Development and Expansion of Research Facilities to Address the Challenges of the Nuclear Industry – 16055

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ABSTRACT

With the wide variety of challenges that surround nuclear decommissioning, bespoke facilities are required to undertake experiments which can inform decision making. Here we describe research carried out at The University of Manchester's Dalton Cumbrian Facility, a custom built research environment which incorporates a 5 MV ion accelerator as well as a self-shielded ^{60}Co irradiator. The ion accelerator allows the investigation in to the radiolytic consequences of various charged particles, including protons, alpha particles and a variety of heavier (metal and nonmetal) ions, while the ^{60}Co irradiator allows the effects of gamma radiation to be studied. We provide specific research examples that demonstrate how this equipment can improve our mechanistic understanding of various aspects of the deleterious effects of radiation in the nuclear industry. These examples include applications in waste encapsulation as well as geological disposal and novel surveying techniques. The outlook for future research and expansion of the facility is also presented.

INTRODUCTION

The UK nuclear program was born during the 1940's, in a war-time era, to provide the nation with nuclear weapons. Immense time pressures did not allow for a complete understanding of the implications of this technological development. Knowledge surrounding nuclear processes has increased immensely since then and the approach to treating radioactive waste has been modified accordingly; much of the legacy waste must be conditioned and old plants must be decommissioned. The Sellafield site, home to some of the original development programs and the location of the world's first nuclear plant to produce electricity on an industrial scale [1] now represents perhaps the greatest hazard within Europe [2]. Despite the expanded knowledge base, a variety of significant challenges remain.

The Dalton Cumbrian Facility (DCF), part of the University of Manchester, was opened in 2013 to investigate these challenges and contribute to the UK knowledge

base. The DCF is located close to the Sellafield site to enhance knowledge and technology transfer between academia and the nuclear industry. Focus areas include decontamination and decommissioning, nuclear waste management and storage, new build plant and both existing and new materials which are relevant to the new nuclear build. The DCF capabilities include a 5 MV NEC 15SDH-4 Pelletron tandem ion accelerator [3, 4] and a Foss Therapy Services Model 812 ^{60}Co gamma irradiator as well as an array of analytical interrogation equipment to characterise any changes as a result of irradiation [5]. Here we provide a brief overview of the irradiation equipment as well as some current applications in radiation chemistry, materials science and mineralogy, and electronics that are directly relevant to the industry. The outlook for the facility is also discussed.

IRRADIATION EQUIPMENT

The Particle Accelerator

The particle accelerator is suitable for inducing radiation damage and conducting radiation chemistry experiments (Figure 1). Two ion sources are available to provide protons (H^+), alpha particles (He^{2+}) and a variety of heavy ions. A high current Toroidal Volume Ion Source (TORVIS) provides up to 10 MeV H^+ at 100 µA and up to 15 MeV He^{2+} at 15 µA; and a low current Source of Negative Ions by Caesium Spluttering (SNICS) providing partially and fully stripped heavy (metal and non-metal) ions. At present, we have cathodes for C, O, V, Cr, Fe, Ni, Cu, Zr and W for the SNICS source, but ease of change of the cathode means that we can exchange the SNICS ion type for virtually any heavy ion type.

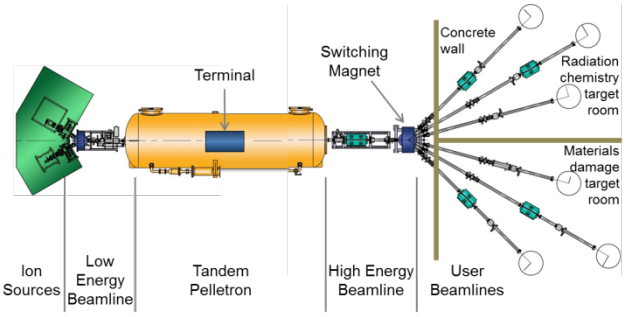

Figure 1: Schematic of the Pelletron tandem ion accelerator at the Dalton Cumbrian Facility.

The ion beam from the accelerator can be directed into one of six different beamlines. Three beamlines are in one concrete walled target room and are designated for radiation chemistry studies at relatively low beam currents of typically 10 nA or less. The other three, incorporating raster scanners for uniform beam illumination over large sample areas – typically 1-2 cm across, are contained in a second target room and are used for radiation damage experiments at much higher currents, employing up to a maximum of 100 µA of protons. This allows

work to be set up in one target room whilst a beamline in the other target room is operational (see Figure 1).

The exact build of the assembly at the end of the beamline varies depending on the nature of the experiment: typically a vacuum chamber is used to house samples for materials damage where as a borosilicate glass cell, complete with magnetic stirrer is often used for radiation chemistry studies. Any assembly typically includes a variety of temperature and dosimetry instrumentation, usually in the form of beam current analytics [5].

The Gamma Irradiator

The gamma irradiator is self-shielded and loaded with capsules of ^{60}Co with an initial total activity of 555 TBq (freshly loaded May 2012). This results in an absorbed dose ranging from below 4 Gy/min to over 400 Gy/min depending on the distance from the source rods. Dose mapping with either Fricke [6] or a Radcal® [7] ion chamber is carried out to provide an accurate dose reading and, with an initial dose rate of 380 Gy/min close to the source rods (both rods raised), the dose rate can be brought down to as low as 0.06 Gy/min with the addition of attenuation and only a single rod raised. This provides the facility with the flexibility to investigate a range of dose rates, from rates characteristic of spent fuel right down to background levels of radiation.

The sample chamber measures 200 by 250 by 270 mm for a total volume of 9 litres and incorporates several turntable positions as well as two 19 mm internal diameter sample ports with scatter shielding. One sample port is directly above a turntable location, the other is to the side of the sample chamber. Three source rod assemblies are available for loading. These are located at the back of the sample chamber and are oriented vertically. Schematic diagrams are provided in Figure 2. This arrangement provides a planar irradiation field and use of the turntable results in a volumetric irradiation of samples, if required. The irradiator is designed to fail safe with the source rods dropping down into the base of the irradiator when the chamber door is open.

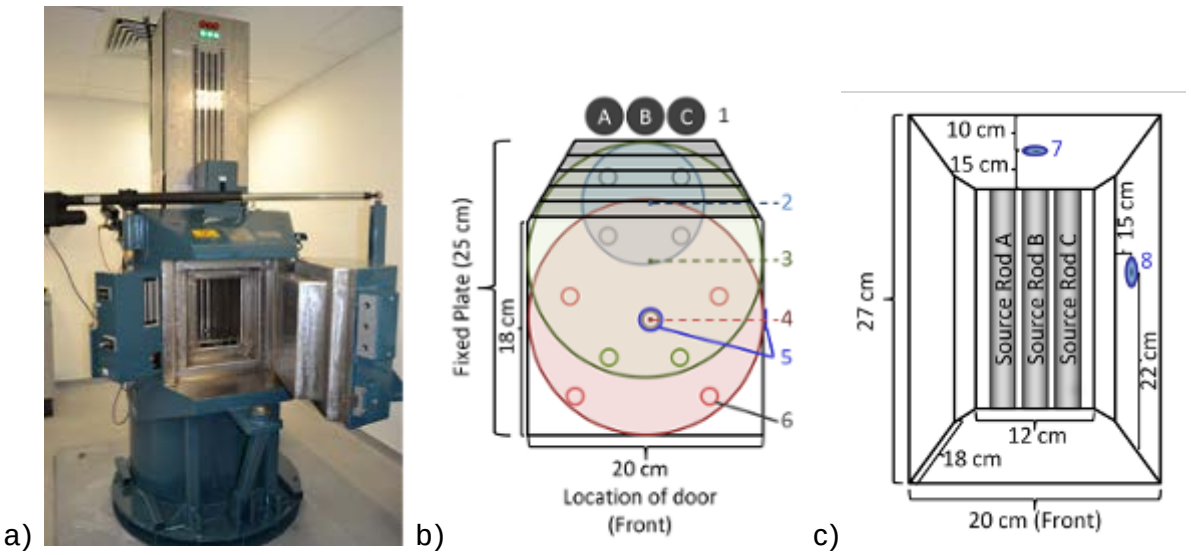

Figure 2: The ^{60}Co gamma irradiator. a) Photograph. b) Top view schematic showing the locations of the various turntables, sample ports and attenuators: 1. Location of source rods; 2. Centre of turntable 1 (source position) 5 cm from sources; 3. Centre of turntable 2 (middle position) 10 cm from sources; 4. Centre of turntable 3 (door position) 15 cm from sources; 5. Service ports (19 mm internal diameter); 6. Turntable bearings. c) Perspective drawing: 7. Top service port; 8. Side service port.

APPLICATIONS

Epoxy Resins for Waste Encapsulation

Epoxy resins are a class of material proposed for waste encapsulation. Conventional waste encapsulants such as cement are unsuitable for certain waste types. Reactive metals such as aluminium, magnesium and uranium react with the high pH cement and corrode, causing them to increase in volume and crack the wasteform. Epoxy resins do not exhibit these limitations and can be selected to display the physical and chemical stability required to be an effective encapsulant and to prevent the release of radionuclides over a long time period (up to 1000s of years) under the conditions of a repository.

Under irradiation, the backbone of a polymer degrades [8] to produce small organic molecules that are capable of leaching from the polymer and dissolving in water. These organic products could potentially form complexes with metallic waste ions, causing them to become mobile and leave the waste package [9]. It is important to gain an understanding of the extent of this leaching and to understand the underlying processes that cause it in order to minimise its occurrence.

Polymer samples were prepared by mixing as-received, stoichiometric quantities of diglycidyl ether of bisphenol A (DGEBA) and tri-ethylene-tetramine (TETA). A curing regime was used that ensured all the components were fully reacted. Preliminary trials with monolithic samples demonstrated a slow leaching rate and further

experiments were carried out with powdered samples. All samples were irradiated in the DCF ^{60}Co irradiator at a variety of dose rates to provide a known total dose, confirmed by Fricke dosimetry.

Analysis of leachate was carried out using an Analytik Jena Multi N/C 2100 S total C/N analyser. Comparison of samples irradiated in calcium hydroxide solution (pH 12.6) with samples irradiated in pure water demonstrated a decrease in concentrations of carbon and nitrogen in the leachate, including organic components, as shown in Figure 3. Changes in the functionality of the polymer were determined using a Bruker Vertex 70 FT-IR with RamII Raman attachment. This found significant changes in the C-N and C-O bonds. Confirmation of minimal leaching of other components of the epoxy resin such as chlorides, and oxalate was provided by ion chromatography using a Thermo Scientific DIONEX ICS-2100**.**

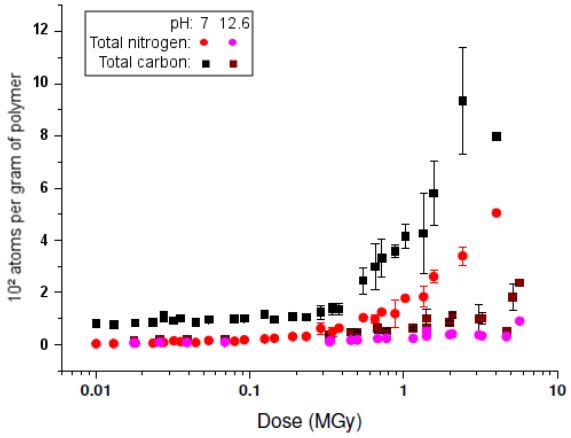

Figure 3: Comparison of total nitrogen and carbon leached from epoxy resin samples under a neutral and alkali conditions (pH 12.6). The latter simulates repository conditions with $Ca(OH)_2$ in solution.

The production of hydrogen was determined using an SRI gas analyser and expressed as G-values: the number of ions or molecules produced per 100 eV of energy absorbed by the medium [10]. These were compared to literature values for pure water and aqueous calcium hydroxide as well as other polymers. Figure 4 shows that the G-values for the epoxy resin are an order of magnitude lower than for the aqueous control samples [11, 12] and no higher than for other polymers such as polystyrene, with a G-value of 0.033 [13], and polyethylene with a G-value 3.1 [14] (not shown in Figure 4 for clarity).

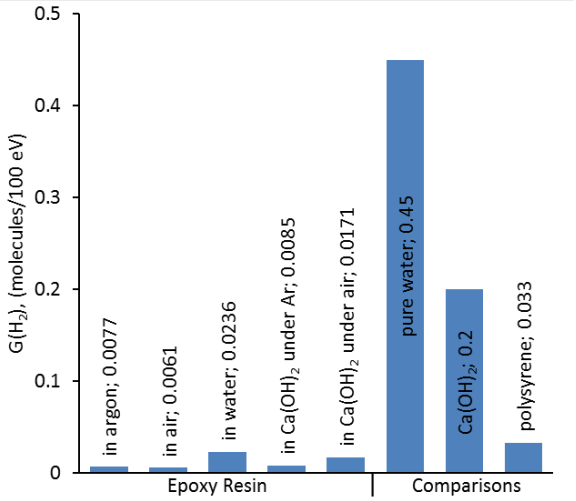

Figure 4: G-values for hydrogen generation from the epoxy resin under a variety of conditions. Aqueous solutions only are provided for comparison as well as polystyrene [13]. G-value for pure water is taken from [11], for $Ca(OH)_2$ from [12].

These results indicate that this class of epoxy resin has a high degree of radiation resistance, and so may be well suited to the encapsulation of waste destined for geological disposal.

Assessment of Potential Host Rock for Geological Disposal

The DCF plays an important role in researching the implications of deep geological disposal of radioactive waste. A detailed understanding of the response of mineral phases to the radiation fields experienced in a geological disposal facility (GDF) is a key aspect of the future UK repository safety case. Of particular importance are the clays that will surround the high level wastes: clays and micas will be ubiquitous in the surrounding geology. Mineral phases will be subjected to controlled amounts of α and γ radiation relevant to the timescales and doses expected in the GDF [15] and will accumulate extensive structural damage as a result [16, 17]. Radiation damage has the potential to affect both the physical integrity of the minerals as well as the oxidation state of redox active elements the phases contain, which in turn will influence their ability to prevent harmful radionuclides migrating from the wasteforms. Furthermore, understanding radiation damage across mineral phases of various structures will provide new insights into the mechanisms and products of radiation damage in complex natural materials [18].

The DCF Pelletron combined with the TORVIS has been used to perform controlled radiation damage experiments across key mineral phases using high energy (5-8 MeV) $^4He^{2+}$ ions to simulate α-particles. Such energies are comparable to α-decay events along the uranium decay chain [19]. Consistent beam currents (50-300 nA) over long time scales (up to 8 hrs) has enabled precise dose determination, and using a diffuse beam spot ($r \approx 0.6$ cm) has allowed for dose/damage gradients across the beam profile to be obtained. Analysis of the radiation damaged samples (Figure 5) using microfocus X-ray diffraction and Fe K-edge X-ray absorption

spectroscopy at Beamline I18, Diamond Light Source [20] has demonstrated significant structural disturbances and chemical changes across the irradiated samples.

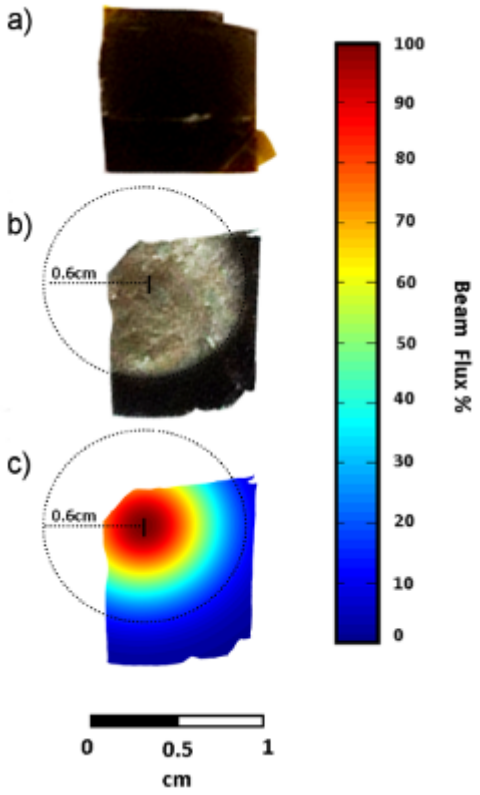

Figure 5: Examples of the optical effects of $^{4}He^{2+}$ ion irradiation on the phyllosilicate mineral biotite. a) Mineral section, ca. 40 µm thick, prior to irradiation. b) Comparable section of biotite following a delivery of ca. 1016 ^{4}He- over approximately a 6 hour irradiation in vacuum. Note the mottled, silver hue developed across the ion beam footprint. c) False colour overlay showing a representative fit of the distribution of the ion beam flux across the sample. Here the beam was focussed to a perfect circle with r≈6 mm, denoted by the dashed circle.

Forthcoming heavy ion irradiation using the SNICS will simulate the effects of α-recoil across minerals and wasteforms containing or sorbing actinides, leading to a change in their capacity to retain radionuclides. In addition to accelerator experiments, high-dose gamma irradiations have been performed upon similar phases (5 MGy) and have yielded chemical changes (paramagnetic defects) and the liberation of free radicals. Combined α and γ irradiations are currently underway.

Electronic Components for Remote Sensing

An active research area at the DCF is in the design and deployment of remote systems, such as submersible vehicles to monitor and characterise the waste stored in legacy ponds and silos. An important consideration when deploying any electronic device in to a radioactive area is the effect that the radiation will have on it.

The Aqua Vehicle Explorer for In-situ Sensing (AVEXIS, Figure 6) has been developed at DCF and is designed to explore and characterise the legacy ponds and silos located on the Sellafield site [21]. The ^{60}Co irradiator has been used to determine what effect exposure to gamma radiation has on this device, with the aim being to determine the expected operating life span of the vehicle when it is deployed in to an active environment. Furthermore, the failure modes for the vehicle will be investigated so that a minimal level of shielding could be introduced to protect sensitive components, increasing the life expectancy if necessary. There has been a considerable amount of research completed on the study of radiation damage to electronic devices used in satellites and other spacecraft [22, 23], with much of this work focusing on the damage caused by high-energy particles at low dose rates over extended lengths of time (40+ years). In contrast, the work at DCF is focused on understanding the damage caused in applications involved in the nuclear industry, where energy levels are lower, dose rates may be higher and exposure times can be hours through to several months or years.

Figure 6: The AVEXIS vehicle used for exploration of nuclear waste storage ponds.

The results presented relate to the analysis of the 5 V, 1 A, Fairchild Semiconductor voltage regulators (KA7805AETU) that are similar to those used on the AVEXIS vehicle. Eight of these components were tested and during the tests a switching circuit, unaffected by gamma radiation, was used to cycle the eight regulators through four different output current levels (100 µA, 250 µA 500 µA and 1.52 mA) while they were irradiated at a dose rate of 220 Gy/min. This dose rate is considerably higher than that experienced in the storage ponds and was used to gain a quick understanding of the long-term effect of radiation exposure within this environment. Throughout the test, the output voltage from each of the regulators,

which under normal conditions should be ~4.3 V (some of the voltage was dropped over a protective diode), was measured in real-time.

Figure 7 shows the average output voltage from the eight regulators during the test. This demonstrates a gradual reduction in output voltage as the total dose the device is exposed to increases. The steps in the graph indicate the repeated switching of the load as the regulators are being exposed, with the peak voltage occurring during the highest load current of 1.52 mA. The nominal output voltage tolerance of this device is +/- 0.1 V and Figure 7 shows that this tolerance is exceeded after a total dose of approximately 1800 Gy, which would represent a time of approximately 2.5 months in the storage ponds being investigated. With shielding of this component, the AVEXIS vehicle could last for longer, if required.

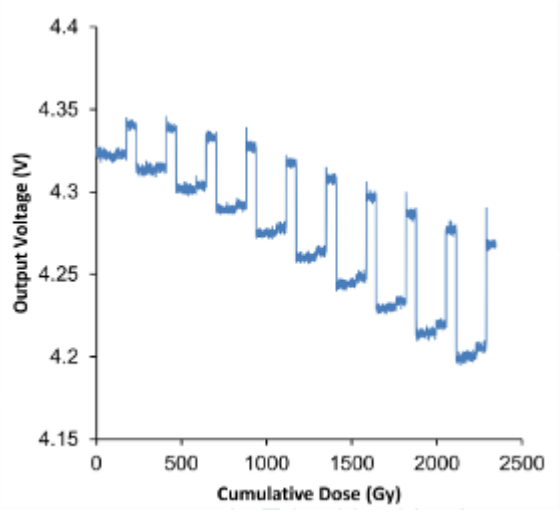

Figure 7: Output from a voltage regulator during gamma irradiation showing a gradual drop with prolonged exposure.

ADDITIONAL WORK

In addition to the technical work outlined above, a recent study in collaboration with the UK's National Nuclear Laboratory, Sellafield Limited and the UK Nuclear Decommissioning Authority has indicated that the current approach to decommissioning and waste packaging can be considered excessively conservative. The results suggested that this may be due to a number of factors including technological limitations, organisational culture and sub-optimal communication mechanisms. This has the potential to open up further avenues of work, to study the organisational behaviours within the industry and, in collaboration with industrial partners, develop robust practises that take into account the risks and challenges inherent to the industry. Alongside this, a program of work is underway which will build on collaborations with industry to identify technical challenges and develop novel solutions such as AVEXIS from academic disciplines such as electrical, mechanical and chemical engineering.

CONCLUSION

As demonstrated above, current capability already allows for a wide variety of work to be performed. Expansion of the facility in the near future will improve on the current technical capability and build on the groundwork laid for organisational cultural studies. This will provide a holistic understanding of the industry which will be of benefit to future operations.

Outlook and Expansion

An additional 2.5 MV NEC 7.5SH-2 single-ended accelerator is currently being installed beside the existing Pelletron. This will provide a dual beam capacity to two of the beamlines in the radiation damage target room: an intense heavy ion beam from the existing 5 MV tandem to induce radiation damage, and a low intensity proton or alpha particle beam from the new 2.5 MV single for ion implantation.

We are also in the process of installing a combined Rutherford Backscattering Spectrometer (RBS) with a silicon drift detector for particle induced X-ray emission (PIXE) and micro-probe optics. This will be installed on one of the beamlines in the radiation chemistry target room to provide an ion beam analysis capability within DCF. In addition, a high temperature and pressure autoclave for in-situ radiolysis and a shielded high dose end station for manipulation of activated samples are currently in development for the beamlines.

This expansion will develop DCF into a Centre for International Nuclear Decommissioning Innovation. This would allow more comprehensive development of a wider range of technologies from a low Technology Readiness Level (TRL) through to deployment on active nuclear facilities. The expanded facility will also conduct analysis of future waste arisings, allowing the identification of optimal management routes and development of new processing techniques. The technical work will be integrated with social science and communication studies to ensure that future decommissioning processes are fit for purpose and that this new approach is actually deployable in practice. The centre will also provide teaching facilities for Master's level and upwards.

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