Institution: University of Bristol

Unit of Assessment: 7 – Earth Systems and Environmental Sciences

Title of case study: Nuclear Waste: Research into corrosion and reactivity of uranium leads to the reduction of operational safety risk and changes in storage protocols

1. Summary of the impact

In the early years of the nuclear industry, numerous different fuels, materials and reactor concepts were tried and tested, building a legacy of varied wastes that have subsequently proven difficult to treat, store and dispose of. This is especially the case for Sellafield in Cumbria, a site home to an extraordinary accumulation of hazardous waste from the UK and abroad requiring treatment. Much of this waste is stored in outdated nuclear facilities. Bristol's research into the corrosion and reactivity of uranium and associated compounds within different storage, treatment and environmental systems, has resulted in the reduction of operational safety risk at Sellafield, CERN and the Atomic Weapons Establishment (AWE) through the alteration of protocols for the storage, retrieval and treatment of uranium and uranium carbide.

2. Underpinning research

Dr Tom Scott, Professor Geoff Allen and their interdisciplinary research unit, the Interface Analysis Centre (IAC; founded to aid research within the School of Earth Sciences), have been working closely with the nuclear industry to predict and assess the risks for storage of nuclear fuel waste – specifically uranium and uranium carbide. After completing a Geochemistry PhD in the School of Earth Sciences in 2005, Scott joined the School as a Research Associate in 2005, and also became an IAC Director in 2009, taking over from Allen, who had founded and directed the IAC from 1989 onwards (and has since stayed as Professor of Materials Science, and academic coordinator for the Bristol-AWE strategic partnership). Scott's research focuses on different aspects of uranium from metallurgy and alloying through to corrosion and environmental geochemistry **[1-6]**. A primary aim has been to develop a fundamental understanding of the mechanisms that control corrosion of uranium metal in different storage environments as the resulting corrosion product, uranium hydride (UH_3), is pyrophoric (flammable), liable to cause thermal excursions, and presents serious safety hazards during the long-term, or even medium-term, storage of metallic waste in almost all storage environments **[a]**.

Scott's research has been focused on identifying and understanding many of the significant factors controlling the onset (initiation) of hydride formation. This understanding has enabled a better prediction of when and if hydrogen corrosion will occur, and thus the implementation of preventative measures that will delay, or even prevent, such an occurrence. Scott's approach to research has been to combine sophisticated surface analysis techniques with specialist isotope-spiked corrosion cells in which the storage environment (pressure, temperature and gas composition) is precisely controlled and monitored **[1-4,6]**. These gas 'rig' systems have enabled Scott to accurately determine the rates of uranium metal and carbide corrosion for different gas mixtures analogous to real-world storage environments. By using the IAC's world-class surface analysis facilities, this research not only provides real time corrosion data, but, more importantly, it reveals the actual mechanisms for corrosion and reactivity of the metal and carbide in different storage environments **[2-4]**. Consequently, Scott and colleagues have been able to develop subsequent methods for treating these materials to (i) improve their corrosion resistance, (ii) transform them into environmentally stable (or acceptable) compounds **[3]**, and (iii) to provide efficient means of remediation in the event of an environmental release **[5]**.

The UK Atomic Weapons Establishment (AWE) plays a crucial role in the defence of the UK, responsible for the manufacture and maintenance of warheads for Trident, a submarine-launched ballistic missile. AWE has been at the forefront of the UK nuclear deterrent programme for more than 60 years, directly supporting the Continuous At Sea Deterrence (CASD). In 2004, the AWE sought to establish a collaborative partnership with Bristol, ultimately funding a continuing programme of research at the IAC in excess of £1M, to provide a detailed investigation directed at better understanding and predicting the corrosion of uranium and its alloys **[a]**. This Bristol-AWE partnership was formalised in 2009 with the establishment of a Strategic Alliance agreement, instigated in response to broadening IAC research activity with the company. This Strategic Alliance presented both organisations with the opportunity to develop new collaborative ventures, combining research, people and skills to ensure both institutions are at the forefront of scientific discovery in years to come. As a measure of Bristol's research excellence, the IAC team has won the research prize at the annual AWE materials science and engineering conference four times, in 2007, 2009, 2011 and 2013 **[a]**.

The Sellafield site is the largest and most hazardous site in the Nuclear Decommissioning Authority's estate and is home to an extraordinary accumulation of hazardous waste requiring treatment, much of it stored in outdated nuclear facilities **[b]**. One of the primary missions of Sellafield is the characterisation, retrieval, treatment, and safe above-ground storage of the legacy wastes produced during earlier operations of the UK nuclear industry. These waste streams contain substantial quantities of uranium metal (from spent nuclear fuel), along with components of the fuel cladding such as Magnox and Aluminium. Waste has been stored under conditions that has led to the formation of corrosion products, therefore leading to substantially different characteristics of materials to those originally consigned to the plants for storage **[b]**. As such, unanswered questions pertaining to the state of uranium in storage are of significant interest at Sellafield for the safe retrieval and treatment of legacy wastes. In 2011, Sellafield embarked on a national search to identify and evaluate academic expertise related to the corrosion of uranium and other reactive metals. Sellafield required a sponsored link between their internal Centre of Expertise on uranium and reactive metals and a suitably qualified UK University that would provide expert advice and research to support Sellafield's activities and research, particularly in relation to the behaviour of uranium in wastes. In 2012, the IAC was recognised as world-leading in the field of uranium research, having a substantial and leading international expertise for research on uranium and its corrosion products which has accumulated over a period of approximately 10 years, in close collaboration with the UK Atomic Weapons Establishment (AWE) **[b]**. Subsequently, in early 2012, through the auspices of the Office for Nuclear Regulation (ONR), Bristol was successfully awarded the CoE-university link with Sellafield, who have acknowledged that the IAC's research directly impacted the company's decision on which University to work with **[b]**. Bristol also gained £500k direct funding from Sellafield over a 5 year period **[b]**.

3. References to the research

[1] Allen, G.C., Brown, I.T. and Harris, S.J. (1994), The matrix dependence of ion emission from uranium oxides. *Nuclear Instruments and Methods in Physics Research* Section B, Volume 88, Issue 1-2, p. 170-173. DOI: 10.1016/0168-583X(94)96099-2.

[2] Scott, T.B., **Allen, G.C.**, Heard, P.J., Lewis, A.C. and Lee, D.F. (2005), The extraction of uranium from groundwaters on iron surfaces. *Proceedings of the Royal Society A*, 461: 1247-1259. DOI: 10.1098/rspa.2004.1441.*

[3] Scott, T.B., Petherbridge, J., Harker, N., Ball, R., Heard, P., Glascott, J. and **Allen, G.C.** (2010), Oxidative corrosion of carbide inclusions at the surface of uranium metal during exposure to water vapour. *40èmes Journées des Actinides conference*, CERN, Geneva. Can be supplied upon request.

[4] Scott, T.B., Petherbridge, J., Harker, N., Ball, R., Heard, P., Glascott, J. and **Allen, G.C.** (2011), The oxidative corrosion of carbide inclusions at the surface of uranium metal during exposure to water vapour. *Journal of Hazardous Materials* 195: 115-123. DOI: 10.1016/j.jhazmat.2011.08.011.*

[5] Scott, T.B., Harker, N.J., Jones, C., Hallam, K., Heard, P. and Catherall, R. (2012), Controlled conversion of uranium carbide fission targets using water vapour. *42èmes Journées des Actinides conference, Bristol, UK*. Can be supplied upon request.

[6] Jones, C.P., **Scott, T.B.**, Petherbridge, J.R. and Glascott, J. (2013), A surface science study of the initial stages of hydrogen corrosion on uranium metal and the role played by grain microstructure. *Solid State Ionics* 231: 81-86. DOI: 10.1016/j.ssi.2012.11.018.*

4. Details of the impact

Globally, the nuclear power industry produces about 14% of all electricity, which equates to 6% of all power in a sector worth hundreds of billions of pounds. There are now over 430 commercial nuclear power reactors operating in 31 countries, with 372 GW of total capacity **[d]**. Waste management from this industry is of high economic and social concern, with considerable emphasis and scrutiny on safety and longevity of sites. Of arguably greater social value and scrutiny is the safe storage and maintenance of the UK's nuclear deterrent. Regardless of the civil or military setting, the occurrence of a storage incident is likely to have significant national and international impact, environmentally, economically and socially. The work of Scott and others at the IAC is part of the effort to make uranium storage safer and better understood in terms of reactive chemistry.

Through collaboration with CERN, Sellafield and the AWE, the IAC has broad reach, connecting with national and international partners. Research efforts **[1-6]** have translated detailed, focused scientific work into tangible outputs and counsel for the nuclear industry, consequently playing a critical part in energy policy. Indirect beneficiaries include other EU research facilities such as *"the SPIRAL2 project at GANIL, Caen, France and the SPES project at INFN, Italy"* **[c]**, as well as the UK public and the US public through trans-Atlantic sharing of information between AWE and the Department of Defence. Impact, as a pull-through, is garnered through the uptake of advice and change to protocols, be it from changing the way uranium (and associated compounds and products) are stored or processed, through to improved understanding of the reactive chemistry of uranium in different possible storage environments. Further detail of the impact provided to direct beneficiaries is provided below:

Atomic Weapons Establishment (AWE)
The UK's *"current nuclear weapons capability costs on average around 5-6 per cent of the current defence budget"* **[d]**, equivalent to approximately £2-£2.4 billion. To date, research has been primarily focused on determining the factors influencing the initiation of hydrogen corrosion on uranium surfaces. This research has had a direct feed-through to the safe storage of uranium and associated materials within the MoD estate, considered to account for a significant proportion of the operational cost of Trident and the cost of running Aldermaston (AWE expenditure between 2008 and 2011 is about £2.6 billion) **[e]**. Over the past 9 years, extensive experience and valuable data relating to the corrosion of uranium metal have been accumulated by the Bristol group; *"This knowledge has furthered our understanding of some uranium corrosion processes (particularly the reaction between uranium and hydrogen) and helped to validate and quantify some important predictive uranium corrosion models developed at AWE"* **[a]**. For example, the fundamental relationship between oxide thickness and the severity of hydride attack, which is an underlying mechanistic assumption within several corrosion models applied at the AWE, was directly verified by Bristol in a 2006 technical report **[a]**. The use of such models enables corrosion predictions which are used unequivocally to reduce the need for physical examination, intervention and measurements of stored materials, all of which account for a heavy financial burden on the UK Government **[a]**. The AWE are strongly voiced in their opinion that the *"contribution to the AWE [from Bristol], and by association the UK, has been significant and much needed"* **[a]**.

Sellafield Ltd
One of the key unanswered questions pertaining to the state of uranium in storage at Sellafield is the safe retrieval and treatment of the legacy wastes. *"It is considered that ongoing research with Dr Scott will allow us, in conjunction with other programmes of work, to better predict the current state and residual reactivity of uranium and reactive metal bearing wastes across a number of the legacy plants on the Sellafield site. This has a direct bearing on UK national level programmes for the safe and cost effective remediation of the Sellafield site"* **[b]**, which was valued at £67.5 billion in February 2013 **[f]**. Scott has recommended that spent uranium fuel should be stored in a semi-dry and open containment system, such that the potential for significant hydrogen accumulation is prevented; Sellafield is currently manufacturing test storage casks, to provide a full-scale demonstration of such a storage concept, and is investing approximately £500,000 in this project **[g]**. Bristol's research on hydride formation has affected the Sellafield safety case for recovery of legacy wastes by reducing uncertainty in quantifying the risk associated with the occurrence of thermal excursions. It has also helped to confirm that there is not a significant safety issue posed by uranium hydride in the recovery of legacy waste from Sellafield storage silos. Sellafield is now continuing with the Magnox Swarf Storage Silos (£387 million) and other legacy retrieval projects, including the silos direct encapsulation plant (£1.28 billion) and pile fuel cladding silo project (£341 million), projects worth a total of ~£1.9 billion **[f,h]**. This work on encapsulation of uranium metal in

cement (using synchrotron X-rays) has proven that hydride can be a developing corrosion product; Sellafield will use this information to inform its safety case for waste encapsulation **[a]**. Although the research relationship between Bristol and Sellafield is relatively new, the technical input provided to date has had *"a positive influence on several Sellafield programmes which predate the formation of the link providing…valuable supporting data and expert advice that have reinforced and influenced decisions relating to waste retrieval and treatment programmes"* **[b]**.

CERN (ISOLDE Facility)

The safe disposal of radioactive waste arising from experimental fission studies at CERN is a key objective for the organisation; *"As a by-product of our experimental activities at the ISOLDE on-line isotope mass separator facility, the site has a stockpile of spent uranium and thorium carbide targets from which we have produced fission product gases…These wastes have historically been stored on site and have gradually accumulated, awaiting the development of a suitable processing route for conversion of the material into a waste form that is acceptable to the Swiss authorities"* **[c].** In 2010, work presented by Scott **[3]**, instigated CERN funding a 2-year research project to develop a process for the controlled chemical conversion of their uranium carbide fission targets into an acceptable waste form. Scott's research has solved this specific problem; *"It is considered that the work conducted by Dr Scott and his team has allowed us to develop and implement (from scratch) a processing route for our radioactive fission target wastes. This has had a direct and positive bearing on the operations at the ISOLDE facility and at CERN in general"* **[c]**. This method for processing ISOLDE fission wastes has *"constituted the building of a specialist hot-cell facility for handling waste targets and a gas treatment system for converting the targets. This investment was in the order of 1.4 million Euros"* **[c]**.

5. Sources to corroborate the impact

[a] Atomic Weapons Establishment (AWE). Factual Statement.

[b] Sellafield Ltd. Factual Statement.

[c] CERN. Factual Statement.

[d] 'David Cameron: We need a nuclear deterrent more than ever'. *The Telegraph*. 3rd April 2013. Available from: http://www.telegraph.co.uk/news/politics/david-cameron/9969596/David-Cameron-We-need-a-nuclear-deterrent-more-than-ever.html

[e] World Nuclear Association (April 2012) 'Nuclear Power in the World Today'. Available from: http://www.world-nuclear.org/info/Current-and-Future-Generation/Nuclear-Power-in-the-World-Today/

[f] 'Sellafield clean-up cost reaches £67.5bn, says report'. BBC News. 4th February 2013. Available from: http://www.bbc.co.uk/news/uk-england-cumbria-21298117

[g] Press Release, Cabot Institute, University of Bristol (March 2013) 'Bristol and Sellafield Ltd to build the future of nuclear waste management together'. Available from: http://www.bristol.ac.uk/cabot/news/2013/282.html

[h] National Audit Office. Managing risk reduction at Sellafield. Nuclear Decommissioning Authority. Report by the Comptroller and Auditor General. Ordered by the House of Commons (2012). Available from: http://www.nao.org.uk/wp-content/uploads/2012/11/n1213630.pdf