

Institution: The University of Manchester

### **Unit of Assessment:** 7 (Earth Systems and Environmental Sciences)

**Title of case study:** Development of radioactive effluent management strategies at Sellafield delivers economic and environmental benefits

### Period when the underpinning research was undertaken: 2013 - present

### Details of staff conducting the underpinning research from the submitting unit:

Name(s):	Role(s) (e.g. job title):	Period(s) employed by submitting HEI:
Katherine Morris	Professor	2010 – present
Sam Shaw	Professor	2013 – present
Jon Lloyd	Professor	2001 – present
Kurt Smith	PDRA	2016 – 2018
Lynn Foster	PDRA	2018 – 2020
Tom Neill	PDRA	2018 – 2019
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Period when the claimed impact occurred: 1 September 2015 – 31 July 2020

### Is this case study continued from a case study submitted in 2014? ${\sf N}$

### 1. Summary of the impact

The Sellafield site, in Cumbria (UK), is one of the most complex and hazardous nuclear sites in the world. Our research in the Effluent Centre of Expertise has led to the following impacts:

- (i) optimised treatment protocols for radioactive reprocessing effluents, allowing Sellafield to achieve a 50 – 90% reduction in actinide discharges during targeted periods of plant operations and significantly reducing alpha radioactivity discharges to the Irish Sea;
- (ii) modified sludge management practices within the spent nuclear fuel ponds has reduced radioactivity within the effluent treatment system by between 69% and 95%, with estimated operational savings of at least GBP22,500,000;
- (iii) informed biomass control strategies, enabling a **40% increase in fuel retrieval** operations from the Pile Fuel Storage Pond (in 2019 compared to 2018), with savings of at least GBP2,400,000.

### 2. Underpinning research

Research in the Department of Earth and Environmental Science at the University of Manchester (UoM) has focussed on effluent behaviour at the Sellafield nuclear site, one of the most complex and hazardous nuclear facilities in the world. This research has been undertaken by UoM academics (Morris/Shaw/Lloyd), PDRAs (Smith/Foster/Neill) and PhDs (Weatherill 2013-16; Konovalovaitė 2013–17; Lopez 2015-18; Winstanley 2016-19; Neill 2015-18; Foster 2015-18), alongside industrial partners working within the Sellafield Effluent Centre of Expertise (ECoE). The ECoE is an ongoing research collaboration between the University of Manchester, Sellafield Ltd. and the National Nuclear Laboratory (NNL) since 2012.

### **Research problem**

At Sellafield, radioactive effluents arise both from fuel reprocessing, and the management of legacy ponds and silos, which are a range of older facilities containing highly radioactive "sludge". The UK Government's Office for Nuclear Regulation (ONR) have instructed the Nuclear Decommissioning Authority, the organisation responsible for operating Sellafield, to prioritise reducing these risks. The optimal management and treatment of the ponds, silos, and reprocessing effluents is crucial to both reduce the hazard, and protect the environment. The legacy ponds and silos are the highest hazards at Sellafield: their safe decommissioning is a programme of the utmost national and international importance.



### Research programme

Since 2013, to address these challenges, a UoM programme of research has covered three areas:

- (i) Iron oxide flocculation and radionuclide removal during reprocessing effluent treatment [1,2]: The Sellafield Enhanced Actinide Removal Plant (EARP) treats acidic, iron containing radioactive effluents from nuclear fuel reprocessing. As Sellafield moves from reprocessing activities to decommissioning (2019 onwards), EARP will need to treat various effluents, including new clean-out effluents with different chemical compositions than previously treated effluents. The EARP process neutralises the acidic, radioactive effluents to form an iron floc that scavenges the radionuclides from solution. The treated aqueous effluent is then safely discharged to the environment under authorisation. UoM research into the EARP floc formation pathway identified that previously undiscovered Fe<sub>13</sub> Keggin moieties were present under very acidic conditions (pH <0.15) [1]. Counter to classic nucleation and growth models, as the pH was increased, the Keggin clusters aggregated to form the iron floc particles [1]. Further research provided detailed insight into the mechanisms of uranium, plutonium and thorium adsorption/removal on the ferrihydrite floc [e.g. 2].
- (ii) Colloid stability and radionuclide behaviour in spent fuel pond effluents [3,4]: The effluent from the Sellafield Legacy Ponds and Silos (SLPS) nuclear fuel ponds is radioactive and treated at Sellafield in the Site Ion Exchange Plant (SIXEP) before authorised discharge to the environment. Effective effluent treatment reduces the high hazard of these facilities by supporting essential operations to empty and decommission the ponds and further leads to reduced radionuclide discharges to the environment. UoM research [3, 4] provided fundamental understanding of uranium colloid stability and speciation in conditions directly relevant to the Sellafield nuclear fuel ponds.
- (iii) Biomass characterisation and control strategies in spent nuclear fuel ponds [5,6]: In order to decommission the SLPS facility, nuclear materials must be removed from the legacy ponds and treated in additional facilities. Within the ponds, microbial biomass growth can reduce pond visibility, which in turn affects the transfer of nuclear material through the facility. For the first time, the UoM research [5, 6] provided detailed information on the microbial ecology of these ponds before, during and after bloom periods, and identified that photosynthetic microorganisms were responsible for the loss in visibility during blooms. Identifying the causative organisms of these blooms is vital to underpin biomass control strategies at Sellafield.

### 3. References to the research

This body of research has been funded through industry (Sellafield Ltd), UoM, EPSRC, BBSRC and STFC, receiving cumulative funding of greater than GBP2,100,000 (EP/R511626/1, ST/N002474/1, EP/G037426/1). UoM researchers are highlighted in bold.

- [1] Weatherill, J, Morris, K, Bots, P, Stawski, TM, Janssen, A, Abrahamsen, L, Blackham, R & Shaw, S (2016) 'Ferrihydrite formation: the role of Fe13 Keggin clusters' *Environmental Science and Technology*. 50 (17) DOI: 10.1021/acs.est.6b02481
- [2] Smith, KF, Morris, K, Law, GTW, Winstanley, EH, Livens, FR, Weatherill, JS, Abrahamsen-Mills, LG, Bryan, ND, Mosselmans, JFW, Cibin, G, Parry, S, Blackham, R, Law, KA, Shaw, S (2019) Plutonium(IV) Sorption during Ferrihydrite Nanoparticle Formation. ACS Earth and Space Chemistry, 3 (11), DOI:10.1021/acsearthspacechem.9b00105.
- [3] Neill, TS, Morris, K, Pearce, CI, Sherriff, NK, Burke, MG, Chater, P, Janssen, A, Natrajan, LS, Shaw, S (2018) 'Stability, composition and core-shell particle structure of uranium(IV)-silicate colloids' *Environmental Science & Technology*. 52 (16) <u>DOI: 10.1021/acs.est.8b01756</u>.



- [4] Neill, TS, Morris, K, Pearce, CI, Abrahamsen-Mills, L, Kovarik, L, Kellet, S, Rigby, B, Vitova, T, Schacherl, B, Shaw, S. (2018), 'Silicate stabilisation of colloidal UO<sub>2</sub> produced by uranium metal corrosion. Journal of Nuclear Materials. 526, DOI: 10.1016/j.jnucmat.2019.151751.
- [5] MeGraw, VE, Brown, AR, Boothman, C, Goodacre, R, Morris, K, Sigee, D, Anderson, L, Lloyd, JR (2018), 'A Novel Adaptation Mechanism Underpinning Algal Colonization of a Nuclear Fuel Storage Pond' mBio, 9 (3). DOI: 10.1128/mBio.02395-17.
- [6] Foster L, Boothman C, Ruiz-Lopez S, Boshoff, G., Jenkinson, P., Sigee, D., Pittman, J.K., Morris, K. and Lloyd, J.R (2020). 'Microbial bloom formation in a high pH spent nuclear fuel pond.' The Science of the Total Environment, 720:137515. DOI: 10.1016/j.scitotenv.2020.137515

### 4. Details of the impact

### Context

The Sellafield nuclear site in Cumbria (UK) has been operating since the 1940s. It is at the heart of the UK's nuclear fuel cycle, and activities at the site are now moving towards the safe packaging and storage of higher-activity radioactive wastes and the clean-up and decommissioning of the site. The Sellafield complex accounts for approximately 76% of the UK's decommissioning legacy, and as of 2019, the estimated cost for decommissioning the site was at least GBP94,000,000,000 with a timeframe of over 120 years [A].

During ongoing site operations and decommissioning, two types of radioactive effluents are generated. Firstly, acidic radioactive effluents from reprocessing spent nuclear fuel, and the subsequent clean-up of legacy reprocessing facilities are treated in the Enhanced Actinide Removal Plant (EARP). Secondly, neutral to alkaline radioactive effluents from the Sellafield Legacy Ponds and Silos (SLPS) are treated in the Site Ion Exchange Plant (SIXEP). The SLPS are the highest nuclear risks and hazards at Sellafield and removing the radioactive material to reduce those risks therefore carries a level of urgency [B].

### Pathways to impact

Formed in 2012, the Effluent Centre of Expertise (ECoE) is a collaboration between UoM, Sellafield Ltd. and the NNL. The aim of ECoE is to "provide fundamental understanding of the underlying processes impacting on effluent management.... which in turn provides direct cost savings and risk reduction and therefore increases stakeholder confidence in operational activities" [C]. Regular (bi-annual) technical meetings are held between the ECoE partners to discuss key results as they are identified, with subsequent publication of the research in papers and PhD theses. State-of-the art research methodologies (Transmission Electron Microscopy, preparation methods and microbial ecology characterisation) have been transferred directly into industry via the ECoE [D][E].

These results have "already been applied to optimise operations in EARP and ponds and silos" [C]. Whilst UoM has several long-standing research collaborations with stakeholders at Sellafield, the impacts described below stem from the fundamental research conducted in the Department of Earth and Environmental Sciences [1-6], and partly build upon research conducted in the Department of Chemistry at UoM which is discussed in an impact case submitted to UOA 8.

### Reach and significance of the impact

In summary, research within the ECoE [1-6] enabled Sellafield to significantly reduce radioactive discharges to the environment from the site [D][F]. Improved understanding of the chemical and microbial processes within these effluent treatment facilities has allowed Sellafield to optimise plant performance thus enabling high hazard reduction [D, F], and to predict the performance of EARP and SIXEP with greater confidence. Sellafield have estimated savings of over GBP24,900,000 as a result of the operational changes underpinned by the ECoE research [D][E][F]. Key highlights are given here:



# (i) Iron Oxide formation and radionuclide removal in EARP: reduction in environmental discharge

UoM research on iron oxide floc formation pathways [1] was discussed with NNL and Sellafield in 2013/14 (prior to publication). This discussion highlighted that effluent streams in EARP with higher initial pH values, may be less effective at removing radionuclides from effluents than previously thought. As a result, Sellafield changed the acid dosing of EARP effluents, specifically increasing the level of acidity in the effluent prior to neutralisation in order to enhance Fe<sub>13</sub>Keggin formation and radionuclide sorption [F]. This change significantly reduced the "alpha radioactivity environmental discharges from this effluent treatment facility to the Irish Sea" [F]. Specifically, in high challenge liquor batches there has been a "reduction in alpha activity discharge to the sea [by] 90%" [F]. In the case of <sup>241</sup>Am, this reduction due to acid dosing changes also has regulatory significance as it "forms part of Sellafield's demonstration of applying BAT (Best Available Techniques) within the Environmental Permit which forms part of [Sellafield's] legal consent to operate." [F].

Additional research on radionuclide removal within EARP [2] provided detailed understanding of the retention mechanisms of key radionuclides on plant. These results directly informed predictive models for radionuclide behaviour, including plutonium, in the EARP system "that will be used to plan future operations and to ensure radioactivity is abated" [F]. Sellafield confirmed this research has "significantly improved effluent treatment processes in EARP... assisting the decommissioning process, overall leading to reduction in discharge and assisting in the clean-up of Sellafield site. These improvements support the optimisation of site decommissioning, which is a multi-billion pound project...These improvements would not have been possible without the research" [F].

## (ii) Uranium colloid stability: improved management of spent nuclear fuel pond decommissioning

During decommissioning of the SLPS, removing highly radioactive sludge reduces the hazard in the ponds, a top priority decommissioning need. During pond retrievals, "settling" must occur to separate liquids from the highly radioactive solids. The associated radioactive liquors are then treated within SIXEP. Previously, these liquors were collected in an effluent collection vessel (ECV) where the radioactivity in the liquors was increasing with each retrieval. The uranium colloid stability research [3, 4] was discussed within the ECoE during 2015/16 (prior to publication). Underpinned by this new understanding of dynamic colloid behaviour [3, 4], in 2017, Sellafield adopted new protocols for plant operations and pond effluents management, and have confirmed *"without the University of Manchester research this new fundamental understanding would not have been developed."* [D].

The new protocols include new ECV mixing regimes implemented to reduce the colloid concentrations in the ECV *"which in turn reduced both alpha activity and turbidity in the system by over 95%* [and] *total beta activity concentrations by 69%"* [D]. This has also reduced the processing time on plant, resulting in estimated cost savings of GBP500,000 over the lifetime of the retrievals programme [D]. Reducing colloid concentrations in the ECV has also reduced the level of monitoring and surveillance required on site, leading to estimated cost savings of GBP2,000,000 over the lifetime of the retrievals programme [D]. Likewise, increasing the rate of waste retrievals has reduced batch processing times, resulting in further estimated cost savings of GBP10,000,000 over SIXEP's operational lifetime [D].

The results in [3, 4] also directly informed decisions to reconfigure the effluent discharge route. This has sustained in-pond visibility at a level that enables retrievals from the SLPS to continue, resulting in a further operational cost saving estimated at GBP10,000,000 over the lifetime of the retrievals programme [D]. Overall, the research has enabled *"high hazard reduction; a reduction in the effluent activity challenge; the avoidance of delays to retrievals that would deliver potential cost savings in the order £10M+ [more than GBP10,000,000]; simplification of effluent treatment; [and] improvements to the characterisation of high activity* 



*samples*" and is integrated into the Sellafield "Alpha Guidelines Document", the primary information source on the behaviour of alpha emitting radioactivity in the SLPS [D].

## (iii) Biomass characterisation: control of microbial bloom events and enabling movement through the Fuel Handling Pond

As discussed already, safely removing highly radioactive sludge from the SLPS is essential to reducing the hazard of these facilities. However, microbial blooms in the SLPS can reduce pond visibility, which in turn can severely delay sludge retrievals [E]. In 2018, UoM research utilising the DNA analysis pipeline demonstrated in [5,6] enabled the predominant photosynthetic species that cause these microbial blooms in the Pile Fuel Storage Pond (PFSP), to be identified [E]. As a result, an optimal, targeted frequency of the ultrasonic biomass control units in the PFSP could be selected to control the species in the pond enabling Sellafield to implement effective bloom control strategies [E].

Sellafield amended these ultrasonic settings in April 2019. After this and for the remainder of 2019, there were 40% more days with sufficient visibility to enable pond retrievals to occur when compared to 2018 [E]. Furthermore, retrievals were possible everyday between 1 January 2020 to 15 March 2020 (74 days), compared to only 26 days in the same period in 2019 [E]. Each day that in-pond retrieval operations are impacted by an algal bloom is *"estimated to cost the programme in excess of GBP50,000"* [E], already representing a cumulative saving of GBP2,400,000. Given the retrievals programme is likely to last 10 years+ [B], the benefit of targeted algal bloom control will run into millions of pounds [E]. Additionally, enabling the PFSP decommissioning programme to run on time, increases confidence in the programme of key stakeholders including the site operators (NDA), regulators (ONR and Environment Agency) and the general public [E].

DNA sequencing using the DNA profiling pipeline demonstrated in [5, 6] has also been used to characterise several hydraulically connected ponds including the First Generation Magnox Storage Pond (FGMSP) and the Fuel Handling Pond (FHP). Controlled movement of material between these ponds is necessary to repackage degraded spent fuel materials and thus reduce hazard. These methods and data showed that each pond has a distinct microbial community adapted to live in that facility, despite being hydraulically connected. Sellafield therefore concluded *"that there were minimal risks of cross contamination [of biomass] causing visibility problems*" [E]. This negated previous concerns that movement between ponds potentially seeds bloom causing microorganisms, and *"helped [Sellafield] with the decision-making process to allow the transfer of fuel between the two facilities and in doing so has allowed the FGMSP high hazard and risk reduction programme to progress"* [E]. Permitting the transfer of fuel from the FGMSP to FHP has led to major cost savings (estimated at greater than GBP10,000,000) [E].

### 5. Sources to corroborate the impact

- [A] Gov.uk online report "Corporate report Nuclear Provision: the cost of cleaning up Britain's historic nuclear sites" Updated 4 July 2019, accessed 15 October 2020
- [B] Sellafield Ltd, 2017/18 "Cleaning Up Sellafield Annual Review 2017/18"
- [C] Sellafield Ltd. 2016/17 Technology Development and Delivery Summary Report
- [D] Letter of Support from SIXEP Plant (Senior Research Manager, Effluents) Sellafield (21 July 2020)
- [E] Letter of Support from Effluent Technical Manager, Retrievals Strategy & Technical Biomass characterisation and control, Sellafield (9 July 2020)
- [F] Letter of Support from EARP Processing Plant, Sellafield (7 January 2020)