A Multi-Faceted Approach to I-129 Abatement Within Off-Gas Capture

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AFCP Advanced Fuel Cycle Programme

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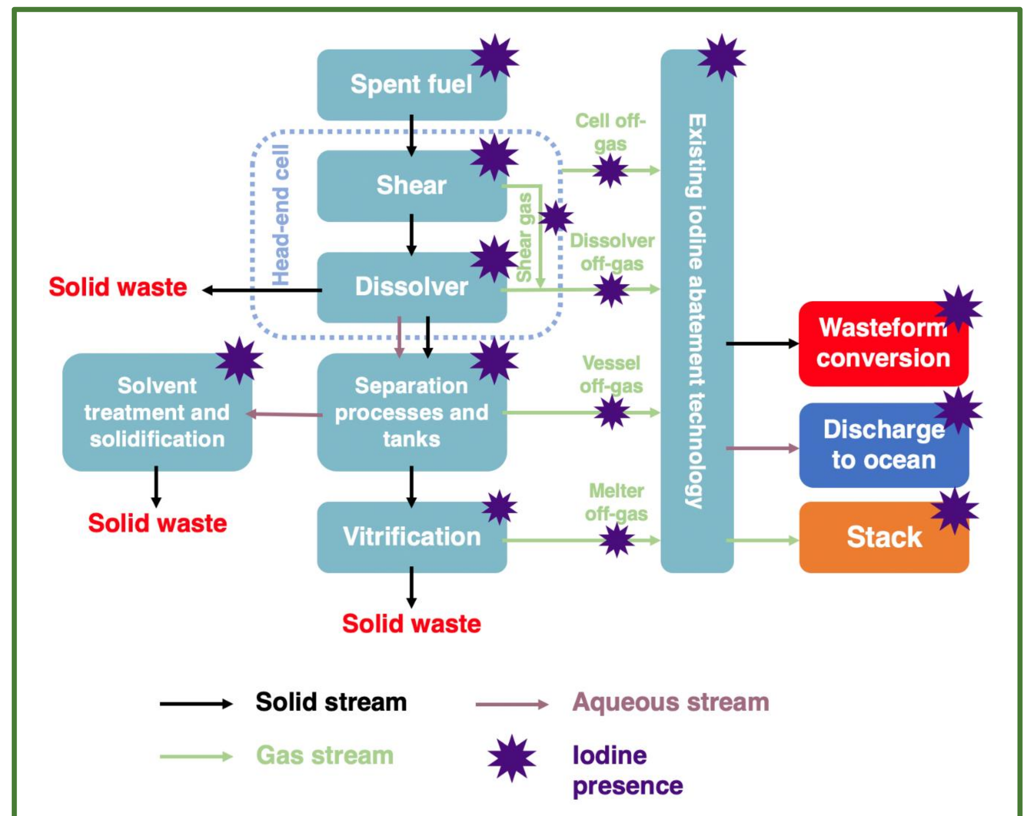
INTRODUCTION

- The UK is investigating off-gas challenges associated with advanced fuel cycles as part of the Advanced Fuel Cycle Programme (AFCP)
- Iodine is the most significant volatile in terms of aerial dose
- Effective abatement crucial in progression to Net Zero by 2050
- To change methodology from **dilute and disperse** to **concentrate and contain**, a credible disposal form is required
- Projects investigate speciation, iodine capture methods and iodine immobilisation

I-129 Fact Sheet

$T_{1/2}$	15.7 million years
Proportion of Thorp aerial dose	~78%
Off-gas speciation	I_2
Thorp abatement method	Caustic scrubbing
Release location	Dissolver Head-end (~98%)



Generalised schematic of radioiodine partitioning in the fuel recycling process [1]

OBJECTIVES

- Reduce iodine discharge from advanced recycling of used nuclear fuel
- Increase iodine retention in wasteforms
- Investigate hot isostatic pressing (HIP) to encapsulate iodine as solid waste
- Progress a next generation of adsorbents for the first effective aqueous iodine capture and synergistic wasteform conversion
- Understand iodine chemistry in dissolver raffinate, to inform removal techniques by sparging
- Construct a test rig for the assessment of solid adsorbents for gaseous iodine

MAJOR PROJECT AREAS

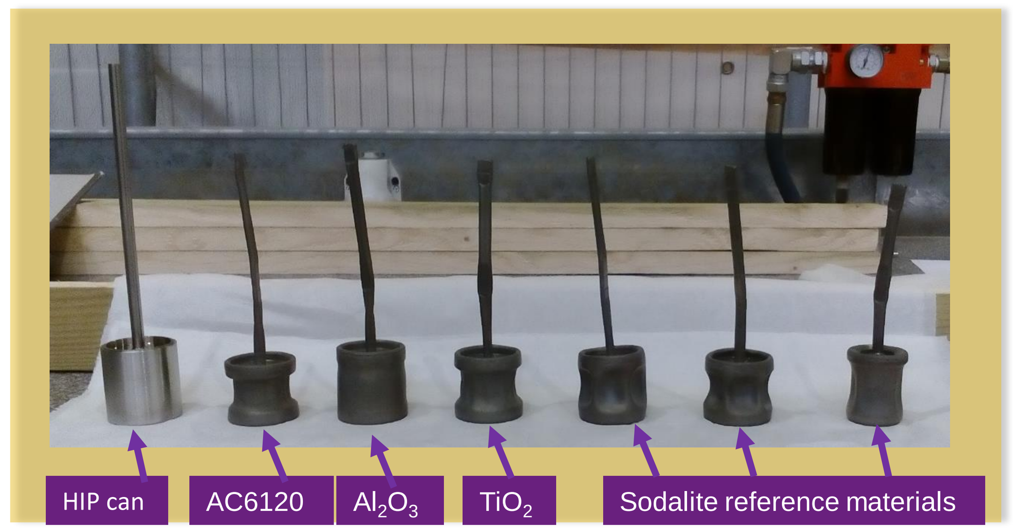
Effective iodine management requires a joined-up approach, with four projects driving four key advances: Adsorption technology | Knowledge of speciation | Stable wasteforms | Control volatility

Using hot isostatic pressing (HIP) to create iodine wasteforms

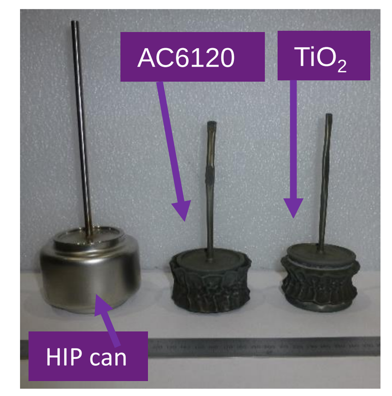
Ewan Maddrell

- **Iodine** in current disposal media has a **high repository dose**
- **Sodalite** has shown to be a good wasteform but is **challenging to manufacture** [2]
- This project investigates using **HIP, to encapsulate iodine**, which is thought to be more easily applied at scale
- **AgI loaded AC6120** (alumina), **Al_2O_3** and **TiO_2** were tested and compared to reference sodalite materials.
- **HIP process retains any volatile iodine** within the container
- Materials have been successfully created and are currently undergoing leach testing

Inactive HIP rig at NNL Workington



Small-scale iodine containing HIP wasteforms



Dumbbell-scale HIP cans

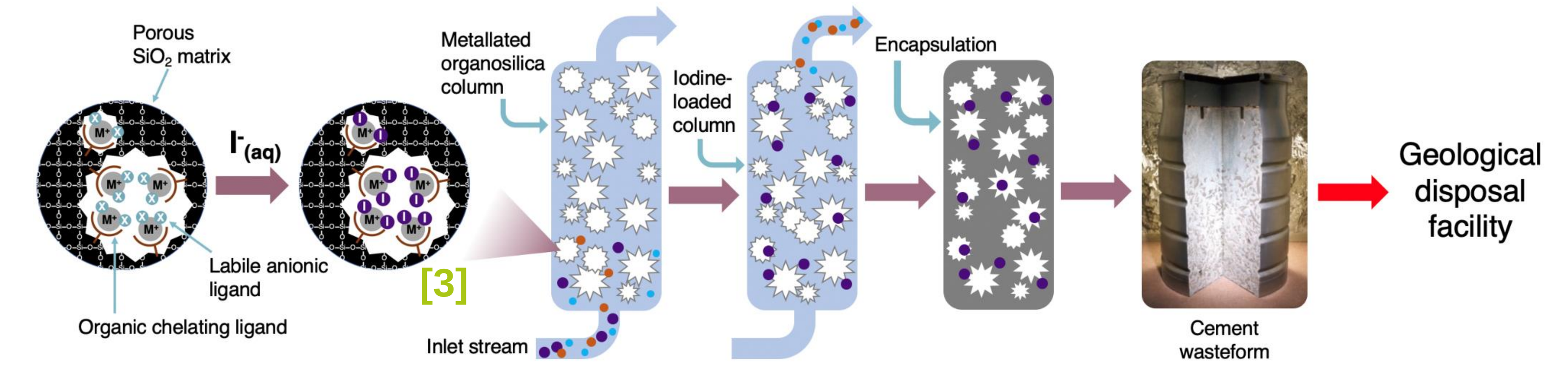
Next Steps

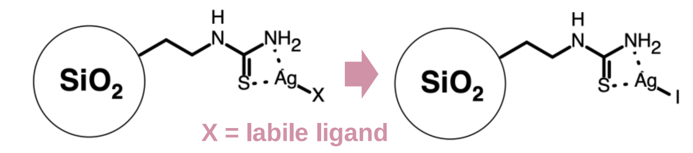
Understand relationship between AgI loading and wasteform performance

Adsorption from the aqueous phase and wasteform conversion

Thomas Robshaw, Sarah Kearney, Brant Walkley, Clint Sharrad and Mark Ogden

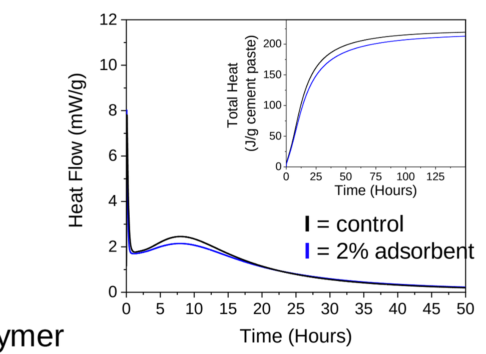
- Dominant I-129 species in off-gas condensates is **iodide anions**, but conventional ion-exchange technology is unfit for purpose and solid waste difficult to treat
- A new generation of adsorbents, suitable for cementation, is proposed
- **Integrated approach**, considering both capture and wasteform-conversion capabilities





- **Thiourea ligand + Ag loading metal** identified as champion system in screening experiments
- Iodide affinity is **4 orders of magnitude higher** than closest competitor system across 17 different aqueous conditions

K-silicate-activated geopolymer formulation with 2% iodide-loaded silica adsorbent



Addition has **negligible effect** on setting

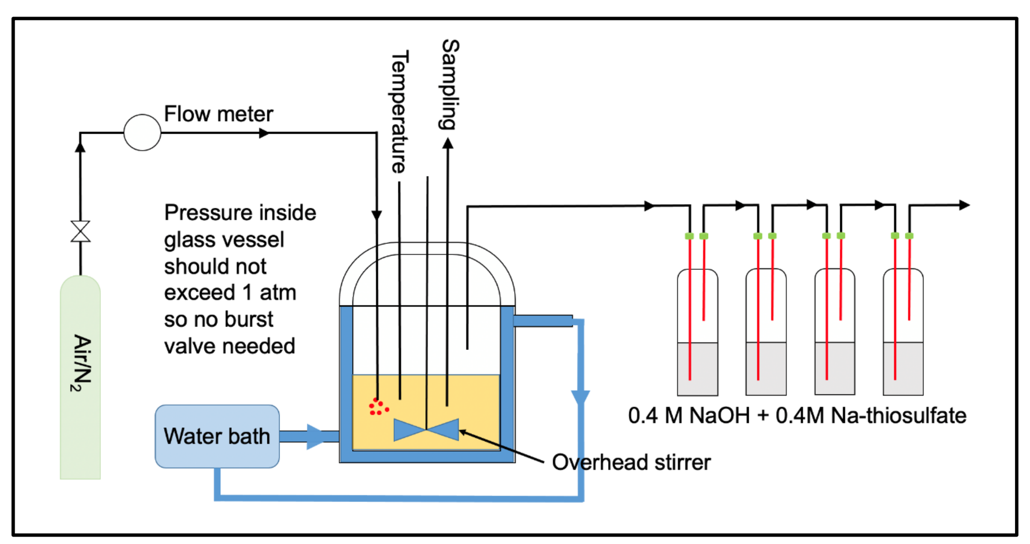
Removal of residual iodine from dissolver solutions

Sarah Pepper and Bruce Hanson

- Dissolution of SNF in nitric acid volatilises >90% of fission-generated iodine
  - Need to remove the rest to prevent it continuing to down-stream processes
- **Chemistry of iodine is complicated** (as is nitric acid and plutonium)
  - In the absence of fission products (FPs), commonly expected species present in solution are 90% iodate (IO_3^-) and 10% iodine ($I_{2(aq)}$)
    - Sparging can remove > 99% of iodine present
  - But in the presence of FPs silver and palladium, **insoluble colloids are formed** and species present are 60% colloidal iodide, 20% $I_{2(aq)}$ and 20% IO_3^- [4]
    - Colloids partially soluble in hot HNO_3 but removal hindered by aging
    - Sparging with up to 10% NO_2 **supresses colloid formation**

$$I^- \rightleftharpoons I_2 \rightleftharpoons HIO \rightleftharpoons IO_3^- \quad (I_2 \uparrow I_{2(g)};\ I^- \downarrow MI_{x(s)}) \quad M = Pd, Ag$$

Experimental setup



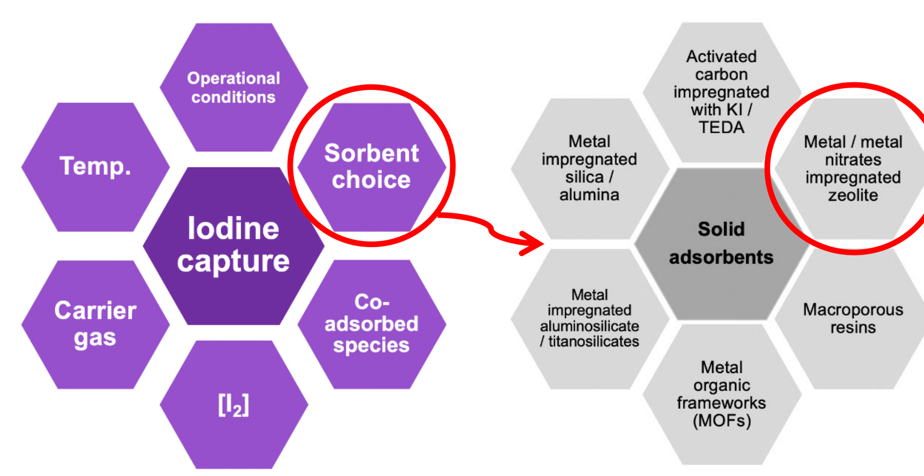
Experimental plan

1. Detection of various species in solution
   - Various iodine species, UO_2^{2+}, V (IV & V), HNO_3, HNO_2, NO and NO_2
2. Speciation of iodine
   - Temperature, [I] and [HNO_3]
   - Presence of FPs Ag and Pd
   - Presence of UO_2^{2+} and V (Pu surrogate)
3. Removal of iodine
   - air or ≤ 10% NOx sparge

Abatement of iodine vapour using silver-based solid sorbents

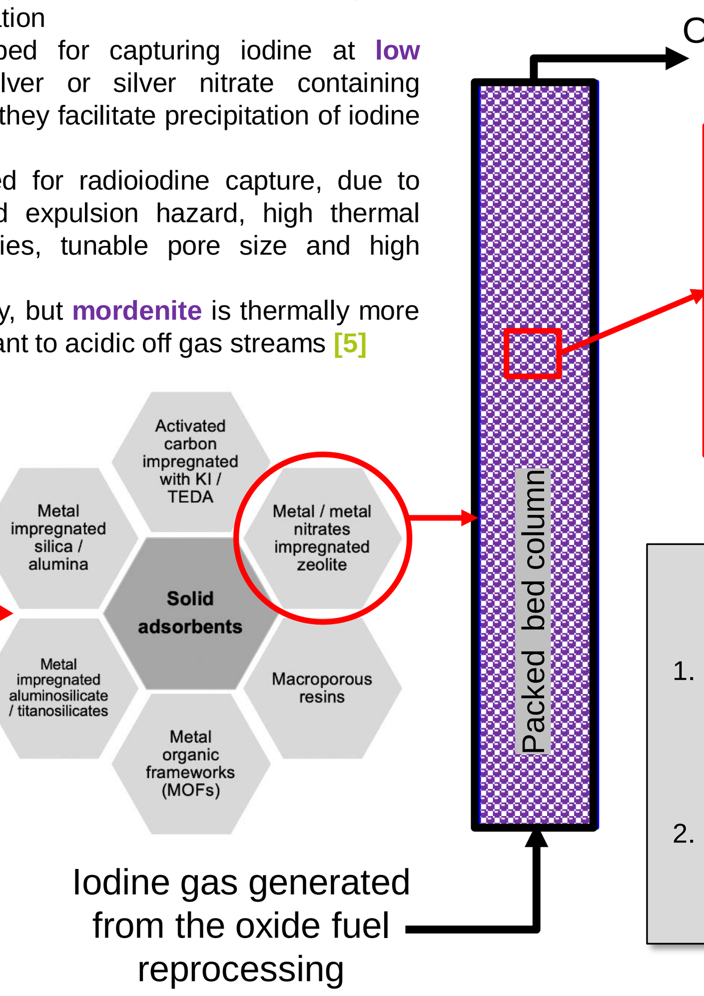
Jabbar Gardy and Bruce Hanson

- **Wet scrubbing** currently the main capture method of radioiodine gases Efficient at high radioiodine concentration
- Solid sorbents have been developed for capturing iodine at **low concentrations** (0.1-0.5 ppm). Silver or silver nitrate containing materials have high performance, as they facilitate precipitation of iodine as **stable AgI** ($K_{sp} = 8 \times 10^{-17}$)
- **Silver-exchanged zeolites** are used for radioiodine capture, due to advantages of low flammability and expulsion hazard, high thermal stability, strong mechanical properties, tunable pore size and high removal efficiency
- **Faujasite** zeolite has greater capacity, but **mordenite** is thermally more stable at high temperature and resistant to acidic off gas streams [5]



Performance-determining parameters

Statistical screening data shows [I_2] and temperature are most important



Ag-loaded zeolite

Aims

1. Design and build a suitable test-rig to accommodate solid media for capture of iodine.
2. Use the rig to assess performance of silver-exchanged zeolites

CONCLUSIONS

- Four iodine projects working together to aim for "near zero" discharge of iodine
- Integrated development of disposal form along with capture method to ensure optimisation across the process
- **Next Steps** → Research into iodine capture methods involving novel materials. Understand interplay between optimal iodine loading in capture material and wasteform performance.

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