‘Petten’ has started world’s first thorium MSR-specific irradiation experiments in 40+ years

On August 10, 2017, a set of concentric sealed tubes was entered into the core of the High Flux Reactor in Petten, Netherlands. In the smallest of these tubes sits an even smaller set of four graphite crucibles, containing a mixture of salts: lithium fluoride and thorium fluoride.

The radiation in the core will provide the heat for the salt sample in the inner tube to melt. Within the salt mixture, due to the presence of thorium in the high neutron flux, over time a fission reaction will start. And although the outer tube is in contact with the hand-warm water of the reactor basin, the concentric set of tubes provides enough insulation to let the temperature of the salt raise to about 620 ^{o}C.

This tiny set of crucibles is the world´s first molten-salt-reactor-related fission experiment in over 45 years. The experiments have been initiated by the Nuclear Research and Consulting Group (NRG) in Petten, Netherlands, and take place at the High Flux Reactor (HFR), the research reactor that NRG operates.

Sander de Groot of NRG explains that the interest for MSR’s at NRG originated while working on several large-scale programs, dedicated to the High Temperature Reactor (HTR) and transmutation of long-living radioactive waste. “There was both internal and external support for our idea to start the SALIENT experiments”, he says. “This is a technology with much potential for large scale energy production. It offers the perspective of a technology suitable for a responsible energy system that is clean, affordable, reliable and safe. It is in principle the safest and most resource efficient solution in nuclear energy. The Netherlands is well positioned to contribute to international R&D towards the realization of the MSR. The program also gave us the opportunity to cooperate with JRC Karlsruhe, a cooperation that we see as very important.”


Aerial view of the research area in Petten, in the north-west of the Netherlands, that houses (amongst others) NRG. The bigger white dot a bit left of the middle of the picture is the dome of the High Flux Reactor (HFR). In the upper left corner, the North Sea. To the right Sander de Groot and Ralph Hania.

Nuclear fuels expert Ralph Hania is the lead scientists at NRG for the SALIENT experiments. For thmsr.nl he explains the experiment, that has its name derived from SALt Irradiation ExperimeNT. And because NRG is planning a series of comparable experiments, this first one is simply called SALIENT-1. The next ones are on the drawing board.

GZ *What would you say is the importance of the SALIENT experiments?*

RH First of all, SALIENT is a way to build up experience with the use of fission fuel in the form of a molten salt. That hasn't been done for decades and we're also doing it to train ourselves. It's important to notice that SALIENT is not a single experiment but really a series, and we'll build that up step by step. On the other hand, it is fundamental research, in which we look at the behavior of salt and the fission products that are formed in the salts.

In this project, we closely cooperate with JRC Karlsruhe. We need highly purified salts for SALIENT, and JRC is one of the very few places in the world that have the expertise to produce them. Here at NRG, we designed the irradiation part of the experiments. After the irradiation, we will send back part of the samples to JRC, and they will be doing several tests, like finding out how stable the fission products are in the salt."

Filling of crucible with sample salt at JRC Karlsruhe (left) and assembly of sample holder at NRG (right)

SALIENT-1: lithium fluoride and thorium fluoride in graphite crucibles

GZ *Why did you choose the lithiumfluoride/thoriumfluoride mixture as a start for your experiments?*

RH That was the salt of choice for the Molten Salt Fast Reactor (MSFR), a European concept for a waste-burning MSR. For SALIENT-02, we will use a different mixture that will also contain beryllium, forming a mixture also known as FliBe, and uranium as the fuel. And we are already considering further experiments, that will focus more on the interaction between the salt and the containment materials. Corrosion resistance is very important for those materials: they should be mechanically strong, and able to resist chemical corrosion and intense radiation. This corrosion resistance will be the next focus of the experiments.

GZ *Could you describe the setup of SALIENT-01?*

RH At the heart of salient 1 are four small crucibles that contain the LiF/ThF mixture. They are placed within a set of concentric steel tubes of about 50 cm high. At the start of the experiment, we will bring this tube in a selected radiation field of the High Flux Reactor. After a while, thorium will be transmuting to uranium and the uranium will start to fission. The salt content of the 4 crucibles is identical at the start, but within one we will place a small nickel sponge, and in another one is a nickel foil. During the fission reaction, fission products will form and a part of these are noble metals. We want to find out if these noble metals precipitate preferentially on the nickel.

GZ *Ah, I see. The forming of noble metals is one aspect that needs to be managed in the salt stream of MSR's. And one proposed way to remove them is the use of nickel sponges. The idea is that the sponge with the noble metal can be replaced periodically…*

RH Yes.

GZ *And the pipes themselves, what are they made of?*

RH Well, of course they must meet all kinds of requirements, but we've made them out of ordinary steel. The idea is to stick to standard materials wherever possible.

GZ *Some of the startups are considering to use steel for their vessels. It's interesting that you use it for the experiments.*

RH Well, as a reactor vessel material the suitability of steel remains to be determined. Corrosion may be a problem there, and we do not yet know if we can manage that by managing the salt chemistry. The high temperatures in MSRs might also be problematic, even if the pressure inside the system is low.

Corrosion tests, including molybdenum alloy, starting at SALIENT 3&4

GZ *Earlier this year I spoke with electrochemist Thomas Steenberg of Copenhagen Atomics. In his view what is needed is adequate management of the redox potential of the salt. If this is done properly, corrosiveness may not be an issue.*

RH He may well be right about that. Even for steel, corrosion may be acceptable, but further investigation is needed. Also, for the purpose of building a reactor, different off-normal situations will have to be covered in addition to normal operation. In later SALIENT tests, the focus shifts from the salt to the interaction between the salt and possible reactor vessel materials, especially corrosion.

GZ *What materials will you be looking at?*

RH For later SALIENT experiments we are planning to do irradiation tests for 316 stainless steel, Hastelloy, the nickel alloy that ORNL used in the 1960s, and TZM – the latter is a titanium/zirconium/molybdenum alloy.

GZ *Wow, that is going to be interesting! I've heard many times that molybdenum may be the ideal reactor material, but nobody has really tested it.*

RH It's true that we could find virtually no experimental data for molybdenum applications in the temperature range in which we intend to test. Neutronically, molybdenum looks much more attractive than nickel.

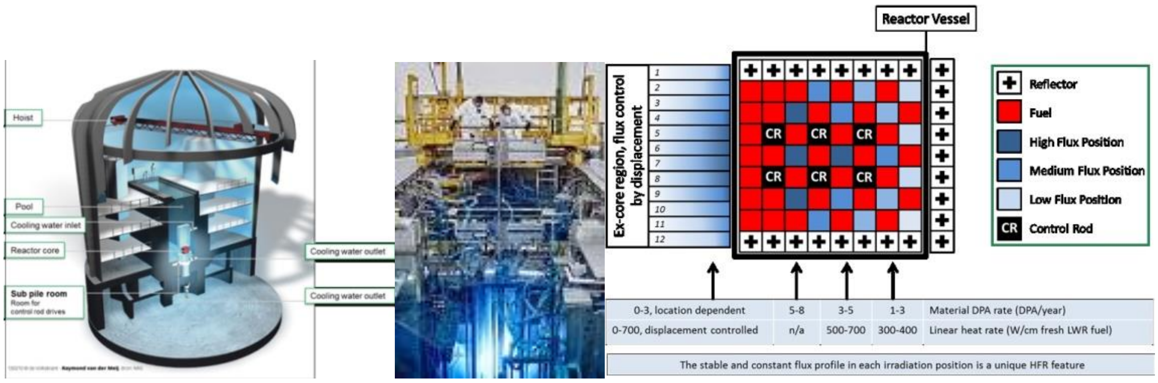
Petten's unique facilities

GZ *One of the things I've been told about the SALIENT is that these experiments can presently only be done here in Petten. That is quite an amazing claim. Is this true and if so, why?*

RH To start with, there is only a limited set of materials test reactors in the world. And only a subset of these can be used for the study of nuclear fuels. And the HFR happens to have a facility for placing larger objects in a high neutron flux zone. That is not yet relevant for the SALIENT experiments, but it will become relevant in a later stage, in which we want to place 'loop type' molten salt experiments that can model the dynamic system of a molten salt reactor. This is a unique facility that will offer us the chance to substantially speed up the process of developing a molten salt reactor.

The main reason that fuel experiments can be done only in a few places is the infrastructure that is needed to handle experimental nuclear fuels. In materials irradiations the neutron irradiation will change some elements, but they usually don't create alpha-emitters. Fuels are a completely different ballgame, that requires a highly specialized additional infrastructure. The point is, once you start fission experiments, you produce fission products and alpha-emitters. Both produce lots of radiation, are hazardous and contagious, and safe handling requires specialized labs that must comply with very strict regulations. That means amongst other things that you need facilities with a wider range of ‘hot cell facilities’, that will allow you to safely handle the samples. And not only that, you also need special other cells that will allow you to completely clean and decontaminate all the materials you’ve worked with, and safely collect and ultimately deposit them. So you need to have the whole ‘train’, all the steps from start to end. And we’re fortunate enough to have all these facilities here in Petten. In the United States, France and perhaps Japan, there are National Labs with materials test reactors that also have the required infrastructure for doing SALIENT type experiments. If these countries would focus on molten salt development, they could definitely do it as well.

Another thing is that you need a regulator that has some flexibility. What we are doing here is new, it hasn’t been done before. If you have a regulator that simply says, ‘this is something we don’t know, we won’t allow you to do it’, than you cannot possibly innovate. We have the good fortune of having a regulator that we can communicate with. That is not to say they’re not strict, they are! Even though we only use a couple of cm3 of salt, they checked every detail.



SALIENT will take place in the High Flux Reactor in Petten, Netherlands. The drawing on the left shows the structure of the reactor building with in the center the reactor pool, containing the core. The photograph in the middle shows this pool, with at the bottom the Cherenkov-light that surrounds the reactor core. The ‘checkerboard’ on the right is the topography of the reactor core, seen from above. The zones on the left of the ‘checkerboard’ form a ‘pool side facility’ that will become relevant in a next phase of experimentation. This pool side facility is a high flux region on the outside of the reactor core with enough room to accommodate a larger molten salt reactor experiment, in which a ‘salt loop’ can be tested. The intention is to fill this ‘salt loop’ with molten salt fuel. Due to the high neutron flux, a fission reaction will start, and the loop will become a test scale molten salt reactor. This pool side facility of the HFR is unique. If the loop experiment proves feasible, this will give the Netherlands a head-start in molten salt reactor development.

GZ *What about the Chinese, they have been assigning several hundred million to MSR programs…*

RH In their case, the main question is when they are going to have a suitable materials test reactor ready for such experiments. Presently, they don’t have that.

Waste treatment and lithium recovery

Future MSR’s are expected to have superior waste properties. This will depend on chemical processes in which fission products can be separated from usable fuel. In molten salt reactors, fission products are the only component that can be called ‘waste’ – all the so called ‘actinides’, the heavy fissionable nuclei, can in principle be recycled back into the reactor core until they fission.

However, these reprocessing facilities need to be developed step by step and are not available at this stage of development. That also means that in the irradiation phase of the SALIENT experiments, the small amount of salt that has been tested will, after it has been carefully investigated, have to be disposed of. And in the case of nuclear waste, regulations require that even cubic centimeters of waste come with a disposal plan.

The NRG team took the opportunity to define several suitable options, and this development process already produced some valuable insights.

GZ *The interesting thing is that regarding the waste, SALIENT-01 already goes a significant step further than the Oak Ridge experiments in the 1960s.*

RH Yes, by starting the SALIENT -01 irradiation, we have also committed ourselves to defining feasible waste routes. Today we cannot simply say 'we're not interested in the waste'. For us, this is a good driver to really sort things out.

GZ *And you came up with some feasible routes…*

RH Probably. Although you have to realize that for this stage of SALIENT experiments our goals for the waste are quite modest. For this experiment, we do not yet separate the fission products from the actinides and into separate waste streams. [Separating fission products out of the fuel salt is one of the developmental goals for the MSR and is not yet practiced in this experiment – ed.] At this stage, it makes more sense to do all processing in a closed system. That being said, we have defined two routes.

The first is quite simple: we dissolve the fluoride salts in strong acid. The fluoride then will precipitate and can be removed. If we then raise the pH, turn the acid into a base, the fission products, and also the uranium and remaining thorium will precipitate as hydroxides or nitrates. That means you can then dry (calcinate) the remaining fraction that contains the actinides and the lanthanides. You have then turned your fluorides into oxides, and that is a kind of waste that everybody in the sector understands. This is quite like the familiar spent nuclear fuel (SNF) coming from many nuclear power plants. The remaining liquid waste can be cemented. You simply add cement and you let it harden. That is a quite straightforward method with which the COVRA is also quite familiar.

A promising feature of the nitric acid route is that in the process of raising the pH, and having the fission products precipitate, the lithium is likely to remain in the solution. That implies that the lithium will be recoverable. You could then simply reuse it for your next reactor.

On the other hand, this route might not be suitable for larger salt quantities because the required volumes of nitric acid are high. The second route is similar but involves distilling off the bulk of the nitric acid solvent. This route would not allow for easy recovery of the lithium.

But again, the goal for these routes apply to the context of the experiments. After solving the waste issue for the SALIENT irradiations we will have to look into scaling up waste treatment. "