

INTRODUCTION

Nuclear data integrate the latest scientific and engineering understanding of nuclear physics to model the underpinning processes of nuclear systems. Nuclear data are relevant across the fuel cycle including fuel fabrication, reactor design and operation, fuel performance modelling and fuel inventory data, waste management and recycling. Data are key to predicting the performance of materials during and after irradiation. These data are drawn from a combination of empirical and theoretical sources and are consolidated into centralised databases, usually as part of international collaborative efforts involving a range of organisations. The Advanced Fuel Cycle Program (AFCP) in the UK focuses on developing capability and capacity, while seeking to reduce costs across the nuclear lifecycle, to ensure that nuclear can play a part in delivering secure, low-carbon energy in the global market. This includes development of Advanced Technology Fuels (ATF) which are targeted at providing increased efficiency within the current generation reactors. In AFCP, this focuses on the development of uranium nitride (UN) fuels to replace existing uranium dioxide (UO₂) fuels. Such a move would have impacts across the nuclear fuel cycle and will require the ability to accurately simulate the performance of fuel in a range of scenarios across the fuel cycle.

Currently, the major challenges when considering this fuel type for deployment are expected to be high fabrication costs and interaction of the fuel with water. Fabrication costs are expected to be significantly impacted by the need to enrich nitrogen in ¹⁵N to minimise ¹⁴N and the subsequent generation of ¹⁴C from the ¹⁴N(n,p) ¹⁴C reaction channel which is significant for thermal neutron reactors.

To date, nitrogen has not been used in commercial reactor fleets as a major constituent of fuel or other significant material. This work is intended to assist understanding where nuclear data needs to be improved and what further nuclear data measurements, simulations and benchmark experiments would be required to allow the safe deployments of such fuels.

BACKGROUND

To model the behaviour of uranium nitride fuels under irradiation in a reactor it is necessary to understand what the nitride consists of in terms of the atomic nuclei of the nitrogen atoms and how these nuclei interact with the neutrons present in the reactor driving the fission chain reaction. The nitrogen nuclei will interact differently with neutrons compared to oxygen nuclei both in terms of capturing neutrons and how they scatter neutrons to different energies altering the spectrum of the neutrons in the reactor, further affecting the rate of energy production.

Neutron interactions with nuclei are described by numerical quantities called cross-sections. These are complex correlated multi-variant parameters that are described in large datasets called evaluated nuclear data libraries. The UK is most involved with the international Joint Evaluated Fission Fusion file project (JEFF) coordinated by the OECD Nuclear Energy Agency (NEA).

These evaluated nuclear data libraries are produced using a combination of experiments and theory. The libraries are then validated against relevant benchmark experiments to confirm they adequately describe important engineering parameter such as criticality safety, fuel performance and reactor operation. This results in a development cycle driven by incremental improvements to reduce uncertainties and to cover new needs. The steps in this process are summarised in Figure 1.



Figure 1: Outline of a simplified nuclear data lifecycle highlighting the key steps in the process.

METHODOLOGY AND RESULTS

An initial literature review identified no existing validation data in the international databases for uranium nitride fuel manufactured with ¹⁵N. Therefore, the only estimate of the accuracy of the nitrogen neutron capture, (n,p) and scattering cross-sections are from experimental measurements of the basic nuclear data. The scatter cross-section at thermal energy was measured as 4.59 ± 0.05 barn as reported by Mughabghab [1]. Above this energy, there are only measurements from 470 to 1959 keV which have no estimated uncertainty. Figure 2 show these measurements and the evaluated data in the energy regions concerned.

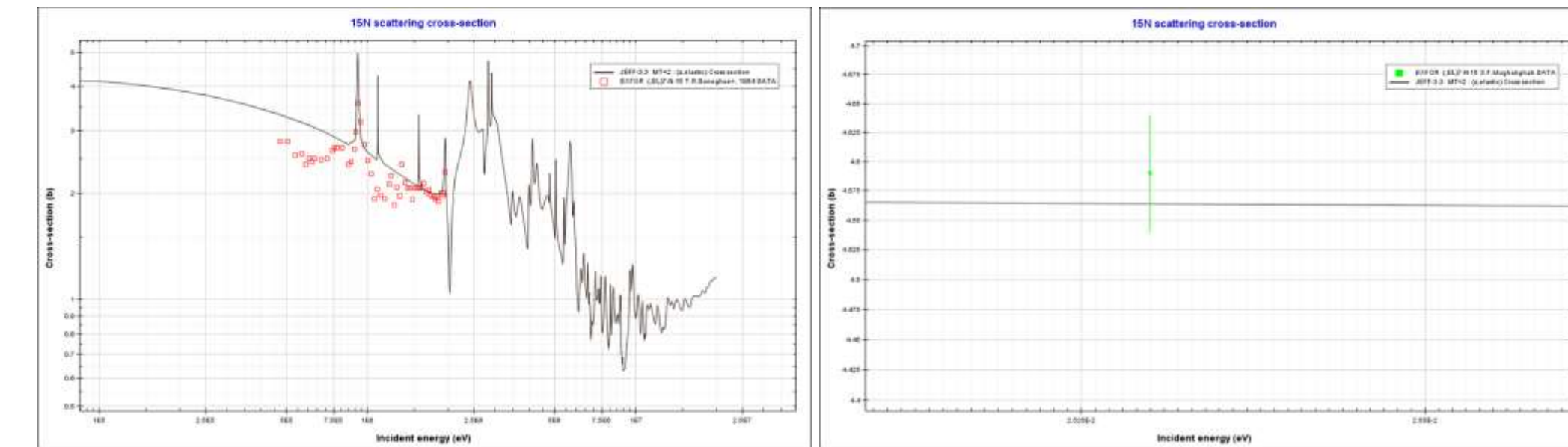


Figure 2: Scattering cross-section of ¹⁵N in the fast and thermal range.

There are a significant number of measurements of the ¹⁴N(n,p)¹⁴C cross-section. However, given the available data, it is expected that the fast neutron spectra and neutron slowing down region covering the neutron resonances will be well covered, especially given the Koehler et al (1989) data in the 61 meV to 34.6 keV region [2]. Looking at the thermal region (Figure 3) the situation becomes less clear. There is a significant amount of data reported at 18 meV, below the traditional 25.3 meV (thermal value). The thermal Mughabghab value (1.86 ± 0.03 barn) appears to be based upon a combination of other data rather than all the available experimental measurements reported.

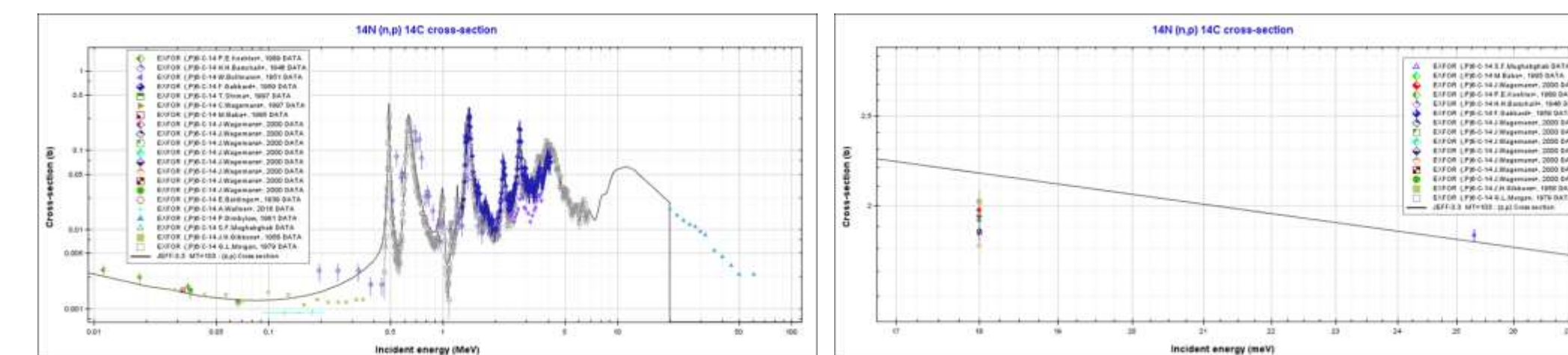


Figure 3: (n,p) absorption cross-section of ¹⁴N in the fast and thermal range

As there is no experience of nitride fuel for Light Water Reactors in the UK, and little published elsewhere, there is a need to study the current nuclear data and its use for simulation within different computer codes to understand how sensitive simulations of these areas are to the nuclear data, to aid understanding of how well we expect important parameters to be predicted and where, if needed, nuclear data needs to be improved to better predict these quantities ahead of commercial deployment.

In this work studies have been made of uranium nitride fuels in comparison to uranium dioxide fuels using MCNP6 (a Monte-Carlo code) and WIMS10A (a deterministic code). Differences between these codes could indicate that approximations for uranium dioxide fuels in certain codes may not be appropriate for nitride fuel.

This study considered the simplest representative model of fuel, an infinite array of stacked fuel pins (known as a pincell model) where geometrical modelling effects are small to study nuclear data sensitivity. Parameters studied included:

- Mass of UN powder required to go critical in spherical configuration (i.e. $k > 1$)
- k in dry pin cell at room temperature (important for fuel manufacture and transport)
- k in wet pin cell at room temperature (important for fuel transport)
- k in reactor conditions during burnup/enrichment (reactor operation)
- Expected life of fuel in reactor with enrichment (i.e. life weighted mean k at end of life ~ 1.02)
- Composition of spent fuel (75 GWd/t) compared to UOX (55GWd/t, 5%) example at 5, 10, 20, 100 and 1000 years.

Analysis of the results by comparing codes showed great differences for UN over UO₂ fuel. Comparison to UO₂ for instantaneous effects (ie. calculation of k) further demonstrate a relative increase in k with increasing ¹⁵N/¹⁴N ratio. More variations were observed in burnup calculations, including an increase in k as a result of reducing packing density and variation in associated spent fuel inventories.

The results showed a significant shift in the neutron spectrum. As an example, the flux at the middle of the irradiation is compared in Figure 4. This shows a significant drop in the thermal flux in the fuel below 100 eV. This will reduce neutron capture and is expected to alter the fission products and higher actinides produced in this fuel during irradiation.

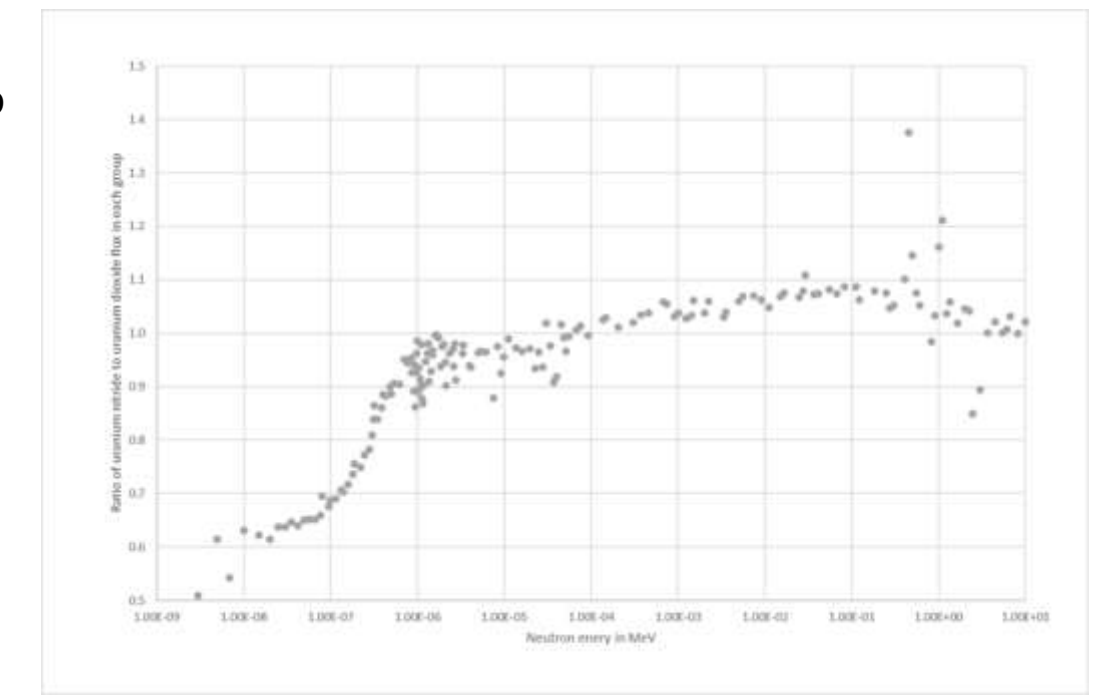


Figure 4: Change in neutron flux in uranium nitride fuel compared to uranium oxide fuel in the middle of irradiation.

CONCLUSIONS

The review of the underlying nuclear data identified that:

- There are no criticality or reactor benchmarks that allow the current nuclear data to be verified for nitride fuels.
- The nitrogen scattering cross-sections for ¹⁴N and ¹⁵N in the thermal range have little justification.
- The ¹⁴N (n,p) ¹⁴C cross-section measurements differ by about 5%
- The ¹⁴N capture cross-section measurements differ by 7%

A review of the literature highlights that there is a lack of experimental data for UN fuels, especially in terms of criticality benchmarks relevant to use in thermal reactors. The very recent publication by Wallenius does not report any available good experimental data for thermal reactors, while there are some data available for fast reactors. Some experimental data are available for uranyl nitrate solution, which will be of value for fuel cycle development [3-6].

Calculations using two reactor modelling codes WIMS10A and MCNP6 show that there are differences in the results that are greater for uranium nitride than uranium oxide. It is expected that assumptions in these codes either in the acceptable model definition or underlying nuclear physics approximations could result in these differences and they need further study at a deeper level to understand.

The calculations for uranium nitride fuels shows that it is expected to show differences in terms of k -eff and the spent fuel composition that is important for reactor operation and reprocessing. These processes are principally driven by the nitrogen cross-sections. Interestingly, these calculations suggest that nitride fuel may have neutronics advantages over oxide fuels but that care needs to be taken as the nitride fuels may be inefficient at higher densities.

The sensitivity studies in this work suggest that better uncertainties on the nitrogen nuclear data are required to better understand the accuracies of this uranium nitride calculations. Further work is now being completed to justify the nuclear data improvement requirements.

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ACKNOWLEDGEMENT AND CONTACT

This work was funded under the £46m Advanced Fuel Cycle Programme as part of the Department for Business, Energy and Industrial Strategy's (BEIS) £505m Energy Innovation Programme. For further information contact robert.w.mills@uknln.com or allan.simpson@uknln.com

INTRODUCTION

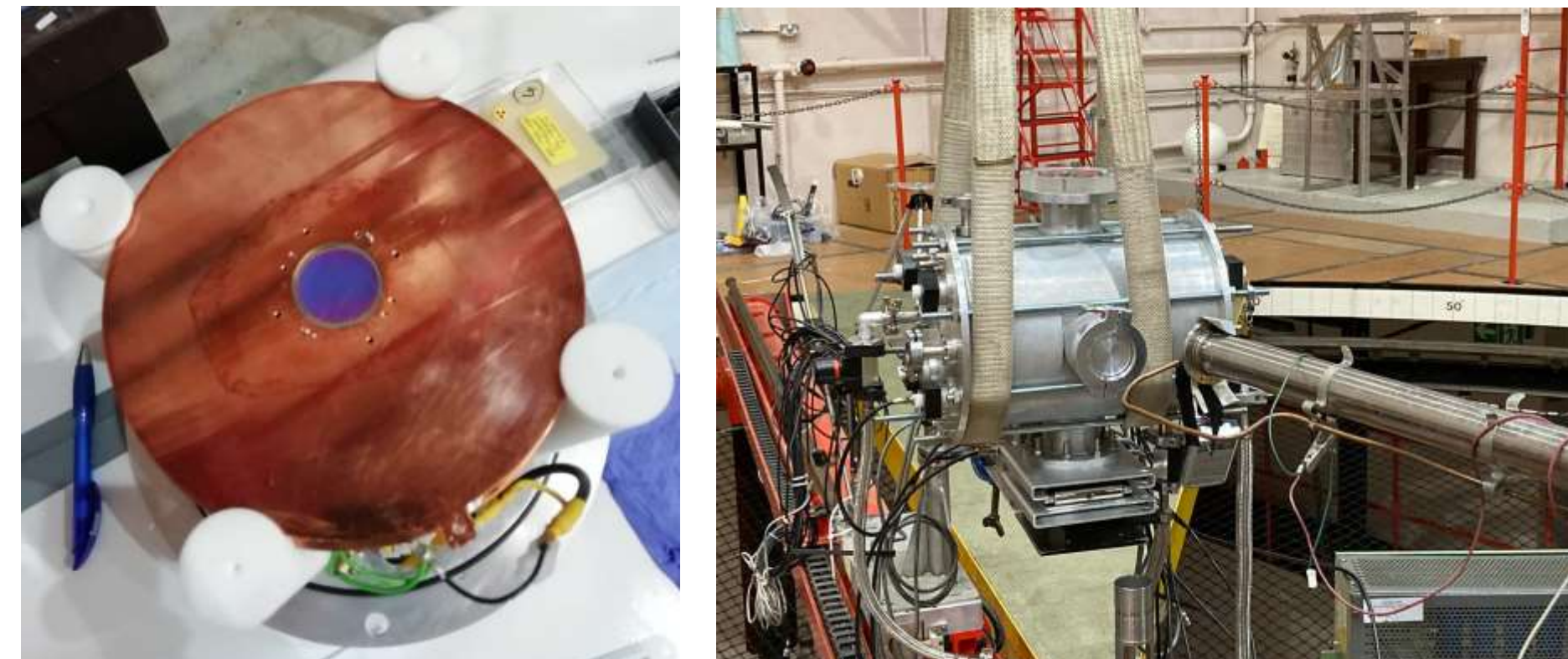
- NPL is the UK Metrology Institute and hosts world-leading facilities such as radiochemistry and neutron metrology that can contribute to Nuclear Data measurements relevant to AFCP
- Our range of accelerator- and source-based calibration fields ensures the continued availability of national standards of neutron fluence and dose equivalent quantities.
- We use a 3 MV Van de Graaff accelerator to generate monoenergetic neutron fields at energies from the keV region to just below 20 MeV (see table below) and, in conjunction with a large graphite pile, we provide an accelerator-based thermal neutron field.
- To make use of these capabilities, we need to establish at NPL new expertise by collaborating with world-leading nuclear-data centers. AFCP has provided such opportunity.

Neutron energy (MeV)	Reaction	Fluence (cm ² s ⁻¹)	Ambient dose equivalent (μSv h ⁻¹)
0.144	⁷ Li (p,n) ⁷ Be	1 10 ⁶	4.5 10 ⁶
0.250	⁷ Li (p,n) ⁷ Be	6 10 ⁶	4.4 10 ⁶
0.565	⁷ Li (p,n) ⁷ Be	1.6 10 ⁷	2 10 ⁷
1.2	T (p,n) ⁹ He	2 10 ⁶	3 10 ⁶
2.5	T (p,n) ⁹ He	6 10 ⁶	9 10 ⁶
5.0	D (d,n) ⁹ He	6 10 ⁶	8.8 10 ⁶
16.5	T(d,n) ⁴ He	4.5 10 ⁶	8.9 10 ³

THE NPL SETUP

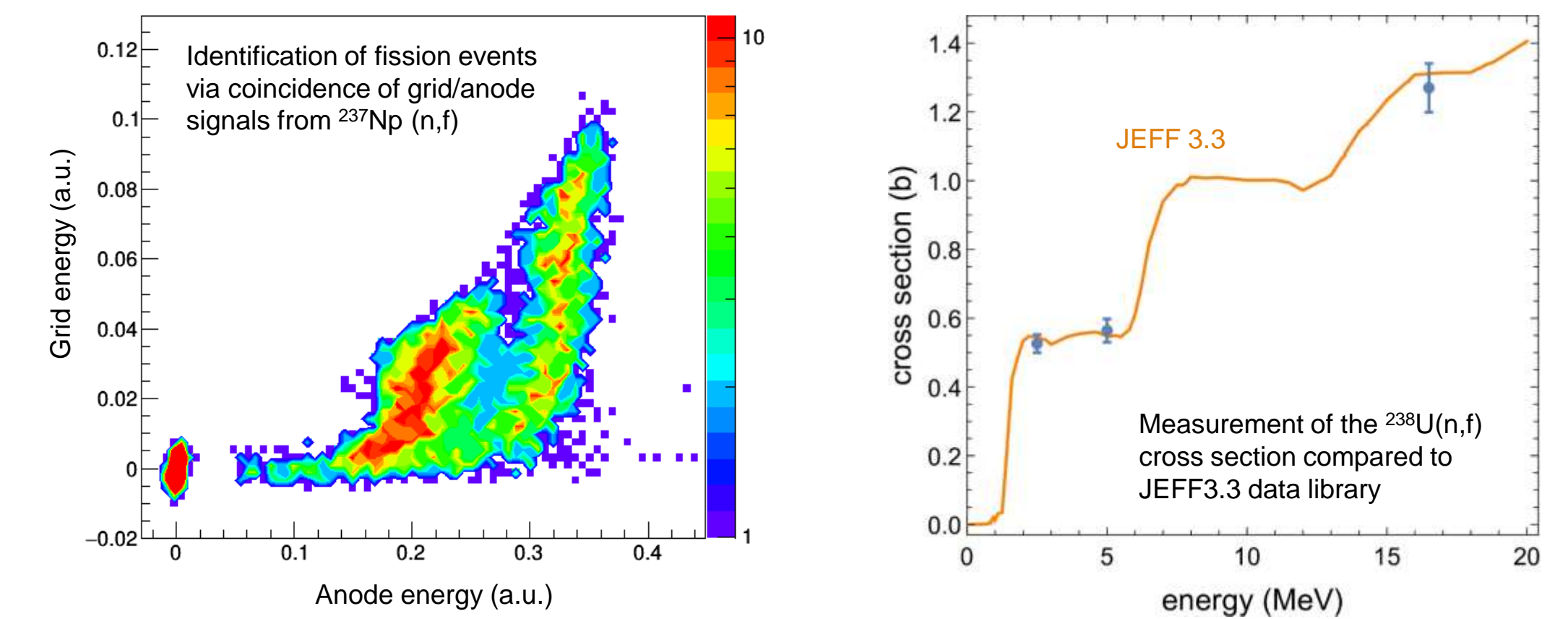


Double-Frisch ionization chamber for fission studies developed by the University of Manchester



RESULTS

- We have tested the UoM fission chamber using the well known ²³⁸U cross-section at 2.5 MeV, 5 MeV and 16.5 MeV. These tests provide valuable data for further detector development, and the first demonstration that the envisioned setup at NPL is suitable for cross section measurements
- A methodology to analyse our data has been developed that includes effects normally negligible in NPL commercial work such as corrections due to the charged particle beam profile and angular distribution of the neutron field
- Tests highlight several details that need improvement and related to the mechanics of the setup, fission chamber efficiency and background characterization
- We have collected data for ²³⁶U(n,f) cross section, which we are currently analysing
- We have identified window-less gas target technologies based on differential pumping and plasma windows as the best way forward to increase our neutron fluence



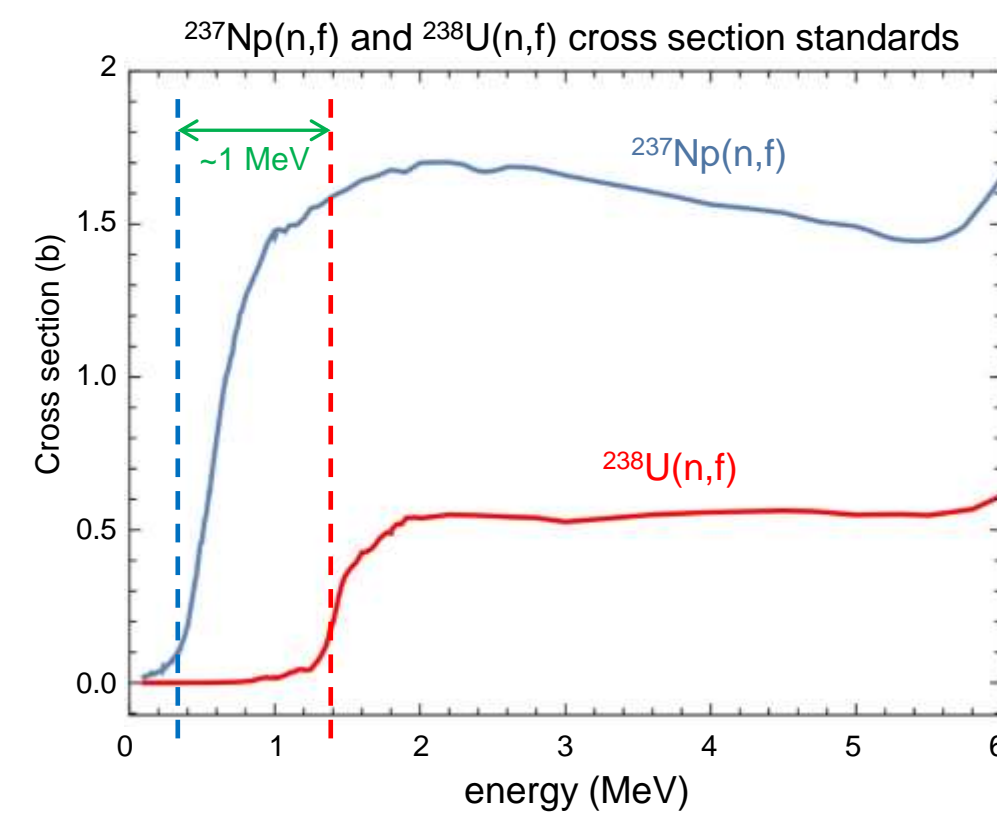
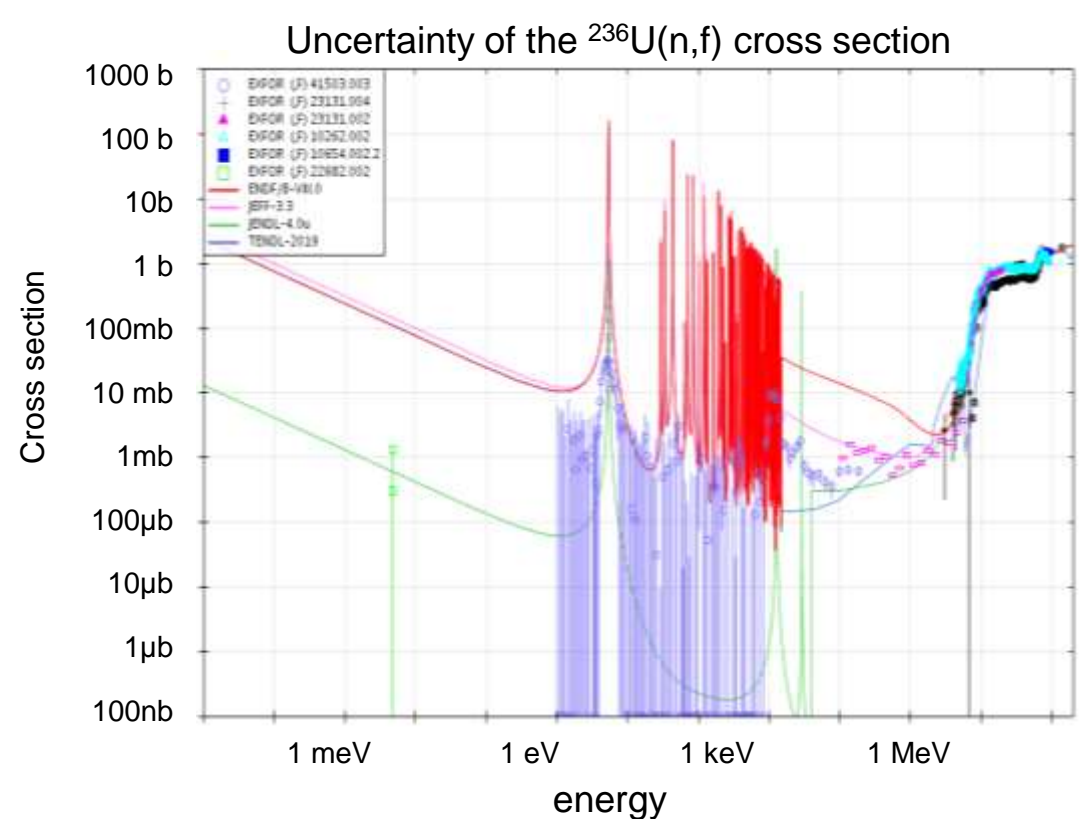
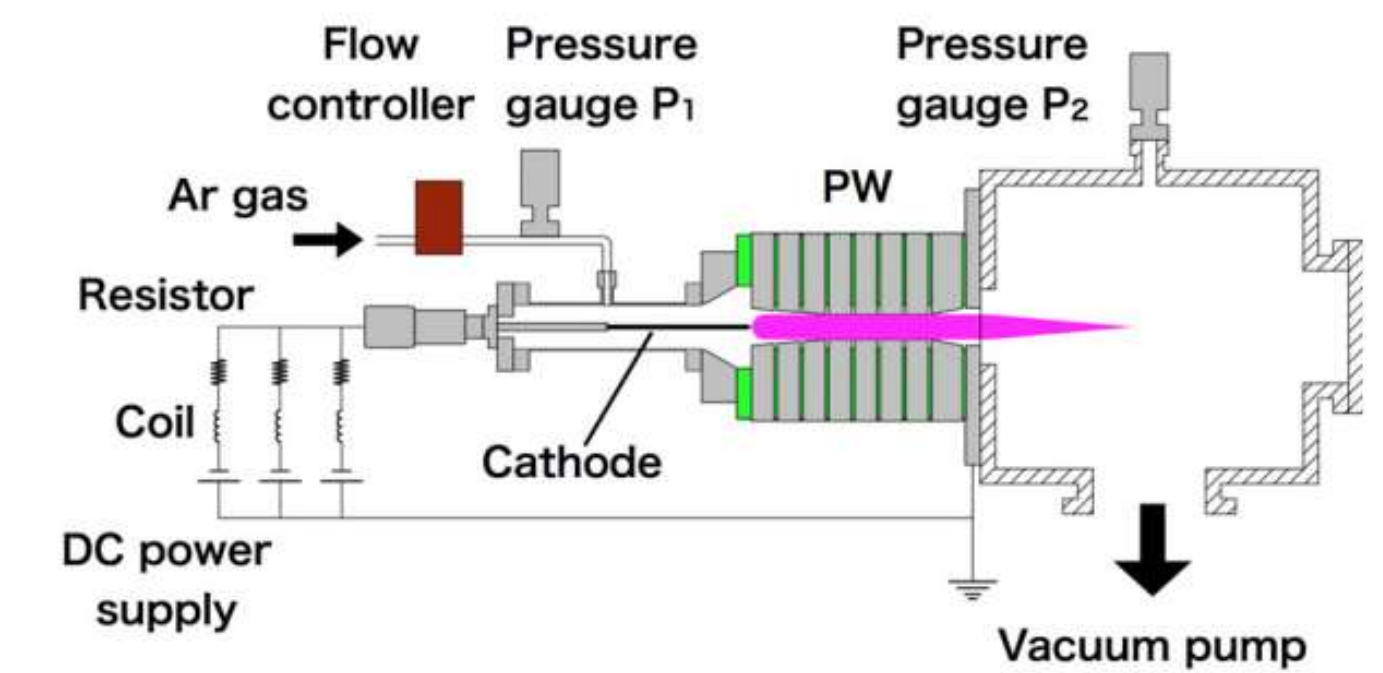
OBJECTIVES

Accurate nuclear data is essential for the assessment of nuclear fuel and understanding waste production. It underpins computer simulations to predict reactor nuclear processes in a safe and cost-effective manner. Our main long-term objective is to establish at NPL a nuclear data capability to provide high accuracy at few selected energies that can contribute to decrease the uncertainties of evaluated data. Within the AFCP our work has been focusing on:

- Tests of a novel fission chamber designed by the University of Manchester and employing segmented anodes to reconstruct fission fragments angular distribution [1].
- Improve the uncertainty of the ²³⁶U(n,f) cross section in the fast region up to 20 MeV. This is important for new fuel cycles such as ²³²Th and fast reactors.
- Perform absolute and relative ²³⁷Np(n,f) cross section measurement toward making ²³⁷Np(n,f) a standard cross section for fast neutron energies. The reaction is of interest because it has a lower threshold than ²³⁸U(n,f) and a higher cross section.
- As a study of current state-of-the-art target technologies that can allow to increase the NPL fluence of monoenergetic neutrons. This investigation includes solid, liquid and gas target technologies.

Plasma-window (PW) gas target

Gas windows degrade the incoming charged particle beam energy resulting in lower neutron energy resolution. Resolution requirements limit the gas cell size and therefore neutron fluence. Heating is also a crucial drawback. The figure on the right is a promising design developed at RIKEN [3].



NPL 3 MV Van de Graff accelerator



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CONCLUSIONS

- Significant steps forward have been made at NPL toward establishing a nuclear data capability thanks to national and international collaborations
- In collaboration with the University of Manchester we have demonstrated nuclear data capability by measuring ²³⁸U(n,f) in agreement with the most recent data evaluations (e.g., JEFF3.3, ENDF-VIII)
- Data analysis is ongoing to measure ²³⁶U(n,f) cross section
- Data analysis is also ongoing to measure ²³⁷Np(n,f) cross section. This will contribute to define a new standard cross section better suited than ²³⁵U(n,f) in the fast neutron range
- We are in the process of identifying new target technologies to boost neutron fluence. We are currently investigating the feasibility of plasma-window gas target as the most promising option.

INTRODUCTION

Pyrochemical processing (or pyro-processing) encompasses a range of high temperature non-aqueous spent fuel separation techniques based around using electrochemistry in molten salt media. The most extensively studied technique is electrorefining and can be implemented to separate U, Pu and the minor actinides (such as Np, Cm, Am) from other fission products. The use of molten salt-based electrolytes provides a high tolerance to radiation, making pyro-processing potentially suitable for short-cooled and high-burn up nuclear fuels such as the Fast Breeder Reactor (FBR) type. See Figure 1 for a description of the electrorefining process.

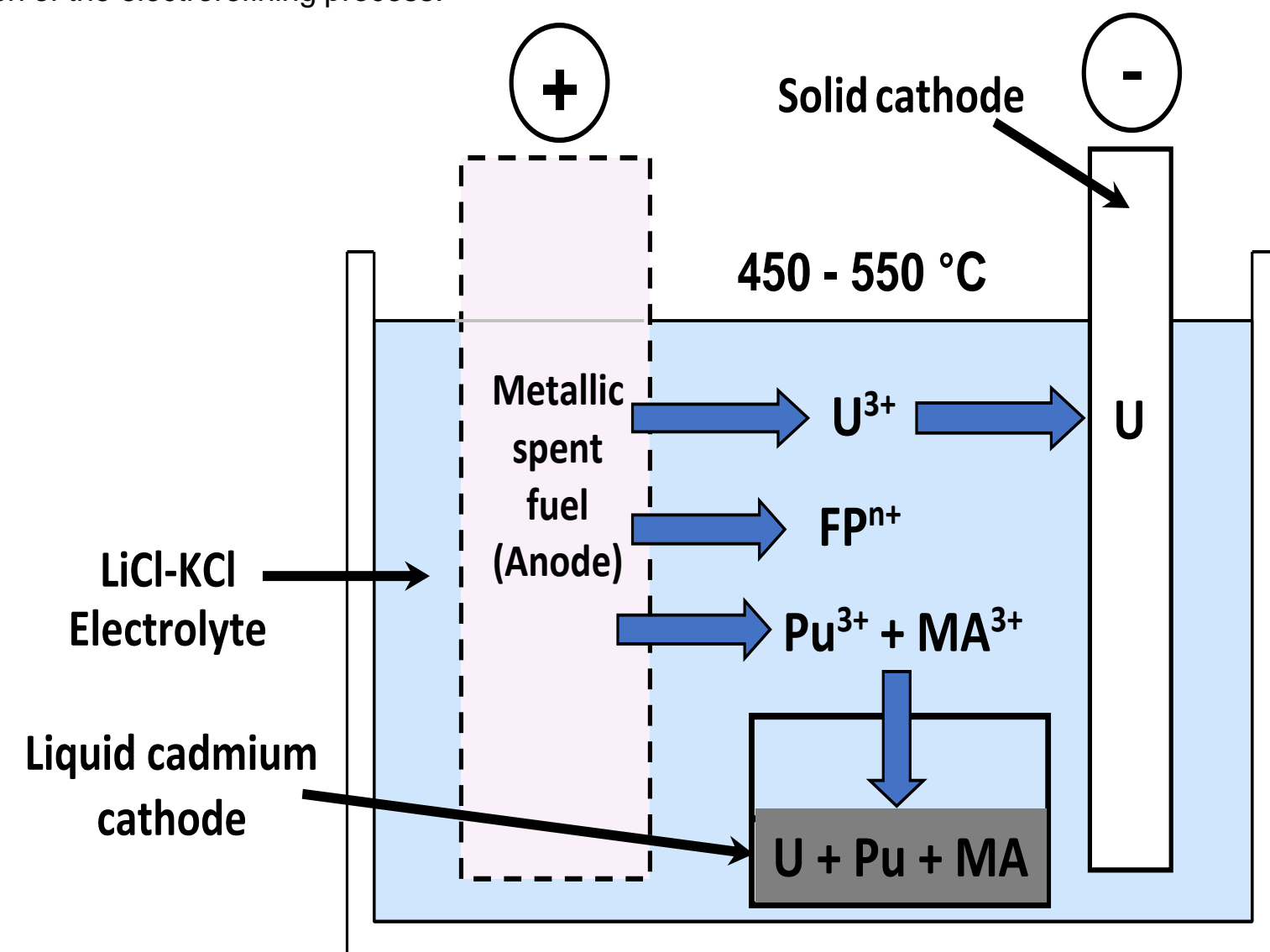


Figure 1: Electrorefining process

- Spent metallic fuel dissolved at the anode;
- Pure U metal electrodeposited at the cathode by applying a suitable voltage;
- Other actinides can also be electrodeposited e.g. to yield U-Pu and/or U-Pu-MA alloys, by selecting a different cathode and voltage;
- The separated actinides are removed from the electrolyte and then cleaned to remove excess salt;
- The cleaned actinide product(s) are then recycled to manufacture new nuclear fuel;
- Fission products (FP) accumulate in the salt requiring periodic removal;

The Advanced Fuel Cycle Programme (AFCP) is part of the Department of Business, Energy and Industrial Strategy (BEIS) investment in nuclear research and development. It builds on the initial phases of Advanced Nuclear Fuels, Nuclear Recycle and Waste Management projects, delivered by an NNL led consortia between 2017 and 2019. One of the main aims of the AFCP is to develop a series of key technologies to support a future Generation IV reactor fuel cycle. One of these technologies is pyro-processing and is the focus of one of the projects under the AFCP [1]. As part of this work, it has been agreed to provide a glovebox (GB) scale, Pu-active, experimental, rig. This will form a UK capability for use in future research, with the key function of providing electrochemical data for developing Pyro flowsheets based around electrorefining (see Figure 1). This capability is known as Pyrochemical Alpha-active Processing Apparatus (PAPA) and is currently undergoing commissioning within the NNL Central Laboratory where it will

DESIGN BASIS

The PAPA design was built on learning from Pu-experiments performed as part of the Engineering and Physical Sciences Research Council (EPSRC)-funded REFINE project [2]. At the outset of the project, an exercise was undertaken to identify shortfalls in the apparatus used for REFINE to form the design basis of the PAPA design and associated functional specification. Improvements are detailed in the table.

Experimental Quality	Safety
Ultra high purity argon required (<2ppm O ₂ + H ₂ O) to prevent oxidation of salt	Engineered mechanical lift lower mechanism - required to prevent electrodes from being immobilized in solidified salt – used as mitigation in safety case for allowing hands-in-gloves operations whilst furnace at temperature
Faraday cage to reduce electrical interference	Insulation and guarding
Sampling system & mechanical stirrer	Gas scrubber to mitigate against mal ops scenarios
Mechanical bellows system to replace dynamic seals - maintain atmosphere quality	Safety control system for gas supply

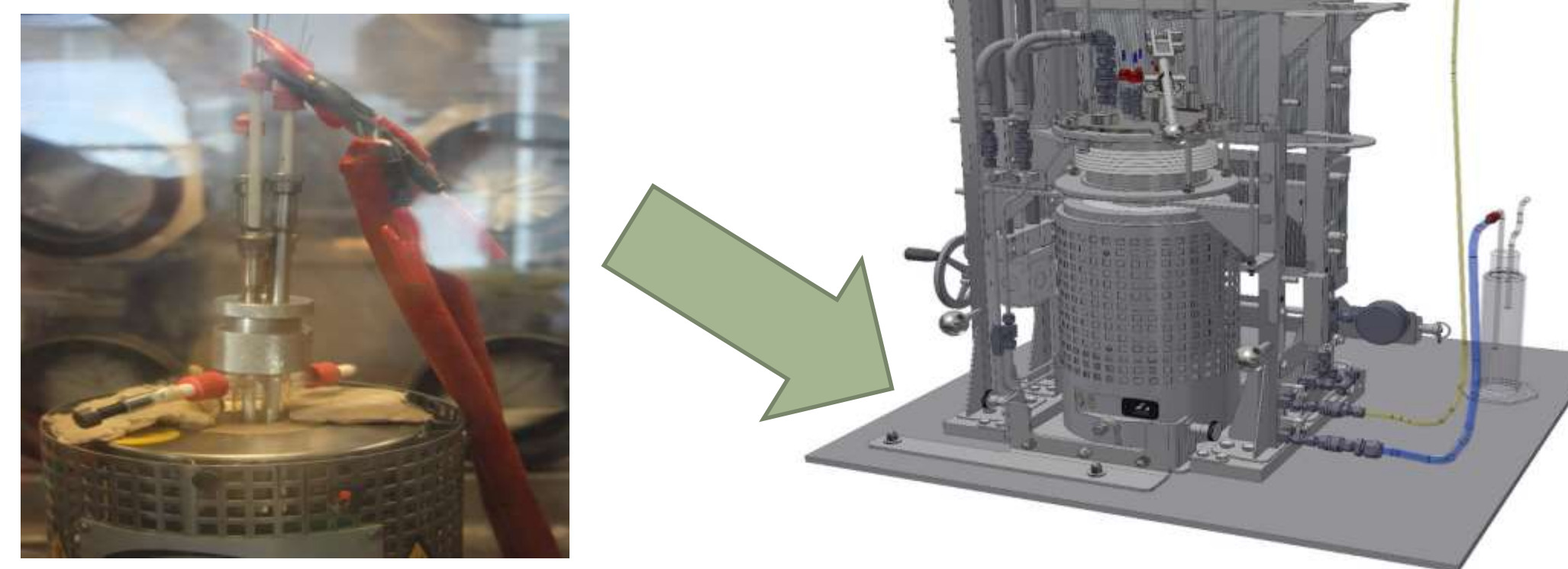


Figure 2: REFINE Apparatus vs PAPA

PROJECT STATUS

The design process has been finalised taking PAPA through preliminary and detailed design and completion of HAZOP 2, see Figure 3 for a project timeline and associated project tasks. Engineering challenges were addressed by a team of two mechanical engineers, a mechanical designer and a process engineer, key challenges were as follows:

- Providing a means to raise/lower electrodes without the use of unreliable dynamic seals
- Development of inert gas feed safety systems to ensure the design met with the safety case whilst providing sufficient flow for purging operations
- Designing the rig in a modular fashion such that it could provide functionality whilst being postable into a 10" posting port
- Providing sufficient instrumentation into the electrochemical cell whilst adhering to space constraints on the top plate
- Fully manual system to negate requirement to adhere to machinery directive
- Designing the rig to the constraints of an existing glovebox environment

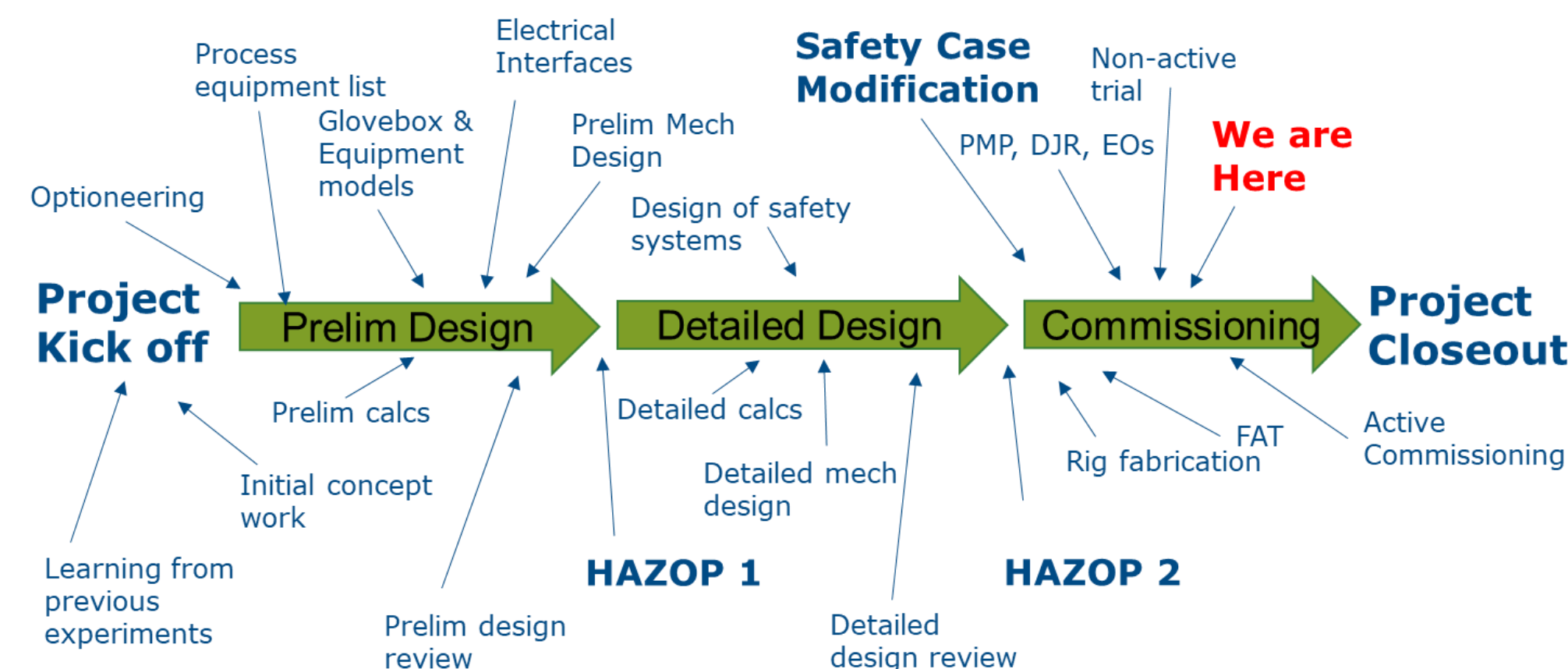


Figure 3: Project timeline



Figure 4: PAPA rig during Workingington glovebox trials

On completion of the design phase, the rig was then fabricated by an external company before a Factory Acceptance Test (FAT) was undertaken. The FAT ensured the rig had been fabricated as per the design specification and resulted in some minor alterations, mechanical fittings bored out etc. before the rig was accepted and transferred to the NNL Workingington Facility.

Here initial trials were performed to assess how the rig could be posted, in its constituent parts and assembled within a glovebox environment making the most of an opportunity to utilize a full scale non-active GB. This resulted in dimensional changes to the base and heat guarding as well as changes to the orientation of the rig within the GB to enhance operability.

The rig was then transferred to NNL Central Laboratory for further trials including running the furnace at temperature and feeding inert gas to the electrochemical cell.

Next stages will involve completion of all necessary safety documentation before conducting an active commissioning trial, this final step will result in the PAPA rig being a fully commissioned UK pyro-processing facility ready for active research programs to be undertaken.

SUMMARY



Figure 5: PAPA rig undergoing non-active trials

- The PAPA rig provides a robust test bed for electrochemical experiments, these can be used to inform future pyro flowsheets
- Learning from the REFINE project was used to define the functional specification
- The design phase of the rig was completed addressing a series of key engineering challenges
- After completion of this phase, the rig was fabricated before undergoing a FAT
- PAPA was then accepted by NNL and underwent glovebox trials which resulted in a variety of minor modifications to enhance operability
- It is now undergoing non-active trials at NNL Central Laboratory
- Once active commissioning is complete, PAPA will be a fully commissioned UK pyro-processing facility ready for active research programmes

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INTRODUCTION

Pyrochemical processing (or pyro-processing) provides a complementary technology to conventional or advanced aqueous treatments for the recycling of used nuclear fuel [1, 2]. However, in an advanced nuclear fuel cycle, pyro-processing may prove to be a key technology, depending on the reactors and fuels adopted, to enable closure of the fuel cycle and thus contribute to sustainable electricity generation to meet the UK's net zero commitment. Therefore, effectively treating and minimizing the waste generated through pyro-processing is essential to support the development of this technology as part of the UK's future clean energy portfolio. However, pyro-processing generates new types of wastes, most notably the highly radioactive and water-soluble salt that is incompatible with current immobilisation technologies such as vitrification [3]. This is mainly due to their low, uneconomic capacity in standard nuclear waste glasses, requiring innovative treatment and immobilisation solutions. This poster provides an overview of the experimental work completed on waste salt treatment in the AFCP 'Processing of Pyro-processing Waste' project.

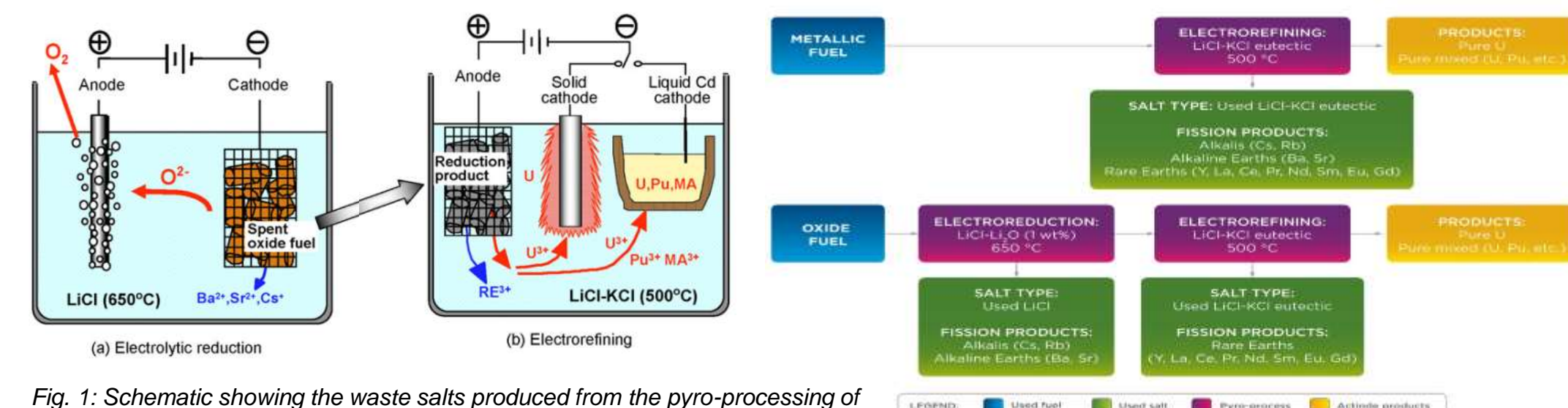


Fig. 1: Schematic showing the waste salts produced from the pyro-processing of metallic and oxide used fuels using an electrorefining-based separation process

OBJECTIVES

As the role of nuclear energy grows to support net zero carbon emission targets, the sector is increasingly considering the long-term legacy of its operations. Ensuring the safe storage and disposal of nuclear waste involves incorporating that waste into durable matrices designed to last thousands of years. One of the advanced separation technologies being considered as part of this programme is pyrochemical processing.

At its simplest, pyro-processing is the use of high temperatures to bring about a physical or chemical change in a material. AFCP's Processing of Pyrochemical Waste (PPW) workstream is investigating how to treat the salt arising from the pyrochemical processing of used nuclear fuel. These treatment options can be divided into three categories:

- a) Direct immobilisation: using wasteforms with high halide tolerance to incorporate untreated salt [4, 5]
- b) Dehalogenation: generation of a halide-free intermediate compatible with a greater range of immobilisation options [6]
- c) Salt clean-up: separation of the fission products from the salt followed by immobilisation of the former in specifically designed matrices [7]

All three approaches may play a role in the overall salt waste management strategy in pyro-processing, with (c) being particularly attractive as it minimizes waste volume and allows cleaned salt to be recycled back to the main pyrochemical processes.

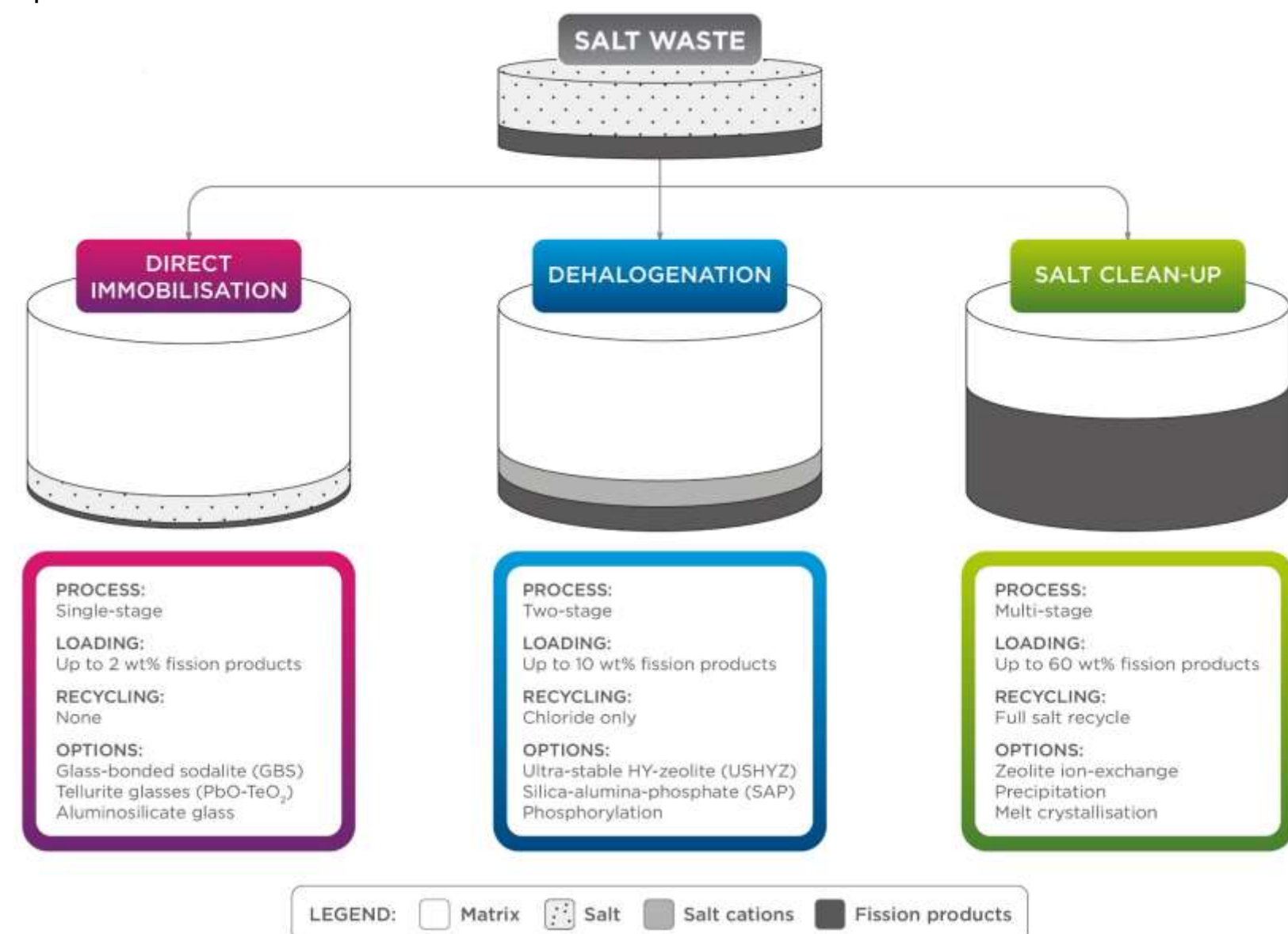
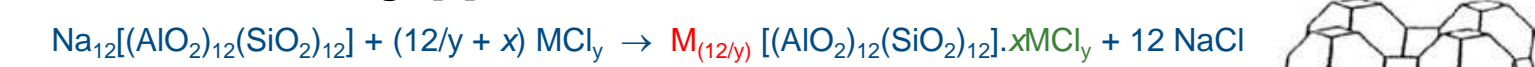


Fig. 2: Options for the treatment of used salt arising from the pyrochemical processing of used fuel showing typical fission product loadings in the final wasteform matrix

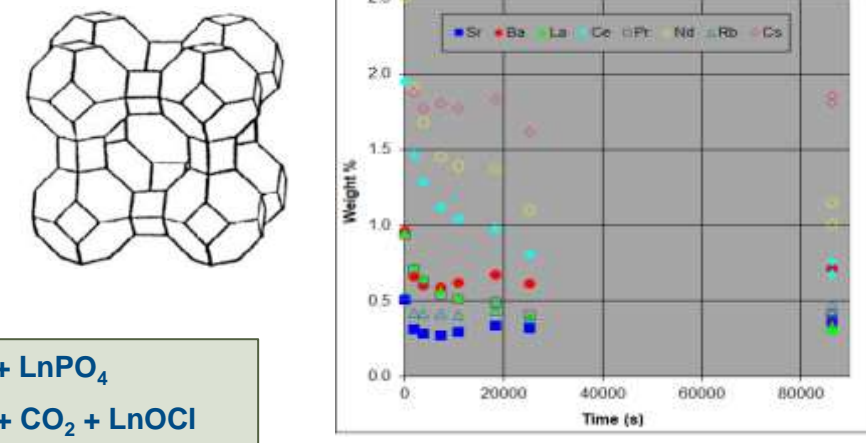
SALT CLEAN-UP

There are several potential salt clean-up methods for the removal of fission products from spent pyro-processing salt. These options include zeolite ion-exchange, precipitation and melt crystallization [7, 8], all three of which have been studied in AFCP.

Zeolite ion-exchange [7]

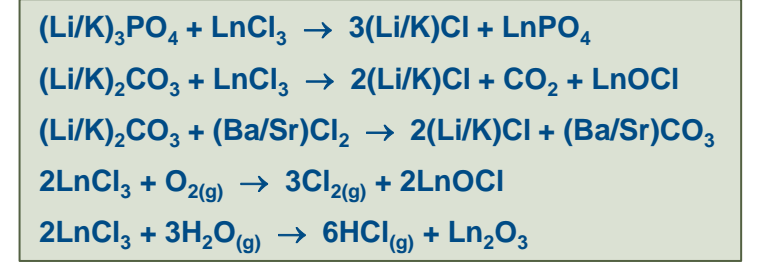


- Salt 'molecules' also occluded in pores
- Works for all FP but rare earths > alkaline earths > alkali metals
- Significant chloride content in waste
- Has to be further consolidated, e.g. glass bonded sodalite (GBS)



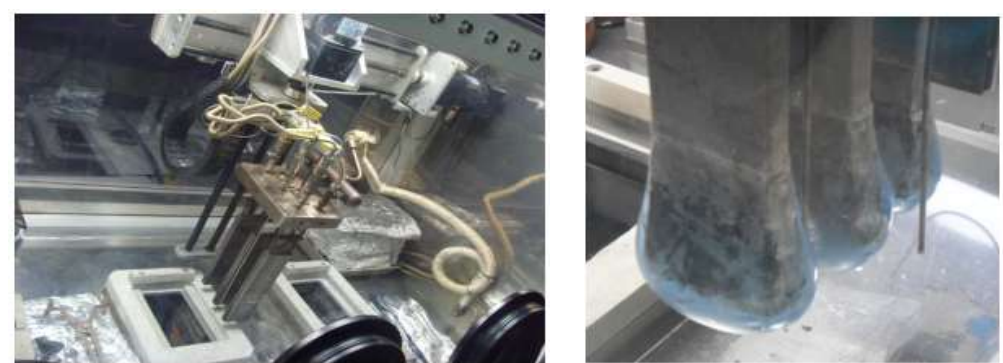
Precipitation [7]

- Reactive with phosphates or carbonates
- Oxidative with oxygen or steam
- Salt and precipitate need separating
- Extra salt / gases need to be managed
- More waste form options, e.g.
 - Monazite ceramic for phosphate
 - Borosilicate / lanthanum borosilicate (LABS) glasses for oxides/oxychlorides



Melt crystallization [8]

- Techniques involving the controlled cooling / freezing of salt
 - Zone refining
 - Layer-melt crystallization
 - Cold finger
- Formation of pure salt crystals with impurities remaining in melt
- Significant progress by KAERI – 20 kg scale demonstrator
- Main technique for LiCl clean up from alkaline earths and alkali metals
- Yields pure LiCl but waste stream still has high chloride-content



METHOD

Salt clean-up and subsequent waste immobilisation experimental programmes at NNL, Sheffield and Hallam universities.

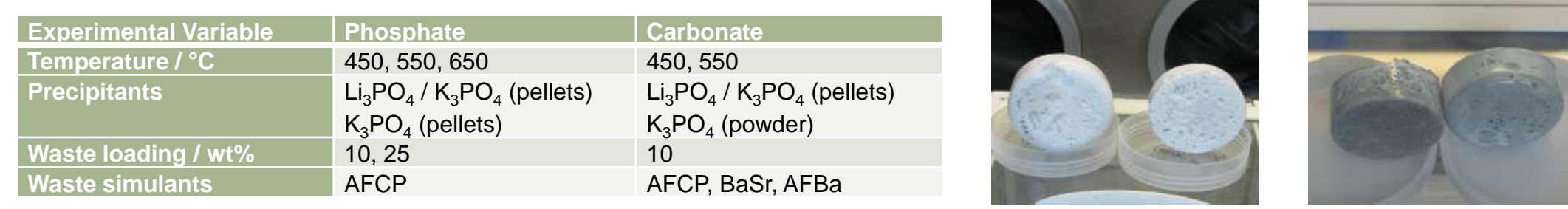
Institution	Salt Clean-Up	Waste Immobilisation
NNL	Precipitation using Li / K phosphates and carbonates	Partitioned rare earth and alkaline earth oxides in borosilicate glasses
UoS	Precipitation and zeolite ion-exchange	Direct immobilisation in phosphate glasses and glass-ceramics
SHU	Melt crystallization / zone refining	Direct immobilisation in alkaline earth aluminosilicate glasses and GBS

NNL salt clean-up: Reactive precipitation

Simulant	Chemical Compound	Target Weight Ratio	Chemical Compound	Target Weight Ratio
'AFCP' rare earth chloride (RECl ₃) mixture	YCl ₃	0.050	PrCl ₃	0.110
	LaCl ₃	0.120	NdCl ₃	0.390
	CeCl ₃	0.230	SmCl ₃	0.100
'Ba/Sr' Alkaline earth chloride (AECl ₂) mixture	SrCl ₂	0.290	BaCl ₂	0.710
	'AFBa' full fission product chloride mixture 50:50 (by weight) 'AFCP' and 'BaSr'			

Experimental Variable	Phosphate	Carbonate
Temperature / °C	450, 550, 650	450, 550
Precipitants	Li ₃ PO ₄ / K ₃ PO ₄ (pellets) K ₃ PO ₄ (pellets)	Li ₃ PO ₄ / K ₃ PO ₄ (pellets) K ₃ PO ₄ (powder)
Waste loading / wt%	10, 25	10
Waste simulants	AFCP	AFCP, BaSr, AFBa

- 25 g of LiCl-KCl eutectic plus simulants
- Heated to temperature in vitreous carbon crucible
- Precipitate added (equi-molar)
- 'Dip rod' samples taken
- Analysis by ICP-OES



NNL waste immobilisation: Partitioned oxides / carbonates, e.g. from oxygen sparging or carbonate precipitation [9]

Oxide Component / wt%	CaZn	MMW	LABS
Li ₂ O	-	4.33	-
Na ₂ O	8.94	9.01	-
SiO ₂	49.72	49.69	42.0
B ₂ O ₃	24.44	18.17	17.0
Al ₂ O ₃	4.38	3.15	41.0
CaO	6.26	-	-
ZnO	6.26	-	-
Gd ₂ O ₃	-	12.50	-

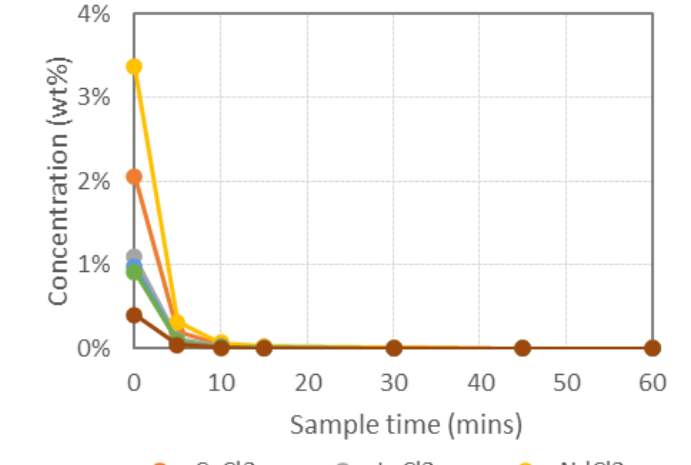
Three borosilicate glass compositions:

- i. CaZn = 1,050 or 1,200 °C
5 - 40 wt% loading of RE₂O₃, BaCO₃, SrCO₃
Silica crucibles
- ii. MMW = 1,200 °C,
10 - 60 wt% loading of RE₂O₃ or (Ba/Sr)CO₃
Alumina crucibles
- iii. LABS = 1,375 or 1,400 °C
30 - 60 wt% loading of RE₂O₃ or (Ba/Sr)CO₃
Alumina / Pt crucibles

RESULTS / CONCLUSIONS

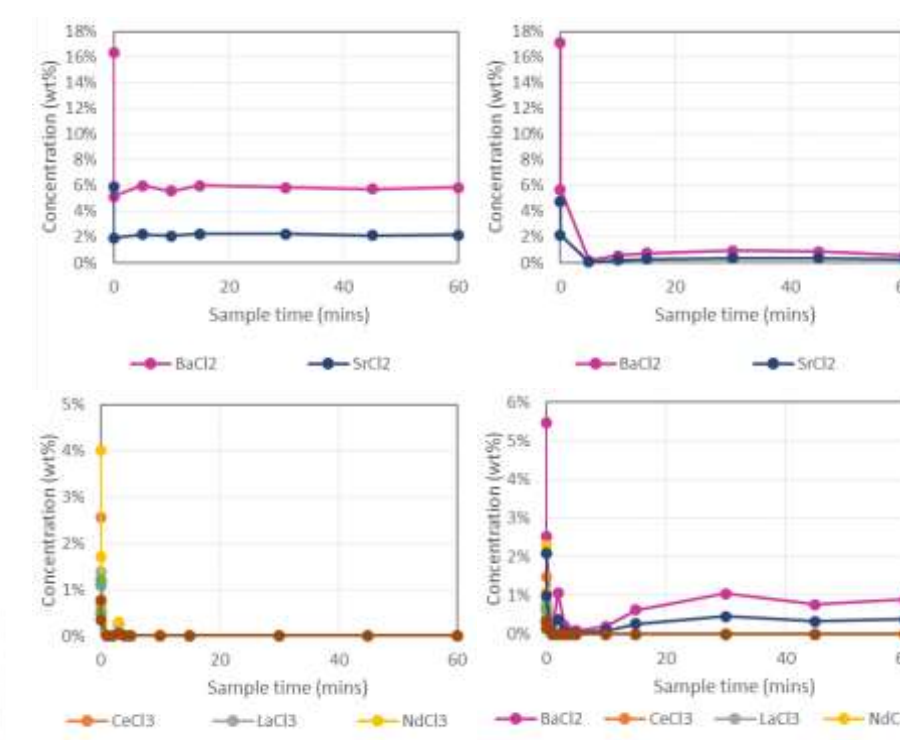
Phosphate Precipitation

- Effective removal of RECl₃ from LKE using single and mixed Li₃PO₄ / K₃PO₄
- Yields >90% in <10 minutes
- Consistent with previous results
- Similar performance of mixed Li₃PO₄ / K₃PO₄ and K₃PO₄ (pellets)
- Yields generally higher at lower temperature (450 °C)
- Agitation has a positive effect (single experiment)
- Pellet and powder delivery method successful
- Undissolved pellets at high waste loading (25 wt%)

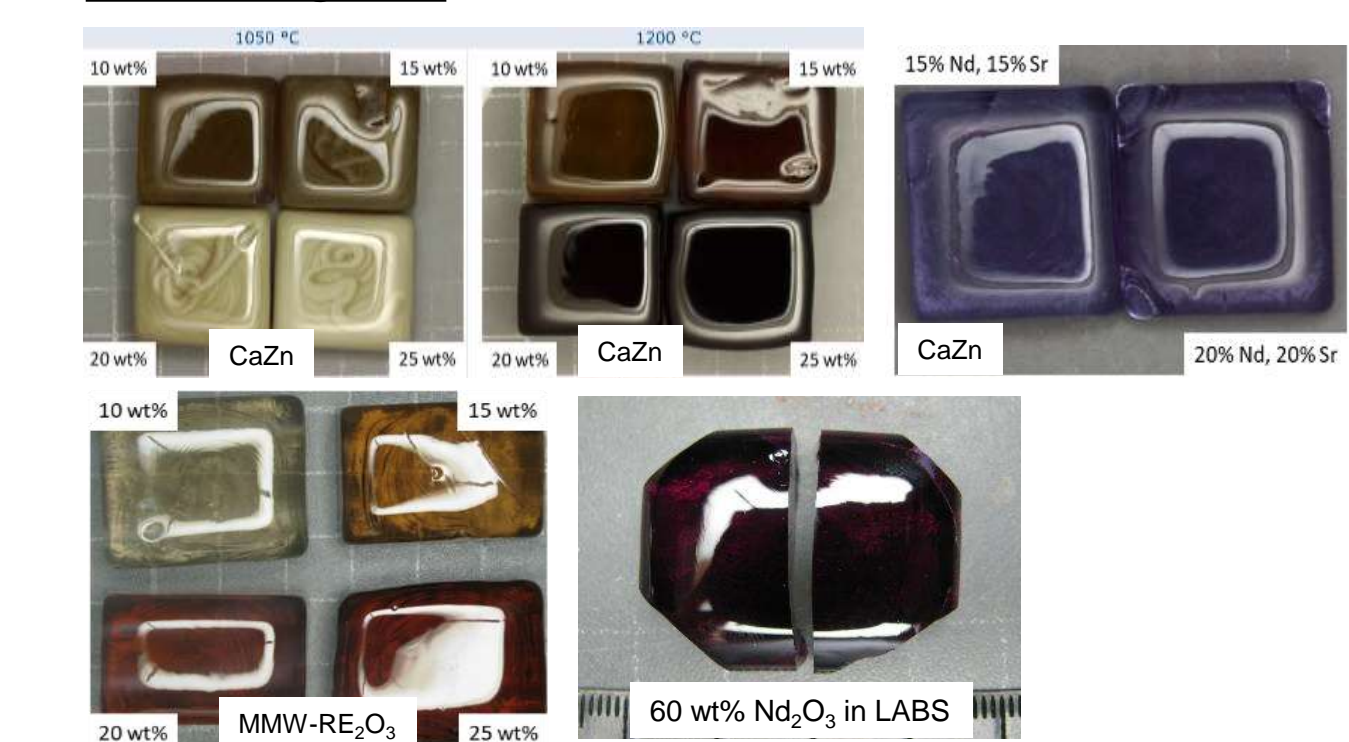


Carbonate Precipitation

- Effective removal of alkaline earth chlorides from LiCl-KCl eutectic
- K₂CO₃ better than mixed Li₂CO₃ / K₂CO₃
- K₂CO₃ powder causes complete removal in <5 minutes
- Complete removal of RECl₃ by K₂CO₃ in 'AFCP' simulant
- Effective removal of RE/AE in mixed 'AFBa' simulant
- K₂CO₃ powder more effective in removing RE and AE than Li₂CO₃ / K₂CO₃
- Very little difference between 450 and 550 °C



Borosilicate glasses



- 1. CaZn (current UK HLW base glass)
 - 40 wt% Ba/Sr carbonates @ 1,050 °C
 - 25 wt% RE₂O₃ @ 1,050 °C, 40 wt% @ 1,200 °C
 - 25 wt% 'AFCP' mixed simulant @ 1,200 °C
 - 40 wt% of Nd₂O₃ + SrO @ 1,200 °C
 - Bulk leach rates < 2 g cm⁻² day⁻¹
- 2. Modified MW (MMW)
 - 60 wt% Ba/Sr carbonates @ 1,200 °C
 - 25 wt% RE oxides @ 1,200 °C
 - 50 wt% mixed AE/RE waste @ 1,200 °C
- 3. Lanthanum borosilicate (LABS)
 - Only produced good glass at high RE₂O₃ (60 wt%) and high temps (1,400 °C)

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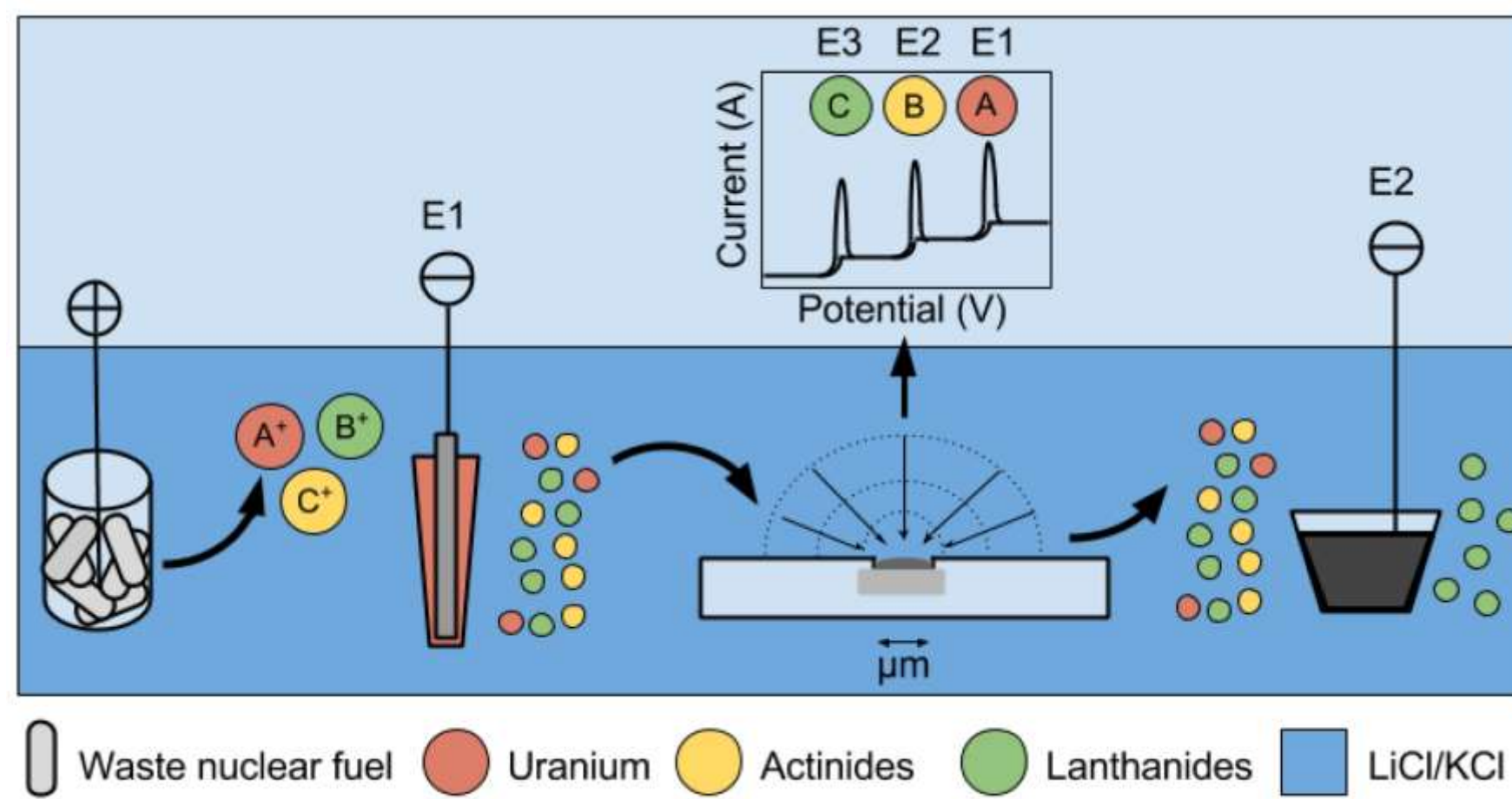
ACKNOWLEDGEMENTS

This research was funded under the £46m Advanced Fuel Cycle Programme as part of the Department for Business, Energy and Industrial Strategy's (BEIS) £505m Energy Innovation Programme

Introduction

Molten salts are an excellent medium for the reprocessing of nuclear fuel because of properties such as:

- High operating temperatures, radiation resilience, and electrical conductivity
- Wide electrochemical window of operation
- Can solubilise many materials

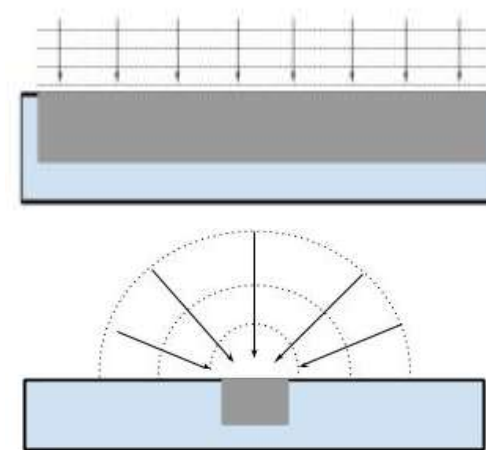


This also makes them very corrosive to construction materials (e.g. steels). Hence, corrosion needs to be monitored for long term safe operation. Electrochemical sensing of low concentrations of corrosion products is being investigated to address this.

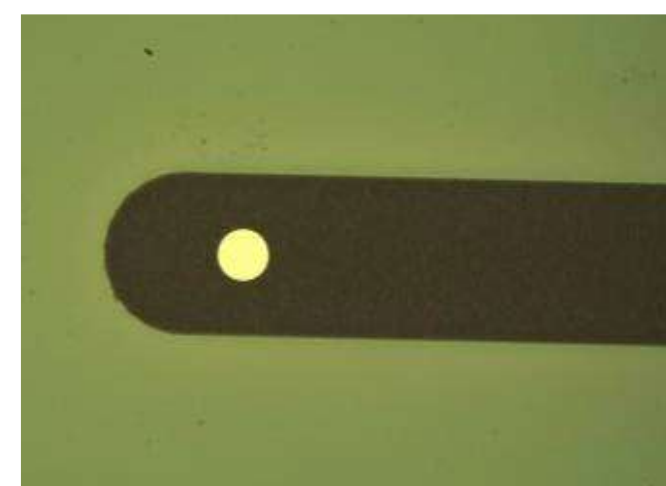
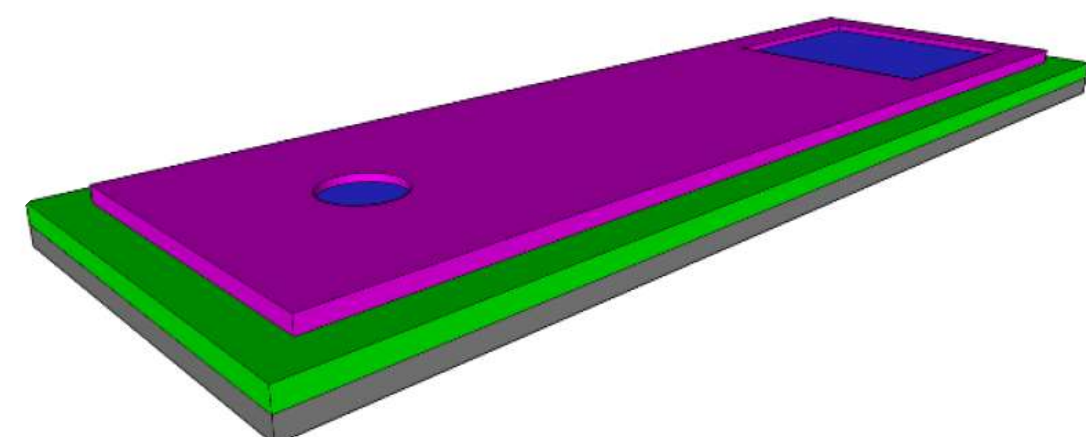
Electrochemical Microelectrode Sensors

Microelectrodes have very favourable characteristics as electrochemical sensors:

- Improved current densities (radial vs. planar diffusion)
- High Signal to Noise Ratio
- Low Limit of Detection
- Low convection susceptibility



We have developed microfabricated microelectrodes that can operate in these conditions.

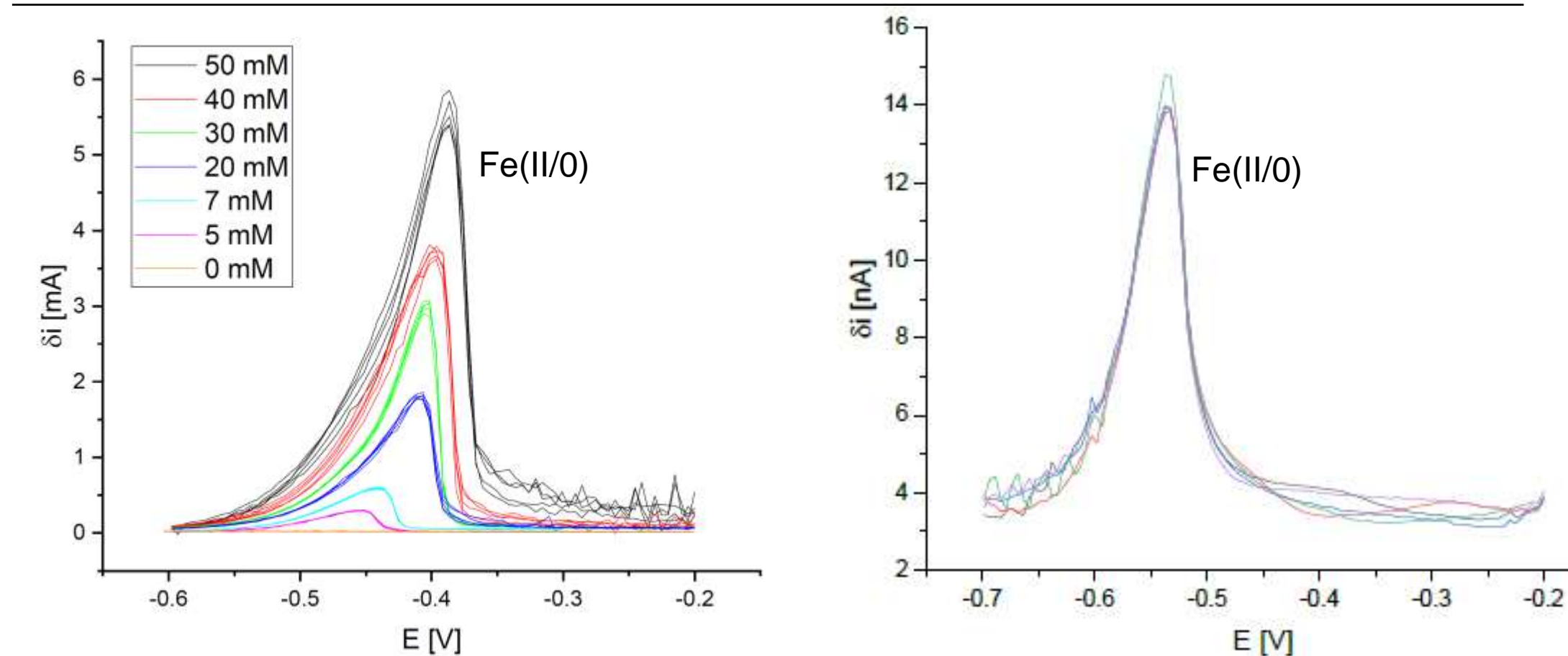


Results – simultaneous detection of Fe, Cr and Ni

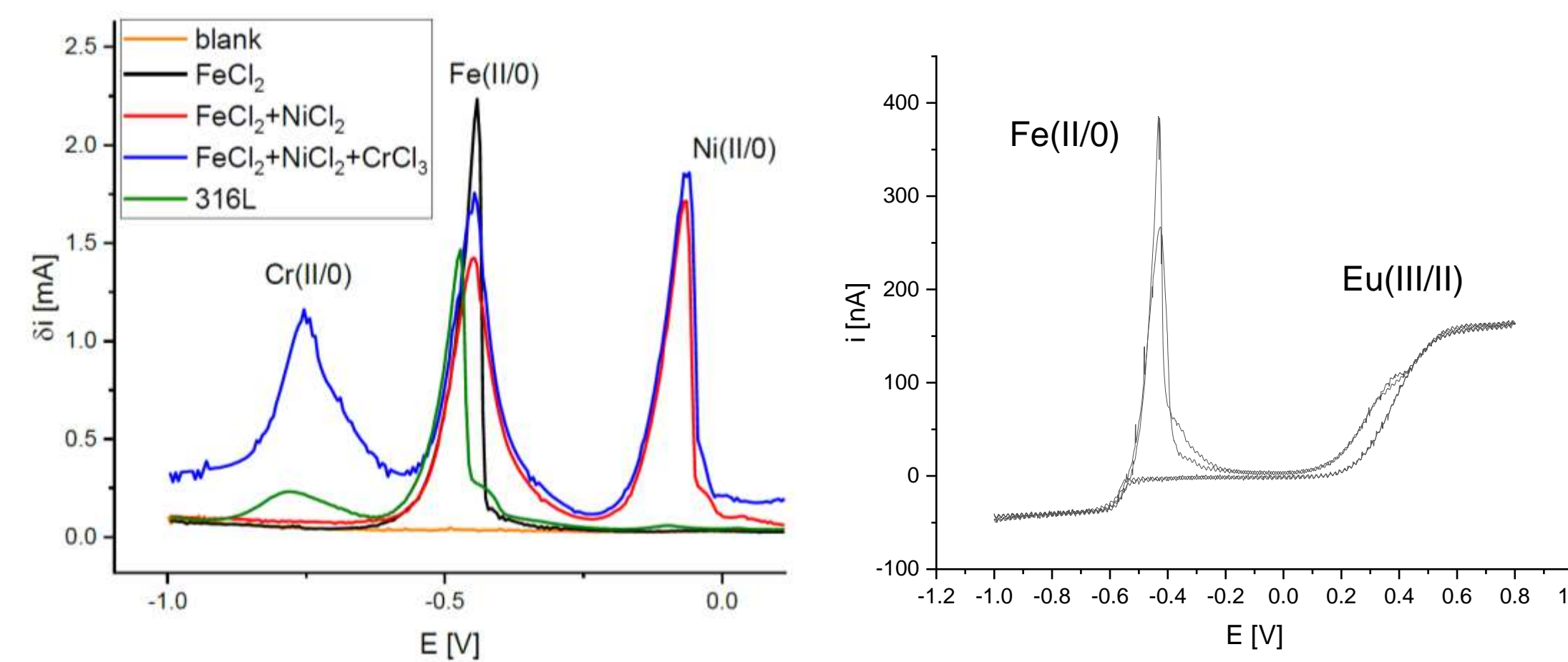
- Most important steel corrosion products are Fe, Ni, and Cr
- Quantitatively monitor Fe(II/O), Ni(II/O) and Cr(II/O)
- Fe is the most abundant and likely to be detected first

Composition of 316L steel:

C	Si	Mn	P	S	Cr	Ni	N	Mo	Fe
0.03	1.00	2.00	0.045	0.015	16.50-18.50	10.00-13.00	0.10	2.00-2.50	balance



- Macroelectrode: serial addition study confirms quantitative electrochemical detection at 5–50 mM (~0.04–0.4 wt%)
- Microelectrode: detection demonstrated at an order of magnitude lower concentration (581 μ M), estimated LOD of 10s μ M



- Simultaneously detect added FeCl₂, NiCl₂ and CrCl₃
- Detect Fe(II), Ni(II) and Cr(II) after electrochemically dissolving 316L steel
- Detection of Fe(II) from 316L steel chemically dissolved by Eu(III)

Materials and methods

- Tube furnace at 400°C, monitored in situ
- Argon atmosphere (<0.1 ppm H₂O and O₂)
- Molten LiCl-KCl eutectic contained in borosilicate crucible in borosilicate cell
- Working electrode: W macro- and microelectrodes
- Counter electrode: W macroelectrode
- Reference electrode: in-house fabricated Ag/Ag+ electrode sheathed in mullite



Objectives

- Confirm ability to electrochemically detect the predicted corrosion products of SS316L in molten LiCl-KCl eutectic
- Determine which electrochemical reaction to use to monitor corrosion
- Demonstrate the detection of corrosion products with microfabricated microelectrode
- Benchmark the sensitivity and LOD of the microelectrodes compared to macroelectrodes

Conclusion

The microelectrodes designed, fabricated and tested at the University of Edinburgh are a highly sensitive and robust electrochemical tool that can be used to monitor corrosion of SS316L in the molten LiCl-KCl salt.

Acknowledgments

- The pyrochemical research laboratory (PRL) at The University of Edinburgh where experiments were conducted
- The Scottish Microelectronics Centre also at The University of Edinburgh where the microelectrodes were fabricated
- Our colleagues at NNL during AFCP: Mike Edmondson, Mike Harrison, Nick Hodge and Angela Jackson and their teams

Introduction

The nuclear physics group at the University of Manchester leads various nuclear data measurement projects, all of which are aimed towards providing more accurate nuclear data sets for applications, including advanced nuclear technologies. These data sets feed into global nuclear data evaluations and thus have direct impact on nuclear data users.

Within the scope of the AFCP, new measurements were performed alongside analysis to help ensure suitable nuclear data is available to underpin the deployment of advanced fuels. on existing data and the work can be summarised in the following subgroups:

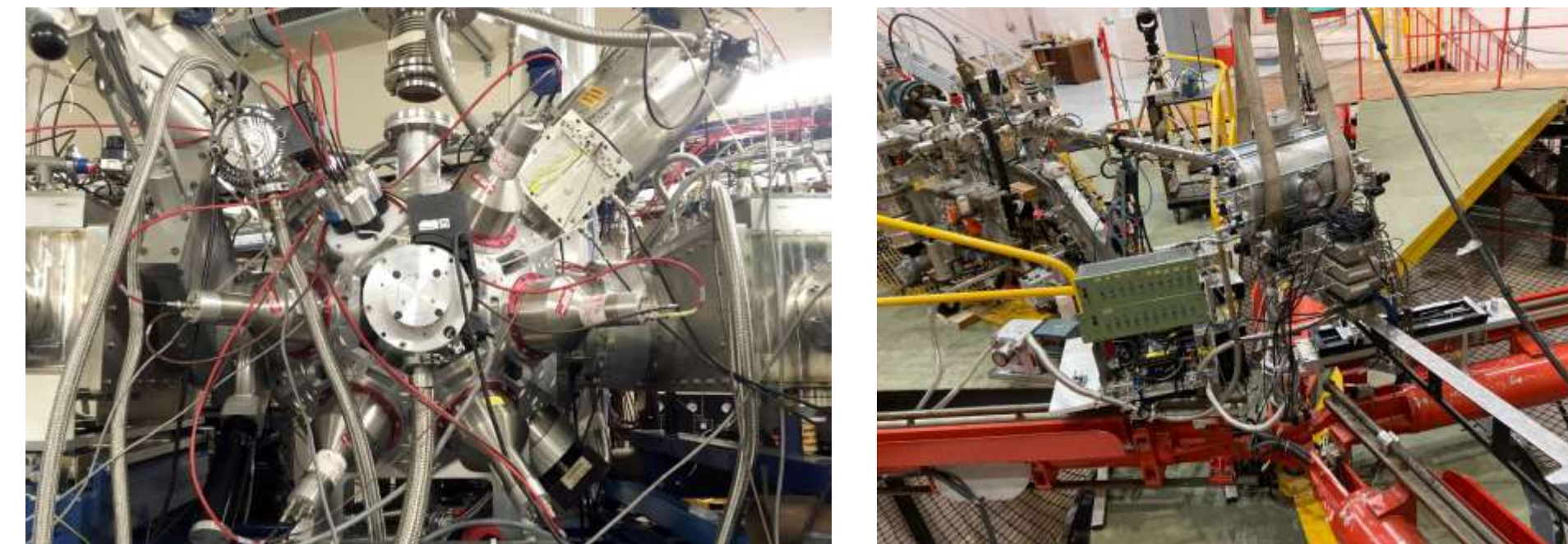
- Measurements at the neutron time-of-flight facility n_TOF
 - Prompt fission γ -ray energies and multiplicities from neutron induced fission on ^{235}U
 - Neutron capture cross section of ^{35}Cl
- Measurements of neutron induced fission cross sections and fragment angular distributions
 - Monoenergetic neutron beam of 2.5, 5 MeV and 16.5 MeV at NPL with a ^{238}U target
 - Thermal neutron beam at ILL with a ^{235}U target
- Measurements of fission fragment atomic charge Z with LOHENGRIN at ILL
- Measurements of properties of fission including the fission barrier and fragment angular, mass and charge distributions using a novel d,pf method in inverse kinematics
 - ^{238}U beam at the Argonne National Laboratory
 - Preparations for measurements using actinide beams with the ISS, CERN

Objectives

- Produce nuclear data sets to improve the quality of existing nuclear data evaluations
 - Data from ^{35}Cl will contribute to future JEFF/ENDF evaluations
 - Results of prompt γ -ray energies and multiplicities from STEFF will help to meet the NEA High Priority Request for these quantities on ^{235}U
- Develop experimental techniques to perform future measurements to meet the nuclear data requirements for Advanced Nuclear Technologies
 - Double-gridded ionization chamber will be used for fission cross section and fragment angular distribution measurements at facilities such as n_TOF, ILL, NPL, STELLAR
 - Fission barrier heights and cross sections of short-lived actinides will be measured using the ISS at ISOLDE, CERN

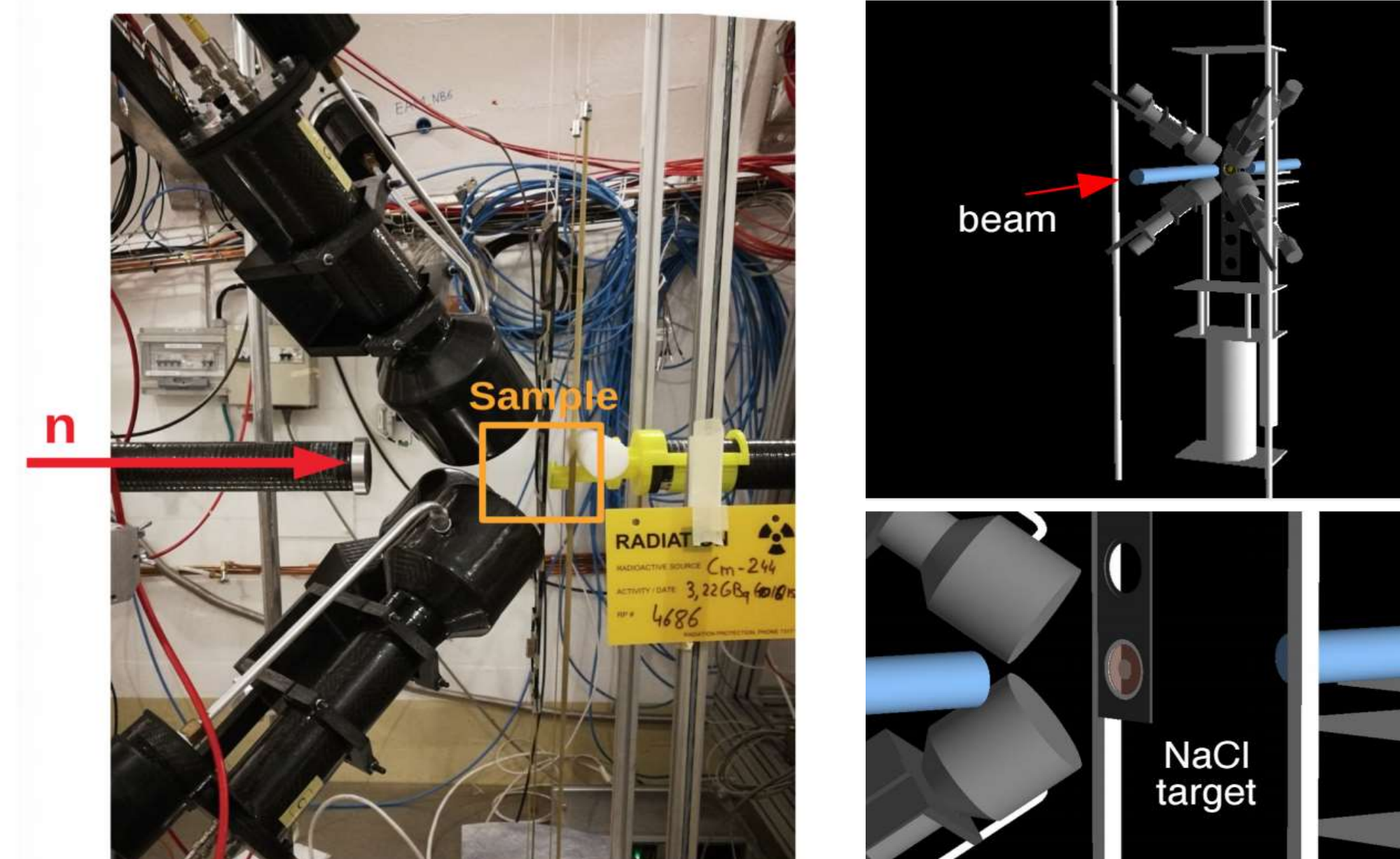
Experimental facilities and setup

A selection of the experimental setups is shown below.



LEFT: STEFF at the n_TOF facility. Neutrons originating from protons impinging a lead target ~20 m vertically beneath induce fission in a ^{235}U target at the centre of STEFF. Fission events in the horizontal axis are registered by fragment timing and energy detectors and the array of NaI scintillator detectors surrounding the target detect prompt fission γ -rays.

RIGHT: The double Frisch gridded ionization chamber at the NPL neutron source. 2.5 MeV, 5 MeV and 16.5 MeV neutrons impinge upon a ^{238}U target and fragments are detected in coincidence in either side of the chamber.

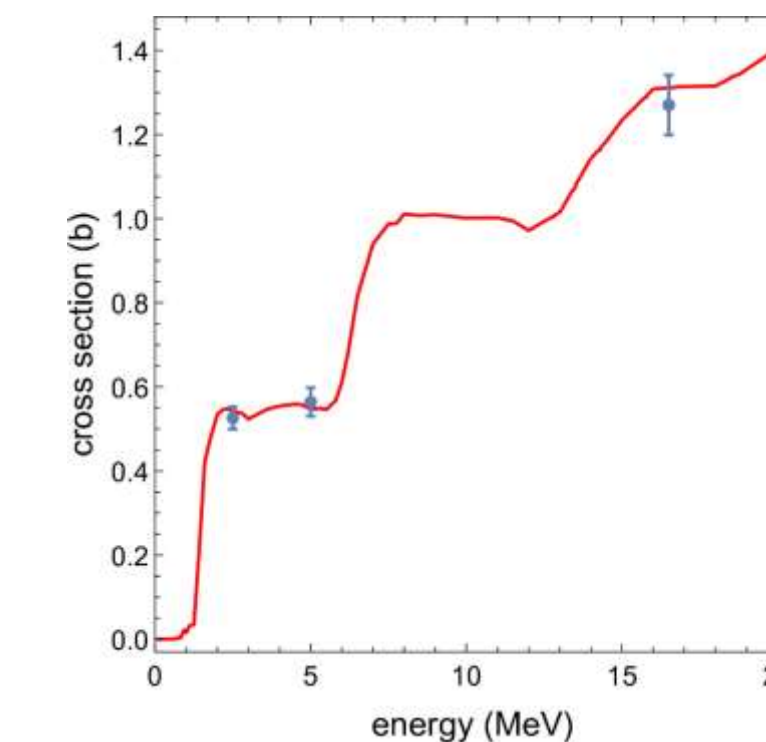


LEFT: The experimental setup for measuring the ^{35}Cl neutron capture cross section at n_TOF. Four C_6D_6 liquid scintillators surround the sodium chloride target and use the Total Energy Deposition technique to detect neutron capture reactions.

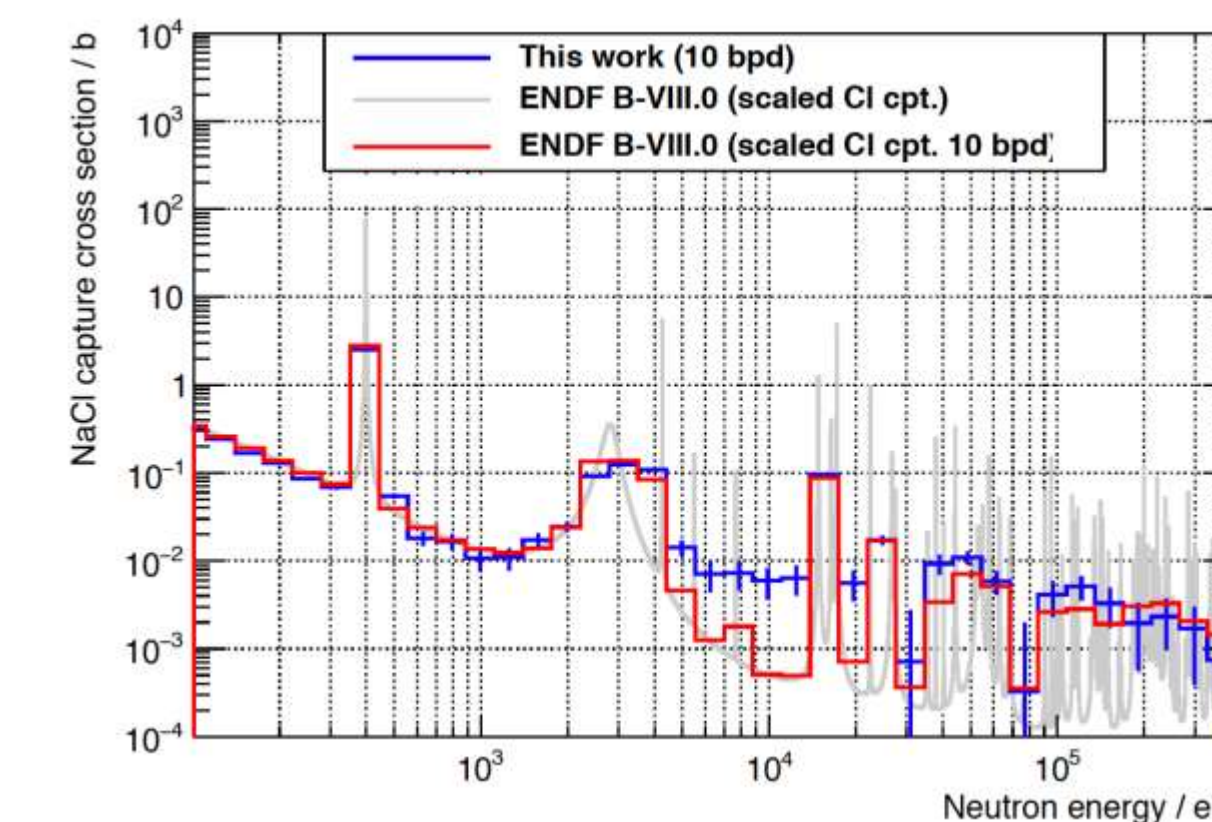
RIGHT: Monte Carlo simulations of the setup as implemented in Geant4 in order to determine the weighting functions, a crucial part of the analysis procedure

Results

The analysis of the ^{35}Cl n_TOF and ^{238}U NPL data is complete and the progress continues to be made on the other projects



The experimentally determined fission cross section of ^{238}U using the chamber at NPL compared to the JEFF 3.3 library. Agreement within uncertainties demonstrates the suitability of this setup for performing fission cross section measurements. There are plans to use this setup to measure the fission cross section of ^{236}U .



The results from the n_TOF ^{35}Cl measurement compared to the most recent evaluated libraries. Our results measure a cross section ~15% higher than the evaluations, in agreement with previous measurements. For more details on this analysis, results and implications, see the thesis of S. Bennett.

Conclusions

A rich and diverse program of nuclear data measurements and analysis has been performed, delivering quality and valuable data sets and also preparing and developing a capability for future measurements.

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Acknowledgements

The authors wish to acknowledge collaborators at NPL, ILL, ANL and the n_TOF and ISS collaborations at CERN.

This work was funded under the £46m Advanced Fuel Cycle Programme as part of the Department for Business, Energy and Industrial Strategy's (BEIS) £505m Energy Innovation Programme.

1. INTRODUCTION

Fast neutron spectrum reactors provide opportunities for a more sustainable route to Net Zero through the efficient use of resources and a reduced need for uranium mining. In addition, they offer a route (along with re-use in thermal reactors and immobilisation to a ceramic waste form followed by direct disposal) for the disposition of the UK's civil plutonium (Pu) stockpile which was acquired through reprocessing of spent nuclear fuel.

Of these options, the manufacture of MOX for use in commercial (thermal) reactors is currently ongoing internationally and was previously conducted in the UK. The alternative re-use option, in "fast reactors", builds on the UK's strong history of fast reactor fuel cycle research and development conducted as part of the UK Fast Reactor Programme (Figure 1). This included the discovery of many world firsts during the operation of the Dounreay and Prototype Fast Reactors, DFR and PFR, respectively.

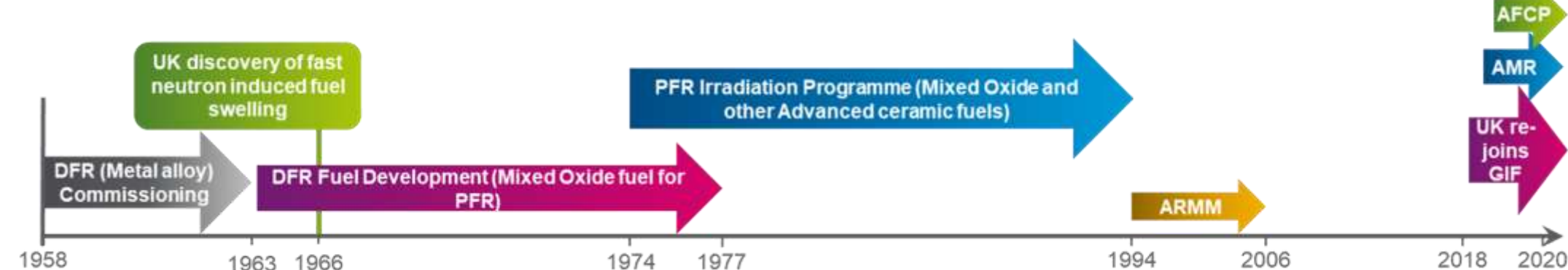


Figure 1. Brief overview of UK experience with Fast Reactor Fuels [1]

While the manufacture of MOX has a long history, including in the UK, there is however a need and opportunity for further optimisation given recent advancements in materials science, process technologies and, in the UK's instance, the available material feeds. This is especially so for fast reactors, where the unique features of liquid metal-cooled systems (higher operating temperatures and higher burn-ups) mean they require fuels with higher (than thermal reactor) fissile material contents while also offering the ability to transmute long-lived radioisotopes such as americium and at the same time producing low-carbon energy.

2. OBJECTIVES

The AFCP Fast Reactor Fuels project focused on re-establishing UK capabilities and capacities to conduct cross-cutting research that would support the development of advanced fuels and, ultimately, the deployment of liquid metal cooled reactor systems. A primary focus has been on the facility upgrades needed to conduct research into the manufacture of fast reactor MOX.

Key objectives:

- Provide the UK with an alpha-active fuel manufacture and research capability to support research into sustainable fuel cycles.
- Develop a flexible facility that enables and supports UK involvement in both national and international research programmes.
- Produce the first "fast reactor" specification MOX pellet in a generation.

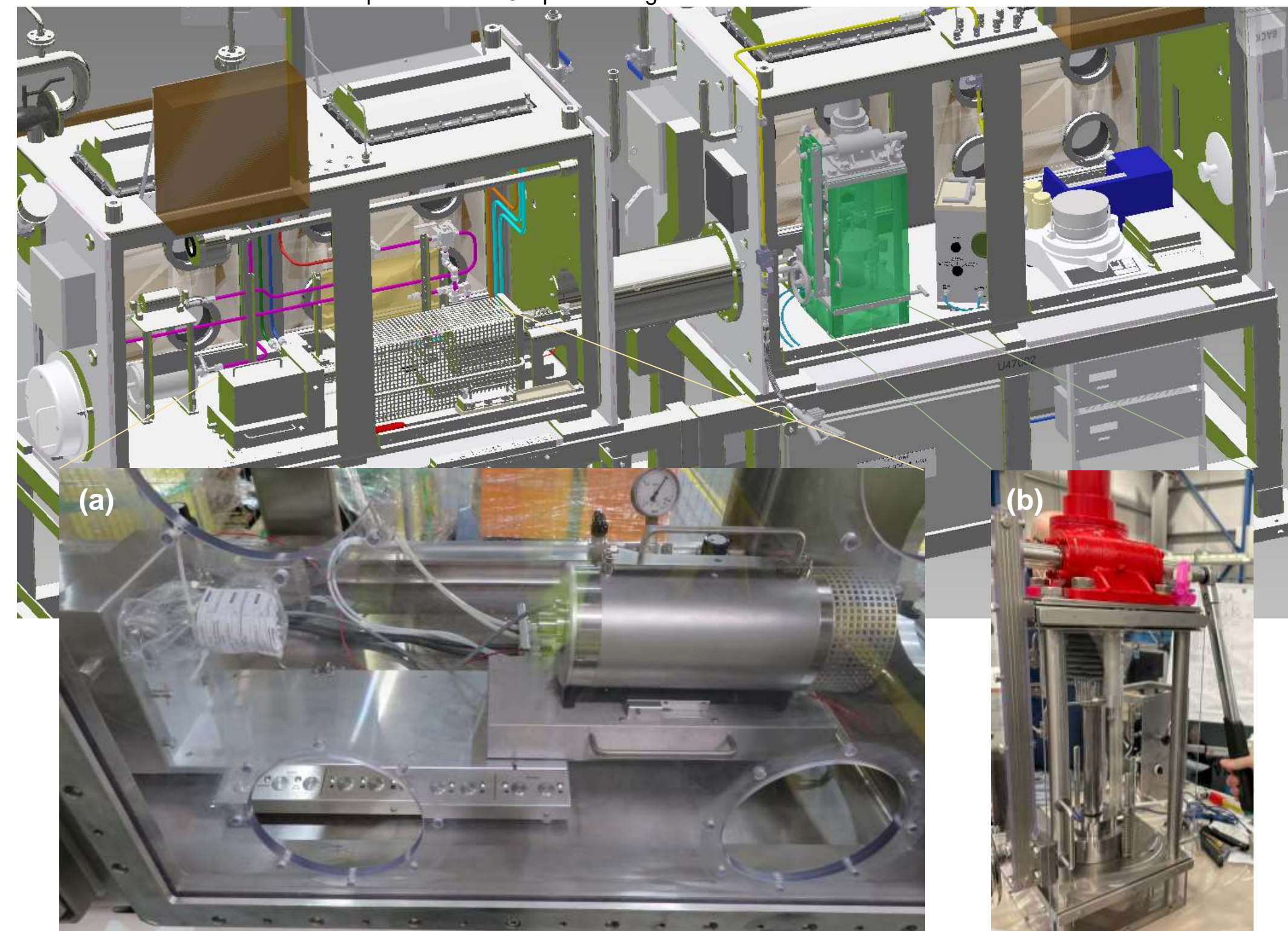


Figure 2. 3D CAD of Flexible Mini Fuel Line (main); dilatometer during trials (a) and press during Factory Acceptance Tests (b)

3. FACILITY OVERVIEW

The (Flexible) Mini Fuel Line (Figure 2) has been designed such that it is contained within two interlinked and inert atmosphere gloveboxes. It contains the necessary equipment to manufacture both solid and annular pellets and Pu-bearing ceramics.

An overview of the pellet fabrication process is presented in Figure 3.

MATERIAL IMPORT

Two processing streams are available:

- the first is for powder feeds which encompasses individual powders (e.g. UO_2 and PuO_2) or co-extracted material (e.g. $(U,Pu)O_2$ or $(U,Am)O_2$)
- the second processing stream recycles previously sintered pellets as the feed, however, the sintered pellets must first be crushed and size reduced (milled) to enable incorporation with the powder feed material.

WEIGHING

The composition of the powders is adjusted to produce pellets of the required composition (e.g. $(U,Pu)O_2$ containing 30% Pu) before further processing.

MILLING & PRESSING

The powder is then milled to yield a homogeneous mixture. This can include the pressing of low density green pellets ('slugs'), which are then granulated by forcing them through a sieve. The granulated feed is then pressed using a high load to produce a green pellet.

SINTERING

The green pellets can then be transferred to the sintering glovebox for sintering at high temperatures in the dilatometer.

4. DESIGN PROCESS & PROGRESS

The (Flexible) Mini Fuel Line (Figure 2) has followed an established and gated process throughout its design and equipment procurement and this will continue as installation in NNL Central Laboratory continues prior to handing over to commissioning. This includes various Design Reviews and Hazard and Operability Studies (HAZOP) that throughout the process are intended to identify design needs and ultimately support active operations. This is pictographically represented for the Mini Fuel Line in Figure 4.

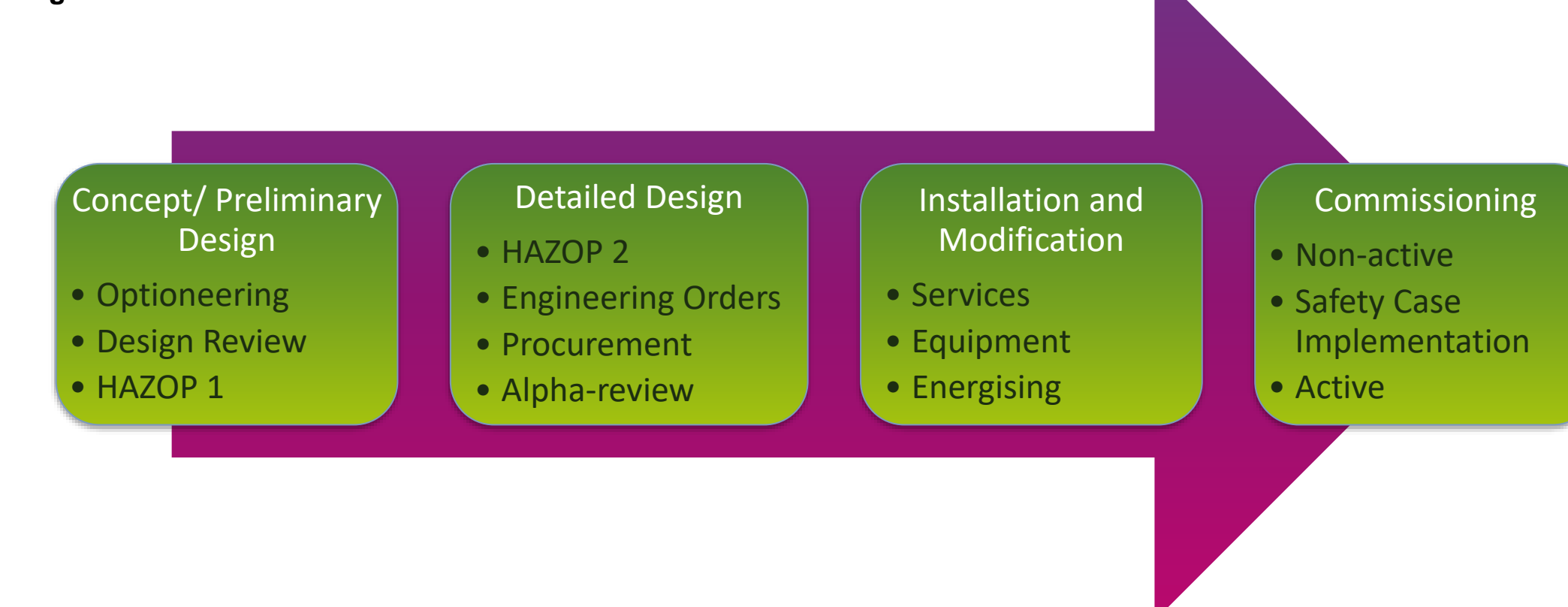


Figure 4. Design process for Mini Fuel Line, adapted from [2]

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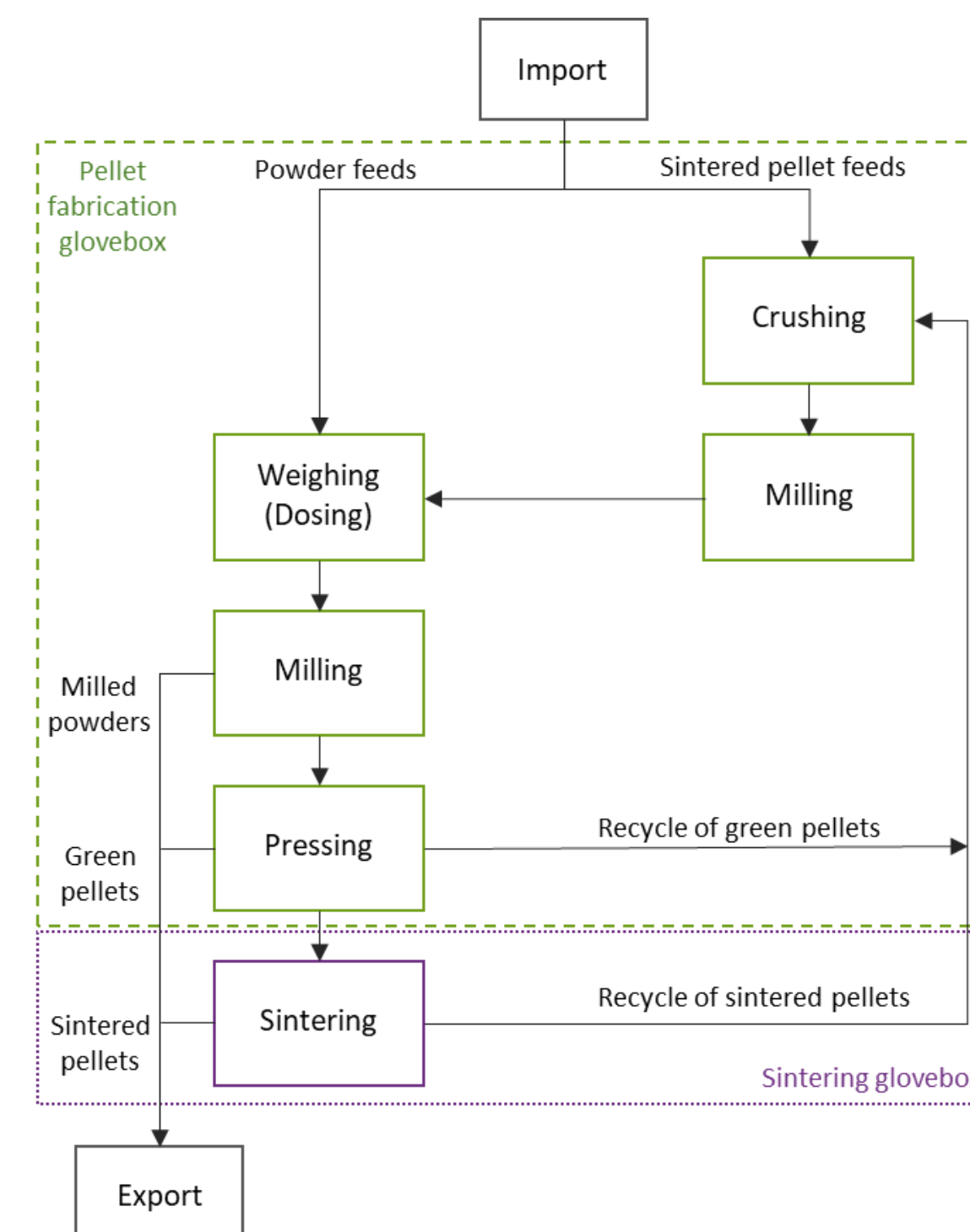


Figure 3. Pellet production process in the Mini Fuel Line, adapted from [2]

5. Results from surrogate MOX STUDIES

Prior to the fabrication of MOX pellets in the FMFL, surrogate studies, using inert or low activity materials have been conducted to evaluate the processes and fabrication conditions. Surrogate MOX pellets, containing cerium dioxide (CeO_2) as a non-active substitute for PuO_2 , have been fabricated at NNL Preston Laboratory, where a suite of equipment, which the FMFL is replicating, is available to allow parallel surrogate and active studies to be conducted.

Six $(U,Ce)O_2$ pellets, containing 70 at% UO_2 and 30 at% CeO_2 , have been fabricated using conventional powder-pellet preparation routes, to validate the procedures that will be used for MOX fabrication within the Mini Fuel Line.

- Four pellets were produced using a granulated UO_2/CeO_2 powder blend
- Two were produced from the as-milled UO_2/CeO_2 powder blend

The intention was to observe the impact of the processing characteristics on the as sintered pellets. The pellets were prepared following the route detailed in the 'Facility Overview' section and were pressed using a pressure of 5.1 tonnes/cm² and sintered at 1730°C for 4 hours in a pure hydrogen atmosphere [3].

The following observations were noted during the trial:

- Pressing characteristics of the granulated feed were superior to the powder feed.
 - Deliberate and premature release of the hold down pressure during pellet extraction (Figure 5) reiterates the importance of this during pellet ejection and validates incorporation of a press with hold-down in the FMFL.
- Pellets pressed from granulated feed achieved a higher green density.
- Defects observed in the sintered pellets indicate that the manufacturing procedure requires further development.
 - All the sintered pellets contained defects, such as cracks, end-capping or chipping, as shown in Figure 6. Some of these defects may have arisen during handling of the green bodies pellets and transfer between the laboratories for sintering (something that will be avoided in the Mini Fuel Line). Alternatively, sintering in a pure H_2 environment may have led to reduction of some of the CeO_2 to Ce_2O_3 , impacting the integrity of the sintered pellet.
 - The sintered pellets from the granulated feed have a textured surface, unlike the smooth surface of the pellets using the powder feed and reference UO_2 pellet. Further studies will investigate the cause of this.

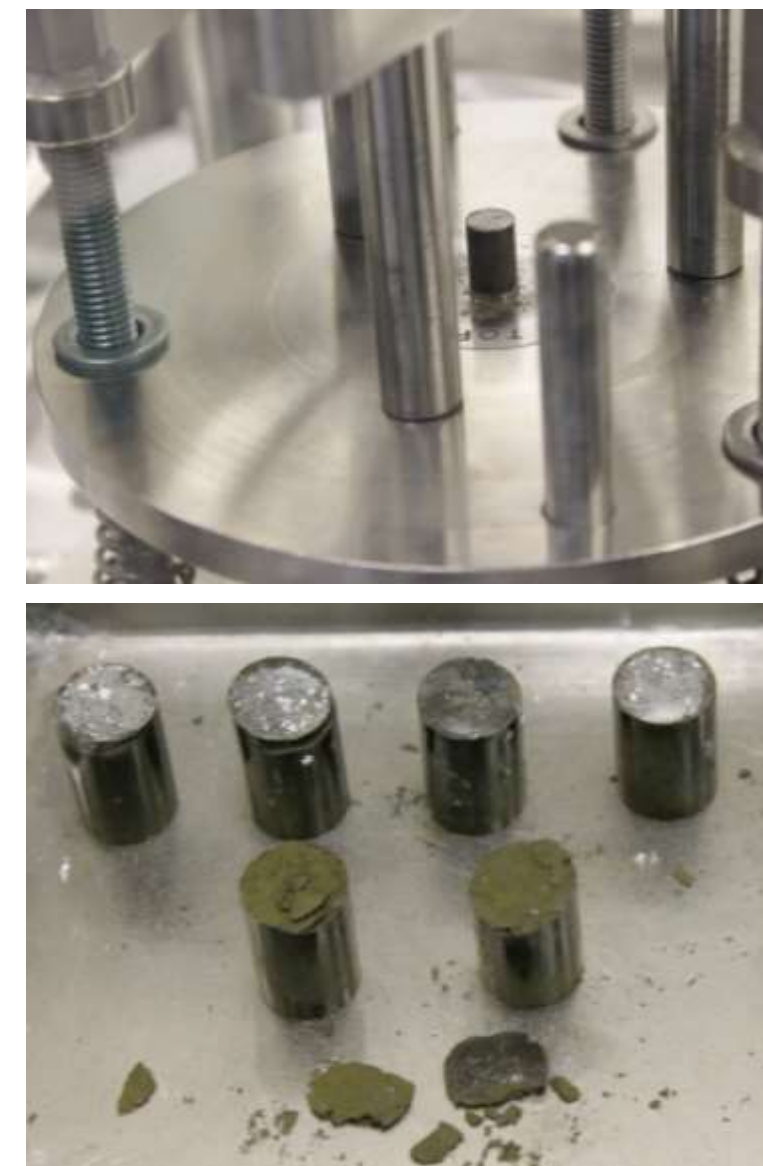


Figure 5. Surrogate green pellets [3]

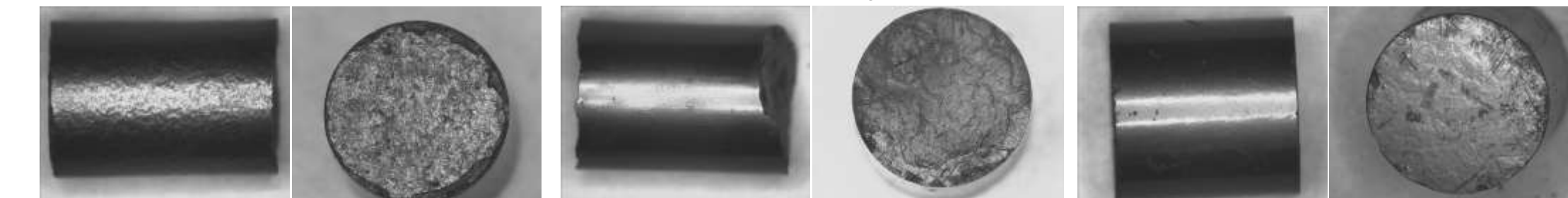


Figure 6. Surrogate sintered (Ce-MOX) pellets [3], produced from the granulated feed (left) and powder feed (centre). A reference UO_2 pellet is shown for comparison (right).

6. CONCLUSIONS

The (Flexible) Mini Fuel Line is a new capability that contains the necessary equipment to manufacture Pu-bearing ceramics and re-establishes UK MOX manufacturing facilities. This has involved determining the needs of current and potentially future users and feeding these requirements into the gated process. Progress has been made towards establishing this, with all key equipment procured and installation scheduled.

Discussions have been held as to the initial experimental programme, which while principally focused on re-establishing UK skills and knowledge in the manufacture of MOX it will expand and evolving, including in the near-term to demonstrate the viability of sustainability and advanced fuel cycles.

7. FUTURE OPPORTUNITIES

Overtime, it is envisaged that this capability will evolve into an innovation led facility able to support multiple national and potentially international customers, with possible opportunities arising for research into plutonium-doped ceramic wasteforms; disposal MOX; and novel fuels for advanced modular reactors and space applications [4].



ACKNOWLEDGEMENTS AND CONTACT

This work was funded under the £46m Advanced Fuel Cycle Programme as part of the Department for Business, Energy and Industrial Strategy's (BEIS) £505m Energy Innovation Programme

Introduction

Mixed uranium-plutonium Oxide (MOx) fuel manufacture is of high interest to domestic research efforts for long-term deposition of the UK's civil Pu stockpile^[1]

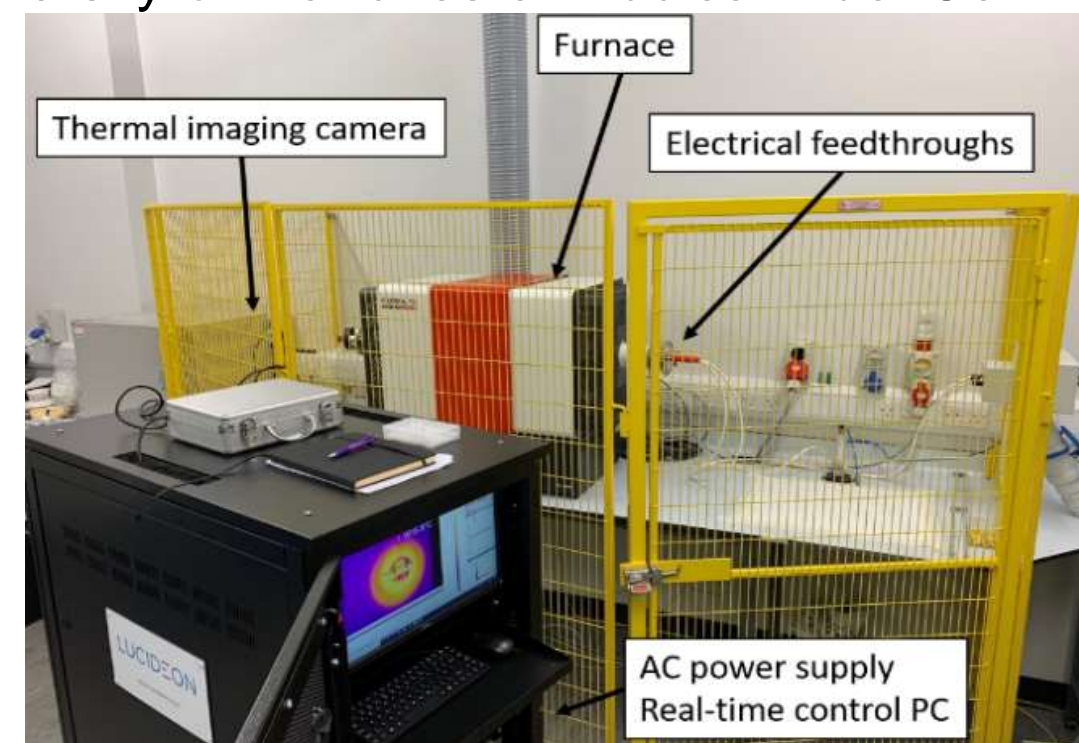
Sintering, is a high temperature process and there are economic, environmental and quality reasons for reducing the temperature. The last is true for pellets containing minor actinides, that could be used in fast reactor designs or non-terrestrial systems^[3] as these can result in pellets that don't meet the required criteria (e.g. through formation of cracks due to volatilisation) as well as present issues with safety & plant design at temperature.

Flash sintering is a novel technique shown to have significant energy and time reductions^[3]

Materials & Methods

As part of Phase 2 of the AFCP, an AC current control flash sintering capability was installed in the University of Manchester Nuclear Fuel Centre of Excellence laboratories.

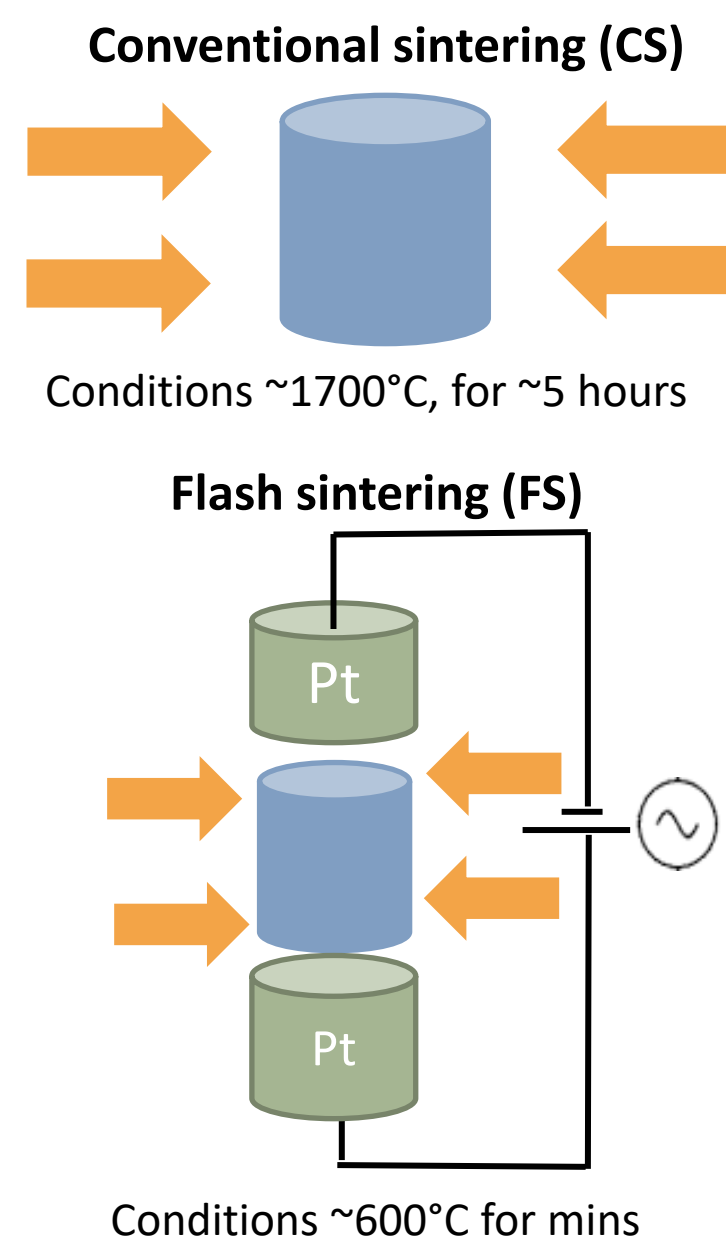
- 135V, 20A, AC power supply
- Bespoke Lucideon non-linear real time control software
- IR camera temperature measurement
- Atmosphere control for inert or reducing conditions



IDR UO_2 and LWR load MOx surrogates, manufactured using the MIMAS and SBR powder processing routes, to be used.

Current work using 6 mm UO_2 green bodies (pellets) for optimisation of a repeatable flash route before MOx surrogate studies can begin. This builds on CeO_2 studies by Lucideon and University of Manchester.

Flash variables to study include: **ambient furnace temperature**, maximum current (I_{max}), **hold time at I_{max}** , current ramp (up and down), frequency, green starting density, electrode material.



Furnace Temperature Trials

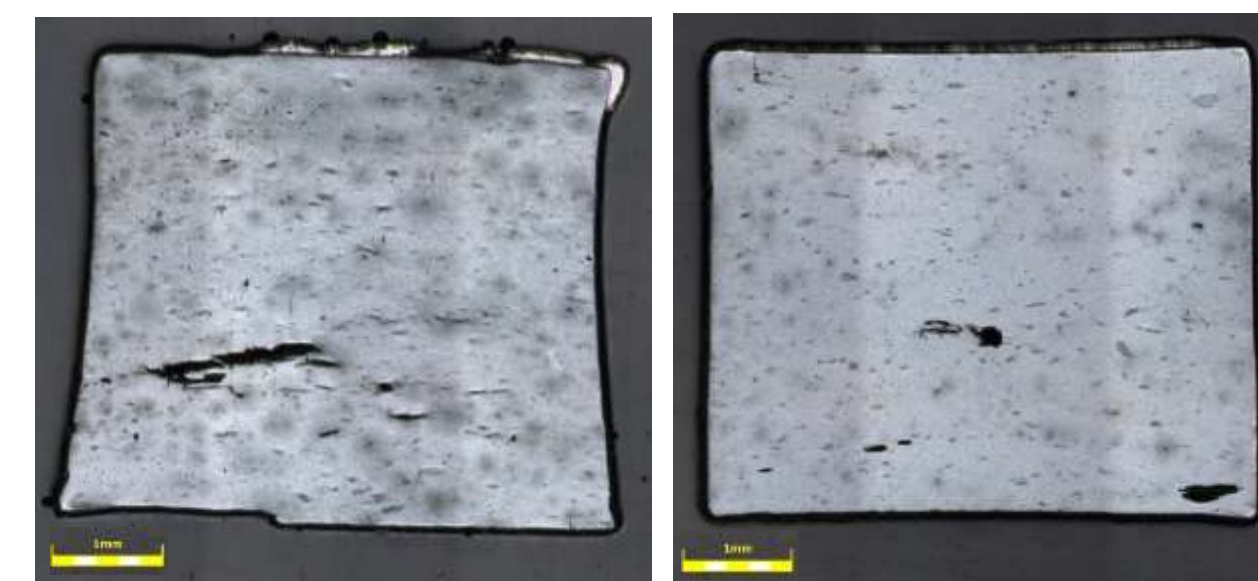
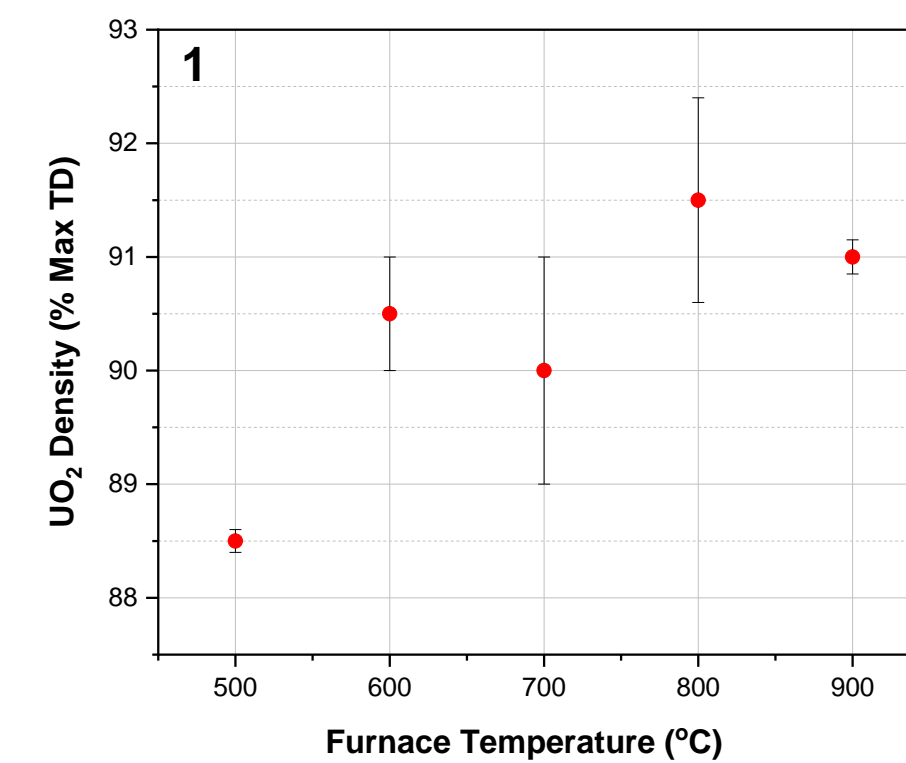


Figure 1a: 500°C Pellet

Figure 1b: 900°C Pellet

Trials studying the effect of furnace temperature on sintered pellet density have been undertaken across a range of temperature ($500\text{-}900^\circ\text{C}$) significantly below those used for conventional pellet manufacture.

All pellets flashed using identical current profile.

Plot 1 shows that above 600°C ambient temperature has only marginal effects on UO_2 density

At 500°C pellets displayed high levels of damage, including an 'hourglassing' effect (shown in **Figures 1a & 1b**)

All pellets found to be near stoichiometric $\text{UO}_{2.00}$

Microstructural Evolution

Scanning Electron Microscopy (SEM) undertaken on all samples to determine heterogeneity in grain size & pore distribution

Figures 2a-d show the typical grain & pore distribution

Little to no observable difference between centre and electrode edge, however significantly finer grains & greater pore content seen at radial edge

Implies the radial edge is colder than pellet centre line during sintering

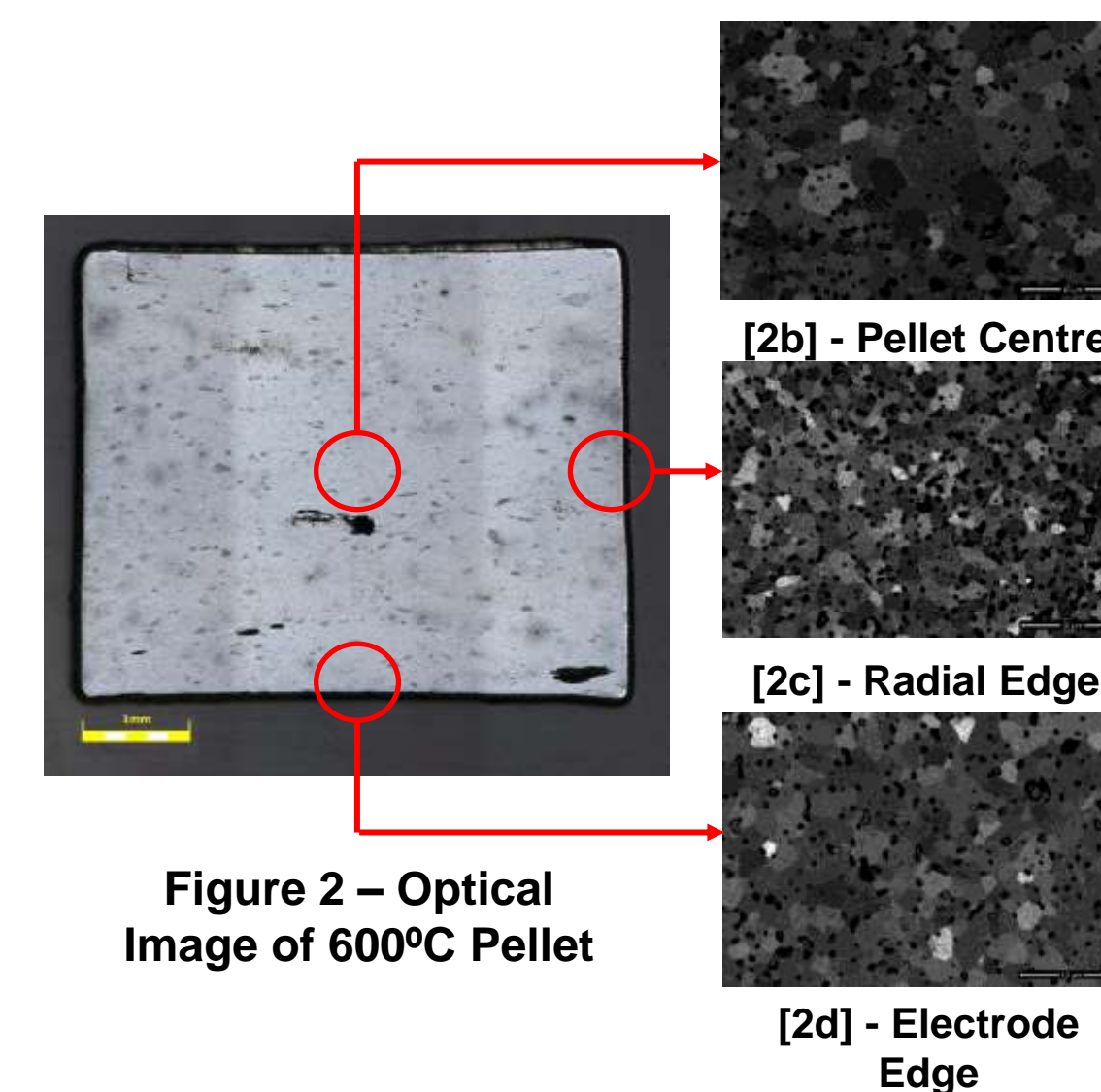
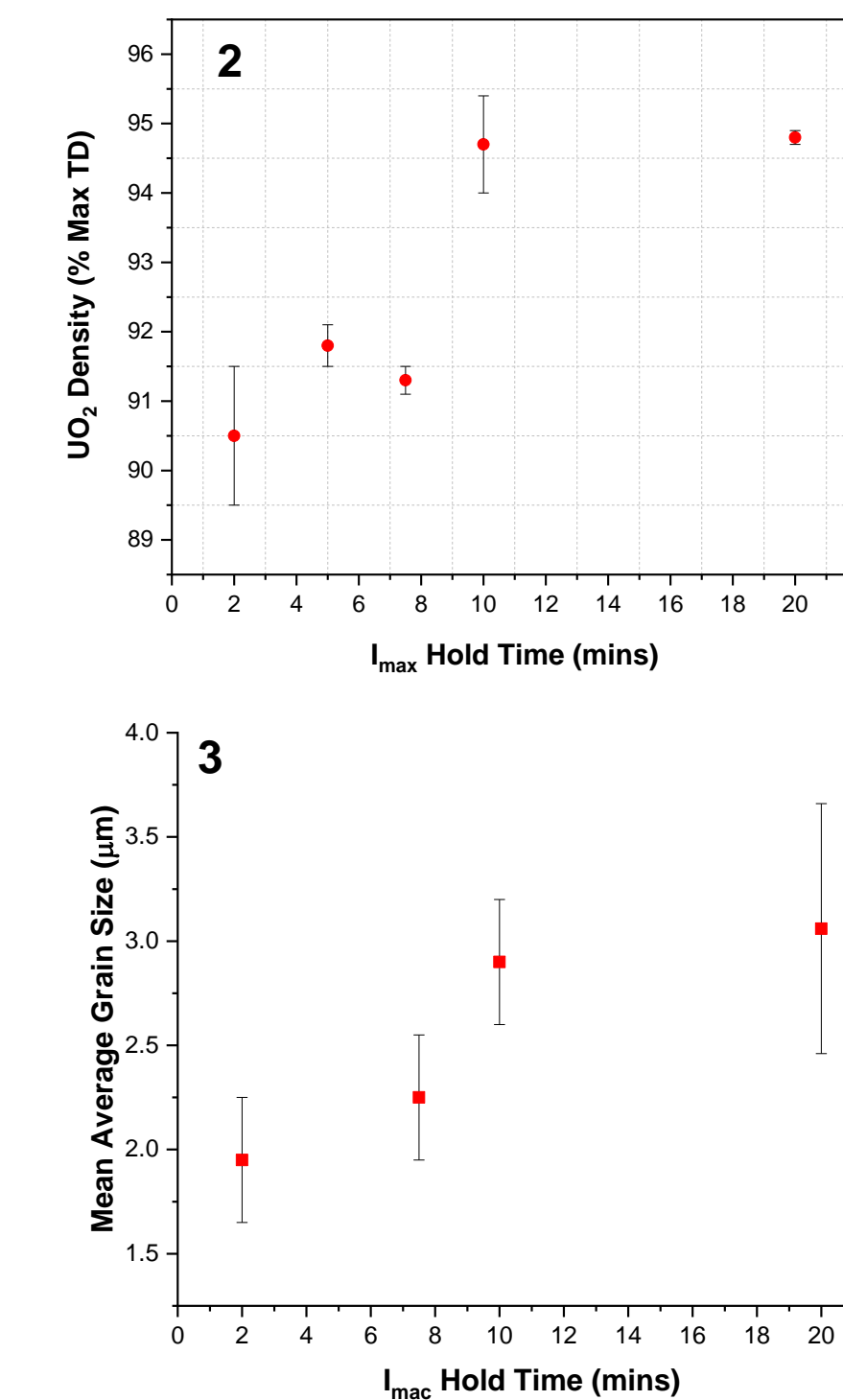


Figure 2 – Optical Image of 600°C Pellet

Optimisation Progress

Hold Time Trials (at 600°C Furnace Temperature)



Following the temperature trials, hold time trials were undertaken at 600°C to try and bring pellets to the targeted theoretical densities of 95-97 %TD.

Hold times from 2-20 minutes at an I_{max} of 8A have been studied

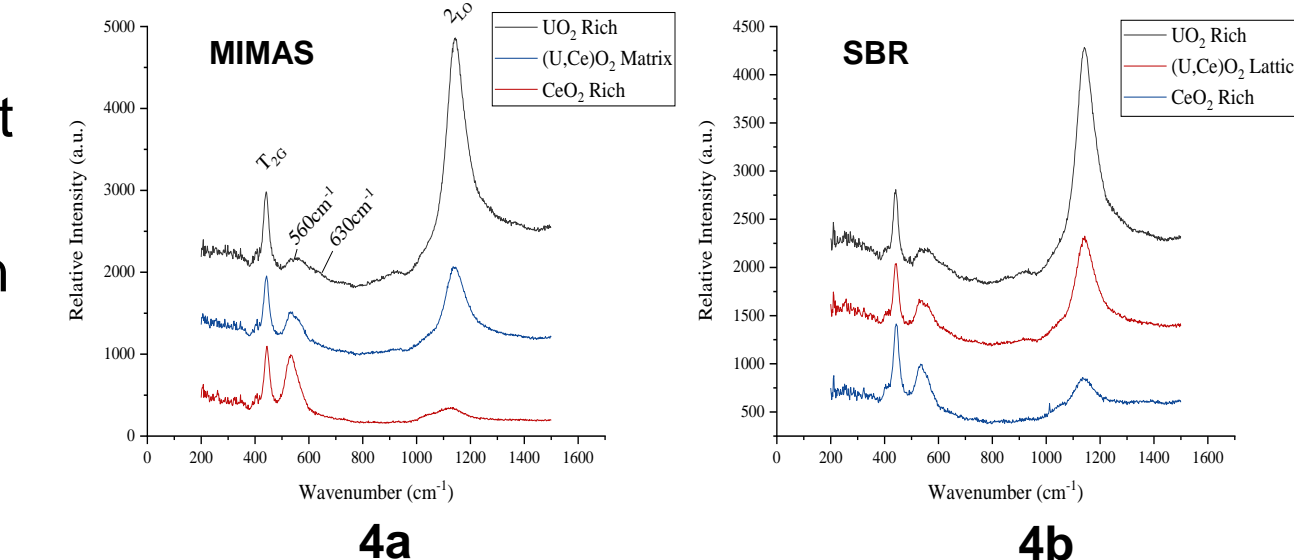
Plot 2 shows the critical influence hold time has on density evolution, with 10 minute holds or higher giving densities close to 95% without further current profile refinement

Furthermore **Plot 3** shows favorable core grain growth in UO_2 pellets when subjected to longer holds, close to the 5-10 μm grains witnessed conventionally

All pellets found to be near stoichiometric $\text{UO}_{2.00}$

Raman Spectroscopy and EDX mapping have found no significant Pt interaction at electrode layers

Plots 4a & 4b show typical raman spectra of MOx surrogates manufactured conventionally



Conclusions

Promising inroads are being made to assess the suitability of flash sintering for UO_2 manufacture. The project has obtained near 95 % theoretical density pellets with an electric current hold-time of 20-minutes at 600°C and minimal further technique refinement. Significantly reducing the sintering time.

Further work will focus on varying **current ramps** and **maximum current ceilings** during flash to try and fully refine density and enhance grain growth prior to surrogate MOx trials.