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MOLTEN SALTS AND PYROCHEMICAL PROCESSING PROGRESS AT THE UK'S NATIONAL NUCLEAR LABORATORY

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Abstract. The UK's National Nuclear Laboratory offers infrastructure and capabilities to support research and development programmes for advanced fuel cycles and partitioning and transmutation (P&T). Our world-leading facilities have been widely used as part the UK government funded Advance Fuel Cycle Programme. Part of the programme involved the development of molten salt and pyrochemical processing capabilities including fuel manufacture, salt science, engineering, online monitoring, and fuel cycle integration. This has led to a resurgence of activity in molten salt technologies for nuclear application.

1. Introduction

Molten salt reactors (MSRs) are receiving increasing attention due to their potential for passive safe, efficient energy production [1]. Combined with pyrochemical processing they offer the potential for closing the nuclear fuel cycle, contributing to energy security and meeting the UK's 2050 Net Zero commitments. MSRs come in a variety of models from those using salt purely as a coolant to those with fuel dissolved in the molten salt [2]. The choice of salt and fuel, their resultant behaviour in the reactor and the energy extraction methods are all fundamental variables. These choices influence the reactor capabilities and more specifically their application and possible limitations for back-end cycle options.

However, molten salts are not exclusively of interest in MSRs, they have several advantages such as high radiation tolerance, high solubility for relevant materials, excellent heat transfer and ionic strength that makes them suitable for different nuclear energy applications [3]. Molten salts can be used in chemical and electrochemical processing for materials recovery [4,5] including fuels for reactors but also radionuclides for medicine, space and other applications [6]. Decontamination and heat storage are further applications [7-9]. Figure 1 shows a schematic of the processes involved in molten salts technologies.

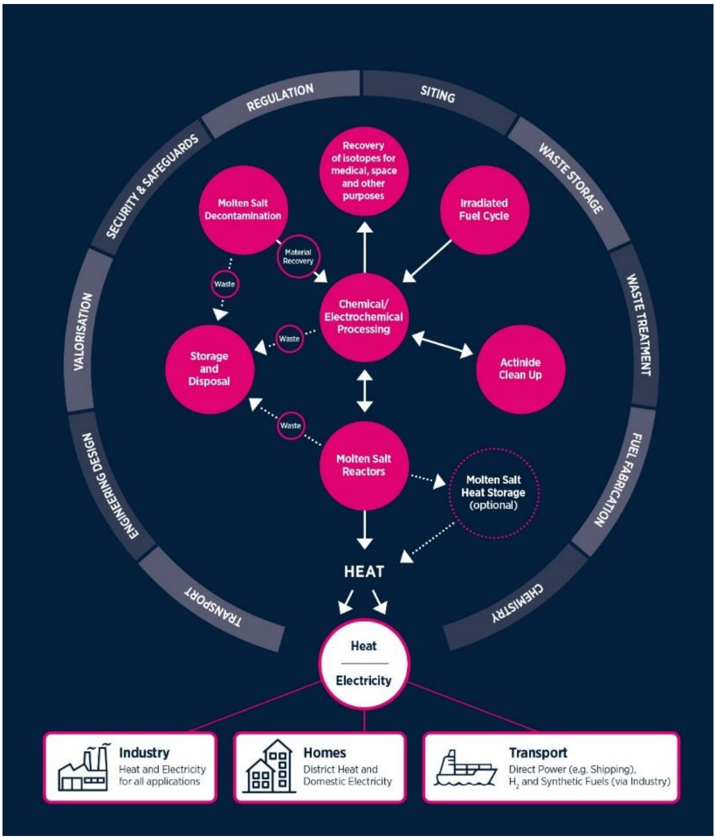

Fig. 1. Molten salts technologies and their application. Influential factors and considerations are listed around the outside.

2. Molten salt chemistry and science

The chemistry of MSRs is complex. The varying composition over time as fissile material is converted to fission products impacts the physical and chemical properties, and neutronics of the system. This leads to challenges in control of corrosion, salt behaviour and the ability to sustain the nuclear reaction. When the fuel is a fluid this can be both an advantage and disadvantage. The ability to add to and remove from the salt allows fuel and redox control agents to be added and poisons removed, but it creates added complexity in modelling, assurance measurement and radiation shielding leading to control and regulatory challenges. The treatment of wastes from MSRs is also far from trivial.

The analysis and measurement of species concentration for process control and accountancy is important. One of our key objectives is to provide high-quality, consistent and comparable data for relevant molten salts systems, which is currently limited, especially when alpha-active material is part of the salt. The data, in areas such as physical properties and materials, will not only be used longer term to specify constraints on future facilities but has already been used to inform areas of NNL's pyro-processing projects such as flowsheet development and pilot scale demonstrations.

In a molten salt based closed fuel cycle fuel manufacture is both the beginning and the end of the process – its manufacture is required to begin operations in the MSR, while the restoration of the fuel by removal of contaminants detrimental to operation closes the loop. NNL have produced a range of fluoride- and chloride-based fuel salts, proving our capability to handle both the salt and control the oxidation state of the uranium. Our ambition is to extend this learning into a wider range of fuels.

Fig. 2. Example of NNL's fuel manufacturing and characterisation capabilities – thermal analyser in argon glovebox (left) and NaF-KF-UF_4 (FUNaK) fabricated at NNL (right).

3. Pyrochemical and waste processing

3.1 Pyrochemical processing

The Advanced Fuel Cycle Programme (AFCP), 2019-2022, was part of a UK government investment in nuclear [10]. It built on the initial phases of Advanced Nuclear Fuels and Nuclear Recycle and Waste Management projects, delivered by an NNL-led consortium between 2017 and 2019. One of the projects under AFCP was pyrochemical processing (pyro-processing) which refers to the use of high temperature, non-aqueous techniques to treat irradiated nuclear fuel. Even though pyro-processing has been extensively used to treat mainly metal fuel [11], NNL's objective in this area is to raise its technology readiness level, TRL [12] through the investigation of methods to close the fuel cycle by restoration of the fuel and recovery of useful biproducts. The production of baseline flowsheet models and demonstration of key engineering aspects such as scalability and on-line process monitoring are also part of our aspirations. Within the AFCP pyro-processing activities led by NNL a glovebox scale, plutonium active research rig, the Pyrochemical Alpha-active Processing Apparatus (PAPA), is being commissioned and tested. This piece of equipment includes a high purity gas feed providing a low oxygen and moisture blanket; it allows the direct study of electrochemical and physical properties of different molten salt media containing actinides (including plutonium). Upon completion, the PAPA rig will provide a platform to extend our active experimental programmes, see Figure 3.

Fig. 3. The pyrochemical alpha-active processing apparatus (PAPA), has been developed to handle and investigate the electrochemical and physical properties of actinides (including Pu) in molten salt mixtures. Note that the protective heat guards have been removed in this commissioning image.

3.2 Waste and decommissioning – salt cleaning

To take advantage of the benefits offered by molten salt technologies the industrial and economic feasibility must be demonstrated, in particular how to manage any legacy material from operations – specifically wastes. Moreover, the production of a qualified wasteform will be essential for the acceptance of these technologies by UK regulators. As with many of the barriers facing adoption of molten salt technologies, the challenge is unlike conventional nuclear systems since the chemistry of the salt is different. The high solubility of halide ions under typical geological conditions provides a unique challenge in terms of treatment and immobilisation for safe long-term storage and disposal.

The direct immobilisation (Fig. 4) of salt containing wastes is possible and, in some cases, may be the most appropriate treatment; however, incorporation rates are low leading to the potential for large volumes of waste to be managed. Increased incorporation can be achieved through dehalogenation (Fig. 4), but incorporation rates are typically still below 10%. NNL continue to investigate how to improve the quality of these wasteforms while reducing the volume. However, to minimise environmental impact it is essential to separate the fission products from the salt as this will reduce the final waste volume and allow the cleaned, potentially expensive, salt to be recycled and re-used.

Currently there is no solitary process effective at removing all fission products efficiently [13]. Therefore, a multi-stage approach (Fig. 4) is required based on selective ion exchange and/or precipitation and melt crystallisation techniques. NNL will persist in its exploration of different waste treatment options with the ultimate goal of a fully integrated process minimising the generation of waste and environmental impact.

4. Integration

The key to successful deployment of molten salt technologies, including pyrochemical processing and MSRs, will be integration. Currently there are a wide range of different salts being studied, but the functional components of the salt system will need to be passed between processes. This will increase in complexity as the different carrier salts and any contamination varies between the component operations. It may be that, to enable the fuel cycle to be closed, compromises need to be made on the choice of salt for some applications, but the goal should be an integrated system of processes that offers the greatest benefit.

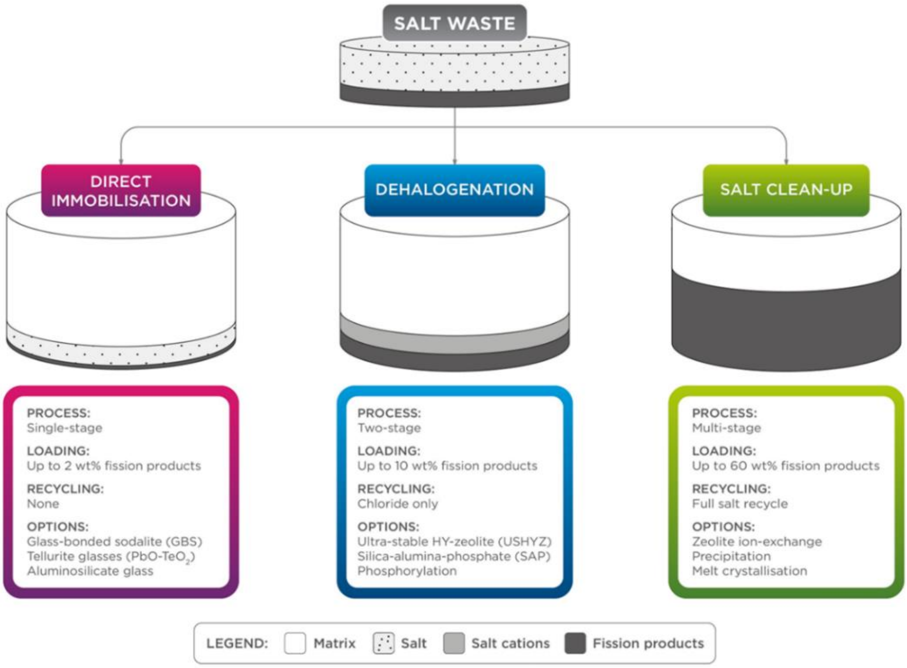

Fig. 4. Chloride salt treatment processes for waste and decommissioning.

4.1 Fuel cycle modelling

Using the 2021 UK Energy System Modelling: Net Zero 2050 paper as a starting point, several scenarios for the future buildout of UK nuclear were developed [14]. It was then investigated, using the NNL proprietary fuel cycle modelling tool ORION [15], what the impact of commissioning fast MSRs, in this case the EVOL MSFR design [16], would be on the UK spent fuel inventory, plutonium management and power generation (see Fig. 5).

In each of the cases shown in Fig. 5, modest means one MSFR commissioned every 10 years from 2045 to 2085. Extensive means one MSFR commissioned every 5 years from 2045 to 2085. The rest of the UK civil nuclear fleet is made up of GW-scale light water reactors, small modular reactors and a large number of high temperature gas reactors, used for providing industrial heat and steam for hydrogen generation.

The EVOL MSFR concept is used due to availability of geometry and material data. This is a design with a binary fluoride salt mixture for fuel, additional fertile blanket for breeding to make use of thorium fuel cycle. Recycling is online, with all fission products removed at the end of each year, and additional fissile material added to maintain criticality.

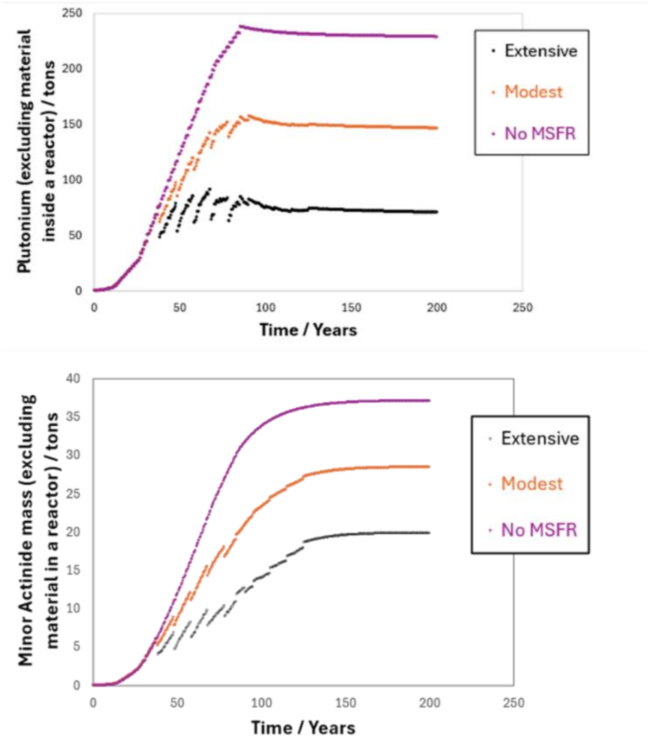

Fig. 5. Impact of commissioning fast molten fast reactors in the UK spent fuel inventory, plutonium management and power generation [14]. Data modelled using the EVOL MSFR design.

The current plan is to expand verification of MSR implementation in ORION: this may also include additional development for usability and coupling of other reactor physics codes. NNL has additional projects building capability in neutronic/computational fluid dynamics modelling of MSRs, which can also be implemented into fuel cycle modelling. Work on the EVOL MSFR is likely to be published in the near future, highlighting particularly the potential of ORION.

4.2 Training and education

NNL has had a long history working with molten salts [4] but in the 5 years preceding AFCP there had been a gradual erosion of knowledge and capability. The AFCP programme enabled a reversal of this trend with new capabilities such as PAPA being commissioned and a new generation of molten salt scientists inducted and educated. It was a feature of programme and work since the completion of AFCP that work was progressed in collaboration with UK universities adding to the pipeline of talent. The repurposing of facilities and engagement with MSR developers through initiatives such as the Molten Salt Technology Platform (MSTP) has further increased the understanding and experience of those engaged.

5. Conclusions and Future Work

To maintain the skills and capability to assess the potential of molten salts technologies it is important that the UK continues to invest in research and development. NNL recognise this and the potential of molten salt technologies to close the fuel cycle – particularly for fast reactor fuels, their use in reactors as a coolant (with or without dissolved fuel), in the processing of nuclear materials, and the separation of usable components.

Significant progress has been made in the upskilling of personnel and re-equipping of laboratory facilities at NNL, which has enabled a contribution to the global effort in this area in both national and commercial programmes. The continuation of this work will enable NNL to provide an informed view to UK government and regulators while advancing the technology.

Acknowledgements

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