NATIONAL NUCLEAR LABORATORY

Electrochemical Enhancement of Nuclear Decontamination Solutions (ELENDES)

NATIONAL NUCLEAR LABORATORY

ELENDES Technology Overview and Demonstration

Liam Abrahamsen

C-Tech INNOVATION

NDA Nuclear Decommissioning Authority

Sellafield Ltd

Innovate UK

Overview

- Decontamination: effluents & organics processing
- The ELENDES technology
- C-Tech Innovation
- Technology scale-up
- Active commissioning and trials
- Deployment concepts
- Conclusions, feedback and next steps

Organisations

- NNL has brought chemistry, nuclearisation (effluents, reprocessing chemistry, engineering, safety teams)
- C-Tech has brought decades of experience in electrochemical processing of industrial effluents (rig design, construction, operation)
- Sellafield Ltd. has provided context and samples for experiments

Summary

- Collaborative project, NNL, C-Tech
- Funded by Innovate UK and NDA
- Contribution in-kind from Sellafield Ltd.
- Objective: to enhance flexibility during decommissioning (wider range of decontamination reagents)
- Developed an electrochemical method for treating problematic effluent waste streams
- Organic complexants, chloride, other misc. organics
- Proved concept non-actively, actively, successfully demonstrated scale-up (lab-scale 1L rig
- pilot-scale 10L rig, including HAZOP-1

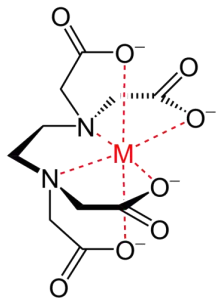
ELENDES Pilot Rig

NATIONAL NUCLEAR LABORATORY

Decontamination

Organic complexants

- Help solubilise radionuclides
- Prevent subsequent re-deposition
- Types include small organic species, e.g. formic, acetic acids
- And larger, polydentate ligands, e.g. EDTA
- These can be effective in decontamination processes



- Organic complexants disrupt incorporation of activity in the ferric flocculation process
- Limits their use in decontamination agents

Decontamination

NATIONAL NUCLEAR LABORATORY

Chloride

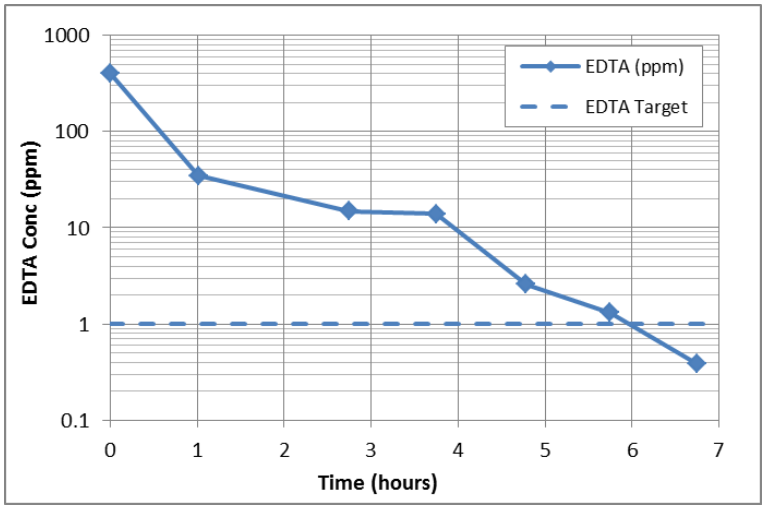
- Effective at enhancing stainless steel decontamination by removing a surface layer
- However, the residual chloride could lead to corrosion of pipework and downstream plant, e.g. effluent plant

There's a barrier to using these species in decontamination

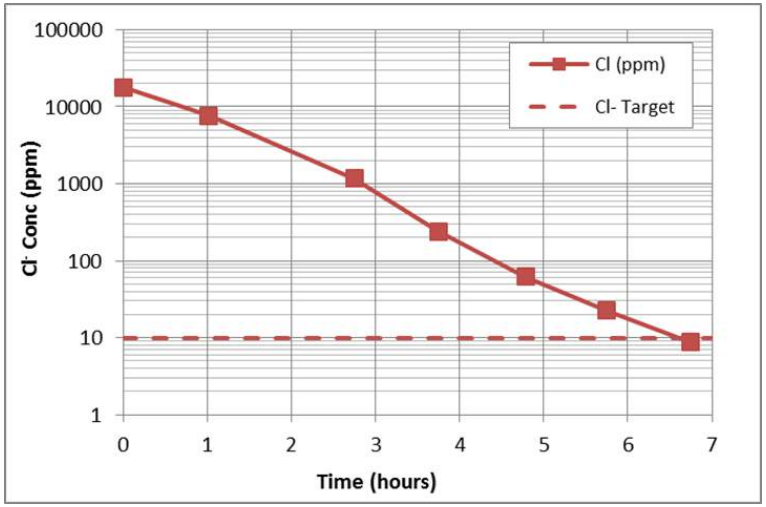
- ➢ Larger volumes effluent generated
- ➢ Ineffective decontamination, not achieving reclassification
- ELENDES designed to treat effluents, remove these species
- Defined treatment targets for complexants and chloride concentration (< 50 ppm TOC, < 10 ppm chloride) Targets for throughput

C-Tech INNOVATION

ELENDES Operation
EDTA and Cl^- Removal



Results of 1L rig active trials
EDTA reduced to < 1 ppm in under 7 hours (1 shift period)



ELENDES process also removes Cl^- to < 10 ppm in same period

Flexibility

NATIONAL NUCLEAR LABORATORY

As well as organic complexants and chloride, process can treat miscellaneous aqueous wastes containing a range of soluble organics (though not bulk organic phases):

- Short and long-chain fatty acids (carboxylics)
  - *formate, acetate, propionate, butyrate, etc.*
- Detergents
- Aromatics
- Polymer wastes

Offers a flexible effluent treatment process

C-Tech INNOVATION

NATIONAL NUCLEAR LABORATORY

C-Tech Innovation

Presented by: Dr Steve Brewer

C-Tech INNOVATION

NDA Nuclear Decommissioning Authority

Sellafield Ltd

Innovate UK

C-Tech Innovation

NATIONAL NUCLEAR LABORATORY

- Origins in ‘ECRC Capenhurst’, a research centre for the electricity industry. Independent since 2001.
- Turnover > £5M.
- 65 people with around 40 degree qualified engineers and scientists.
- Organised in 3 groups: Industrial Products, Research & Consultancy.
- Core technical strengths are:

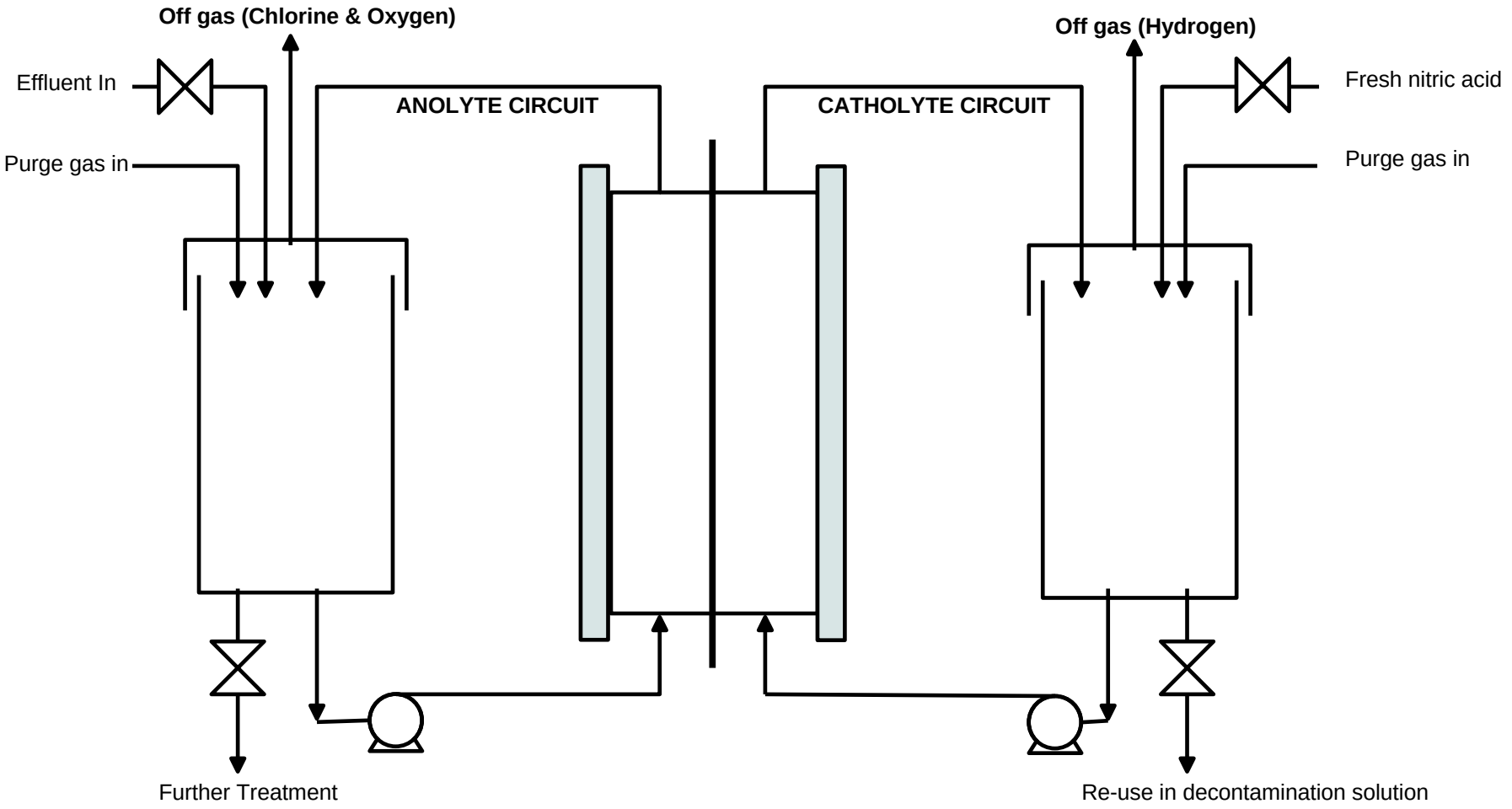
1. Electrochemical and chemical engineering
2. Industrial RF and Microwave Processing
3. Resource Efficiency and recycling technologies

Electrochemistry Fundamentals

NATIONAL NUCLEAR LABORATORY

- Chemical reactions by electron transfer at the electrodes
- Charge balance required
- ANODE – addition of electrons to Anolyte species » OXIDATION of species (e.g. Cl^- and EDTA)
- CATHODE – loss of electrons from Catholyte species
- REDUCTION of species (e.g. H^+ and NO_3^-)
- Applied potential determines the reactions that happen – function of standard reference potentials, overpotentials, mass transport and kinetics

Electrochemistry – ELENDES cell



Inactive Lab Trials - Summary

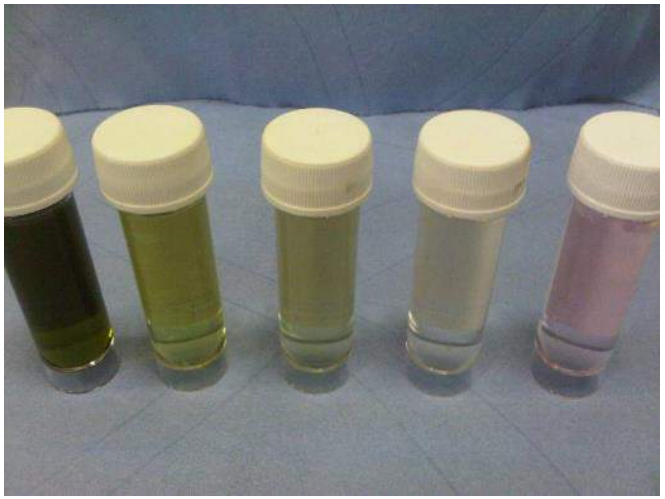
NATIONAL NUCLEAR LABORATORY

Chloride removal

- Target less than 10 ppm in final solution
- Target achieved for all solutions, with sub 1ppm achieved with extended runs

Organics destruction

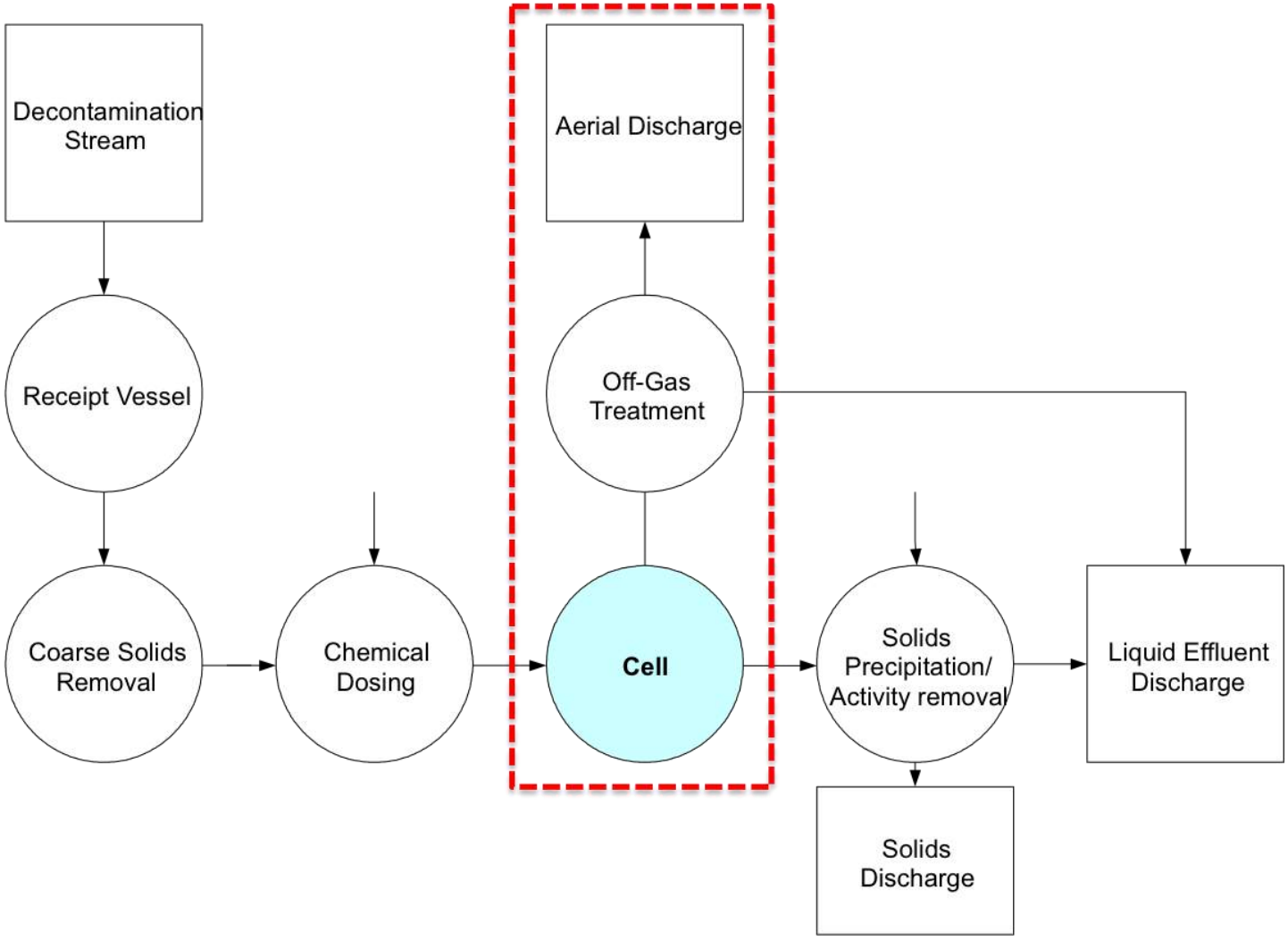
- Analysis is more difficult
- TOC (total organic carbon) standard measure
- Relying on TOC means process is required to mineralise all organics (Target 50mg/L)
- Overriding target is for final solution to have no remaining complexing capability
- New analytical method developed to measure complexant concentration



- EDTA, formic acid destroyed by direct electrochemical oxidation

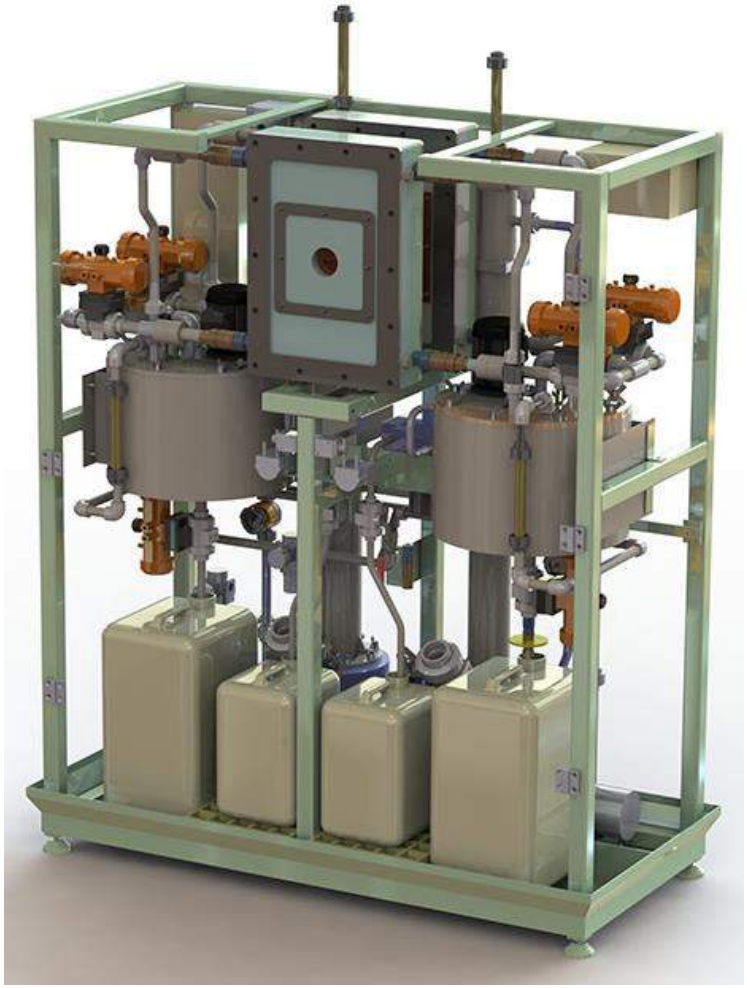
Pilot Scale – 10L Rig Scope

NATIONAL NUCLEAR LABORATORY



10L rig – Design & HAZOP

- Process description, P&ID and D mechanical model prepared
- HAZOP-1 chaired by NNL for operation in a walk in fume hood in the central laboratory
- Mechanical and electrical designs updated to close out all ‘current’ HAZOP actions
- All materials of construction suitable for use with active solutions
- PLC controlled, but all safety systems are hardwired



10L rig – Commissioning & trials

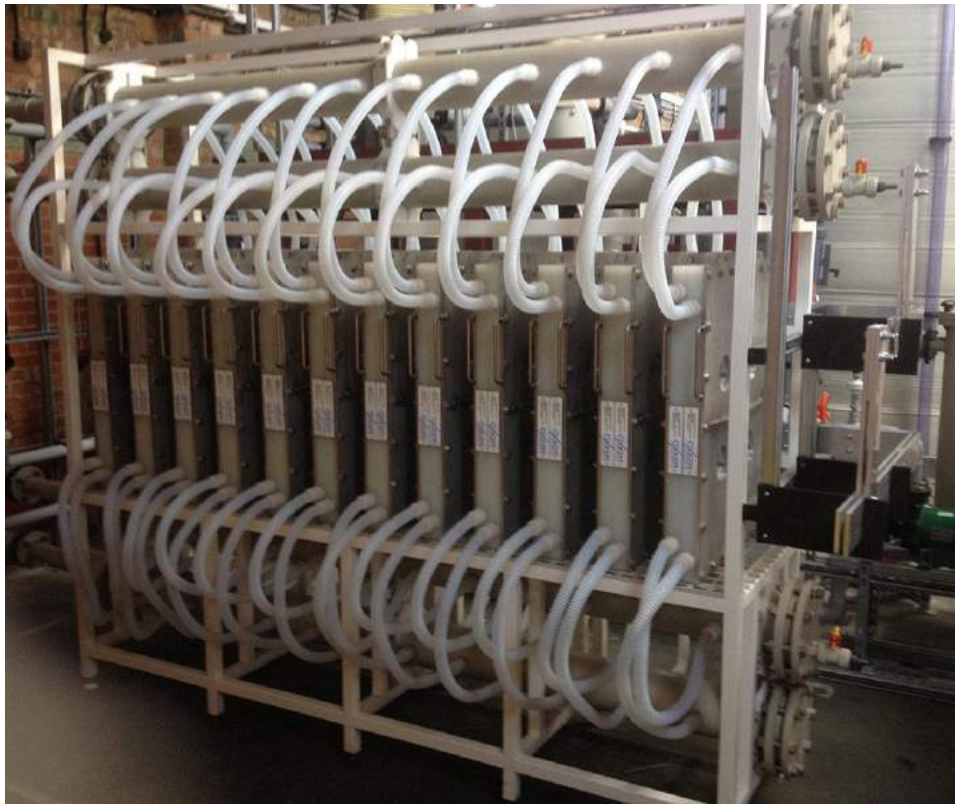
NATIONAL NUCLEAR LABORATORY

- Rig initially commissioned with air and water to test instrumentation pumps, and safety system function
- Initial trials on anolyte containing EDTA and nitric acid to check performance of the electrochemical cells
- Trials with Cl containing solutions confirmed scrubber function
- Batch of model decontamination solution achieved Cl & TOC targets more rapidly than lab scale

Scale-up of pilot design

NATIONAL NUCLEAR LABORATORY

- 10L ELENDES pilot rig has 25*25 cm electrodes and operates at 250A
- Pilot cells have been scaled up to 50*50 cm and are already operational at 1,000A for none nuclear operations
- A 12 cell system has been installed for one of C-Tech's customers for chemical synthesis
- This scale would be suitable for processing $1m^3$ batches of decontamination solution



NATIONAL NUCLEAR LABORATORY

First active demonstration of ELENDES process

Presented by: Dr Chris Maher

C-Tech INNOVATION

NDA Nuclear Decommissioning Authority

Sellafield Ltd

Innovate UK

Aim of active work

NATIONAL NUCLEAR LABORATORY

- Demonstrate ELENDES process in a radioactive fumehood (C3).
- Assess the success of the ELENDES process for the reduction in the complexing ability of solution.
- Demonstrate the value of the use of aggressive decontaminants.

How do we demonstrate the success of the ELENDES process?

NATIONAL NUCLEAR LABORATORY

1. Analysis of chloride by ion-chromatography
2. Analysis of total carbon by combustion method

Is this enough? Does this demonstrate success?

3 .Ferric flocculation of alpha activity.

- general method for assessing complexing ability of the solution
- Ultimately what we're interested in, minimising effect upon EARP

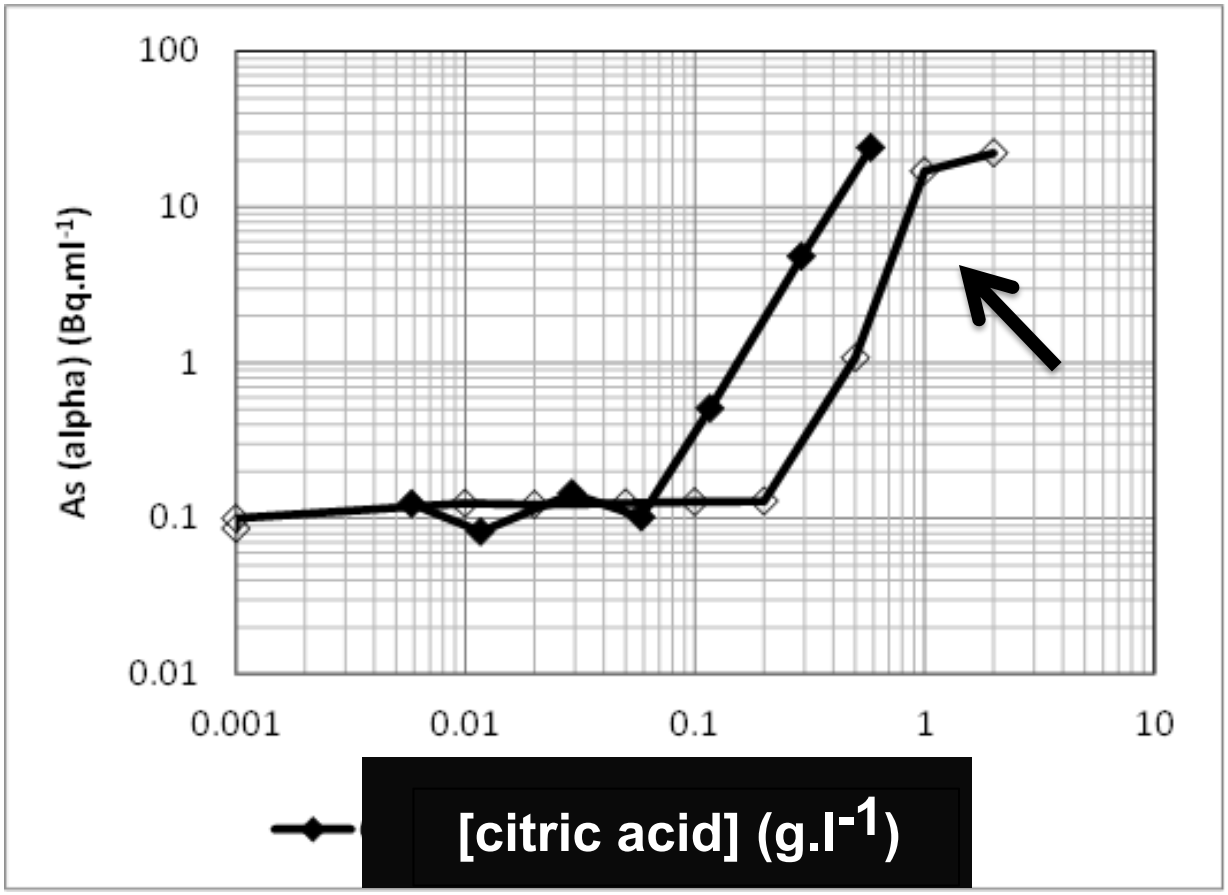
4. Value of using hydrochloric acid and complexants

- Decontamination of plant steel sample

Effect of complexants upon ferric flocculation filtrate activity

NATIONAL NUCLEAR LABORATORY

1980's work demonstrates the SDG3 containing citric acid and EDTA increase filtrate alpha activity.



1 $g.l^{-1}$ citric acid causes high filtrate alpha activity

Testing the ELENDES cell with spiked simulants

NATIONAL NUCLEAR LABORATORY

- **Active experiments completed with spiked**

➢ Plutonium alpha activity

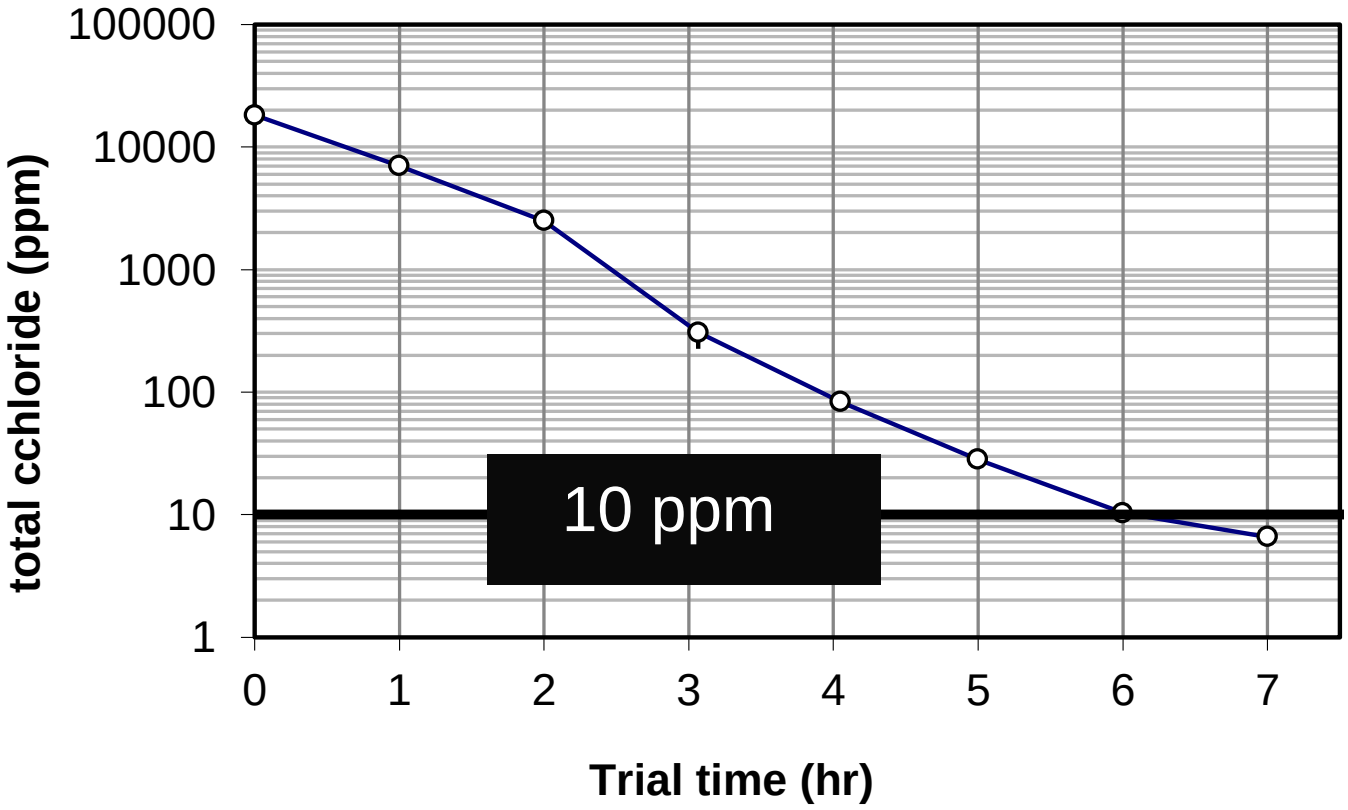
➢ Mixed Sr, Cs, Co, alpha activity

- ***With either:***
- 3 mol.l^{-1} nitric acid, 0.5 mol.l^{-1} hydrochloric acid, 1 g.l^{-1} EDTA
- 3 mol.l^{-1} nitric acid, 0.5 mol.l^{-1} hydrochloric acid, 1 g.l^{-1} SDG3
- 3 mol.l^{-1} nitric acid, 1 g.l^{-1} citric acid

Electrochemical chloride decontamination

NATIONAL NUCLEAR LABORATORY

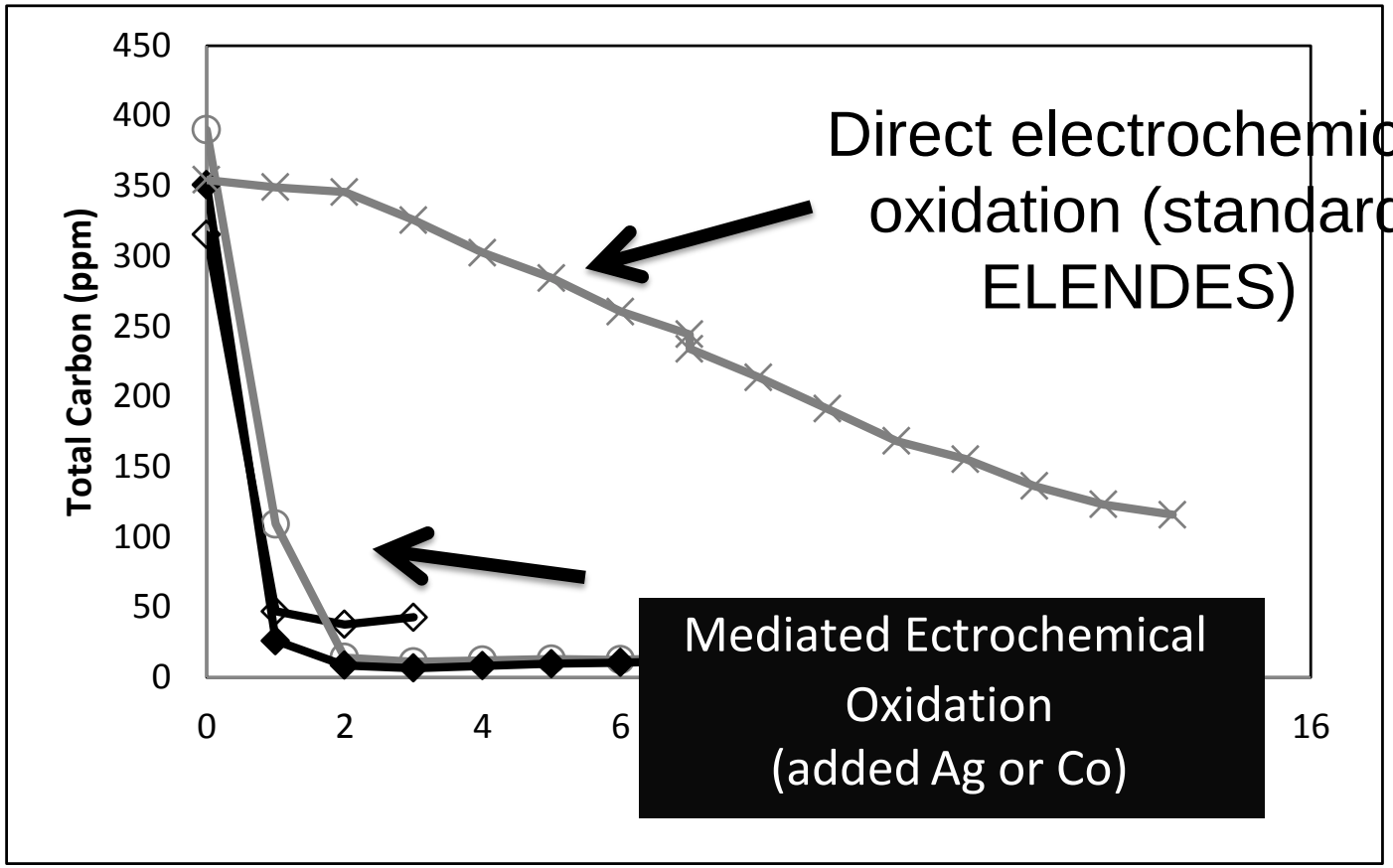
Chloride can be removed efficiently



Electrochemical destruction of citric acid

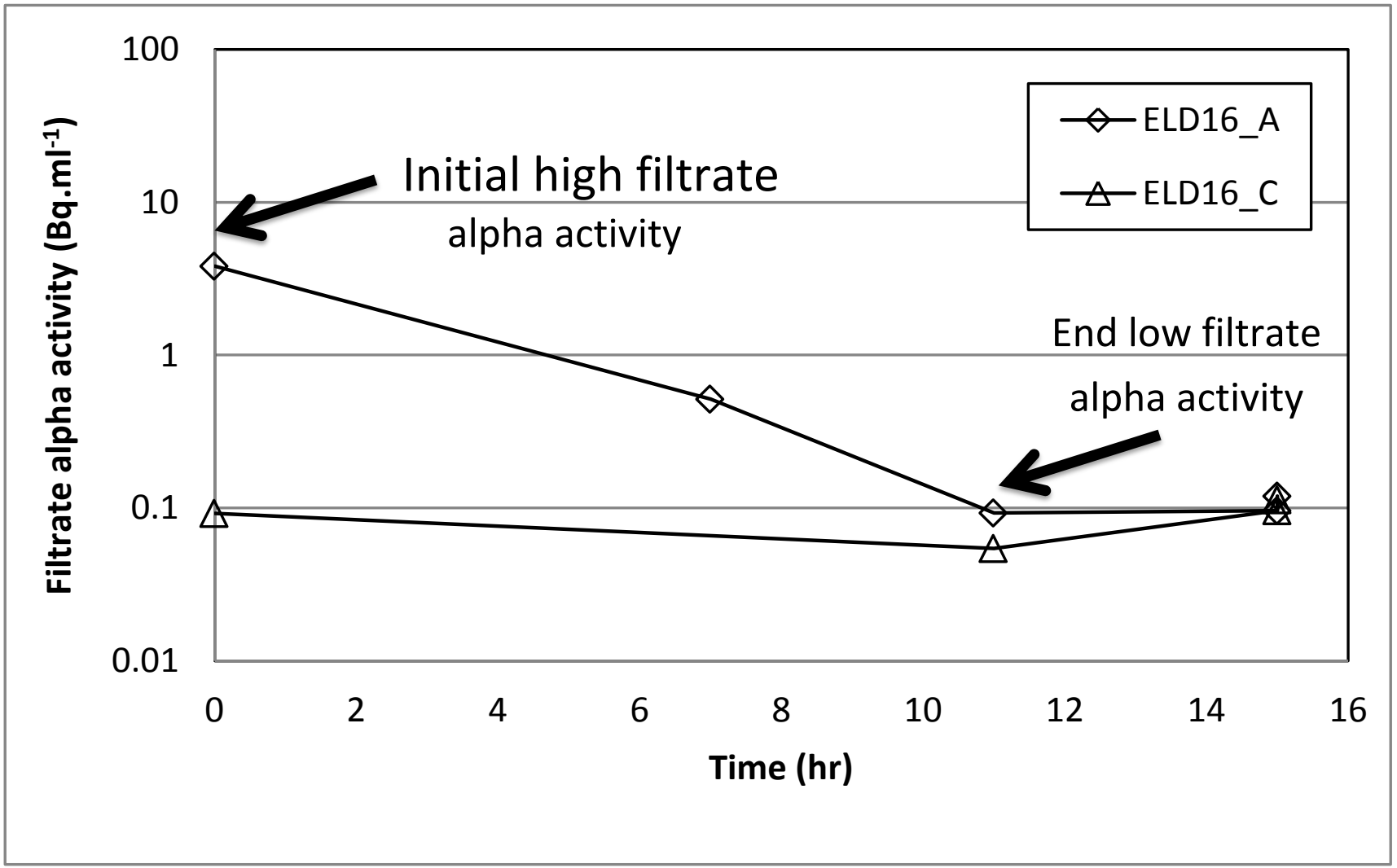
NATIONAL NUCLEAR LABORATORY

Experiments show that citric acid, a 'difficult' small molecule can be destroyed



Citric acid destruction test - ferric flocculation filtrate alpha tests

NATIONAL NUCLEAR LABORATORY



Decontamination of B6 mild steel sample

NATIONAL NUCLEAR LABORATORY

- Steel sample received very low contamination ^{137}Cs (initially 11 $\mu Sv.hr^{-1}$ $\beta\gamma$)

➢ *2 hours leach using 3 mol.l^{-1} nitric acid – 0.5 mol.l^{-1} hydrochloric acid – 1 g.l^{-1} EDTA*

➢ *Decontamination factor of 16 for ^{137}Cs (determined by γ-spec)*

➢Chloride and EDTA efficiently removed

➢Low ferric flocculation filtrate alpha activity achieved.

Other experiments with the ELENDES rig

NATIONAL NUCLEAR LABORATORY

- EU FPVII ASGARD project – advanced head-end option for UC reprocessing.
  Destruction of soluble organics generated during uranium carbide dissolution in nitric acid.

- EU FPVII SACCESS project – destruction of complexants using in advanced solvent extraction processes prior to Actinide finishing
  (ongoing) destruction of citric acid, acetic acid and DTPA.

- NNL signature research – destruction of organics
  (planned) destruction of oil, TBP/OK and TODGA

➢ **Destruction of many other organics published in the scientific literature**

Summary of active demonstration of ELENDES process

NATIONAL NUCLEAR LABORATORY

- Efficient chloride decontamination
- Efficient at destruction of organic complexants
- Destruction of complexants allows decontamination of alpha activity using ferric flocculation
- Successful decontamination of plant steel using hydrochloric acid and complexants
- Used in other experimental programmes demonstrating the adaptability and flexibility of ELENDES rig.

C-Tech INNOVATION

NATIONAL NUCLEAR LABORATORY

Project Overview, Deployment Concepts and Summary

Liam Abrahamsen

C-Tech INNOVATION

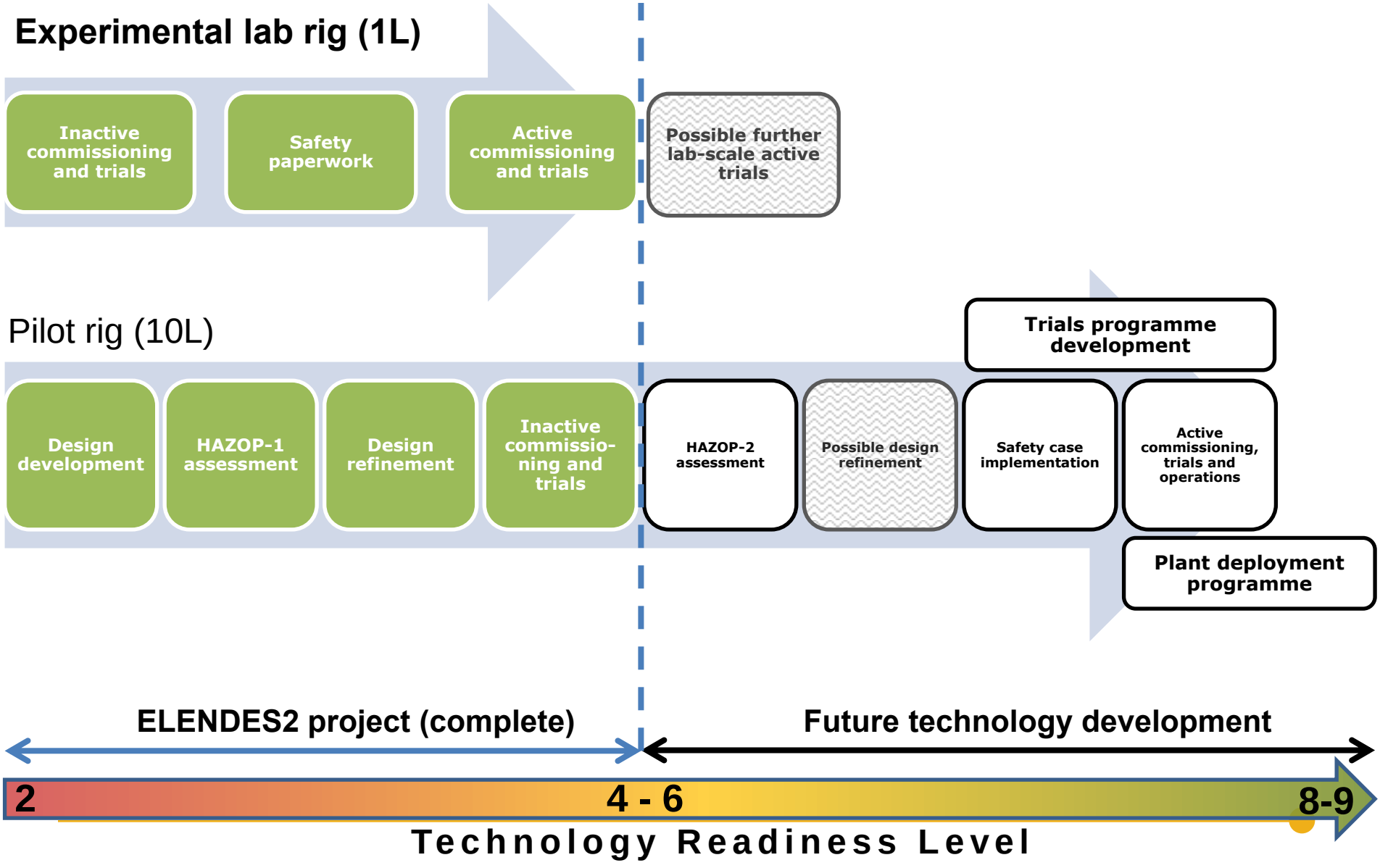
NDA Nuclear Decommissioning Authority

Sellafield Ltd

Innovate UK

Project outline

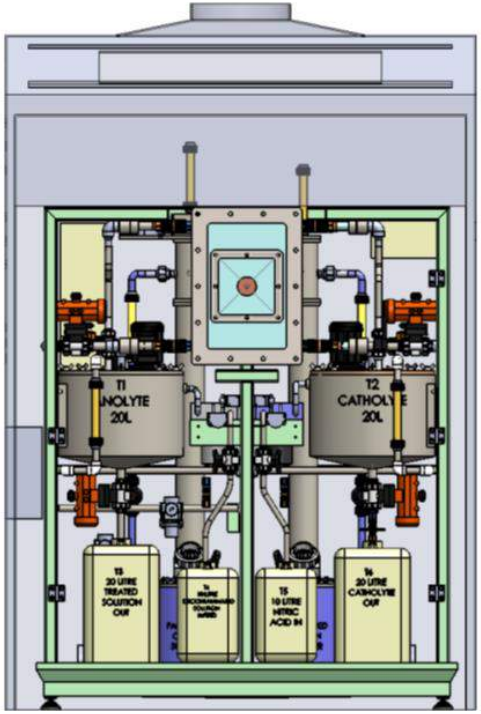
NATIONAL NUCLEAR LABORATORY

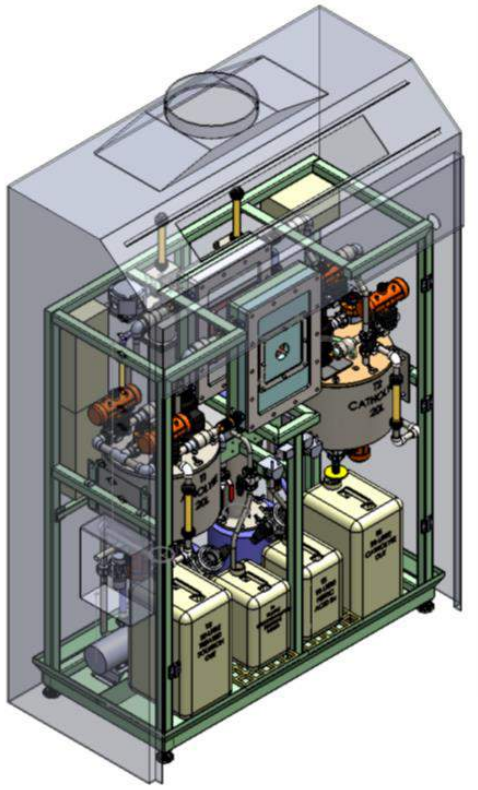


Deployment Concepts

NATIONAL NUCLEAR LABORATORY

- 10L scale static installation, e.g. for lab waste processing
- 10L mobile unit, e.g. for in-cell treatment
- Centralised unit to treat bowsered wastes from cross-site decommissioning / decontamination operations





Summary and discussion

- Saw an industry requirement
- Established an effective partnership
- Making best use of available funding streams
- Treats organics, complexants, chlorides down to levels acceptable for discharge or disposal – allowing the use of these in decontamination
- Proven concept through to scale-up
- Infrastructure and team available for future trials
- Now need to establish way forward to plant deployment