ATLANTIC: Accident ToLerANT fuels In reCycling

Advanced Technology

PI – Prof. Bruce Hanson

Nuclear Academics Discussion Meeting

7th to 8th September 2021

Cambridge

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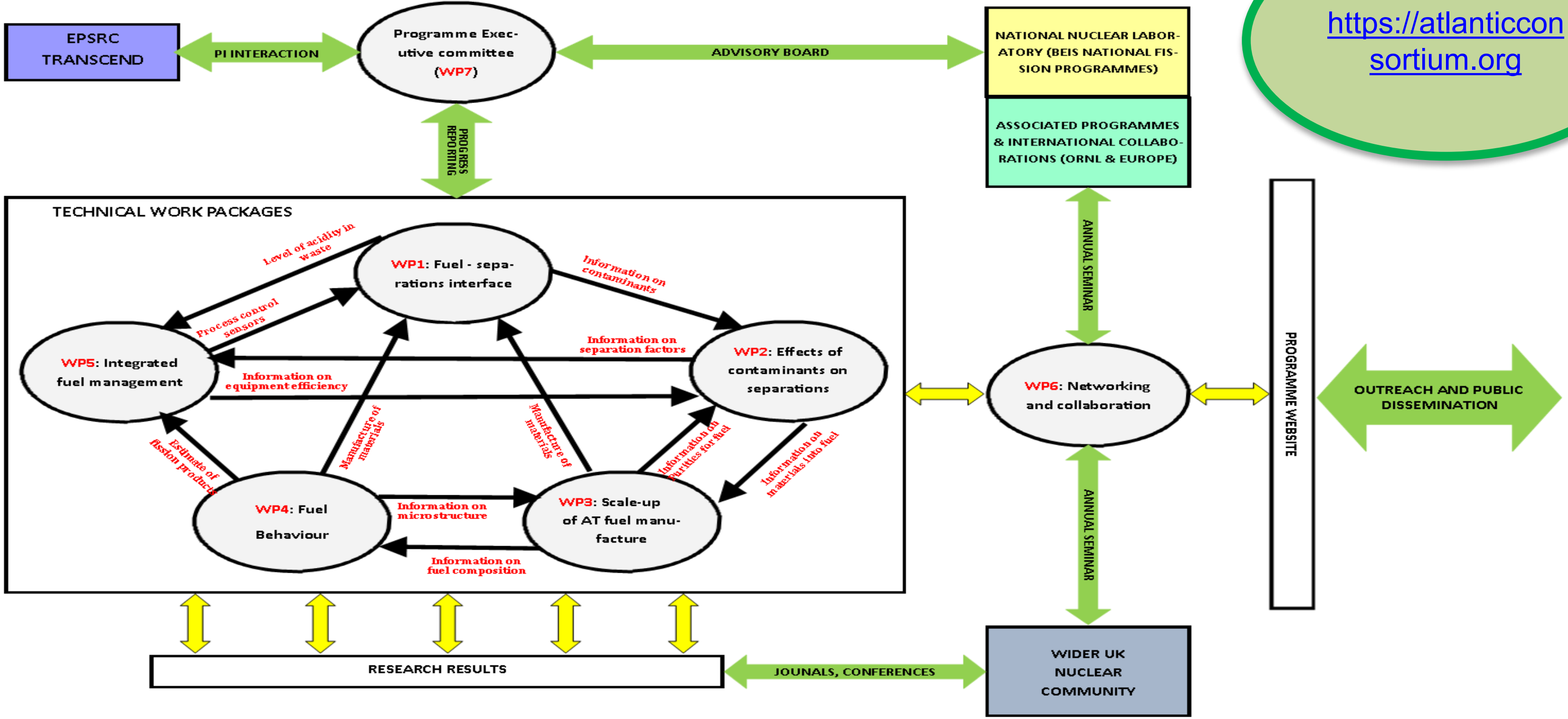
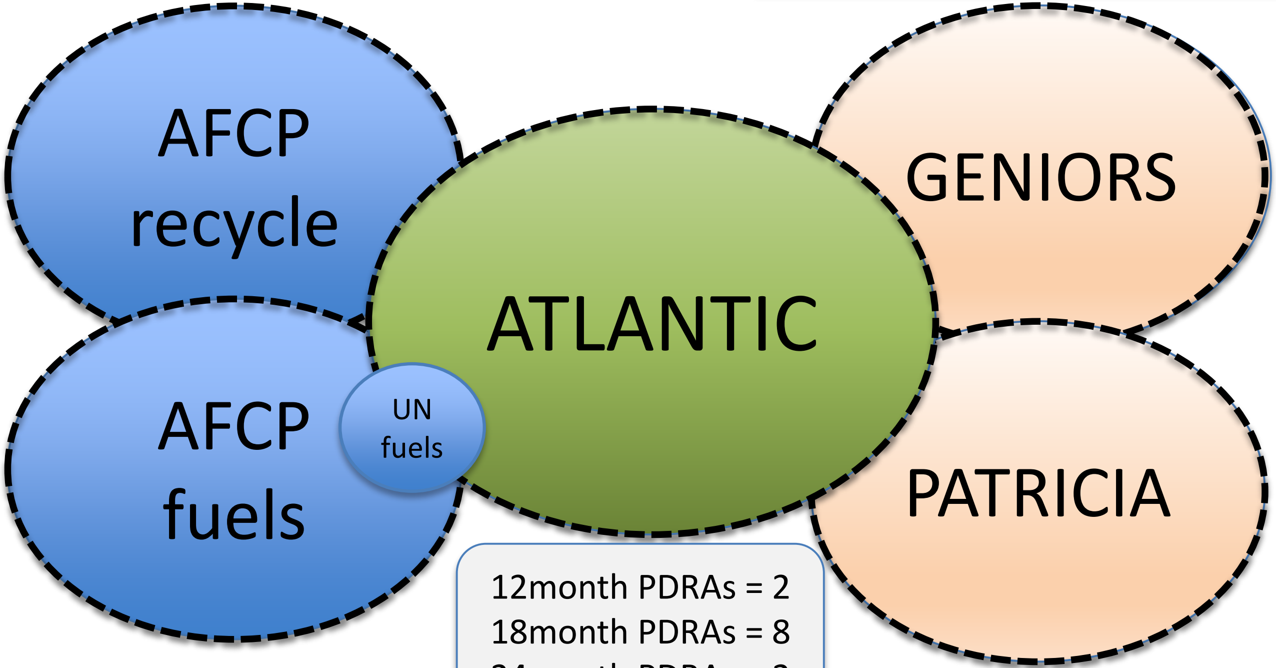
Atlantic – PI update



https://atlanticconsortium.org

Programme Links

GENIORS + ATLANTIC + AFCP = SUBJECT OF STUDY



12month PDRAs = 2
18month PDRAs = 8
24month PDRAs = 2

1st annual meeting at IWM had representatives from many related programmes

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Atlantic – technical highlights

•WP1 – **Julio Vasquez-Chavez (UoL)** is investigating voloxidation of fuels as a pretreatment for reprocessing; using air and steam on Zr. At >900^{o}C the cladding forms a brittle oxide.

•WP2 - **Steve Faulkner (Oxford)** is measuring speciation by deconvolution. Developing methods to separate signals using time-, wavelength-, and temperature dependent luminescence spectroscopy.

•WP3 - **Rob Harrison (UoM)** working on Ce_3Si_2 oxidation as U_3Si_2 surrogate. TGA, XRD, HRTEM, STEM-EDS and EFTEM have confirmed the formation of CeO_2, SiO_2 and Si up to 750^{o}C in air.

•WP4 - **Eleanor Lawrence Bright (UoB)** is characterising corrosion and oxidation of UN surfaces using TEM, XRR, XPS. UN surface passivates at room temperature (U_2N_3 interlayer forms).

•WP5 - **Ilka Schmueser (UoE)** is developing electrochemical sensors for process control.
Use electrochemistry to generate a signal that tells you something about a target chemical.

WP1.3 UN Fuel Inventory at Higher Burnups Using FISPIN

Work carried out in AFCP Fuels Programme

- Attempting to create the first UN SIMFuel that replicates the chemical properties of this material after removal from a reactor.

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Model inventories used for the thermodynamic calculations

In spent fuel with the highest BU (60 MW d kg^{-1}) the solid solution comprises:

- 87 mol% UN,
- Transuranics nitrides (AnN) 1.4 mol% (with 1.2 % PuN),
- Lanthanide nitrides (LnN) 2.5 mol% (with 1 mol% NdN)
- Transition metals (TrN) 1.7 mol%.

#	BURNUPS (GWd/tU)	IRRADIATION TIMES (YEARS)
1	5	½
2	10	1
3	15	1½
4	20	2
5	25	2½
6	30	3
7	35	3½
8	40	4
9	45	4½
10	50	5
11	55	5½

Nitride and silicide SIMFUELS (AFCP)

Dissolution Trials (ATLANTIC)

WP2.2 Practical Statistical Modelling of Spent Fuel Compositions

The University of Manchester

Create a dataset for Gen III(+) reactor systems (e.g. EPR) for ATLANTIC SF targets

- To support our separations work, various key parameters required for unknown SNF compositions:
  - Elemental (g/tHM)
  - Decay Heat (W/tHM)
- Both can be derived from isotopic concentrations
- Calculate from basic input parameters:
  - Initial Enrichment (%235U or %Pu)
  - Burnup (GWd/tHM)
  - Post- Reactor Cooling Time (y)

Isotopes modelled

Light Fission Products	Heavy Fission Products	Actinides
^{4}He	^{117}Sn, ^{118}Sn, ^{119}Sn, ^{120}Sn, ^{122}Sn, ^{124}Sn, ^{126}Sn	^{234}U, ^{235}U, ^{236}U, ^{238}U
^{77}Se, ^{78}Se, ^{79}Se, ^{80}Se, ^{82}Se	^{121}Sb, ^{123}Sb, ^{125}Sb	^{237}Np
^{81}Br	^{125}Te, ^{126}Te, ^{128}Te, ^{130}Te	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu
^{82}Kr, ^{83}Kr, ^{84}Kr, ^{85}Kr, ^{86}Kr	^{127}I, ^{129}I	^{241}Am, ^{242m}Am, ^{243}Am
^{85}Rb, ^{87}Rb	^{128}Xe, ^{130}Xe, ^{131}Xe, ^{132}Xe, ^{134}Xe, ^{136}Xe	^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm
^{86}Sr, ^{88}Sr, ^{89}Sr, ^{90}Sr	^{133}Cs, ^{134}Cs, ^{135}Cs, ^{137}Cs	
^{89}Y	^{134}Ba, ^{135}Ba, ^{136}Ba, ^{137}Ba, ^{138}Ba	
^{90}Zr, ^{91}Zr, ^{92}Zr, ^{93}Zr, ^{94}Zr, ^{95}Zr, ^{96}Zr	^{139}La	
^{95}Nb	^{140}Ce, ^{141}Ce, ^{142}Ce, ^{144}Ce	
^{95}Mo, ^{96}Mo, ^{97}Mo, ^{98}Mo, ^{100}Mo	^{141}Pr	
^{99}Tc	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd	
^{100}Ru, ^{101}Ru, ^{102}Ru, ^{103}Ru, ^{104}Ru, ^{106}Ru	^{147}Pm, ^{148m}Pm	
^{103}Rh	^{147}Sm, ^{148}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{154}Sm	
^{104}Pd, ^{105}Pd, ^{106}Pd, ^{107}Pd, ^{108}Pd, ^{110}Pd	^{153}Eu, ^{154}Eu, ^{155}Eu	
^{109}Ag, ^{110m}Ag	^{154}Gd, ^{156}Gd, ^{158}Gd, ^{160}Gd	
^{110}Cd, ^{111}Cd, ^{114}Cd	^{159}Tb	
^{115}In		

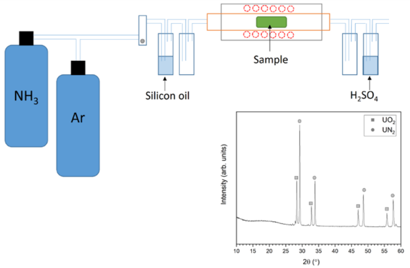
WP3.2 Synthesis of uranium nitride

Synthesis by reaction with ammonia gas

- An ammonolysis reaction furnace has been designed, tested and operated successfully – Fig 1.
- Ammonolysis of UF_4 proved partially successful, yielding phase assemblage of UN_2 and UO_2,
- Exploring alternate route starting from NH_4UF_8 precursor

Synthesis by low temperature with $NaNH_2$ molten salt

- Method used to synthesise transition metal nitrides from oxides by reaction in $NaNH_2$ molten salt at ca. 240°C for 24 h.
- Reaction between uranium oxides and $NaNH_2$ investigated; failed to form nitride phase, but reaction of UO_{2+x} with $NaNH_2$ effects a low temperature reduction to stoichiometric UO_2



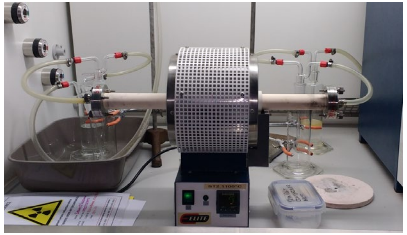


Fig 1: Ammonolysis reaction furnace

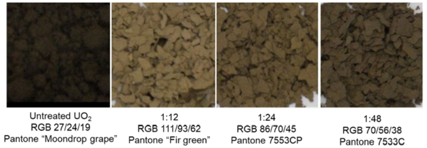
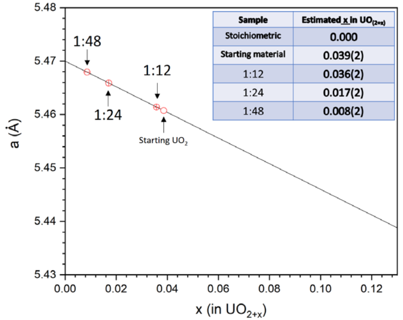


Fig 2: Products of reaction from UO_2 and $NaNH_2$, in 1:n mole ratio at 240°C, 24h. Oxygen determination by internal standard XRD and unit cell parameter calibration



Sample	Estimated x in $UO_{(2+x)}$
Stoichiometric	0.000
Starting material	0.039(2)
1:12	0.036(2)
1:24	0.017(2)
1:48	0.008(2)

Recent publications from ATLANTIC and PACIFIC:

- A. Mason *et al.*, molten salt synthesis of Ce doped zirconolite for the immobilisation of pyroprocessing wastes and separated plutonium, Ceramics International, *in press*.
- S. Sun *et al.*, On the existence of the compound "$Ce_3NbO_{7+\delta}$" prepared under air atmosphere, Journal of Rare Earths, *in press*.

WP4.2 Fabrication and testing of uranium nitride thin films

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University of BRISTOL

Work carried out in AFCP Fuels Programme

- Fabrication of poly epitaxial and single crystal thin films of UN and U_2N_3

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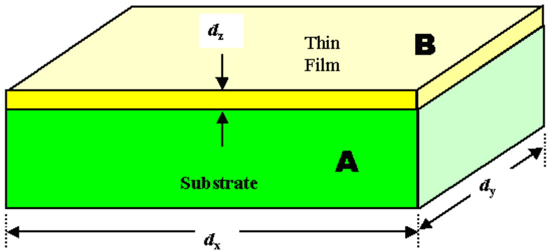
Next stage

- Steam rig experiments – Novoclave at 500 °C, 500 bar
- Dopants co-deposition within the films, e.g. Cr to investigate the effect on corrosion resistance
- Fuel-cladding interaction - Deposition directly onto cladding materials
- Comparison between nitrides, oxides and silicides



Advantages:

- Precisely controlled thickness, microstructure, stoichiometry and impurity concentrations.
- Simple model surface,
- Reduced radioactivity making them significantly easier to handle

Nitride Thin Films (AFCP)

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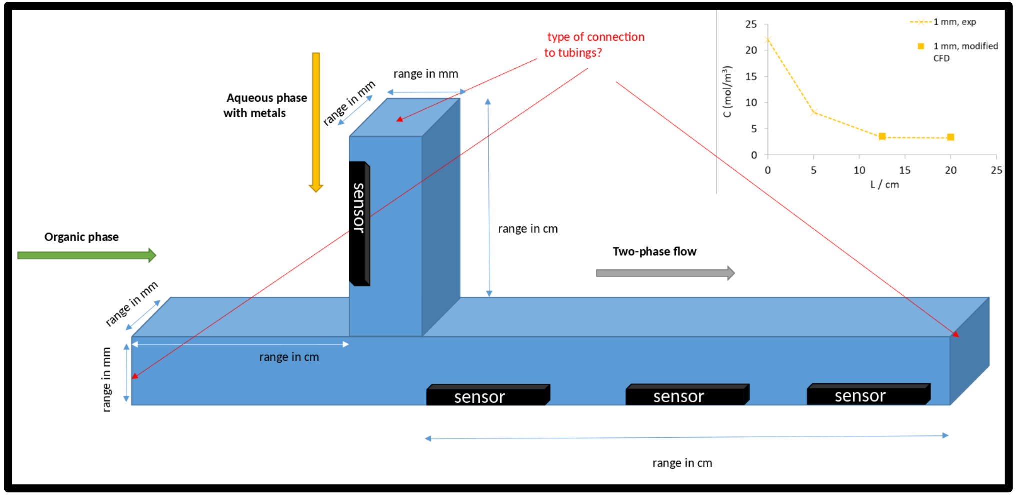
Corrosion Trials (ATLANTIC)

WP5.1 Sensor placement and quantification of uncertainty of readings

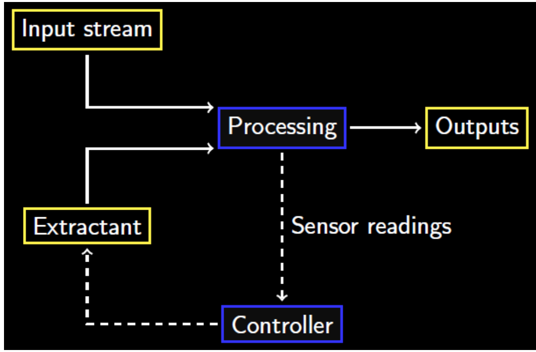
Determine number and placement of sensors in intensified operating units to enable

- Monitoring of performance for quality assurance and safety and
- Real time control of the processing.



Wish to design and operate an intensified processing unit:

- **Decision variables:** size, configuration, extractant, sensors, flow regime, . . .
- **Measured variables:** concentration in aqueous phase, flow regime ?
- **Criteria for optimization:** economic, number of sensors, performance, safety



Model-based control

Wish to implement a model based controller so need a predictive model that we can invert to determine appropriate changes in operation.

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Atlantic – project status

Mid Term Review 25th March

	University		In Post	Started
WP1 Fuel-Separations interface				
WP 1.1 Voloxidation as a pre treatment for accident tolerant fuels	Leeds	PhD		
WP 1.2 The effect of scale up of dissolution kinetics	Leeds	PDRA		
WP 1.3 Corrosion and Dissolution of Accident Tolerant Fuels under Conditions Relevant to Head End	Lancaster	PDRA		
WP 1.4 Molecular Simulation of the Corrosion of Accident Tolerant Fuels: A Modelling Study	Lancaster	PhD		
WP2 Effects of Contaminants on Separations				
WP 2.1 Manufacture of functionalised BTPhen ligands	Reading	PDRA		
WP 2.2 Radiation Stability Testing of Ligands	Manchester	PDRA		
WP 2.3 Development of new tools to analyse speciation	Oxford	PDRA		
WP3 Investigation and Optimisation of Accident Tolerant Fuel Materials				
WP 3.1 Manufacture, Characterisation and Testing of Uranium Nitride Fuels	Sheffield	PDRA		
WP 3.2 Manufacture, Characterisation and Testing of Uranium Silicide Fuels	Manchester	PDRA		
WP 3.3 Radiation Effects in Novel Accident Tolerant Fuels	Liverpool	PDRA		
WP 3.3 Accident Tolerant Nuclear Fuels – Options and Designs (tbc)	Liverpool	PhD		
WP4 Fuel Behaviour: non-stocihiometry and the fuel-water interface				
WP 4.1 High Resolution NMR analysis of Uranium Silicides and Nitrides	Cambridge	PDRA		
WP 4.2 Corrosion Tests of Thin Film Uranium Silicides and Nitrides	Bristol	PDRA		
WP 4.3 Characterisation and Oxidation of Uranium Silicide Phases	Bristol	PhD		
WP 4.4 Atomic scale modelling of UN fuel	Imperial	PDRA		
WP 4.5 Ab initio random structure searching to improve fabrication routes for U3Si2 and UN fuels	Cambridge	PhD		
WP5 Integrated Management of Accident Tolerant Fuels				
WP 5.1 Development of High Efficient Separation Technologies	UCL	PDRA		
WP 5.2 Sensor Development and Optimal Placement	Edinburgh	PDRA		

Conclusions

How has the research landscape, and therefore the role of ATLANTIC within it, changed since funding was awarded?

The proposal was submitted on the 28th March 2018 = AFCP + COVID + Brexit Deal on EU Research

Aim to provide a clear view on the Accident tolerant fuels and claddings (ATFC) technology of choice

- From a fundamental science perspective = proof of concept on manufacture of uranium nitride and silicide fuels;
- From a technological perspective = work so far (fuels) has raised the TRL to 2-3 and AFCP is taking over;
- From a strategic perspective = too early to call, but through AFCP Quarterly Meetings, BEIS are aware of ATLANTIC and its aims

Acknowledgements

EPSRC
Engineering and Physical Sciences Research Council

University of Bristol
University of Cambridge
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Imperial College London
Lancaster University
University of Leeds
University of Liverpool
Manchester 1824 The University of Manchester
University of Oxford
University of Reading
The University Of Sheffield.
UCL

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