Thorium as fuel in molten salt reactors

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1 Abstract

Thorium molten salt reactors offer significant advantages over the currently operational uranium pressurized water reactors. They are safer due to their negative feedback mechanism, lower core radioactivity and fewer long lived radioactive waste elements. The reactor design uses a molten salt of LiF with the fuel ThF_4 dissolved in it. The use of a liquid fuel-salt mixture allows continuous processing of the fuel to remove neutron poisoning isotopes and waste products through fluoride volatization and electrochemical extraction. The difficulty of designing and implementing this process reliably is the main obstacle towards commercial thorium molten salt reactors.

2 Introduction

Thorium has recently often been named as a potential alternative for uranium fueled nuclear reactors. One possibility is using thorium in a light water reactor, but in this paper, the use of thorium in a molten salt reactor will be treated. After a section on thorium as a natural actinide, the practical design and the design considerations of a thorium molten salt reactor (TMSR) will be discussed, a short overview and history of research on TMSRs is given and the author's personal prediction for the future of the TMSR.

3 Thorium mining and extraction

Together with uranium and protactinium, thorium is one of the only naturally occurring actinides, due to the long half-life time of its main isotope. Relatively speaking, thorium is quite abundant on earth, more so than uranium, and almost exclusively in the form of fertile ^{232}Th. For the continental earth crust, a 9.6 ppm concentration of thorium, and a 2.7 ppm concentration for uranium is estimated.[1] Like uranium, a large part of the earth's thorium reserves are dissolved in sea water, however for thorium, the fraction is very low, at 0.01 parts per billion, compared to 3 parts per billion for uranium.[6] All thorium isotopes are radioactive, and its main isotope, ^{232}Th, has by far the longest half life (14 billion years) of all isotopes of thorium. Thorium was first identified as a new element by Jöns Jacob Berzelius in Sweden.[2] It has never had widespread applications, but some niche products have used thorium or thorium oxide owing to its optical properties and the high melting point of the oxide. Because all thorium isotopes are radioactive, nowadays thorium is only used for applications utilizing its radioactive properties.[3]

There are 60 known minerals that contain thorium in some concentration, summing up to 6.2 million tons in thorium reserves on earth. Countries with some of the largest thorium reserves are India, the United States, Australia and Brazil.[3] Thorium is mainly extracted from a mineral called monazite, $(Ce, La, Nd, Th)PO_4$, containing 10% thorium.[4] Another raw material that can be used to obtain pure thorium is thorite $(Th, U)SiO_4$.[5] Less significant thorium containing minerals are thorianite (ThO_2) and bastnasite ($(Ce, La)CO_3F$ with 0.2 weight% thorium oxide contents).[6 7] As currently, thorium is not used on a large scale, there are no operations solely focused on thorium extraction. For larger scale thorium production, thorite would be a better source of

Isotope	Half-life
^{227}Th	18.68 days
^{228}Th	1.9116 years
^{229}Th	7917 years
^{230}Th	75400 years
^{231}Th	25.5 hours
^{232}Th	14.05 billion years
^{234}Th	24.1 days

Table 1: Isotopes of thorium and their corresponding half-lifes. Source: `https://en.wikipedia.org/wiki/Isotopes_of_thorium`

thorium than monazite.

To extract thorium from monazite, a process called *alkaline cracking* is used. Monazite powder is mixed with an highly concentrated aqueous NaOH solution at 240 °C for several hours. This leads to the formation of components such as $Th(OH) \cdot H_2O$ and Na_3PO_4. The next step is to boil this mixture with concentrated hydrochloric acid, which will dissolve the lanthanides (Ce, La, Nd), and leaving solid $ThO_2 \cdot H_2O$.[8]

Another process used for extraction from monazite is acid cracking, while the rare earth elements are dissolved in sulfuric acid, the thorium is converted into a solid compound:[7]

$$ThO_2 + 2\,H_2SO_4 \longrightarrow Th(SO_4)_2 + 2\,H_2O$$
$$Th(SO_4)_2 + H_4P_2O_7 \longrightarrow ThP_2O_7 + 2\,H_2SO_4$$

Yet another way for separating thorium from rare earth elements is by making use of their different precipitation pH. At low pH values (2.5 - 5.5), thorium will precipitate, but the rare earth elements and uranium will not.[7]

4 Thorium fuel cycle

Thorium itself is nonfissile. By converting it into $^{233}_{92}$U through neutron absorbtion, it becomes a fissile atom. The thorium fuel cycle starts with the the isotope $^{232}_{90}$Th. By capturing a neutron in the reactor, this transforms into $^{233}_{90}$T, which is very unstable,

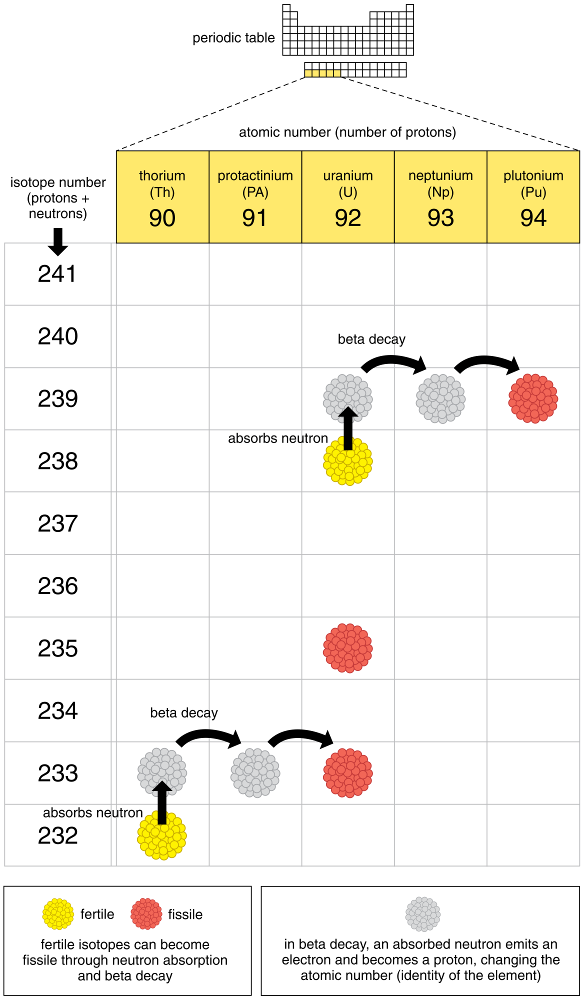

Figure 1: Thorium-232 decay chain via uranium-233[9]

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having a half-life of 21.83 minutes. Therefore it will quickly transform by beta decay into ${}^{233}_{91}$Pa which has a half-life of 27 days. By another beta decay step, this becomes ${}^{233}_{92}$U, a fissile isotope.

The main advantage of thorium as a nuclear fuel is in it's decay chain as shown in figure 1. The conventional ^{235}U fuel cycle produces long-lived radioactive fission products as waste, such as ^{239}Pu. In contrast, in a thorium molten salt reactor, no long lived ^{239}Pu is produced, and other radioactive actinides are produced in lower numbers.[10 9] This makes the waste of thorium nuclear reactors radioactive on a length scale of a few hundred years, compared to ten thousands of years for used uranium fuel. Another advantage of the thorium fuel cycle is that ^{233}U has a high neutron reproduction factor, and it is therefore easier to keep a chain fission reaction going.[11]

In comparison to ^{235}U, the advantage of thorium is its abundance, and almost all of the natural occuring thorium is the fertile isotope, suitable for power generation. Compared to ^{238}U, which is also fertile, the advantage of thorium is that no ^{239}Pu is produced in the reactor, which limits nuclear proliferation.[12]

Unlike the uranium fuel cycle, the thorium cycle does not produce any isotopes that are suitable for nuclear weapons of mass destruction. Although ^{233}U is a suitable isotope for making these weapons, ^{233}U always co-exists with some ^{232}U in the thorium cycle. The latter has decay products, especially ^{208}Tl that emit strong gamma radiation (2.6 MeV). This makes the material very dangerous to handle, and therefore unsuitable for weapons production. A thorium reactor should thus have shielding to protect the environment from this radiation from the fuel mixture.[13]

There are also some caveats in the fuel cycle of thorium. One is that ^{233}Pa is quite radioactive on the timescale of weeks and months, and is therefore a danger to safety and environment on the short term after a thorium reactor is stopped (during continuous operation the protactinium is fed back and fissioned into other decay products).[11]

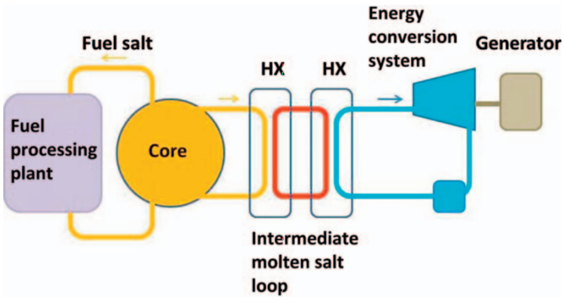


Figure 2: Schematic overview of a thorium reactor design with all main components.[12]

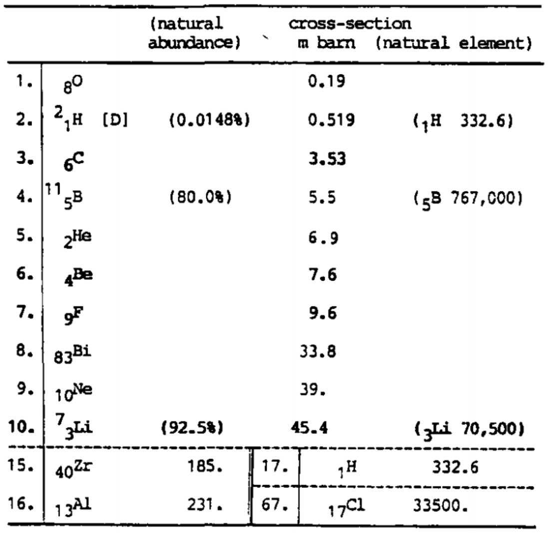
5 Practical design

Molten-salt-fueled reactors differ significantly from conventional water cooled reactors. In this section, the relevant components of a molten salt reactor design, which make it differ from conventional power plants, are discussed.

5.1 Carrier salts

The choice of carrier salt depends on solubility of the actinide compounds and fission products, the melting temperature should be low enough, on its reactivity and finally on it's neutron capture diameter, for moderation purposes. That is why the light lithium salts are excellent carrier salts. All lithium salts have to be enriched to remove any ^{6}Li contents, which is a good neutron absorber, leaving only ^{7}Li, which has a smaller neutron capture cross-section.[14] In practice the carrier salt that is often used is ${}^{7}\mathrm{LiF-BeF_8}$, or 'Flibe'.[15]

The choice of fluoride salts is explained by their good stability at the high operating temperatures ($650\,^{\circ}\mathrm{C}$ - $750\,^{\circ}\mathrm{C}$),[12] as well as nuclear stability in the radioactive environment of the reactor.[16] For example, chlorine has many similar properties to fluorine, such as it's bonding abilities with actinides and the high boiling point (and lower meling point) of chloride salts, but under influence of radioactivity the natural ^{35}Cl and ^{37}Cl will form radioactive ^{36}Cl,



		(natural abundance)	cross-section m barn	(natural element)
1.	${}_{8}O$		0.19	
2.	${}^{2}_{1}H$ [D]	(0.0148%)	0.519	(${}_{1}H$ 332.6)
3.	${}_{6}C$		3.53	
4.	${}^{11}_{5}B$	(80.0%)	5.5	(${}_{5}B$ 767,000)
5.	${}_{2}He$		6.9	
6.	${}_{4}Be$		7.6	
7.	${}_{9}F$		9.6	
8.	${}_{83}Bi$		33.8	
9.	${}_{10}Ne$		39.	
10.	${}^{7}_{3}Li$	(92.5%)	45.4	(${}_{3}Li$ 70,500)

15.	${}_{40}Zr$	185.	17.	${}_{1}H$	332.6
16.	${}_{13}Al$	231.	67.	${}_{17}Cl$	33500.

Figure 3: The elements with smallest neutron capture cross sections. Li, Be and F are the smallest elements of these that can form salts.[15]

LiF	78.6%
ThF_4	12.9%
UF_4	3.5%
$TRU-F_3$	5%

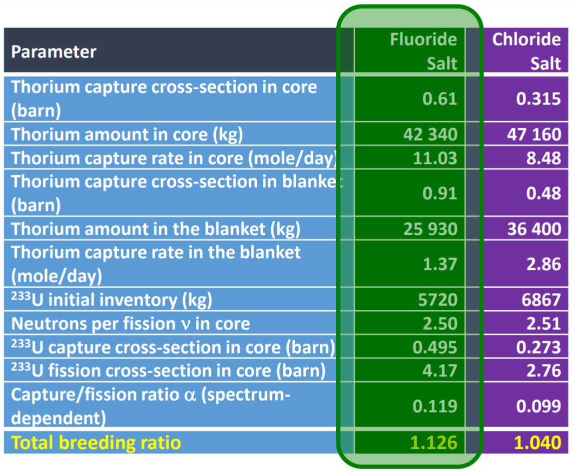
Table 2: Possible fuel-salt composition. TRU stands for transuranium elements Np, Pu, Am etc.[6] Here, the LiF is the carrier salt.

which is not desired. Bromide or iodide based salts have been considered too, but those have weaker bonding and will mix with fission products of their own kind.[17]

The molten salts can corrode metals however, such as pipes and reactor vessels. This issue can be reduced by removing any oxygen compounds in the salt-fuel mixture.[18] A special alloy was developed to withstand the environment within the reactor, Hastelloy-N.

5.2 Fuel processing

The fission processes introduce some components that are detrimental to the operation of the reactor. Those elements are either not well soluble in the salt, or capture neutrons that are important in keeping the nuclear reactions alive.



Parameter	Fluoride Salt	Chloride Salt
Thorium capture cross-section in core (barn)	0.61	0.315
Thorium amount in core (kg)	42 340	47 160
Thorium capture rate in core (mole/day)	11.03	8.48
Thorium capture cross-section in blanke (barn)	0.91	0.48
Thorium amount in the blanket (kg)	25 930	36 400
Thorium capture rate in the blanket (mole/day)	1.37	2.86
${}^{233}U$ initial inventory (kg)	5720	6867
Neutrons per fission ν in core	2.50	2.51
${}^{233}U$ capture cross-section in core (barn)	0.495	0.273
${}^{233}U$ fission cross-section in core (barn)	4.17	2.76
Capture/fission ratio α (spectrum-dependent)	0.119	0.099
Total breeding ratio	1.126	1.040

Figure 4: This figure shows the qualitative advantages that fluoride salts have over chloride salts.[19]

A major advantage of molten salt reactors where the fuel is dissolved in the salt is that the fuel can be reprocessed during plant operation. For helium bubbling, this can be done within the fuel loop, the other reprocessing techniques require part of the salt-fuel mixture to be tapped off to an on-site reprocessing plant.

5.2.1 Helium bubbling

During the fission process, Xe and Kr gas is produced. This is removed using helium bubbling, which can be operated while the fuel is being pumped during operation. This greatly reduces costly downtime of the reactor.[21] The gas bubbles also partially remove small solid dust of noble metals like Nb, Mb and Ru, however, the remaining metal dust remains an issue in test reactors.[12]

5.2.2 Fluorination

Fluorination serves to remove volatile fluorides from the stream. By reaction with fluoride gas, they

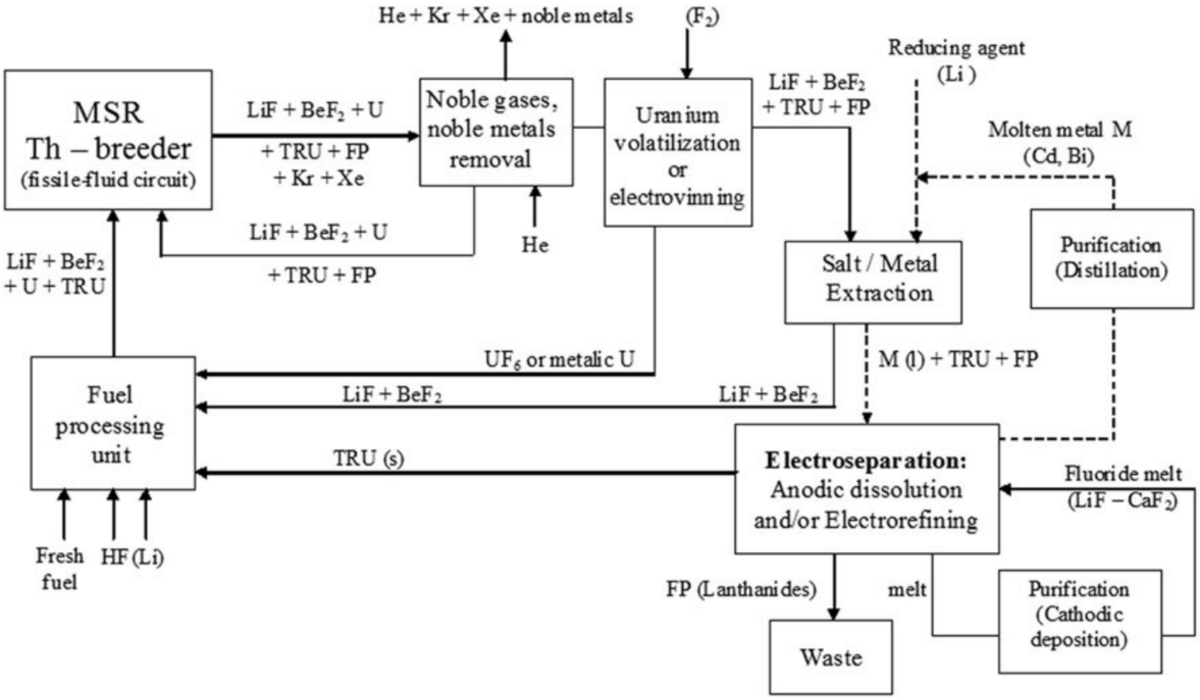


Figure 5: Schematics of the fuel processing plant.[20]

form the gaseous fluorides and can thus be captured, as most other components cannot form gaseous fluoride at low temperatures. By distillation, they can be seperated further.[22] Mainly uranium is removed, in the following reaction scheme:[12]

$$\mathrm{UF_4 + F_2 \longrightarrow UF_6\,(g)}$$

5.2.3 Actinide extraction

Actinides (Am, Cu, Np, Pu) are removed by reduction with mettalic thorium dissolved in bismuth, which serves as a solvent.[21] The reactions are:[23]

$$\begin{gathered}\tfrac{4}{3}\,\mathrm{AcF_3 + Th\,(Bi) \longrightarrow ThF_4} + \tfrac{4}{3}\,\mathrm{Ac\,(Bi)}\\ \mathrm{Ac\,(Bi) + 3\,F^- \longrightarrow AcF_3}\\ \mathrm{ThF_4 \longrightarrow Th + 4\,F^-}\end{gathered}$$

5.2.4 Lanthanide extraction

By electrochemical separation, lanthanides are extracted from the salt. The process is similar to the actinide extraction but with different reaction potential to make use of their difference. The waste products are $\mathrm{LnO_2}$.[21] The total reaction is:[23]

$$\tfrac{4}{3}\,\mathrm{LnF_3 + 2\,H_2O(g)} \longrightarrow \tfrac{2}{3}\,\mathrm{Ln_2O_3 + 4\,HF}$$

5.3 Reactor core

Because the fuel is dissolved in the fluid coolant salt, the reactor core does not need any solid components such as fuel rods or pellets. The high radioactivity does break down metal alloys however. Whereas for the piping of the molten salt cycle, metal alloys have been developed (Hastelloy-N), for the core, a graphite reflector is required.[15] To control the power of the core, the pumping speed, the new thorium influx and the influx of fresh or processed salt is utilized. The heat at which the salt is kept externally (by the heat exchange from the reactor core) is also a way to change the reaction rate, as the salt expands when it heats up, and this reduces the number of fissions. And lastly, helium or other inert gas bubbles can be injected into the core to further reduce the density and the fission rate.[25]

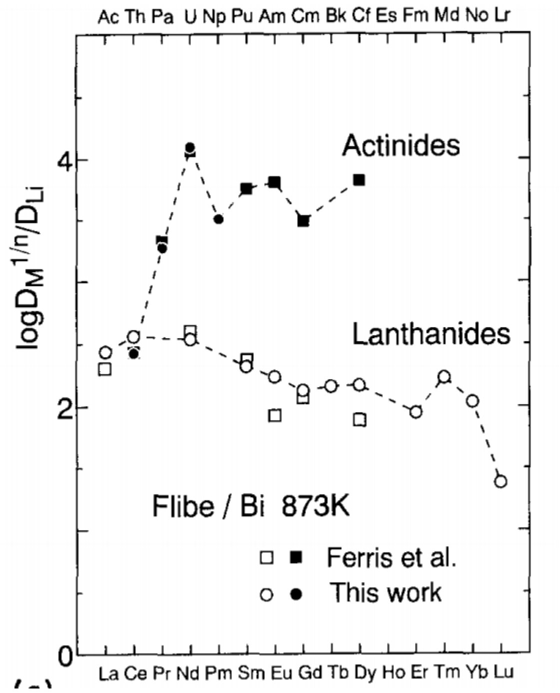


Figure 6: Extractability values for lanthanides and actinides in lithium-beryllium salt.[24] The figure describes that the actinides are reduced from the salt into a metal phase more readily than the lanthanides. This mechanism can be used to remove actinides from the salt-fuel mixture.

5.4 Safety mechanisms

Because the molten salt reactor operates at atmospheric pressure or lower, no risk of explosion exists. The most severe accident scenario would be a leakage of fuel-salt mixture. Since this is not volatile, the risk of contamination of the environment is relatively low. By placing a capture basin below the reactor, it is prevented that the fuel-salt mixture enters the outside world. Without management, the fuel-salt mixture will cool down to solid state by itself.[15]

Another safety feature is the use of a freeze plug. At the lowest point of the fuel loop, a side channel leading to capture tanks is present. The entrance of the side channel is kept blocked by a solid block of salt, kept frozen using external equipment. Should the power fail, the salt will melt and the fuel-salt mixture will drop into the capturing tanks.[?] [?] This also shows that a meltdown scenario is not possible for the TMSR.

6 Development locations and pilot plants

There are, and have not been any (commercial) TMSRs as described in this paper in operation, but a number of test setups that mimic some parts of a TMSR design have been built.

A first test molten salt reactor, not using thorium, was operating in 1954 at the Oak Ridge National Laboratory (ORNL) in Tennessee.[16] The first working concept 'Aircraft Reactor Experiment' showcases that the ultimate goal of this project was to develop a nuclear reactor to power aircraft. The plant operated only as a burner plant, using ^{233}U directly, produced from thorium externally.[26] A larger scale reactor operated at ORNL between 1966 and 1969 without any significant incidents. This reactor implemented gas bubbling to process the fuel continuously.[27] The research was eventually abandoned because the system was not suitable to be used in aircraft.[28] At the time, pressurized water nuclear reactors had become commercial already, and were not in need for replacement. In 2006, the research reports of the ORNL on its molten salt reactor experiments were declassified, which was a catalyst for new interest in molten salt reactor development. The ORNL experiments have been of massive importance to all following research and are still relevant today. However, due to changing research standards and lack of insight in the research methods of ORNL in the 1960s, eventually all of this research will have to be repeated in some form.[26]

SALIENT (Salt Irradiation Experiment) is a research setup started in 2017 in Petten, The Netherlands, where a very small quantity of LiF/ThF mixture is being heated and kept in the high neutron flux reactor. This causes the salt to melt and the thorium fuel cycle to start. The main aim of this setup is to study how the mixture behaves as the nuclear reactions occur. Corrosion of possible material for piping in a molten salt reactor is also studied.[29]

The Nuclear Research Institute in Řež, Czech Republic, is active in research on the fluorination process and lanthanide extraction.[20]

In Grenoble, France, the Laboratory of Subatomic Physics & Cosmology (LSPC) has a research group occupied with molten salt reactor physics.[30] [19] [25]

SAMOFAR (Safety Assessment of a MOlten salt FAst Reactor) is a joint project of European universities and research laboratory, led by TU Delft, to research the safety features of the TMSR, such as the freeze plug, suitable materials to hold the salt, lanthanide and actinide extraction chemistry and fluid dynamics of the salt.[19] [25]

Very recently (December 2017), China announced US$3.3 billion of investments in a large-scale molten salt reactor experiment.[28] [27]

As can be noted, between the end of the ORNL experiments and a few years ago, little experimental progress has been made on molten salt reactors, not because of technological limitations, but due to reduced research interest, which led to lack of funding and research groups moved on to other topics.

6.1 Conclusion

The thorium molten salt reactor offers the promise of many advantages over current commercial nuclear reactors. Most importantly it offers greatly improved operational safety by design. There is no radioactive material under high pressure, and fissile products are recycled and removed during operation, such that better up-times and more efficient operation is possible. Other advantages are the low operating pressure, reducing explosion risk, and the higher operating temperature, which can result in a higher thermodynamic efficiency.

The molten salt loop includes many safety features.

- New fuel influx can be stopped at any time, as new fuel is added continuously in the form of dissolved thorium.
- Molten salt expands as it heats up, so a runaway reaction is prevented. As the salt's density decreased, so do the number of fission events.
- Molten salt does not have to be pressurized at high operating temperatures, removing the risk of explosions.
- The salt-fuel mixture can be drained to a holding tank at any moment, this can even be done passively in case of a power-loss incident, by using a freeze plug. The equivalent of a meltdown does not exist for molten salt reactors, as molten salt will cool down without fuel management.

The combination with thorium as a fuel offers further advantages:

- Waste products of the thorium cycle are radioactive on a relatively short timescale of a few hundred years.
- Thorium, it's decay products and their fissile products, are not suitable for production of nuclear weapons.

The main obstacle is the practical operation of the fuel processing plant. There is still more experimental research needed to test if the removal of various fission products is reliable and safe as the theory describes. A fundamental disadvantage of the TMSR is that the entire fuel loop contains radioactive material and thus becomes radioactive. This includes the processing loop, and piping between the heat exchanger and the core. The result is that maintenance requires large safety precautions or even use of remote machinery.[12] To start a thorium reactor, an initial batch of fissile material is required, which does negate on of the advantages. However, the fissile material production can be limited to a smaller number of higher security breeder reactors, and delivered to smaller scale burner reactors.

It can be concluded that thorium is a very promising fuel for use in molten salt reactors. With no fundamental obstacles in the way, it was more of a fate in history that delayed the TSMR from widespread use today. Only a few decades were between the discovery of nuclear fission and the first commercial nuclear reactors. With no fundamental technological issues, it is not unthinkable that new interest in molten salt reactors could result in a commercial molten salt reactor in another few decades away from now. Practical TSMR designs have been worked out in detail, and new, larger scale experimental TSMRs can be expected in the near-term feature. The results of those experiments will decide if the concept of a TMSR will be abandoned again or if it will turn out to be the reactor type the world should have chosen in the 1960s already.

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