactors being developed in South Africa and China, specifically in combation with plutonium. Adding thorium to plutonium fuel allows going to higher burnups and thus higher plutonium destruction rates. The future prospects for thorium is looking bright.

4.2 Molten salt reactors

In the 50's researchers at Oak Ridge National Laboratory lead by Alvin Weinberg turned the tables on nuclear power. Instead of using solid fuel elements and flowing coolant they examined the idea of having the fuel itself as a circulating liquid. After looking into aqueous, molten metal and other liquids they realized the best liquids are molten fluoride salts⁶. Fluoride salts have low neutron absorption and all the characteristics of a good coolant like, high heat capacity, high boiling point, low fusion temperature and medium heat conductivity. As added bonus, the salts are chemically inert and the ionic bonding in fluoride salts makes the fuel totally impervious to radiation damage.

When the US air force started its aircraft reactor experiment, the molten salt

reactor was chosen which lead to the construction of the first molten salt reactor. The reactor itself was a success, but the idea to put a reactor on an air plane was not $^{7 \ 8 \ 9}$. After the termination of the aircraft reactor experiment work on molten salt reactors continued with the molten salt reactor experiment(MSRE)¹⁰



Figure 2: MSRE reactor vessel

The molten salt reactor experiment was a 7.4 MW_{Th} single fluid reactor moderated by graphite. Over the course of the experiment the reactor was operational for over 9000 equivalent full power hours and various fuels where tested all based on a LiF-BeF₂-(HN)F₄ at 70-17.5-12.5 mole% eutectic mixture where HN is heavy nuclide. At that time the main idea for a full power producing reactor was to have one fuel fluid and another blanket breeding fluid containing the majority of the thorium, the blanket fluid was however omitted in the reactor experiment for simplicity of design. The first fuel load was with 33% enrichened uranium but that was later switched to U233 and a uranium, thorium mix. During the experiment the inherent stability at all power levels where confirmed, the viability of online reprocessing was shown and low levels of corrosions was observed ¹¹. After the positive experience of the molten salt reactor experiment researches at Oak Ridge continued to develop a design for a full scale, $1000MW_e$ molten salt breeder reactor. They abandoned the idea of a two fluid design for reason of simplicity and assumed one fluid in which both breeding and power production takes place. However, later re-evaluations of the design has shown it would possibly have positive reactivity coefficients¹² which would be unacceptable for reactor safety reasons. The problem can be corrected however by tweaking the fuel channel radius.

The main advantages of molten salt reactors are in safety and resource utilization. Accident scenario like LOCA and core meltdown has no meaning in a molten salt reactor since the fuel is also the coolant and it is always molten. Excessive temperatures can be passively protected against by having a "'freeze plug"' that melts at a certain temperature and drains the salt into a passively cooled storage tank with a geometry that makes criticality impossible. Furthermore a MSR can be properly designed to have negative temperature coefficients at all power levels, by making use of fuel expansion and the doppler reactivity feedback effect. Since the fuel is continuously reprocessed there is no need to have excess fissile material in the core to compensate for burnup or poisoning. Due to the MSR being a breeder the only fuel being needed after initial startup is thorium. The online reprocessing means separation of waste products is done at the reactor while its running, thus the only waste leaving the installation is fission products. Both fuel consumption and waste leaving the installation is reduced by a factor of 50-100 compared to a light water reactor. Since the MSR is a thermal reactor the initial fissile load for a MSR is far lower than for a fast breeder reactor. Another advantage for the MSR is that no fuel fabrication is needed, fuel fabrication is a significant part of the fuel cost of regular reactor fuel.

Today the MSR concept is primarily worked on by a few research groups in different parts of the world. The most active group is one at Laboratoire de Physique Subatomique et de Cosmologie(LPSC), Grenoble. The group at LPSC has drifted towards a promising unmoderated MSR design working in the epithermal range due to the weak moderation of fluoride. A group head by Kazuo Furukawa has been developing the FUJI molten salt reactor for quite some time. Other efforts are being made around the world, but there are no immediate plans for a research reactor. Compared to other gen-IV reactors the MSR is probably the reactor receiving the least attention.

The main challenges the MSR technology is facing is mostly chemical in origin. A wide variety of different fluoride compounds are formed from the fission products and the chemical behavior of them all is not well understood. There are fears that corrosion will be a significant problem in large scale molten salt reactors. The reprocessing technology needs to be researched further and there are some issues with expected graphite lifespan under the conditions that exist in the reactors. According to the gen-IV technological roadmap¹³ research is needed atleast until 2020 before a commercial design can be expected. As with all projections however this is highly dependent on funding for the research.

4.3 Heavy water moderated reactors



Figure 3: A CANDU reactor vessel

Heavy water reactors have been around since the birth of nuclear power, as it was early recognised that deuterium has prominent features as a neutron moderating material. Two factors determine the merit of a moderator from a reactor physics perspective. Firstly it must be made of light nuclei since they slow down neutrons most efficiently, secondly they must not absorb to many neutrons while slowing them down. A ratio between those two properties, macroscopic slowing down power and macroscopic absorption cross section can be defined for each material and it decides how good a moderator a material is. Heavy waters moderating ratio is in fact some 80 times higher than light water and 20 times higher than graphite. Heavy waters superior moderating ratio directly translates into superior neutron economy and makes possible the use of natural uranium as fuel.

Today heavy water moderated reactors make up roughly one tenth of all power producing reactors. Canada is the dominant nation in heavy water reactor technology and has long been developing its CANDU line of reactors. Besides being heavy water moderated CANDU differs from light water reactors in that they don't have a large pressure vessel. The fuel is inserted into individual pressure tubes that are housed inside a large vessel, the calandria, filled with heavy water held under normal atmospheric pressure. India also has a very active program for heavy water moderated reactors, the design they use today is virtualy identical to the CANDU design. More advanced design are beeing worked on that deviates more from the CANDU.

5 Thunder - the concept

The THUNDER (molten salt Thorium UraNium DEuterium reactor) concept reactor developed in this thesis is a heavy water moderated molten salt reactor. This work is a conceptual study with a focus limited to the neutronic properties and basic reactor physics. Material considerations, thermal hydraulics and reactor kinetics, all very important aspects of reactor design, are not covered in this work. The goal of this work is to investigate the possibility to design a compact, inherently safe, THUNDER that utilize resources in a efficient way. It is also of interest to see if THUNDER has any distinct over other similar concepts, in particular a graphite moderated MSR.

THUNDER is based on a similar design as the CANDU reactor. Just like the CANDU, THUNDER is composed of fuel channels going through a calandria filled with heavy water. The fuel channels are composed of graphite or alternatively silicon carbide(SiC) to be able to withstand the high temperatures of the salt. The graphite is thermally insulated from the heavy water moderator by a gas layer contained within a tube made up of steel as shown in the figure below.



Figure 4: To the left a cross sectional view of a fuel channel from the top, to the right a cross section of the reactor from the side. Fuel reprocessing unit and second coolat loop not shown on the picture.

In an ordinary MSR, consisting of large graphite moderator blocks, the graphite has to be replaced regularly due to swelling and cracking from neutron irradiation. In THUNDER the only thing that needs to be replaced are the graphite fuel channels. properly design that can possibly be a much simpler procedure. The salt mixture chosen for THUNDER is LiF-(HN)F₄, the reason for choosing this over LiF-BeF₂-(HN)F₄ is primarily simplicity. A simpler salt will most probably mean easier reprocessing and the selection simplify the defining of materials in reactor codes. LiF-(HN)F₄ has a higher melting point than LiF-BeF₂-(HN)F₄ with the eutectic mixture at 22 HN mole percent with a melting point at 820 Kelvin. Uranium content has been varied from 1-2 percent of the HN content, the rest being Thorium.



Figure 5: Melting point vs HN fraction

The fuel salt flows through the vertical channels and out to a heat exchanger. A small part of the flow is diverted to a reprocessing unit that separates out ²³³Pa and fission products. Minor actinides can be separated in the reprocessing unit, but due to the low concentrations of minor actinides, that has been neglected in this work. Overall the rational for the selection of most parameters has been simplicity of calculations based on rough estimates of what might be plausible. Many of the selections might be unfeasible since no material properties has been taken into account, except the properties that has a direct influence of neutronic behavior. The size of the system is arbitrarily selected between the limits of being so small that neutron leakage becomes a significant effect and so large that the system becomes unfeasible. Temperature is selected to give some margin above the melting point of the salt. Graphite is selected as piping material due to being compatible both with the chemical properties of the salt and the high temperature of the system, the thickness of graphite in the pipes is an arbitrary choice.

Table 1. Summary THUNDER.

| Power | 530 MWe |
|---------------------------------|--------------------|
| Salt composition | $LiF-(22\% HN)F_4$ |
| Reprocessing time | One month |
| Salt temperature | 900 K |
| Calandria diameter | 230 cm |
| Calandria length | 500 cm |
| Fuel channel radius | 11.5 cm |
| Fuel channel pitch | $22.5~\mathrm{cm}$ |
| Graphite channel wall thickness | $0.95~\mathrm{cm}$ |
| Gas annulus thickness | $1 \mathrm{cm}$ |
| Steel tubing thickness | $0.15~\mathrm{cm}$ |

5.1 Design performance parameters

For THUNDER to be a viable reactor it has to fulfill some basic performance parameters. The parameters focused on in this work is. Acceptable reactor safety parameters, efficient conversion of fertile thorium and reasonable fuel reprocessing times. Other aspects that might pose additional, important conditions on the design, like proliferation resistance, waste minimization, minimization of fissile inventory and thermal-hydraulics are however disregarded in this study.

5.1.1 Conversion ratio

The conversion ratio in a pure thorium fuel cycle, with fissile plutonium nuclei omitted since they make a negligible contribution, can be defined as.

$$CR = \frac{\Sigma_c(^{232}Th) - \Sigma_a(^{233}Pa) + \Sigma_a(^{234}U)}{\Sigma_a(^{233}U) + \Sigma_a(^{235}U)}$$
(1)

A conversion ratio less than one implies that more fissile nuclei are consumed than what is produced by transforming fertile nuclei. In such a case fissile nuclei has to be continuously fed into the core to keep it critical. A conversion ratio greater than one implies that more fissile nuclei are created than what is consumed, no external input of fissile nuclei is needed and the reactor can be fuelled only with fertile nuclei. The excess fissile nuclei created can be extracted and used as fuel in other reactors. A condition on THUNDER is a conversion ratio equal to or greater than one to fulfill the goal of effective resource utilization.

5.1.2 Reactor safety

The safety parameter studied in this work is the temperature reactivity coefficient, $\frac{dK}{dT}$. Other safety parameters connected to heat flow, fuel flow, loss of delayed neutrons due to fluid fuel etc has not been examined. The temperature reactivity coefficient determines how the neutron multiplication factor, K, behaves when the

temperature of the reactor changes. A positive temperature reactivity coefficient implies that the neutron population and thus the power will increase with increasing temperature, a very unstable condition to be in since a temperature increase can cause a runaway chain reaction. A negative temperature reactivity coefficient on the other hand implies that the power will decrease with increasing temperature, the decrease in power will decrease temperature and thus the reactor is self regulating and inherently safe.

The coefficient can approximately be split into partial additive coefficients, each connected to a specific parameter that changes with temperature. In this case there are two dominant parameters, thermal expansion of the fuel salt and doppler broadening.

$$\frac{dK}{dT}_{total} = \frac{dK}{dT}_{doppler} + \frac{dK}{dT}_{salt\ expansion}$$
(2)

A condition on THUNDER is a negative total temperature reactivity coefficient. In cases where the different reactivity coefficients are associated with different time spans it might be a requirement that the individual reactivity coefficients are negative by themself. But in the case of THUNDER both the doppler and fuel salt expansion coefficient are connected to fast processes and such a stringent condition is not neccesary.

5.1.3 Fuel reprocessing

No closer look into the details of chemical reprocessing has been done in this work. It is generally believed that a slower reprocessing is more feasible. In the original MSBR program at Oak Ridge the envisioned reprocessing speed was to reprocess the entire salt volume once every 10 days, today that is considered unfeasible¹⁴. One condition on THUNDER is thus to be able to have a conversion ratio above one with a reprocessing speed no faster than the core volume once every month. All elements reprocessed are assumed to have the same reprocessing speed, except the nobel gases that are assumed to be removed in a few seconds.

6 Methods and tools

The aim of this work is to examine the neutronics of the THUNDER reactor, the standard neutron transport code MCNP has been used to calculate criticality and find reaction rates. Burnup and reprocessing calculations has been done with the code MURE developed by Olivier Meplan and others at LPSC, Grenoble¹⁵.

The main focus has been to examine the behavior of one single cell with neutron reflecting boundary conditions. The cell consist of a fuel channel, the graphite tubing and a square section of the D2O moderator with a side length equal to the pitch. Since the reflecting boundary conditions eliminate leakage, all values of Keff and conversion ratio are a bit higher than would be the case in a more realistic model. If the core is large the difference should not be very large. However, when calculating temperature reactivity coefficients the reflecting boundary conditions gives unphysical results since the expansion coefficient is very sensitive to neutron leakage. For this reason a whole core made up of a lattice of cells was used to calculate temperature reactivity coefficients.

6.1 MURE and MCNP

MURE(MCNP Utility for Reactor Evolution) is a c++ based reactor evaluation program developed by Olivier Meplan and others at LPSC, Grenoble ¹⁶. MURE couples to MCNP¹⁷ to perform burnup and evolution calculations between MCNP criticality calculations. MURE is capable of simulating reactor evolution under various different conditions. Most importantly for this work is MUREs ability to simulate quasi continuous reprocessing, a requirement for MSR evolution.

After creating an input file for MURE consisting of materials, geometry, cells and evolutions options an executable is compiled that automatically creates MCNP input files, run MCNP, extract data from MCNP output files, perform evolution of the materials based on a Runge Kutta method and finally includes the evolved materials into a new MCNP input file and repeaters the process over the wanted evolution time period.



Figure 6: General scheme of a MURE evolution

MURE is a work in progress and therefore, perhaps not surprisingly, we ran into some troubles with the installation and some methods in the program. The ability to control K_{eff} during the evolution by adjusting the concentrations of fissile isotopes was unfortunately not working in the version used, description on how to construct own control methods are included with the MURE package but due to time constraints no attempt was made to do so. Overall MURE is a very powerful program that is perfectly suited for MSR calculations.

The inability to control fissile concentration creates problems for burnup calculations. The conversion ratio is highly dependent on how fast protactinium-233 is removed from the core. The faster protactinium-233 is removed less of it will be destroyed by neutron capture and more will have a chance to decay into uranium-233. In reality the uranium- 233 that is formed by decay outside of the core is then feed back into the core. However, since there is no working method in MURE to insert fissile material into the core during burnup calculations, faster removal of Pa will mean less uranium-233 gets created in the core and K_{eff} drops much faster than it would in a realistic case.

The problem forced us to construct an alternative ad hoc method. Every evolution is started with a large excess reactivity, K_{eff} is allowed to drop below 1 due to fissile material depletion. When Keff has dropped to around 0.95 evolution is stopped, all nuclei of significant concentrations is extracted, put into a new MURE input file along with an extra addition of uranium-233 and another evolution is started. This is repeated until the desired evolution time has passed. This should be a reasonable approximation of the real behavior.



Figure 7: Keff over time

Conversion ratio is then calculated from the last evolution as the average of the conversion ratios at different time steps where Keff on average is slightly above one. This should give a good approximation of the conversion ratio when the system is close to equilibrium.

6.2 Parametric design study

The impact of the most important design parameters on breeding capabilities and reactor safety features of the THUNDER have been calculated. Table 2 summarises the parameter ranges examined. Not all effects on the performance parameters due to changes in the above parameters has been examined. Tabled 3 outlines what combination of parameters and effects has been examined. In each case the parameter in question is varied, while all other parameters is held constant at the values given in table 1 in the "'THUNDER - the concept" chapter.

The **Fuel channel radius** has a large impact on the neutron spectrum. When modifying fuel channel radius the pitch has been kept constant. This makes the fuel channel radius the sole determinant of the moderator to fuel ratio, a larger channel lowers the ratio and makes the neutron spectrum harder. Radius between 5-18 cm has been examined

The **Pitch** has a similar effect as fuel channel radius, when the pitch is decreased the spectrum becomes harder. Pitch between 14- 21 cm has been examined

The **Reprocessing time** has a large impact on conversion ratio, faster reprocessing giving a higher conversion ratio. At longer reprocessing times the main effect is from reducing parasitic neutron absorbers, at short reprocessing times the dominant effect is from having more and more Pa out of the core. Changes in reprocessing time is predicted to have a very small impact on temperature reactivity coefficients and that effect has not been examined closer. The following reprocessing times has been examined, 1 week, 1 month, 6 months and one year.

The **fuel salt composition** has a small impact on the hardness of the neutron spectrum. Its effect on temperature reactivity coefficients is negligible. HN fractions from 16 percent to 28 percent has been examined.

| Tuble 2. Design parameter range. | | |
|----------------------------------|---|--|
| Fuel channel radius | 5 - 18 cm with pitch fixed at 22.5 cm | |
| Fuel channel pitch | 14 - 22.5 cm with radius fixed at 11.5 cm | |
| Reprocessing times | 1 week, 1 month, 6 month, 1 year | |
| HN mole $\%$ | 16 - 28% | |

Table 2. Design parameter range.

Table 3. Combination of examined effects and parameters

| | Expansion coefficient | Doppler coefficient | Conversion ratio |
|---------------------|-----------------------|---------------------|------------------|
| Fuel channel radius | Х | Х | Х |
| Fuel channel pitch | Х | Х | Х |
| Reprocessing time | - | - | Х |
| HN mole $\%$ | Х | Х | - |

7 Results

In this chapter, the effect of fuel channel radius, fuel channel pitch, reprocessing time and HN fraction on temperature feedback coefficients, conversion ratio and neutron spectrum is shown in graphs. A complete evolution series has not been performed for each radius and pitch in the graphs below, instead equilibrium concentrations from the standard design parameters has been used as salt composition. Then radius/pitch has been varied and the differences in reaction rates has been used to calculate the conversion ratio. Therefore the results concerning conversion ratio should be considered qualitative, not quantitative. The margin of error on all temperature reactivity coefficients is on the order of 0.2 pcm/K. The erratic behavior of most of the graphs are due to statistical fluctuations in the MCNP calculations.

7.1 Effect of fuel channel radius

The effects the fuel channel radius has on temperature reactivity coefficient, conversion ratio and spectrum is shown in the graphs below. The positive expansion coefficient is due to the thermalizing effect on the neutron spectrum of the fuel expansion. When the fuel expands more fast neutrons will reach the moderator before being absorbed in the fuel. The fast neutrons that reach the moderator returns to the fuel as thermal neutrons, leading to a more thermalized spectrum and higher reactivity. The larger the fuel channel the harder the spectrum is and this makes it more sensitive to additional thermal neutrons. For small radius the spectrum is already so well thermalized that additional thermal neutrons has low influence. The fuel expansion also leads to more leakage of neutrons out of the core reducing reactivity, but this effect is much smaller.

It is seen that the total temperature reactivity coefficient is negative for small and large fuel channel radius. Radius between 8 to 14 cm has positive total coefficient. The doppler coefficient becomes increasingly negative with increased radius, but it is almost completely canceled by the increasingly positive expansion coefficient.

The conversion ratio is improved with increasing channel radius but flattens out when the radius becomes greater than 10 centimeters.



Figure 8: Fuel channel radius vs feedback coefficients, conversion ratio and spectrum





Figure 9: Pitch vs feedback coefficients, conversion ratio and spectrum

The effect on the temperature reactivity coefficients of increasing the pitch is similar to the effect of decreasing the radius. In both cases the spectrum becomes softer and this seems to make both the expansion and doppler coefficients smaller in magnitude. Other effects that probably effect the reactivity coefficients is the increasing homogenization of the core with decreasing pitch(or increasing radius). A more homogeneous core has a lower resonance escape probability and this increase the doppler coefficient since the doppler coefficient is due to the broadening of a capture resonance in thorium.

Surprisingly no effect on the conversion ratio with changes in pitch can be seen, aa increase in conversion ratio with decreasing pitch was expected since it makes the neutron spectrum harder. No explanation for the fairly constant conversion ratio with changes in pitch can be presented at this time.

7.3 Effect of HN fraction

The effect of different heavy nuclide fractions on the temperature reactivity coefficients seems small if not negligible entirely. The influence on the spectrum hardness is noticeable but weak, increasing HN fraction makes the neutron spectrum harder. The explanation is probably the decrease in fluorine concentration with increasing heavy nuclide concentration, fluorine is a weak moderator.



Figure 10: Heavy nuclide fraction in the fuel salt vs feedback coefficients and spectrum

7.4 Effect of Reprocessing time

The graphs below show the influence reprocessing time has on conversion ratio and K_{eff} without protactinium-233 removal. Without protactinium-233 removal reprocessing time only influence conversion ratio by determining concentrations of fission products, and to a lesser extent, transuranics. Lower fission product concentrations leads to less parasitic neutron absorption and higher conversion ratio and K_{eff} . It can be seen that the effect on conversion ratio is not very large. The effect on K_{eff} is significant. Shorter reprocessing times greatly increase K_{eff} at equilibrium. Only the case with reprocessing time of one week leads to an equilibrium state with K_{eff} larger than 1. This implies that if fission products are quickly removed from the fuel salt, then conversion ratios above one might be possible without protactinium-233 removal. That is desirable from a proliferation point of view. Separated protactinium-233 decays into pure uranium-233 which is a very efficient weapons material.



Figure 11: Time evolution of Keff and conversion ratio with different reprocessing times

7.5 Optimal choice of parameters

The optimal configuration found in this work for THUNDER is presented in the table below. Based on the results presented in the previous chapter a set of parameters has been selected that gives the highest conversion ratio while keeping the total temperature reactivity coefficient negative. It should be said that this is probably not the best combination of parameters that can be found, it is just the best combination found so far through trial and error.

| Fuel channel radius | 11.5 cm |
|-------------------------------|--------------------|
| Fuel channel pitch | $16.5~\mathrm{cm}$ |
| HN mole fraction | 17% |
| Uranium fraction of HN | 1.8% |
| Fraction of thermal fission | 75% |
| Total temperature coefficient | -0.8 pcm/K |
| Doppler coefficient | -3.1 pcm/K |
| Expansion coefficient | 2.2 pcm/K |
| | |

Table 4. Optimal design and its temperature reactivity coefficients

7.6 Comparison between reflectors

Two way to construct the reflector for the THUNDER core is considered. In one case the calandria diameter is expanded and the extra heavy water is used as a reflector. In the second case the calandria is surrounded by graphite slabs. The feedback coefficients for a fuel temperature increase for the two cases is shown in table 5.

Table 5. Comparison between reflector materials

| | D2O | Graphite |
|-------------------------------|-------------|-------------|
| Doppler coefficient | -2.6 pcm/K | -3.1 pcm/K |
| Expansion coefficient | 2.3 pcm/K | 2.2 pcm/K |
| Total temperature coefficient | -0.3 pcm/K | -0.77 pcm/K |

7.7 Comparison between moderator materials

To get a sense for what difference there is between D2O and graphite with regards to basic reactor parameters D2O has been replaced with graphite in the THUN-DER design. However, if nothing is done except to replace D2O with graphite the reactor is not critical and the neutron spectrum becomes much harder. Therefore the pitch has been increased in the graphite case to soften the spectrum and the uranium concentration has been increased to achieve criticality. As a further comparison the conversion ratio and temperature coefficients for the unmoderated TMSR molten salt reactor developed by LPSC(ref 11) is included. The fields for the TMSR marked with - is due to the fact that all design data isn't available for the TMSR and some parameters are not applicable to the TMSR design.

| TMSR | | | |
|-------------------------------|---------------------|-------------------|--------------|
| | D2O | Graphite | TMSR |
| Fuel channel radius | 11 cm | 11 cm | - |
| Fuel channel pitch | $16.5 \mathrm{~cm}$ | $21 \mathrm{~cm}$ | - |
| Reprocessing time | 1 week | 1 week | 10 days |
| HN mole fraction | 17% | 17% | - |
| Uranium fraction of HN | 1.7% | 2.14% | - |
| Fraction of thermal fission | 75% | 73% | - |
| Conversion ratio | 1.06 | 1.02 | 1.062 |
| Doppler coefficient | -3.1 pcm/K | -2.2 pcm/K | |
| Expansion coefficient | 2.2 pcm/K | 2.4 pcm/K | |
| Total temperature coefficient | -0.77 pcm/K | -0.36 pcm/K | -2.25 pcm/K |

 Table 6. Comparison between D2O and graphite moderated THUNDER and

 TMSR

7.8 Protactinium-233 vs reprocessing time

Complete evolution series has been made with the optimal design for three different reprocessing times, 1 week, 1 month and 6 months. Unfortunately an as of yet unknown error in MURE makes the results useless. As can be seen in the graphs below the time behavior for the concentration of several nuclides in the case of one week reprocessing time is very erratic. The protactinium-233 concentration is also highest for the shortest reprocessing times. Those results are clearly unphysical and has to be discounted.



Figure 12: Nuclide concentrations vs reprocessing time

Unfortunately this means any conversion ratio calculated based on these evolutions are practically useless. To approximate how the conversion ration should depend on protactinium-233 removal times a analytic treatment has been done instead to estimate protactinium-233 concentrations. To a first approximation the protactinium-233 concentration should behave as

$$\frac{dN_{Pa}}{dT} = -\frac{N_{Pa}t_{eff}}{ln2} - N_{Pa}\sigma_{abs}\phi + S \tag{3}$$

Where σ_{abs} is the absorption cross section for protactinium-233, S is protactinium-233 production rate, t_{eff} is effective half life. The effective half life is determined by the radioactive half life of the nuclei and the reprocessing time.

$$\frac{1}{t_{eff}} = \frac{1}{t_{1/2}} + \frac{1}{t_{repro}}$$
(4)

The solution to equation 3 is, if $t \to \infty$

$$N_{Pa}(t) = \frac{S}{\frac{ln2}{t_{eff}} + \sigma_{abs}\phi}$$
(5)

The values of $\sigma_{abs}\phi$ and S are approximately the same for different reprocessing times. Assuming the earlier evolution series without protactinium removal is correct then the values of $\sigma_{abs}\phi$ and S can be calculated since t_{eff} and N_{Pa} are known in those cases. The resulting relation between protactinium-233 concentration and reprocessing time can be seen in the plot below, normalized as fraction of the 6 month reprocessing time concentration. For a reprocessing time of one week and



Figure 13: Protactinium-233 concentration vs reprocessing time

one month the protactinium- 233 concentrations should be roughly 40% and 75% of the 6 months concentration. These values has been inserted into a MCNP file and the conversion ratios has then been calculated from the reaction rates given by MCNP.

Table 6. Comparison between D2O and graphite moderator.

| Reprocessing time | Conversion ratio |
|-------------------|------------------|
| 1 week | 1.06 |
| 1 months | 1.017 |
| 6 months | 0.90 |

8 Conclusions

From the above results THUNDER looks like a promising concept without any fatal flaws, further study is needed to clarify some issues. The most important conclusions are.

- Conversion ratio is better for heavy water moderation, conversion ratio seems to be almost as good as for the unmoderated molten salt reactor. However the result is based on the analytic treatment of protactinium concentration so it needs to be validated.
- Safety parameters appears to be better for heavy water moderation, but the margins of error in the feedback coefficients are to large for it to be possible to draw a final conclusion.
- The unmoderated TMSR is superior in safety parameters, its major drawback is the need for a larger fissile inventory since the spectrum is in the epithermal-hard range.
- The graphite moderated reactor require a roughly 25% larger fissile inventory due to larger neutron losses in the moderator.

From a neutronics point of view THUNDER is roughly equal to graphite moderated molten salt reactors, in addition THUNDER has some structural, and possibly economic, advantages that warrants further studies on the concept. The most attractive features of THUNDER is the ease by which fuel channels can be replaced compared to the large effort needed to change moderator block in a traditional molten salt reactor. THUNDER is modular by design, the reactor is composed of identical fuel channels that can be mass produced in factories. Changing the size of the reactor only means changing the number of fuel channels in the core, making THUNDER very flexible. Heavy water moderation has the added advantage that the creation of large amounts of radioactive graphite waste is avoided.

9 Future work

Since this is a first look at a new reactor concept there are plenty of areas that needs to be investigated further.

- Due to errors in MURE a more detailed study of how protactinium reprocessing effects the system is needed.
- A better understanding of how the different parameters effect the temperature reactivity coefficients would be needed to improve the safety parameters. An elongated core could for instance possibly shrink the positive fuel expansion coefficient but at the cost of worse conversion ratio. Different pitches and fuel channel radius at different zones of the reactor might be another way to improve safety parameters.
- A look at how sensitive the conversion ratio is to different power densities is needed, it can be done either by increasing the size of the core or the total amount of salt.
- A two fluid system with separate fertile and fissile salts improves the safety parameters and conversion ratio in the standard graphite moderated MSR. A detailed investigation into a two fluid THUNDER system would be interesting.
- Other salt compositions should be investigated, especially LiF-BeF₂-(HN)F₄ but also more exotic salts. Since THUNDER doesn't use hastalloy as a structural material salt compositions excluded from MSR research because of hastalloy incompatibility should be examined.
- The impact the loss of delayed neutrons have on reactor kinetics, due to the flowing fuel salt, should be investigated.

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References

¹Generation IV forum http://nuclear.inl.gov/gen4/

²Norwegian government Thorium report committee, Thorium as an energy source

³V. Jagannathan et al, Energy Conversion and Management, April 2008

⁴G. L. Olson et al , Fuel Summary Report: Shippingport Light Water Breeder Reactor - Rev. INEEL/EXT-98-00799, Rev. 2 September 2002

⁵World Nuclear Association's information paper on Thorium http://www.worldnuclear.org/info/inf62.html

⁶R. C. Briant and Alvin M. Weinberg, Molten Fluorides as Power Reactor Fuels, Nuclear Science and Engineering, 2, 797-803 (1957)

⁷E. S. Bettis et al, The Aircraft Reactor Experiment-Design and Construction, Nuclear Science and Engineering 2, 804-825 (1957)

⁸W. K. Ergen et al, The Aircraft Reactor Experiment-Physics, Nuclear Science and Engineering 2, 826-840 (1957)

⁹E.S Bettis et al, The Aircraft Reactor Experiment-Operation, Nuclear Science and Engineering 2, 841-853 (1957)

¹⁰R. C. Robertson, MSRE Design and Operations Report Part I: Description of Reactor Design, ORNL-TM-0728, 1965-01

¹¹Haubenreich et al, Experience with the Molten Salt Reactor Experiment, Nuclear Applications and Technology 8, 118136, 1970

¹²L. Mathieu et al, The thorium molten salt reactor: Moving on from the MSBR,

Progress in Nuclear Energy, 48, Issue 7, 664-679, 2006

¹³http://gif.inel.gov/roadmap/pdfs/gen_iv_roadmap.pdf

¹⁴L. Mathieu et al, The thorium molten salt reactor: Moving on from the MSBR,

Progress in Nuclear Energy, 48, Issue 7, 664-679, 2006

¹⁵Olivier Meplan et al MURE : MCNP utility for reactor evaluation, ENC 2005,

 $http://democrite.in2p3.fr/action/open_file.php?url = http://democrite.in2p3.fr/docs/00/05/39/11/PDF 53911$

 $0 \mathrm{pt}$

¹⁶Olivier Meplan et al MURE : MCNP utility for reactor evaluation, ENC 2005,

 $http://democrite.in2p3.fr/action/open_file.php?url = http://democrite.in2p3.fr/docs/00/05/39/11/PDF 53911$

 $^{17} \rm http://mcnp-green.lanl.gov/$

The place to look for almost every major historic article ever published on Molten Salt Reactor technology is Kirk Sorensens document repository. http://www.energyfromthorium.com/pdf/