A Novel Approach for the Study of Corrosion and Ageing of Spent Nuclear Fuel - Looking Inside the box; From a Distance - 16467

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ABSTRACT

The management and safe long-term storage of fuels and waste products is one of the most important challenges that faces the commercial nuclear industry. Understanding how these materials behave and evolve during wet and dry storage will then have significant impact on our ability to understand and predict the long-term impact of storing spent nuclear fuel.

In many research laboratories the infrastructure required for the safe handling and examination of spent nuclear fuel materials is simply not available, thereby limiting the number of research organisations able to contribute to this field of nuclear research. The current article details a novel approach being developed to provide in-situ studies of corrosion and ageing in uranium-based systems relevant to the UK nuclear industry but without the need for hands on experimentation with real spent fuel materials.

INTRODUCTION

The majority of the UK's nuclear waste is currently stored at Sellafield in Cumbria, where a number of different storage strategies are applied depending on the type and radioactivity of each waste. Also present are legacy nuclear wastes, which have arisen from previous complacencies preventing the management of ex-military and civil waste. Urgent action is needed in some cases, e.g. the Magnox Swarf Storage Silo (MSSS), which requires extraction from current storage, followed by reprocessing and packaging in order to transform the material into a safer state, with both effective monitoring and environmental control. Due to the high radioactivity and unpredictable corrosion products exhibited by intermediate and high level nuclear wastes, it is exceptionally difficult to accurately analyse simple parameters which describe the waste contents and therefore define the risks posed by particular storage scenarios. This is due to either the opacity of the physical containments or an inaccessibility to the waste, even for some well managed and controlled stores. Such parameters include the morphology, radioactivity, reactivity

and chemical composition of the material, and any associated hazards, such as gaseous hydrogen production. For example, intermediate level nuclear waste (ILW) arising from the processing of spent fuel from Magnox reactors consists of 500 L stainless steel drums infilled with Magnox cladding, aluminium, uranium and steel, encapsulated in a grout mixture of Ordinary Portland Cement and Blast Furnace Slag. Although originally designed to last for 'at least 50 years' [1, 2], recent inspections have shown that after ~30 years in storage a small portion of the containers are beginning to exhibit signs of degradation and deformation as a result of metallic corrosion of the encapsulated ILW. Management of these particular containers is thwarted by the inability to identify the exact cause of the deformation and, therefore, validate their suitability for future interim storage (≤150 years) without repackaging. In addition, deformation of the packaging, due to the volume expansion via metallic corrosion, greatly increases the probability of a containment breach. This is most likely during transportation, which carries with it the associated risk of an impact event. Similar storage environments have shown that the potential production of pyrophoric material is a recognised risk within these containers [3, 4].

Spent fuel held in legacy ponds and other metallic wastes present in the MSSS are yet another example where the limited material identification has hindered the processing of waste and its subsequent storage.

Current monitoring methods

As the time for preparing a geological disposal facility draws closer, there is a requirement to accurately characterise and evaluate the stability and suitability of current nuclear waste packaging, repackaging and storage methods for the safe future storage, transportation and eventual disposal of waste. Currently, there are two approaches for accomplishing this:
(i) Direct investigation of the waste; and
(ii) Analytical studies of simulated waste to determine predictive corrosion rates and mechanisms occurring. The following manuscript details the initial development of a new analytical approach for examining the internal contents of nuclear waste packages or structure of spent fuel materials, aiming to deliver a step-change in capability for direct characterisation of wastes using a novel stand-off methodology.

A novel approach to stand-off analysis of nuclear waste packages

At present, direct analysis of the waste, including clad-less spent fuel, requires expensive hot-cell capability. In addition, existing stand-off analysis includes conventional radiation detection and visual or video monitoring over long periods of time to detect external changes in the system. These stand-off methods do not have the capability of characterising the waste morphology or chemistry and are, therefore, incapable of identifying potential risks within the waste form. Additionally, some studies have proceeded to simulate waste environments either computationally, or in the laboratory, in order to predict specific metal behaviour, or the long term stability and integrity of the waste package systems. Finite element modelling has become a useful tool for understanding the mechanical

behaviour of waste containers with progressive corrosion of metallic wastes held within. For example, Kallgren et al. investigated how material flow and heat affects the formation and distribution of cracks and voids during friction stir welding used to seal copper radwaste canisters [5]. However, successful predictive models require parameters collected from real world experimentation e.g. [6, 7]. In particular, Wellman et al. highlighted the need for a greater number of in-situ studies of metals retained in grout media [8]. This is due to the fact that grout is a chemically and physically evolving system with solubility limiting metal-mineral phases, metal mobility, corrosion phases and mineral components all changing over time. For example, the presence of quartz (SiO_2), a primary component of grout, was neglected in grout behaviour studies by Glasser [9] and Atkins [10], when studying the Ca-UO_3-H_2O system, even though valuable data were collected regarding the formation of sodium, calcium and uranium mineral phases. The main issue with creating analogous in-situ corrosion experiments is the required time frame to mimic current waste forms which have been evolving for up to 60 years. Consequently, the only accurate way of characterising a waste form is to perform a destructive physical examination which, in the case of radioactive waste packages, is far from trivial. Owing to the potential masses of pyrophoric and radioactive material that would be exposed to oxygen and the environment, this option is clearly not permissible. This leaves only one possible solution: determining a method of looking through the waste package, with good spatial resolution, and creating a computed tomography (CT) scan of the waste container.

Muon tomography can partially achieve this and utilises penetrating natural cosmic rays to produce 3D reconstructions of opaque objects via attenuation of the flux or coulombic scattering of particles from a single point [11, 12, 13]. Muon tomography is non-destructive, is sensitive to the materials atomic number and produces high z-number contrast images. However, at the current technological status the resolution is poor, 2-12 cm per pixel (corrosion products form on the micrometre scale), scan times are long (hours to months) and it can only comparatively identify elements by their z-number [11]. Furthermore, since muons are naturally occurring, there is no control over the muon flux, which limits development of the technique, particularly if larger, more complex, objects require analysis. Nevertheless, it is expected that these technologies will undergo a rapid change in the next few years and demonstrate good applicability for homeland security and for large scale scanning application where fine resolution is not required.

As an alternative approach to more accurately identify the waste chemistry and arising hazardous corrosion products, recent investigations by the authors have explored the use of synchrotron x-rays [14] capable of analysis at micron length scales. Synchrotron x-ray powder diffraction and tomographic analysis were used to explore the rate and mechanisms of metal corrosion, successfully characterising the developing morphology of the arising corrosion products and residual metal to a 1 μm per pixel resolution. Important corrosion characteristics of uranium metal were determined using this method. For example, formation of UH_3 on uranium encapsulated in grout was observed to initiate and propagate in large, protruding, blisters instead of forming a continuous layer across the metal surface (which is the typical case with unenclosed uranium metal).

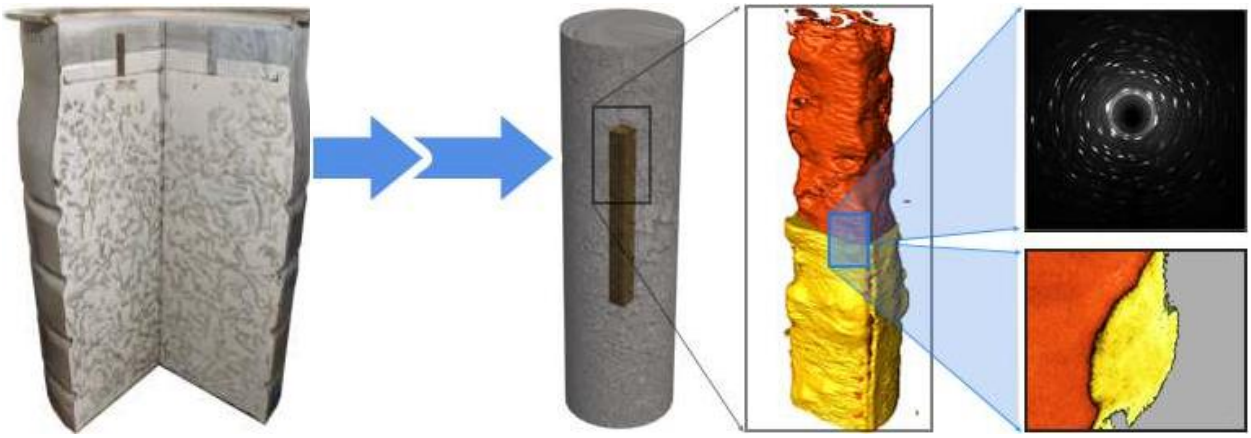

Figure 1. The concept of using synchrotron x-ray tomography and selected volume diffraction to provide direct investigation of uranium metal as it corrodes inside a miniaturised simulant waste package. Tomogram data and diffraction patterns are taken from the work of Stitt et al [14].

Furthermore, it was shown that these blisters persisted whilst submersed in water for at least 18 months [14, 15]. However, due to uranium's high density (18.95 g/cm^3), the x-ray attenuation was too great to analyse samples thicker than 1 mm, even utilising the highest beam energy achievable at the synchrotron (130 keV). Therefore, synchrotron x-rays are considered a useful tool for small scale detailed investigations of uranium behaviour and corrosion when encapsulated in grout, but are not suitable for the monitoring of real waste containers.

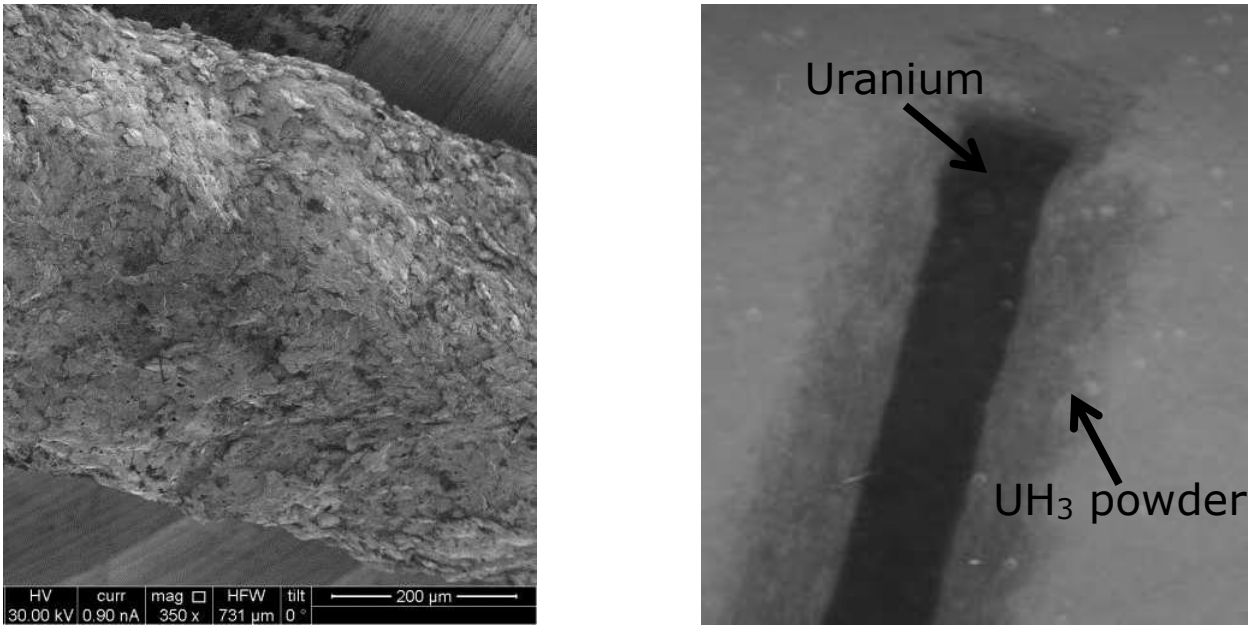

Figure 2. An electron micrograph image (left) and synchrotron captured projection radiography shot (right) of a 0.5 mm thick uranium rod subjected to a limited period of surface hydride (UH_3) formation.

Computer simulations indicated that in order to penetrate typical large waste packages currently in storage, and provide the resolutions necessary to identify corrosion products, photon energies of 6 MeV are required. Commercially available accelerator-based technology (e.g. linacs) or radioactive sources are capable of generating x-ray beams with the required energy for this application (>6 MeV). However, the dose delivered limits tomographic imaging to long exposure time-scales per drum (days - months) and the x-ray source size is restricted to a lateral resolution of ~1 mm. For example, using an 8 MeV linac source in conjunction with translation-rotation acquisition, a horizontal slice image through a 500 mm diameter drum will take approximately 20 mins, with up to 1.5 hours required for larger drums [16]. High power laser driven photon sources can also generate x-ray beams with the required energy and have the favourable beam qualities for this imaging challenge (high photon brightness and small source size <<1 mm).

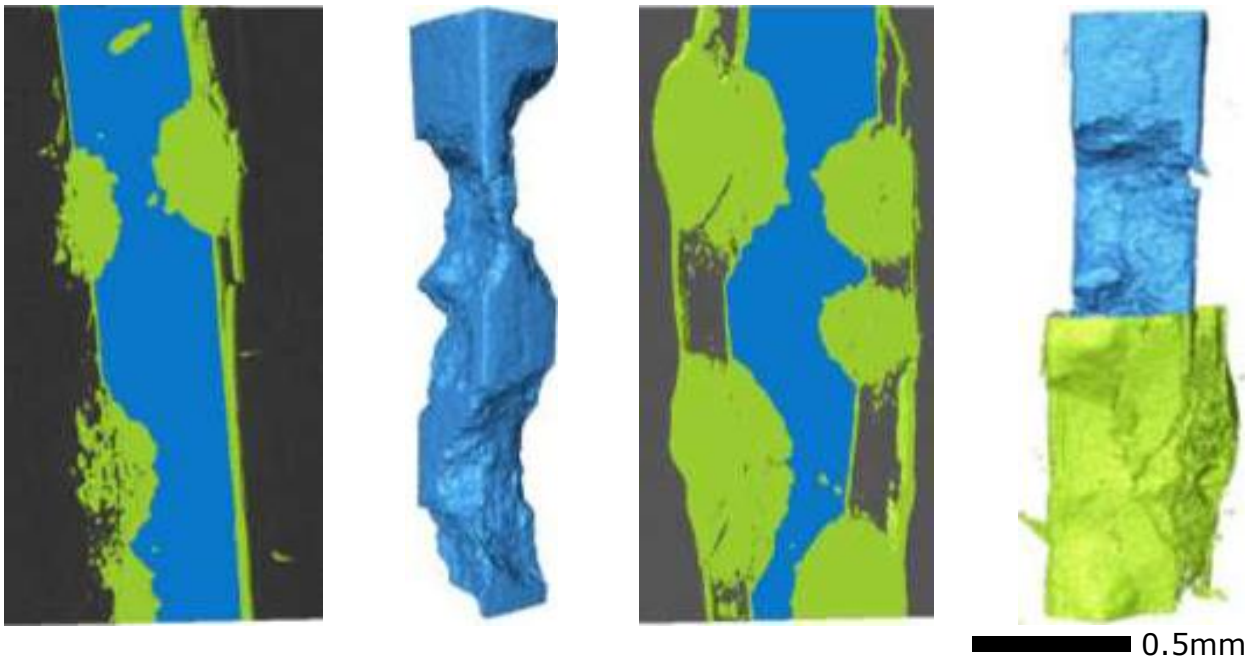

Figure 3. A series of computed tomography reconstructions of corroded uranium encapsulated in ILW-composition grout. Blue represents uranium metal whilst green represents corrosion product. Hydride (UH_3) blisters are clearly resolved in addition to delamination of the surface oxide layer. Due to both UO_2 and UH_3 having an extremely similar density, the synchrotron projection radiography cannot differentiate between the two phases.

In May 2014 we successfully utilised the Vulcan laser at the Central Laser Facility (Rutherford Appleton Laboratory, STFC, UK) to demonstrate the potential for using laser driven photon sources as a future means of examining the internal state of radioactive waste containers. The feasibility experiment produced radiographic data of near life-size simulated Magnox ILW waste material, without modification to the laser apparatus. Improvements and modifications required for producing a stand-

alone waste monitoring facility for Sellafield and other similar nuclear facilities were also discussed. The experiment (Figure 4) utilised a laser pulse with a peak intensity greater than 10^{18} W/cm^2 incident onto a solid tantalum target foil to generate a laser driven electron beam which generates a bright burst of Bremsstrahlung x-ray radiation as the electrons interact with the atomic structure of the target material. This interaction results in a particularly high peak intensity and energy of Bremsstrahlung radiation, thus generating a large flux of high energy x-rays. A review by Giuletti and Gizzi [17] provides a detailed introduction to x-ray emission from laser produced plasmas. The Vulcan experiment utilised P-polarised, 1.054 nm wavelength pulses of 10 picoseconds duration to successfully deliver ~150 J of laser energy for each shot onto 100 μm thick tantalum foil targets. The arising Bremsstrahlung energy peaked at 200 keV, above which the photon flux logarithmically decreased but achieved energies >2 MeV with sufficient photon intensity to achieve projection radiography through the 25 mm lengthwise thickness of a solid uranium Magnox fuel penny encapsulated in cement.

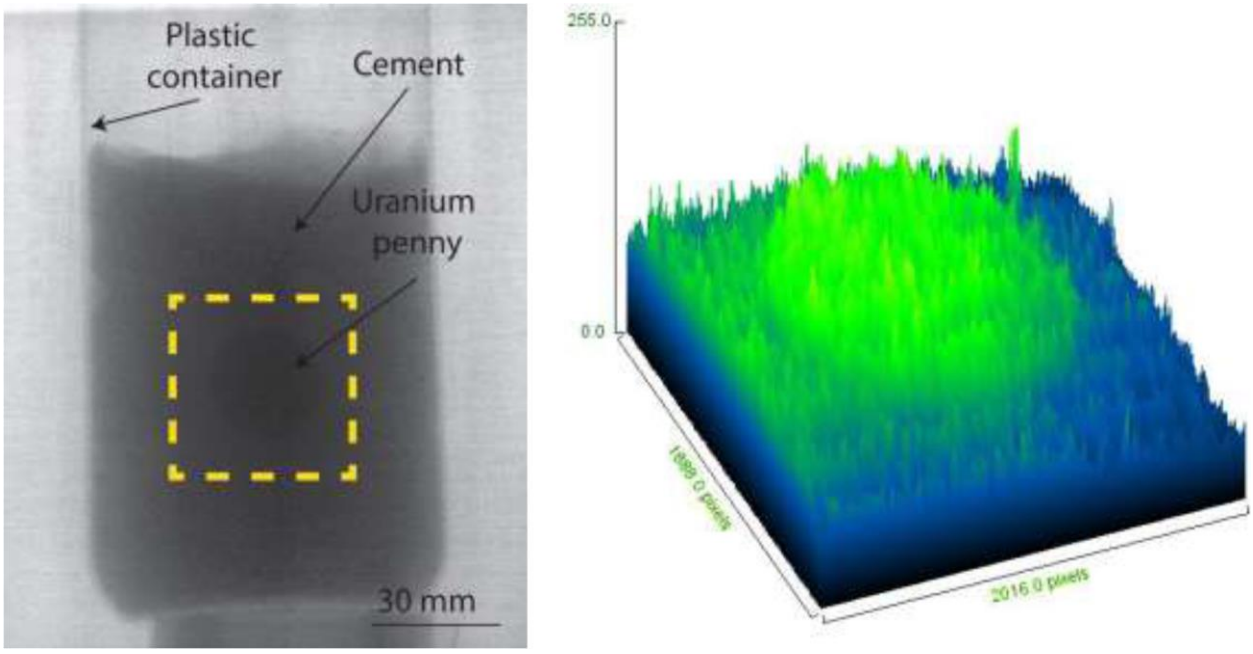

Figure 4: a) Single pulse exposure radiograph of the whole sample using image plates (IPs) without filtering. The outline of the plastic container, cement and uranium penny are clearly shown. The yellow dotted line outlines the area used for the transmission profile in figure b. b) Transmission profile of raw signal (no background subtraction) showing the change in greyscale between the grout and uranium. Green areas coincide with high density material (i.e. uranium) whilst blue areas correspond to lower density material (i.e. cement). The anisotropy in the background signal corresponds to the Gaussian angular distribution of the x-ray flux across the analysed sample.

DISCUSSION

This 'proof of principle' study has successfully demonstrated non-destructive radiography of encapsulated high density nuclear material without modification to the Vulcan laser system. In the current configuration the laser pulse driven x-ray source was capable of penetrating medium masses of material, successfully attaining imaging resolutions well below 1 mm over wide scan areas of ~30 cm^2. Furthermore, this technique has exhibited potential for further development of additional techniques including x-ray tomography, gamma-ray tomography, neutron imaging and criticality testing.

Advantages of using a laser driven source

Laser driven, pulsed, x-ray sources have exhibited properties well suited for radiography and non-destructive testing applications in security and high value sectors, such as aerospace, nuclear and advanced manufacturing. The high resolution imaging of large, dense, objects is achievable due to the high energy (up to 10 MeV), high flux and short pulsed laser driven photon source. These characteristics are unique to laser driven sources when compared to their commercially available counterparts, such as linac systems, which cannot achieve these qualities in a single source. This tunable, laser driven, system provides the flexibility to image a range of components and materials in a vast range of situations, and is thereby well suited for the advanced inspection techniques required in nuclear waste management, thereby offering the potential for a ground-breaking analytical technique, most notably in the nuclear sector.

Other authors have previously conducted studies to characterise x-ray emission from high power laser solid interactions across a variety of laser systems [18, 19], as well as radiography demonstration of the imaging qualities of the beams [20, 21]. In the present experiment the x-ray beam demonstrated large divergence which permits a large field of view, thus providing a suitable foundation for the development of tomographic imaging of large objects. Simultaneous production of high energy, bright neutron beams provides the potential for neutron radiography; a non-destructive, element specific, imaging technique, which is particularly sensitive to large variations in atomic number.

Potential for developing a nuclear waste scanning facility

Adequate nuclear waste scanning requires the identification of the approximate mass and spatial distribution of i) high density material, ii) sensitive corrosion products, iii) fissile material, iv) pyrophoric material and v) any gaseous products.

The Vulcan experiment has shown that laser driven photon sources can successfully identify the distribution of material based on relative mass, from the stark difference between uranium and cement attenuation of the x-rays. Furthermore, real ILW containers often exhibit external deformation which is likely caused by expanding material located near the circumference. This is ascribed to volumetric expansion driven by corrosion of contained metal waste located close to the drum walls. The point source and divergent nature of the photon beam should prove

particularly capable at identifying this errant material since the transmission path through the drum margins is shorter than the centre of the container (Figure 5). Thus radiographs are expected to retain better absorption transmission contrast in these marginal areas with evidence of this observed in Figure 3, which displays the entirety of the sample.

Further experimentation is required to determine if additional low density corrosion products can also be identified using this method. However, previous lower energy x-ray studies (115 keV) have shown that this should be easily achievable [14]. Tuning the laser setup to generate high fluxes of x-rays in the 6 MeV range requires modification of the interaction conditions to increase the laser intensity (by increasing the laser energy and shortening the pulse duration to ~1 ps), thereby transferring the Bremsstrahlung energy profile peak further towards the MeV range.

Alternatively, this can be achieved by using different methods to accelerate the electrons. Other studies have passed the laser through gas capillaries and utilised Bremsstrahlung converter foils to generate x-ray emission with very short pulse lengths of 30-40 fs and source size down to ~30 μm [22]. The laser accelerated electron energy in this case is tuneable from several hundred MeV up to GeV, generating average Bremsstrahlung radiation of 10-100s MeV when the electron beam is passed through the converter foil [22, 23]. However, the beam emission is much more collimated and the photon density is greatly reduced, rendering large field-of-view single pulse exposure image acquisition infeasible.

Currently, progression and development of imaging at this energy is limited by detector technology and instrumentation cooling. Detection of high energy gamma-rays requires development of a high density, high resolution, large 2D scintillator detector with rapid acquisition and processing times. Although acquisition of a single image yields rapid data acquisition (for the CsI detector testing in this work), multiple shots over a long time period (e.g. when performing tomographic analysis) is time limited to one pulse per 20 minutes since the laser system is flash pumped and, therefore, requires cooling after each shot.

Cooling restrictions may be overcome in the future by transitioning to using a high power diode pumped laser, such as the DiPOLE system [24]. This not only offers higher power but can run at 10 Hz operation and is more efficient, more compact, and permits deployability - potentially with a footprint sufficient to fit inside an ISO sized freight container.

Efficient cooling of diode pumped systems potentially allows for multiple shots per second, permitting higher quality image processing via frame averaging. In addition, a rapid acquisition rate enables the development of a tomography system, which would provide accurate volumetric measurements of large mass systems containing both high and low density materials.

Materials posing the greatest risk to the safe confinement of nuclear waste are either hydrogen based, (e.g. hydrogen gas or metal hydrides) and, therefore, potentially flammable, or are highly radioactive, thus providing the potential for large accumulations of the material to reach criticality. Neutrons are highly sensitive to hydrogen based compounds. Thus, neutron imaging will be able to efficiently identify their location and relative quantity [25]. Neutrons have also

previously been utilised for criticality testing, for example by the nuclear materials identification system (NMIS) [26]. In this method, a time resolved analysis is achieved; neutrons that pass through active material induce fission, generating additional particles that are detected later than directly transmitted neutrons.

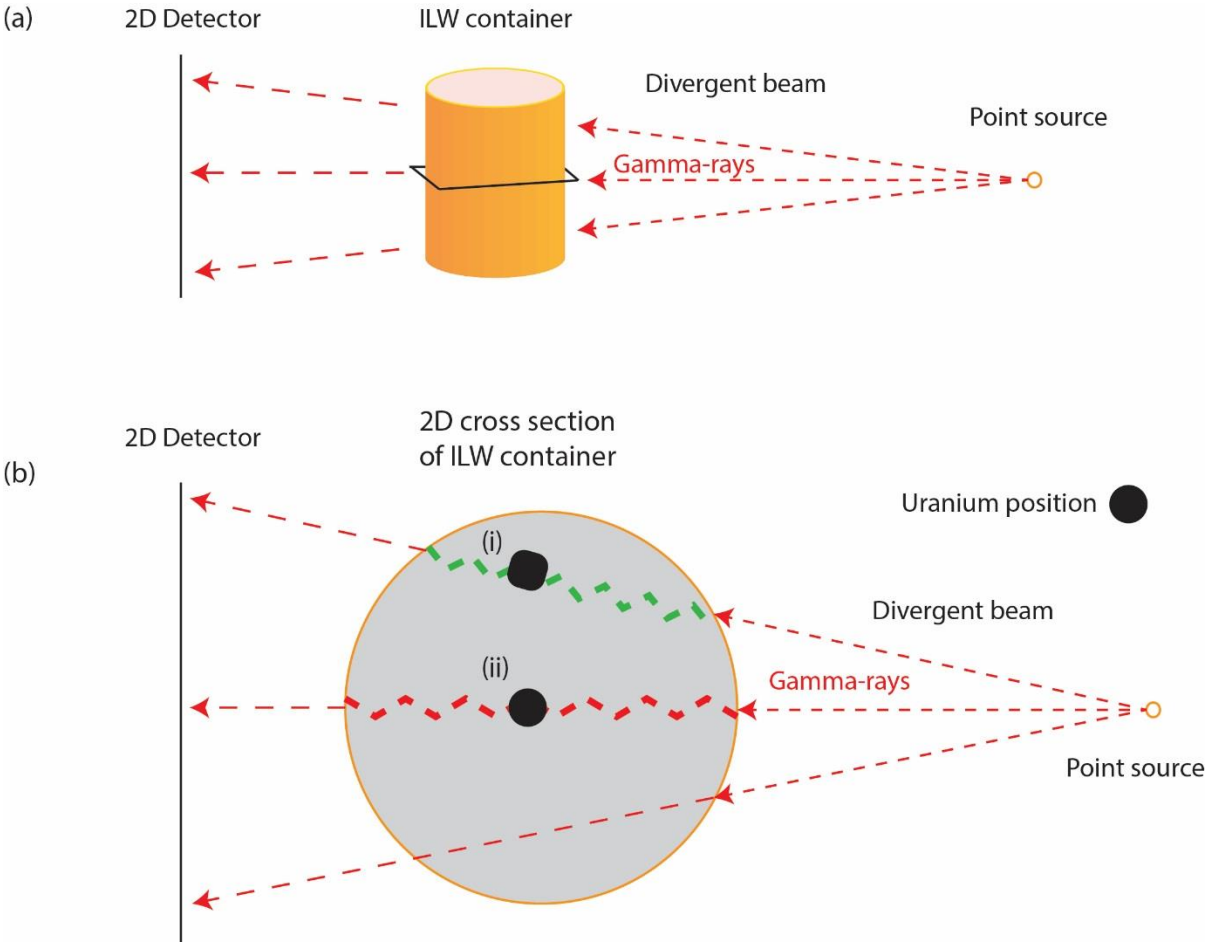

Figure 5: Schematic diagram demonstrating the advantages of a divergent beam from a point source for imaging of ILW containers. Deformation of ILW containers is often observed at the circumference of the container, where the gamma-ray transmission path is shorter (green), producing an image that has potentially higher contrast compared to the middle of the container where the path length is longer (red) and more likely to be scattered.

CONCLUSIONS

Understanding how spent nuclear fuel and associated ILW materials behave and evolve during wet and dry storage and repackaging is of significant importance for better predicting and managing long-term safe storage.

The work detailed in this manuscript has provided proof of concept for the development of laser induced gamma-ray burst tomography and in-situ detection of

high density materials in life-sized nuclear waste packages.

By transitioning to the use of diode pumped lasers, gamma-ray bursts may be achievable at rates of up to 10 images per minute, allowing for both gamma and neutron tomography. The quality of the gamma-ray projection radiography demonstrated here presents the possibility of providing a sub-millimetre resolution of observation, invaluable for examining fine features including corrosion product thicknesses, void spaces and cracks or defects.

The development and construction of a diode pumped gamma-ray scanning instrument is feasible on a 10 year timescale and holds potential for an in-situ waste scanning capability at nuclear waste storage facilities. Once operational, waste packages displaying errant behaviour will be prioritised for scanning and analysis, whilst standard packages will undergo routine product control and assessment before transportation to a geological disposal site.

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