

Theme 1: AGR, Magnox and Exotic Spent Fuels

Theme Leads: Scott and Evans

DISTINCTIVE Roadshow Meeting
1st July, 2014
Sellafield

The Technical Challenge

- Current, declared lifetime for AGR power stations will result in ~8800 t of AGR fuel across whole fleet
 - >2,300 t has been reprocessed to date
- NDA's preferred option for AGR SF is to keep it in interim storage prior to packaging for disposal in the UK GDF in *ca.* 2075
- Risks with long-term wet storage of AGR SF
 - So transition to dry storage may be preferred
- Also Magnox SF in Sellafield ponds
 - Where retrieval and repackaging is needed
- For both SF types this transition, as well as dry store environment carries risks
 - e.g. to integrity of AGR cladding or hydride formation on U metal
 - So, better understanding is required before implementation

The Technical Challenge (2)

- Goals of this SF theme:
 - To increase knowledge and develop mechanistic understanding of the processes involved
 - Quantify the physiochemical evolution of SF and different waste forms in storage environments with specific emphasis on aqueous (pond) as well as dry storage
 - Develop an internationally respected and integrated team of experimentalists, theoreticians and modellers capable of tackling SF waste management problems

Aims and Objectives

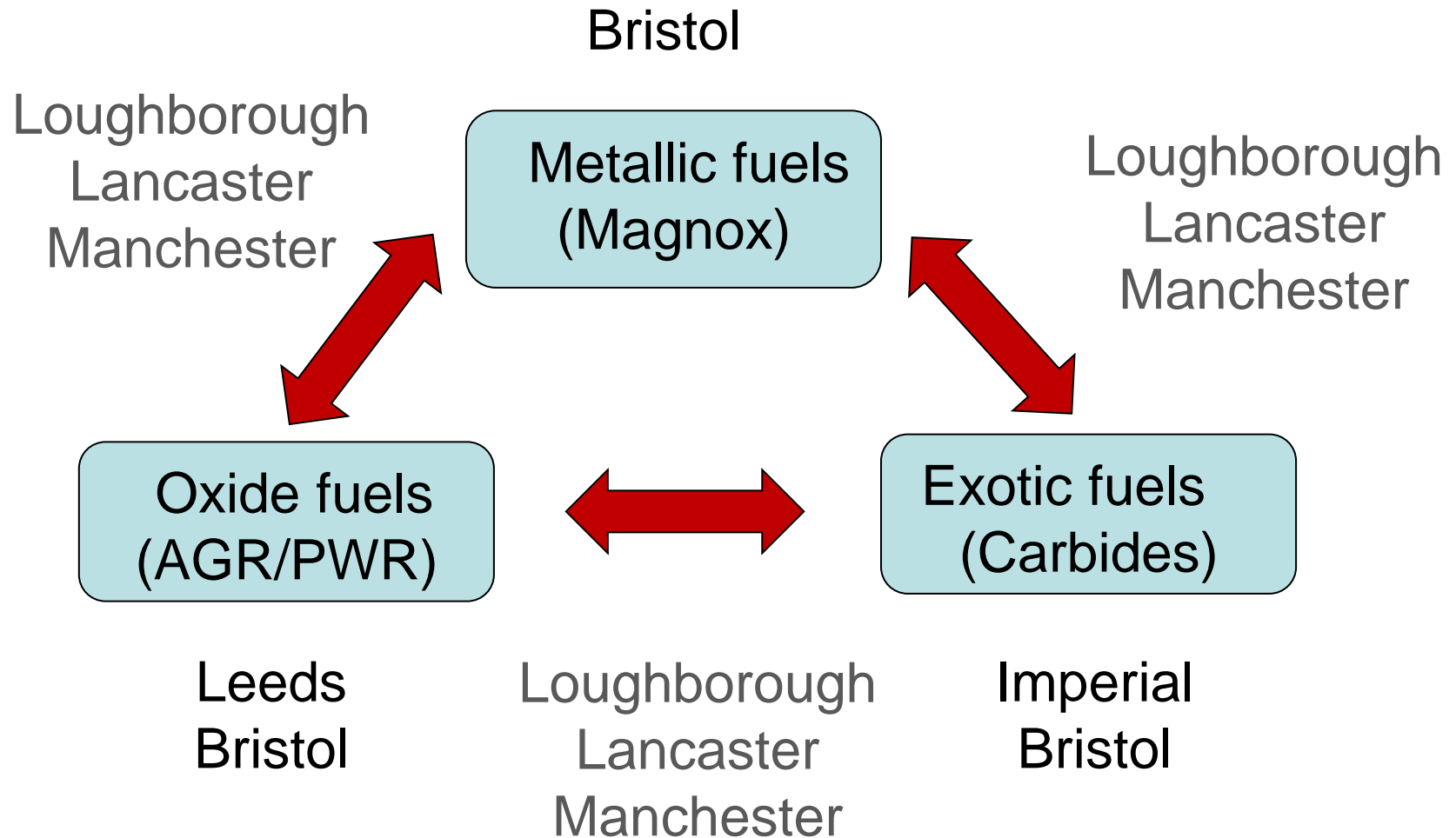
Aim:

To provide technical underpinning to the options for the management of the UK's AGR, Magnox and Exotic Spent Fuels

Objectives:

1. Understand evolution of Magnox and exotic SF during recovery from aqueous storage, drying and repackaging
2. Develop spectroscopic methods for improved determination of SF dissolution and corrosion rates in water.
3. Determine optimum drying conditions for AGR fuels and subsequent surface reactivity and alteration of unclad UO_2 in dry storage
4. Determine consequences of radiation damage in SF, cladding and other waste forms for safe long term storage
5. Determine suitable waste management options for spent carbide fuels

Interconnected study



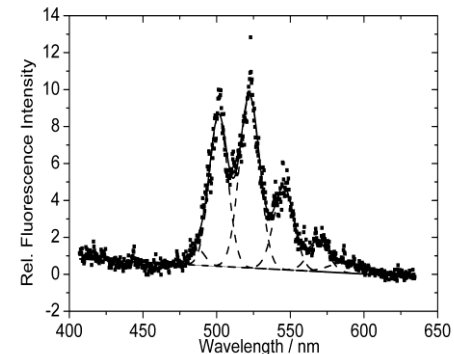
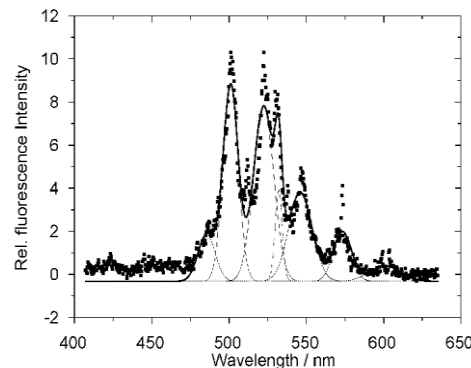
WP 1.3.1 Wet Fuel Storage Issues

Evans & Read (Loughborough), Boxall (Lancaster)

- Time Resolved Laser Fluorescence Spectroscopy (TRLFS) – technique for fundamental actinide & lanthanide speciation studies
- Corrosion and dissolution rates in water will be investigated using TRLFS by a PDRA working at Loughborough
- System will be modified/optimised to investigate dissolution and corrosion rates of uranium fuels
 - U, UC₂ and UO₂ will be investigated
 - Particular focus on the solid-solution interface
- TRLFS is unique in being able to determine in-situ metal speciation at environmentally relevant (pM) concentrations
 - Essential for incipient corrosion of speciality metals or alteration of ceramic and other materials

Wet Fuel Storage Issues (2)

- TRLFS is a very sensitive technique providing information on oxidation state
 - U(VI) phases emit characteristic fluorescence signals and can be distinguished from an unaltered U(IV) subsurface
- Final part of this project will be to extend investigations to mixed U/Pu and SYNROC materials in conjunction with Sellafield Ltd.
- Active working in NNL's Central Laboratory will be a key part
 - Where there is a state-of-the-art, Am-active TRLFS



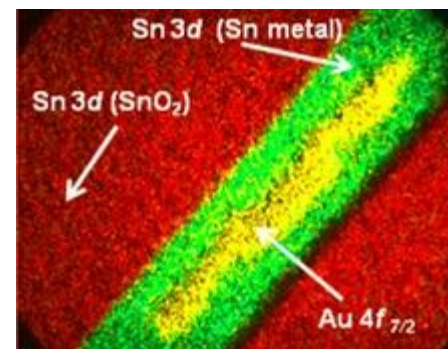
WP 1.3.2 Transitions to Dry Fuel Storage

Scott & Hallam (Bristol), Hanson (Leeds)

- PDRA project at Bristol will use surface analysis techniques to investigate physiochemical changes in SF - specifically uranium metal and exotics – during recovery from aqueous storage, forced drying and repackaging in a dry engineered containment system
- Key aspect will be determination and evaluation of reactions occurring at material surface
- Links directly to planned recovery and repackaging of spent U in Sellafield ponds
 - But also relates to the study of both non-irradiated Magnox U and UC_2
- Ultimate aim is to provide sufficient understanding to underpin safety case for repackaging Sellafield's oldest wastes

Transitions to Dry Fuel Storage (2)

- Bristol PhD will explore surface reactivity of UO_2 in simulated dry and wet environments, and under irradiation
- Project will combine surface analysis and modelling approaches to study oxidation, hydrogenation, hydrolysis and photocatalysis on uranium oxide surfaces
- Programme is supported by a new £1.8M NanoESCA system in Bristol for surface chemical state mapping and quantification



Thin film growth

Grown using Reactive DC
Magnetron Sputtering

Advantages

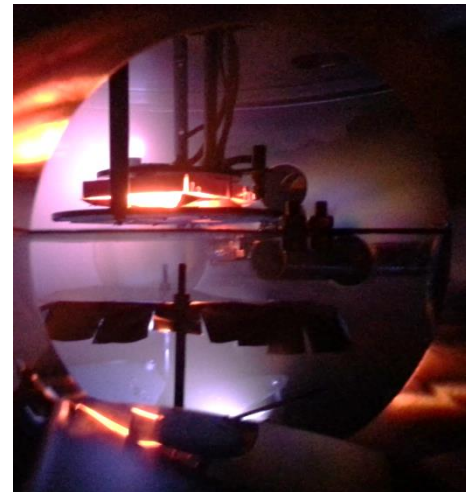
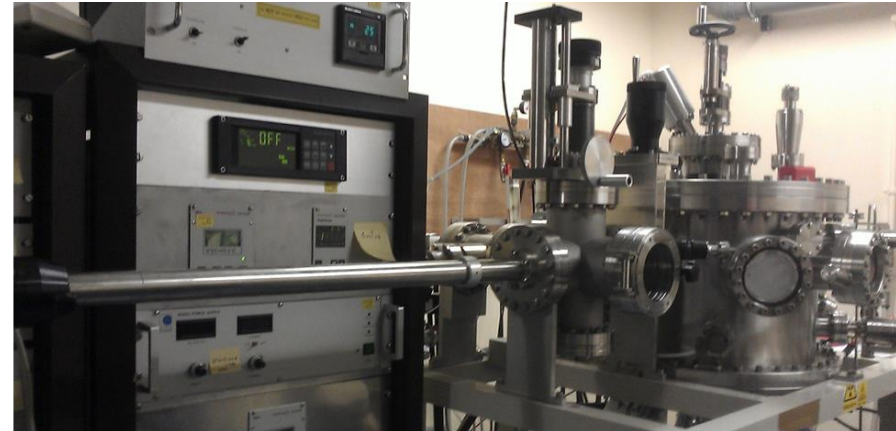
- High deposition rates
- Uniform growth
- Good control of stoichiometry

Growth Conditions

Temperature: $\approx 800\text{ }^{\circ}\text{C}$

Oxygen Pressure: $5 \times 10^{-5}\text{ mbar}$

Argon Pressure: $7.2 \times 10^{-3}\text{ mbar}$



Transitions to Dry Fuel Storage (3)

- Linked PhD at Leeds will explore optimum drying conditions for AGR fuels using small-scale tests on a simulant AGR fuel element
 - Test element will consist of cladding representative of that stored in a wet environment with a sealed simulant pellet inside
- High degree of monitoring to ensure a full mass balance can be constructed and physical and chemical processes identified
- Output will be an experimentally validated process model able to predict optimum conditions for AGR drying

WP 1.3.3 Long-term Storage Effects & Exotic Fuels

Lee (Imperial), Jimenez-Malero, Pimblott & Sherry (Manchester)

- Longer term stability issues associated with storage of SF will be explored by 2 linked PhD projects
- Consequences of radiation sensitisation for AGR fuel cladding will be investigated at Manchester
 - Use new irradiation capabilities at Dalton Cumbrian facility (DCF)
- Key aim is to elicit role of irradiation sensitisation on stability and performance of AGR fuel cladding through radiation damage experiments
- Data will underpin fundamental understanding of corrosion mechanisms and rates

Long-term Storage Effects & Exotic Fuels (2)

- PhD student at Imperial College will explore options for storage of exotic SFs
 - Especially carbides
- Project (supported by Bristol) will examine treatment options
 - e.g. thermal conversion to UO_2 , direct disposal, encapsulation and immobilisation
- Variety of analytical approaches, underpinned by modelling, will be used to assess corrosion rates and mechanisms
 - Ultimately project will provide data to inform waste management strategy for these unusual wastes

LDE programme (STFC-DLS)

- **Scott** - Acceptance onto the **Long Duration Experiment** programme at Diamond; Starting October 2014
- Incorporate the use of thin-film samples to provide time-resolved observations of simulant U, UO_2 waste materials
 - Corrosion rates and products
 - Solid state phase transformations
 - Dissolution
- I11 beamline (powder diffraction)
- Measurements weekly for up to 2 years

Experimental design is underway