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Programme

Tuesday 9 July

16:00 Early registration, networking and light buffet
   Foyer

18:00 Close

Wednesday 10 July

08:30 Registration

Lecture Theatre 1
Chair: Roy Awbery, AWE, UK

09:00 Welcome
   Roy Awbery, AWE, UK

09:10 Keynote 1
   Ensuring Nuclear Forensics
   David Kenneth Smith, Nuclear Security Coordinator (Forensics),
   International Atomic Energy Agency, Austria

09:40 Remarks from the Chief Scientific Officer
   Robin Grimes, Imperial College London, UK

09:45 Keynote 2
   Nuclear Forensics at AWE
   Daniel Thomas, AWE, UK

10:15 Coffee break, exhibition and posters
   Foyer

Analytical Techniques 1

Lecture Theatre 1
Chairs: Marianne Wilkerson, Los Alamos National Laboratory, USA and Thomas Scott, University of Bristol, UK

10:45 Isotope Analysis of Particles by SIMS for Nuclear Forensics: Analogues and Applications
   Travis Tenner, Los Alamos National Laboratory, USA

11:00 Internal structure and composition of Fukushima-derived particulate examined through
   complementary synchrotron and mass-spectrometry techniques
   Peter Martin, University of Bristol, UK

11:15 Laser-induced breakdown spectroscopy of actinides: elemental and isotopic analysis
   for nuclear forensics
   Gregory Hull, The University of Manchester, UK
11:30  **Morphological analysis of Uranium oxides**  
Luther McDonald, University of Utah, USA

11:45  **Relationships between experimental signatures and processing history from a variety of Plutonium Oxide materials**  
Brian Scott, Los Alamos National Laboratory, USA

12:00  Lunch, exhibition and posters

  *Foyer*

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**Environmental**  
*Lecture Theatre 1*

*Chair*: Anya Keatley, University of Bristol, UK and Matthew Stapleton, AWE, UK

13:00  **Radiological source localisation using multiple unmanned aerial vehicles (UAVs) and survey heights**  
Thomas Scott, University of Bristol, UK

13:15  **Environmental nuclear forensics applied to Fukushima fallout: From bulk environmental samples to individual particles**  
David Richards, University of Bristol, UK

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**Data Analysis & Statistics**  
*Chair*: Anya Keatley, University of Bristol, UK and Matthew Stapleton, AWE, UK

13:30  **Pitfalls of data quality and how to avoid leaping into them**  
Peter Hiller, National Nuclear Laboratory, UK

13:45  **Uncertainty and confidence in nuclear forensics: theory and practice**  
Matt Stapleton, AWE, UK

14:00  **Don’t Believe Everything You Think: Investigating Forensic Signatures Materials from Early in the Uranium Fuel Cycle**  
Enrica Balboni, Lawrence Livermore National Laboratory, USA

14:15  Tea break, exhibition and posters

  *Foyer*

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**Radiochemistry**  
*Lecture Theatre 1*

*Chairs*: Brian Bucher, Idaho National Laboratory, USA and Olivia Marsden, AWE, UK

14:45  **Application of plutonium age dating to samples of known provenance**  
Matthew Higginson, AWE, UK

15:00  **Variable geometry: The art of measuring the unknown**  
Alanna Downing, AWE, UK
15:15  **Rapid isolation of fission products from a UO$_2$-salt target**  
Jacquelyn Dorhout, Los Alamos National Laboratory, USA

15:30  **Determination of weight distribution ratios of protactinium and neptunium on TrisKem ZR (hydroxamate) resin**  
Jack Mullen, AWE, UK

15:45  **Improved nuclear data from synthetic fallout production for post-detonation forensics exercises**  
Brian Bucher, Idaho National Laboratory, USA

16:00  **Nuclear Forensics: Why should I want to be involved?**  
Paul Thompson, AWE, UK

16:15  Posters, exhibition and reception

18:00  Close

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**Thursday 11 July**

*Lecture Theatre 1*  
**Chair:** Paul Thompson, AWE, UK

09:30  **Keynote 3: Nuclear Forensics and International Engagement – The NSDD Perspective**  
Mansie Iyer, Department of Energy, USA

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**Materials & Processing**  
*Lecture Theatre 1*  
**Chairs:** Neil Hyatt, The University of Sheffield, UK and Matthew Gilbert, AWE, UK

10:00  **On using the natural uranium isotopic signature in ores for nuclear forensic investigation**  
James A Dunne, AWE, UK

10:15  **Chemical and morphological analysis of Ammonium Diuranate compounds**  
Jacquelyn Dorhout, Los Alamos National Laboratory, USA

10:30  **Coffee break, exhibition and posters**  
*Foyer*

11:00  **Effects of aqueous processing conditions and thermal decomposition on forensic signatures of Uranyl Oxalate**  
Nathan Thompson, University of Sheffield, UK

11:15  **Hydration of UO$_3$ materials following storage under controlled conditions of temperature and relative humidity**  
Marianne Wilkerson, LANL, USA
### Modelling & Simulation

**Chairs:** Neil Hyatt, The University of Sheffield, UK and Matthew Gilbert, AWE, UK

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<thead>
<tr>
<th>Time</th>
<th>Title</th>
<th>Speaker(s)</th>
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<tbody>
<tr>
<td>11:30</td>
<td><strong>Water layers on Actinide Oxide surfaces</strong></td>
<td>Bengt Tegner, Heriot-Watt University, UK</td>
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<td>11:45</td>
<td><strong>Spent fuel modelling for Actinides and fission products in pebble bed high temperature reactors</strong></td>
<td>Bing Xia, Tsinghua University, China</td>
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<tr>
<td>12:00</td>
<td>Lunch, exhibition and posters</td>
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**Foyer**

13:00  **Keynote 4**  
Chair: Daniel Thomas, AWE, UK  
Investigations and forensics science support to a nuclear security event  
John Simm, UK Metropolitan Police, UK

13:30  **Good Day Mr Naxos**  
Paul Thompson, AWE, UK

14:00  Tea break and posters  
**Foyer**

### Analytical Techniques 2

**Lecture Theatre 1**

**Chairs:** Travis Tenner, Los Alamos National Laboratory, USA and Luther McDonald, University of Utah, USA

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<tr>
<th>Time</th>
<th>Title</th>
<th>Speaker(s)</th>
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<tbody>
<tr>
<td>14:30</td>
<td><strong>Material exercises of the International Technical Working Group (ITWG) for nuclear forensics</strong></td>
<td>Olivia Marsden, AWE, UK</td>
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<td>14:45</td>
<td><strong>Advances in rapid nuclear forensic preparation techniques</strong></td>
<td>David Reading, GAU-Radioanalytical, UK</td>
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<td>15:00</td>
<td><strong>Passive, non-intrusive assay of depleted uranium</strong></td>
<td>Helen Parker, Lancaster University, UK</td>
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<td>15:15</td>
<td><strong>Progress in ultra-low-level $^{238}$Pu measurements by Thermal Ionization mass spectrometry</strong></td>
<td>Jeremy Inglis, Los Alamos National Laboratory, USA</td>
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<tr>
<td>15:30</td>
<td>Closing remarks</td>
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<td>15:45</td>
<td>Close</td>
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Posters

P1. **Is Thorium Truly a “Proliferation Resistant” Nuclear fuel: A Nuclear Forensics Perspective**
Erin Holland, University of Bristol, UK

P2. **The application of Isochrons to evaluate radiochronometry model age assumptions in UOC**
Lauren Harrison, Lawrence Livermore National Lab, USA

P3. **Assessing High-energy, Laser-driven, X-ray Sources as a Technique to Analyse ILW Containers**
Christopher Jones, University of Bristol, UK

P4. **A compact laser ion source for in situ mass spectrometry for rapid detection of nuclear materials in environmental samples**
Ankur Chaudhuri, Canadian Nuclear Laboratories, Canada

P5. **Employing a Fractional Factorial Matrix to Study the Effects of Solution Processing Parameters on Morphology of Studtite and UO$_3$**
Nathan Thompson, University of Sheffield, UK

P6. **Thorium procurement, policy and disposal**
Sam Cross, AWE, UK

P7. **Rapid Radiochemical Separations of Americium For Nuclear Forensics**
Joe Mahmoud, University of Manchester, UK

P8. **Deformation and Strain Concentration in Cast Uranium**
Phil Earp, University of Oxford, UK

P9. **Using a high-performance ICP-QQQ in MS/MS mode for challenging nuclear applications**
Raimund Wahlen, Agilent, UK
Keynote: Ensuring Nuclear Forensics

David Kenneth Smith
International Atomic Energy Agency, Austria

Utilized by States to criminalize the unauthorized use, production or storage of nuclear and other radioactive materials, as well as prevent these occurrences, nuclear forensics has emerged at the forefront of the fight against nuclear terrorism. A priority is the development of national capabilities through a risk informed approach consistent with requirements to respond to a nuclear security event. Nuclear forensics relies upon technical expertise crossing the boundaries of analytical, materials, forensics, chemical, isotopic, environmental and nuclear science to identify nuclear and other radioactive material out of regulatory control. Resulting signatures are utilized by law enforcement, nuclear security and regulatory officials to determine if violations of criminal or civil laws have occurred as well as the integrity of measures, at the State and facility level, to secure these materials. Essential is the recognition of the technical results from the examination as a key piece of legal proceedings. The conduct of the examination must be fully planned and documented, meet the requirements of the leading competent authority and must be undertaken by trained personnel using scientifically accepted methods recognized by a court or regulator.

Fortunately, cases involving nuclear or other radioactive material out of regulatory control occur infrequently. The challenge then becomes sustainability of robust nuclear forensic capabilities already in place and their availability when required. Access comes from investigators, examiners, scientists, responders, regulators, prosecutors and policy makers who understand the relevance and application of nuclear forensics as part of a nuclear security regime. Training and exercises involving responders and examiners maintains readiness. Research is a magnet to provide peer-reviewed solutions and build confidence in techniques relevant to a nuclear forensics examination. Conferences and international forums such as NuFor 2019 further promote outreach and awareness. This collective favourably positions nuclear forensics as an enduring and effective component of a State’s national response plan.

Nuclear Forensics at AWE

Daniel Thomas
AWE, UK

Nuclear Forensics is an important part of our national capability to respond to an incident involving radiological or nuclear material. We seek in particular to be able to determine what an unknown material is, what its intended uses might be, whether or not the material originated from one of our facilities and processes, and whether individuals can be linked to the material in question. Our holistic approach to unpredictable scenarios is intended to provide an agile ability assist investigators and officials. Our understanding has evolved significantly over the last decade or so, and AWE has been at the forefront of this evolving area of scientific capability. We are part of a broad community of technical organisations seeking to help our Government and legal systems to respond and assist, and we call upon a wide array of scientific disciplines in doing so. This talk will aim to give a brief overview of what we’re doing at AWE, why we are doing it, and highlight the amazing technical challenges we face now and into the future.
Analytical Techniques 1

Isotope analysis of particles by SIMS for nuclear forensics: analogues and applications

Travis Tenner, Todd Williamson, and Stephen Lamont
Los Alamos National Laboratory, USA

Characterizing pre-detonation material and post-detonation debris is a problem of interest for nuclear forensics. A challenge is that such samples are often in particulate form, micron-sized or less. Additionally, particulate samples may be heterogeneous, as a mix of device material and environmental debris, and/or having multiple isotope populations. To survey such samples, we rely on secondary ion mass spectrometry (SIMS), as it is an in-situ analytical technique with the ability to produce isotope ratios of individual particles with micron-scale fidelity.

To understand the nuclear forensic capability of SIMS, we’ve been characterizing U-oxide reference materials at Los Alamos National Laboratory. We’ve developed techniques to evenly disperse particulates for analysis, suspending materials in solvents by ultrasonication, and pipetting suspensions onto a substrate. We produce spatially resolved maps of particle isotope ratios by rastering the primary ion beam over regions of the sample surface. We’ve evaluated the precision of individual particle data from a homogeneous reference material source, and find they scatter symmetrically about the averaged isotope ratio. Furthermore, the extent of scatter among a particle dataset can be modelled as a function of counting statistics uncertainties (Fig. 1a). These error envelope models provide a measure of confidence in determining if a sample has one or more isotope sources present.

By analyzing samples with mixed isotope sources, we also evaluate the quality of uranium three-isotope plots (Fig. 1b). Specifically, higher counts from individual particles generate improved mixing lines between endmember isotope sources. This is demonstrated for both reference material samples, as well samples from the ITWG CMX-5 exercise.

Fig. 1 caption: (a) particle data from two reference materials and error envelope models of their isotope ratios; (b) corresponding uranium three-isotope plot. Through Los Alamos National Laboratory, this document is approved for unlimited release under LA-UR-19-23425.
Internal structure and composition of Fukushima-derived particulate examined through complementary synchrotron and mass-spectrometry techniques

Peter Martin¹, Silivia Cipiccia², Darren Batey², Chris Jones¹, David Richards¹, and Tom Scott¹
¹University of Bristol, UK, ²Diamond Light Source Ltd., UK

Keywords: Synchrotron, Fukushima, Spectrometry, Isotopics

Through the application of both laboratory and synchrotron radiation (SR) x-ray tomography (XRT), the internal structure of a representative 450 μm × 280 μm × 250 μm particle was shown to be highly-porous – with 24% of the internal volume constituted by void space, exhibiting limited interconnectivity between these bi-modal sized pores. The use of coherence x-ray diffraction imaging (iterative ptychography) to investigate sub-regions of the particulate sample enabled nanometer spatial resolutions beyond those attainable using traditional x-ray optics. Alongside this extensive internal gaseous volume, the fibrous morphology of the Si-rich particle was also illustrated using these non-destructive x-ray-based methods. Compositional (elemental) analysis of the particulate material through SR x-ray fluorescence (XRF) detailed the peripheral enrichment of several elements (including Sr, Pb and Zr) – with the existence of Cs associated near-exclusively with the location of several highly-angular Fe-based fragments, extruding from the materials surface. Many larger fragments of cement composition were additionally observed embedded into the particles surface, however, these were conversely shown to conform to the exterior morphology of the particle. The component of fissionogenic Cs was determined to account for most of the elemental abundance within the particle with limited contribution from natural ¹³³Cs.

SR x-ray absorption near edge structure (XANES) analysis on several high atomic density particles located within the bulk particle confirmed them to be U in composition, existing in the U(IV) oxidation state, as UO₂. The isotopic analysis of this micron-scale U material enclosed just below the surface of the particle was subsequently determined using secondary ion mass spectrometry (SIMS), having spatially referenced their co-ordinate positions between the different techniques. SIMS mapping revealed the U-rich particle to be ~1 μm in maximum dimension, consisting of enriched U with 3.54 wt% ²³⁵U – analogous to that used in the FDNPP reactor Unit 1 fuel assemblies.

Laser-induced breakdown spectroscopy of Actinides: Elemental and isotopic analysis for nuclear forensics

Gregory Hull
The University of Manchester, UK

Keywords: Laser-Induced Breakdown Spectroscopy (LIBS); Isotopic Analysis; Multivariate Analysis

Laser-induced breakdown spectroscopy (LIBS) permits real-time elemental analysis of a material irrespective of its nature, physical state or environment. The ability to perform contact-free or stand-off analysis of materials by this method offers significant advantages for applications which require interrogation of hazardous materials, and those contained within controlled or inaccessible environments. This capability is a particularly attractive feature for applications in the nuclear industry and in the field of nuclear forensics. The LIBS technique demonstrates many of the attributes required of a field-deployable method: accurate and rapid analysis, minimal sample preparation and the ability for contact-free or remote operation. Whilst the technique is capable of identifying all elements in a sample on the basis of their atomic / ionic emission spectra, isotopic LIBS require the use of high resolution spectrometers – limiting the throughput and portability.

This project has focused on the development of tuneable diode laser absorption spectroscopy (TDLAS), in conjunction with LIBS, for isotopic analysis of uranium. This ‘hyphenated’ approach (LIBS-TDLAS) provides complementary information about a sample: LIBS can fingerprint major and trace constituents using
multivariate data analysis, whilst TDLAS can quantify the precise $^{235}\text{U}:^{238}\text{U}$ isotopic ratio. This approach offers potential applications in nuclear forensics investigation, as it could immediately indicate a seized material’s intended use (as fuel or weapon) and provide background information on where and when it was processed and mined. Two studies were completed to demonstrate the feasibility of the hyphenated LIBS-TDLAS approach. Firstly, LIBS was used to quantify rare earth elements impurities in alkali metal salt matrix using multivariate data analysis (Fig. 1). Secondly, lithium D-lines at 670.78 nm was detected using TDLAS and a laser-ablation plasma (Fig. 2). Lithium was selected for the latter study because the combination of the $^6\text{Li}:^7\text{Li}$ shift and the D-line spacing provides a good surrogate for the isotopic shift of uranium.

Morphological analysis of Uranium oxides
Luther McDonald, Alexa Hanson, Sean Heffernan, and Cody Nizinski
University of Utah, USA

Keywords: Morphology; Scanning Electron Microscopy; X-ray Diffraction

Particle morphology is an emerging signature that aids in safeguarding and preventing the illicit trafficking of nuclear material. Work by our team has proven that quantitative analysis is possible, and that particle morphology can be used as signatures of process history. These initial studies quantified the surface morphology of $\text{Am-}\text{UO}_3$, $\alpha-\text{UO}_3$, $\text{U}_3\text{O}_8$, and $\text{UO}_2$ based on synthetic temperature and precipitation route. It was found that five different precipitation routes including: uranyl peroxide, ammonium diuranate, sodium diuranate, magnesium diuranate, and ammonium uranyl carbonate, all produced precipitates with unique morphologies which were maintained through the calcination process to $\text{UO}_3$, $\text{U}_3\text{O}_8$ and $\text{UO}_2$. Statistical analysis of the morphological results will be presented illustrating the potential of this technique in identifying processing history.
**Relationships between experimental signatures and processing history from a variety of Plutonium Oxide materials**

Brian Scott, John Joyce, Alison Pugmire, Jared Stritzinger, Laura Wolfsberg, and Marianne Wilkerson
Los Alamos National Laboratory, USA

**Keywords:** Plutonium oxide, X-ray diffraction, Raman spectroscopy, Aging

The primary oxide of plutonium, PuO₂, is a complex material due to ongoing change in lattice structure resulting from radioactive alpha decay. In addition, the element plutonium is complex, existing in various allotropes and oxidation states which can influence formation of oxides due to different nucleation and growth mechanisms. The juxtaposition of these unique properties, along with formation conditions of the initial oxide, makes for PuO₂ materials with a wide variety of local and bulk experimental signatures. In addition, the presence of PuO₂ on metal surfaces often leads to Pu₂O₃ through self-reduction, which provides additional signatures that can be related to sample history. This work measures plutonium oxide signatures with X-ray diffraction, X-ray and neutron reflectivity, Raman, and X-ray photoelectron spectroscopy from a variety of different oxide materials. These materials include PuO₂ powders from nuclear facilities, synthesized PuO₂ and Pu₂O₃ materials, and laboratory fabricated PuO₂ epitaxial thin films used as aging standards. This talk will describe the origin of these materials, experimental signatures, and their relation to specific PuO₂ and Pu₂O₃ production and aging routes. This information is of potential use in nuclear fuels processing and production, nuclear non-proliferation, forensics analyses, and interplanetary power sources.

**Environmental**

**Radiological source localisation using multiple unmanned aerial vehicles (UAVs) and survey heights**

Thomas Scott, Dean Connor, Peter Martin, Sevda Goren, Yannick Verbelen, and Kieran Wood
University of Bristol, UK

**Keywords:** UAV, Radiation Mapping, Disaster Response, source localisation, Gamma-spectrometry, Radionuclides

Following the release of a significant amount of radiological material into the environment, or for locating isolated ‘interest’ sources, quantifying the spatial distribution of the radioactivity and its isotopic composition, is one of the most important first steps for responding authorities. At present, first assessments of radiation distribution are conducted via manned aircraft (aeroplanes or helicopters) operating at altitudes of approximately 100 m – 200 m (Pitkin and Duval, 1980; MEXT, 2011; Sanada and Torii, 2014). While this method has been used effectively following both Chernobyl (1986) and Fukushima (2011), there are limitations associated with its implementation for smaller incidents, e.g. radiological dispersion devices. Unmanned aerial vehicles have the potential to replace manned aircraft within first response radiation surveying. The use of these platforms is not only considerably less expensive and more readily available at short notice but present additional advantages. UAVs are also able to operate at much lower survey altitudes than the equivalent manned aircraft system, which increases the spatial resolution of the survey (Connor, Martin, and Scott 2016; Sanada and Torii 2014; Sato et al. 2018). This makes them potentially much better at identifying isolated sources of radioactivity.

A dual UAV approach (combining fixed-wing and multi-rotor UAV surveys) was conducted over the Red Forest area of Ukraine, which received radioactive contamination from the Chernobyl incident. Initially, a fixed-wing UAV mapping system surveyed at 50 m, and then 40 m altitude, over an approximate area of 4 km², with a smaller area mapped at 20 m and 10 m altitude conducted by a multi-rotor UAV. By combining multiple...
flights at different height, the interpolation of on-the-ground radiation is improved versus single in-plane survey flights at a single altitude.

**Environmental nuclear forensics applied to Fukushima fallout: From bulk environmental samples to individual particles**

David Richards¹, James Dunne¹, Peter Martin¹, Thomas Scott¹, Yukihiko Satou², and Yosuke Yamashiki³

¹University of Bristol, UK ²Japanese Atomic Energy Agency, Japan ³Kyoto University, Japan

**Keywords:** Plutonium, Caesium Isotopes, Fukushima,

To explore the fingerprint of contamination in fallout in areas affected by the Fukushima nuclear accident, we have developed new mass-spectrometric methods (TIMS and MC-ICPMS) for the determination of isotopic ratios at ultra-trace levels of radiocaesium and plutonium in soil, sediment and plant material (Dunne et al. Sci. Rep, 8, 16799, 2018)

Since the accident, a great deal of effort has been expended on a comprehensive survey of gamma-emitters at a wide range of spatial scales, but the analysis of additional, longer-lived isotopes adds much more forensic detail. The terrestrial environment affected by the accident provides a unique place to put new methods for separation of isotopes from soils and vegetation samples to the test and assess whether a distinctive signal for the 2011 events could be separated from the widespread background signal from atomic weapons testing in the 1960s.

This is challenging work because of the very low levels of radioactivity, but it provides useful information about the distribution of material in the areas that were evacuated. We observe a wide range of values and no consistent pattern, but we have identified a Fukushima nuclear accident signal. To take this one step further, we need to identify the form of such material, consider the partitioning of signal within environmental samples and the significance of specific individual particles. We will focus on very high-resolution techniques that can characterise material at the nanoscale. To facilitate this effort, we have teamed up with researchers at JAEA and elsewhere in Japan, to work on high activity individual particles from the contaminated region. We intend to use mass-spectrometric methods to identify $^{239}$U, elevated $^{235}$U/$^{238}$U and Pu isotopes in such material. In addition, we intend to compile a database of particles to share information more widely and promote community engagement.

**Data Analysis & Statistics**

**Pitfalls of data quality and how to avoid leaping into them**

Peter Hiller and Nick Smith

National Nuclear Laboratory, UK

**Keywords:** Data, Quality, Interpretation

The interpretation of data and any subsequent decisions made are completely dependent on the quality and robustness of the underpinning data set. Assuring the quality of data and ensuring all necessary data is collected in any scenario is key to avoiding delays in interpretation and decision making or worse, making a wrong decision or incorrect determination.

This paper considers how the quality of data collected can be assured (both in the field and laboratory environments), common pitfalls which may occur ultimately challenging data quality and how these pitfalls can be avoided.
Uncertainty and Confidence in Nuclear Forensics: theory and practice
Matt Stapleton, Matthew Gilbert, and Roy Awbery
AWE, UK

Keywords: Statistics; Uncertainty; Confidence; Assessment

A key aspect of nuclear forensics is the assessment of the provenance and intended authorised use of nuclear materials found outside of regulatory control. Much of the assessment is based on materials data, where properties such as the isotopic or impurity content of the material are measured and compared to data from other sources, such as historical records, process models, or measurements of other samples of material. In reality of course, measurements are noisy, historical records can be incomplete or biased and processes may not be fully understood. Such difficulties mean that assessments carry an inherent level of uncertainty which needs to be accessed and communicated effectively to law enforcement and other decision-makers. Assessing this uncertainty and communicating what confidence we have in the assessment is therefore a crucial component to a nuclear forensics investigation.

Probability and statistics provide a theoretical framework for assessing uncertainty and this will be presented, with a focus on how specific challenges in nuclear forensics dictate the statistical formulation of the problem. For instance, a rigorous assessment of the probability that material has a particular origin requires detailed knowledge of all potential sources of nuclear material in the world, which is often unachievable. This necessitates an alternative approach based on forming a smaller set of hypotheses about the sample which can be ‘ruled out’ in turn.

The developed framework is demonstrated through two examples: the first being how it has been used in recent exercises carried out at AWE, the second in the development of a statistical methodology for assessing the origin of spent nuclear fuel based on plutonium isotopic measurements.

Don’t Believe Everything You Think: Investigating Forensic Signatures Materials from Early in the Uranium Fuel Cycle
Naomi Marks, Rachel Lindvall, Martin Robel, Lars Borg and Enrica Balboni
Lawrence Livermore National Laboratory, USA

Keywords: UOC; Yellowcake; Signatures; Trace Elements

Uranium ore is mined and milled to produce uranium ore concentrate (UOC, also referred to as yellowcake) prior to the conversion to uranium hexafluoride (UF₆) for enrichment. The high uranium concentration (>60%) and relative ease of transport of UOC, has made it a commonly traded commodity on the worldwide market (Kristo and Tumey, 2013). As a result, UOC is an attractive material for diversion from civilian power purposes to possible weapons production, and the presence of UOC can be a leading indicator of proliferation. Modern UOCs are comprised of approximately 60-80% U₃O₈, and can be primarily composed of any of the chemical compounds including ammonium diuranate (ADU), sodium diuranate, uranyl hydroxide, uranyl peroxide, or U₃O₈. Various trace element and organic impurities are associated with UOCs. The high concentration of elemental impurities in UOC has triggered a number of studies aimed at defining impurity signatures. These signatures could potentially be applied to tracing UOC to their mine or mill of origin. In this talk I will discuss recent work conducted at LLNL to understand the behavior of isotopic and elemental impurities during U processing and implications for their use as forensic signatures.
Radiochemistry

Application of plutonium age dating to samples of known provenance
Matthew Higginson
AWE, UK

Keywords: Age Dating; Radiochemistry; Mass Spectrometry; Nuclear Forensics

The Actinides Analysis group at AWE specialises in the bulk chemical analysis of actinide materials, with a particular emphasis on plutonium and uranium metals. We have developed a bulk Pu age dating capability using validated analytical techniques (TIMS, HRGS, Potentiometric Assay) typically used for the quality assured analysis of plutonium metal. We use this expertise to serve as the UKs bulk nuclear forensic laboratory. This work presents the application of this capability to a variety of plutonium samples from various processing routes (reprocessing/pyro-processing) to research the utility of the signature for forensics. We study the 241Pu-241Am-237Np and Pu-U chronometers in detail and use concordant and non-concordant chronometers to predict processing histories.

Variable geometry: The art of measuring the unknown
Alanna Downing
AWE, UK

Keywords: Gamma Spectrometry; Variable Geometry; Nuclear Forensics; UKAS

What would happen if a radioactive sample was found at an airport? Nuclear forensics aims to accurately and safely analyse radiological samples outside of regulatory control to help law enforcement by using analytical methods such as gamma spectrometry. However, gamma spectrometry has a downside: for quantitative analysis, the sample must adopt a strict set of geometries to gain meaningful results. Within AWE, this limits analysis to a 5 mL vial, a 250 mL bottle or a coin sized disc due to these efficiencies being characterised.

My research aimed to change this, making the geometries more flexible by using mathematically modelled efficiencies. These two main areas were focused on by looking at the quantity of liquid within the source holders and the distance between the detector and radiological sample. Focussing on these areas was necessary to allow greater flexibility within the sample preparation stage and greater distance between the detector and source. The latter ensures that if (for example) a sample is highly radioactive, the increased distance results in decreased dead time, the time in which no quantitative data is collected. Research in this area has been successful, leading to an accredited United Kingdom Accreditation Service (UKAS) process at AWE.

Rapid isolation of fission products from a UO₂-salt target
Jacquelyn Dorhout¹, Marianne Wilkerson¹, and Kenneth Czerwinski²
¹Los Alamos National Laboratory, USA ²University of Nevada Las Vegas, USA

The ability to compare fission products from a radiological event to a known library is imperative for post-detonation nuclear forensics. Current methods for the creation of these fission product libraries include dissolution of an irradiated actinide target, removal of any unreacted actinide, and subsequent separation of any fission products of interest. A new isolation method that does not require the complete dissolution of the target will decrease the time, cost, and associated liquid radioactive waste required to remove the fission
products. Uranium samples were irradiated, and gamma spectroscopy was used to determine extraction yields. The results from the irradiation and isolation of fission products from a novel uranium-based target material will be discussed.

**Determination of weight distribution ratios of protactinium and neptunium on TrisKem ZR (hydroxamate) resin**

Jack Mullen, and Chris Gilligan

AWE, UK

**Keywords:** Neptunium, ZR Resin, separation techniques

Neptunium (Np) is produced in a nuclear reactor that uses natural uranium. Low energy neutrons convert $^{238}$U to $^{239}$U and Beta decay produces $^{239}$Np.

Zirconium (Zr) and Np can be separated through anion exchange or extraction chromatography if the oxidation state of Np is controlled. Fe$^{2+}$ is used to reduce neptunium to Np$^{4+}$; the sample is then loaded onto an anion exchange column under nitric conditions to separate the Zr. However, this method leaves a significant amount of niobium 95 ($^{95}$Nb) and $^{95}$Zr still mixed in the Np fraction.

A $^{237}$Np tracer is used, this decays to protactinium 233 ($^{233}$Pa) via alpha decay. $^{233}$Pa has a gamma emission at 86KeV, this is the same energy as the main emission band for the $^{231}$Np tracer. Despite the long $^{237}$Np half life of 2.17E+6 years, the gamma activity from $^{233}$Pa is enough to cause significant interference to the measurement of the tracer.

ZR Resin is being used to try to improve the separation of $^{237}$Np and $^{233}$Pa. Zr, Nb and Pa are strongly adsorbed under nitric and hydrochloric conditions, while Np is weakly adsorbed. The weight distribution ratios of Pa and Np were determined, and a rapid separation method was developed for neptunium from a mixed fission product sample with increased decontamination factor compared to traditional methods using anion exchange or extraction chromatography.

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**Improved nuclear data from synthetic fallout production for post-detonation forensics exercise**

Brian Bucher, and Mathew Snow

Idaho National Laboratory, USA

Realistic synthetic fallout debris is critical for testing and assessing the reliabilities of new technologies relevant to post-detonation nuclear forensics. The most useful test debris must incorporate as many fission product radioisotopes as possible through a process that is easily tunable to simulate varying degrees of chemical fractionation. In recent years, the forensics group at Idaho National Lab has made significant strides towards this goal, utilizing a photonuclear production mechanism that can be tuned to produce a range of fission products along with development of rapid chemistry for blending short-lived isotopes in varying amounts into a realistic glass-like matrix. Additionally, as production and chemical processing of fission product isotopes continue to improve, possibilities to fill gaps in nuclear data become increasingly likely. A recent example of this has been the revision of decay gamma-ray energies in the fission product Sb-127 ($T_{1/2}=3.85$ days), where two of the three most intense lines were found to have near-1 keV deviations from the values listed in nuclear databases [1]. This talk aims to provide an overview of current research efforts at INL and discuss prospects for further improvements on nuclear data relevant to forensics.
Nuclear Forensics: Why should I want to be involved?

Paul Thompson
AWE, UK

Keywords: Challenge; Future Career

The presentation will explain via examples from the history of nuclear forensics how challenging the problems to be solved can be, and how a career in the subject can be exciting, challenging and rewarding.

Keynote: Nuclear Forensics and International Engagement – The NSDD Perspective

Mansie Iyer
IB3 Global Solutions - Contractor to the U.S. Department of Energy, USA

The subject of nuclear forensics is a priority within the U.S. Government and a focus of international engagement, e.g., Nuclear Security Summit, Poneman-Kiriyenko Nuclear Energy and Security Working Group, Global Initiative to Combat Nuclear Terrorism and the Joint U.S.-Japan Statement toward a World without Nuclear Weapons. Nuclear forensics capacity serves to strengthen nuclear security and material accounting measures, and contributes to the global mission of combating illicit trafficking by supporting investigations of incidents involving nuclear and other radioactive materials found outside of regulatory control. This is a field where the worlds of policy, technology and law enforcement collide. One needs to deal with technical issues relating to nuclear forensics, managing nuclear activities, interacting with the interagency and in the international nuclear forensics community.

Defense Nuclear Nonproliferation’s (DNN) Office of Nuclear Smuggling Detection and Deterrence (NSDD) in the U.S. Department of Energy/National Nuclear Security Administration (DOE/NNSA) is working to achieve nuclear security goals in which states’ appropriate authorities are able to use technical nuclear forensics to prevent, identify, and deter nuclear security breaches. My presentation will discuss how NSDD works with international capacity building in nuclear forensics, challenges and limitations, attracting and sustaining the skill base and career pathways and in strengthening global technical nuclear forensics capacity and best practices as a potential deterrent to prospective proliferators.

Materials & Processing

On using the natural uranium isotopic signature in ores for nuclear forensic investigation

Anya C Keatley1, James A Dunne2, Roy Awbery2, Morten B Andersen3, Tom B Scott1, and David A Richards1
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Keywords: Uranium Ore; High Precision Uranium Isotopic Analysis; Nuclear Forensics

Recent nuclear forensic investigations have identified 234U/238U and 235U/238U variation as a potential key signature for provenance determination of materials from the early stages of the nuclear fuel cycle (Keegan et al. 2014), in particular, uranium ore concentrates (UOCs). Previous studies have shown that these
variations are a function of the geological setting of the uranium ore deposit (Brennecka et al. 2010). However, relatively little work has been performed on within-region and within-mine variability of different deposit types and isotopic fractionation caused during conversion of the uranium ore to UOC.

Here, high-precision U isotopic analysis has been applied to 42 vein type U ores collected from mines located across southwest England and central Portugal. Within-mine U isotope heterogeneity has been assessed by collecting multiple samples along the same vein. To simulate the early stages of processing, we conducted a simple batch leach experiment on a variety of ores using dilute sulphuric acid and undertook U-isotopic analysis on the extractions and residues.

For total digests of sub-samples collected from the same mineral vein, we observe a range in $\delta^{238}$U values of 0.190 ‰ and $\delta^{234}$U values of 63.1 ‰ (typical 2σ uncertainties of ±0.01 and ±0.5 ‰, respectively) indicating highly localised within mine heterogeneity. The effects of acid leaching caused $\delta^{234}$U to change by as much as 62.5 ‰.


Chemical and morphological analysis of Ammonium Diuranate compounds

Jacquelyn Dorhout, Gregory Wagner, Christine Anderson-Cook, Amy Ross, Brian Scott and Marianne Wilkerson
Los Alamos National Laboratory, USA

Ammonium diuranate (ADU) is a common precursor to UO₂ in nuclear fuel production[1], but the chemical and morphological characteristics of it are largely under-studied because of its complicated chemical composition[2]. We synthesized a series of ADU compounds using various temperatures, pH values, reaction stir rates, reagent addition rates, and uranium concentrations in order to identify physical or chemical changes related to the synthetic method. A variety of techniques, including powder X-Ray diffraction (pXRD), scanning electron microscope (SEM) imaging, thermogravimetric analysis (TGA), Fourier-transform infrared (FTIR) spectroscopy, and image analysis using the Morphological Analysis for Materials Attribution (MAMA) software were used for characterization. The differences between the ADU compounds and their potential correlations to synthetic methods will be discussed.


Effects of aqueous processing conditions and thermal decomposition on forensic signatures of Uranyl Oxalate

Nathan Thompson¹, Neil Hyatt¹, and Matthew Gilbert²
¹The University of Sheffield, UK ²AWE, UK

Keywords: Signatures; Uranyl; Oxalate; Aqueous; Conditions

At the back-end of the nuclear fuel cycle, uranyl nitrate solution may be converted to uranium oxide powder for eventual interim storage; this can be achieved via precipitation of an intermediate compound. Various aqueous processing parameters, such as concentration of reagents and reagent addition order, may affect the physical and chemical properties (signatures) of the product, thus, serving as a fingerprint of their provenance.
In the experiments presented, we have precipitated the uranyl oxalate intermediate ($\text{UO}_2\text{C}_2\text{O}_4$) from uranyl nitrate solution, to finally produce triuranium oxide ($\text{U}_3\text{O}_8$) by thermal decomposition of the oxalate. Both of these powders have been examined for their forensic signatures by X-ray diffraction (XRD) and scanning electron microscopy (SEM), to examine how aqueous processing parameters and thermal decomposition affect the crystal phase and morphology of the products.

**Hydration of UO$_3$ materials following storage under controlled conditions of temperature and relative humidity**

Marianne Wilkerson$^1$, Sarah Hernandez$^1$, W Tyler Mullen$^1$, Andrew T Nelson$^2$, Alison L Pugmire$^1$, and Brian L Scott$^1$

$^1$Los Alamos National Laboratory, USA, $^2$Oak Ridge National Laboratory, USA

**Keywords:** uranium, speciation, oxide

Chemical speciation and morphology of actinide oxide materials may provide information important for understanding provenance and history of unknown samples.$^{1,2}$ We have conducted controlled experiments to measure the phase and chemical speciation of $\text{U}_3\text{O}_8$ materials prepared in our laboratories and then stored the materials under controlled conditions of temperature and relative humidity in order to correlate any changes in chemical speciation.$^3$ We have extended these studies to identify chemical signatures in UO$_3$ materials using tools commonly employed to measure chemical structure, such as powder X-ray diffraction analysis and X-ray Absorption Spectroscopy. Thermogravimetric analysis and development of models has facilitated our understanding of difference observed in the chemical changes of UO$_3$ materials as a function of temperature and relative humidity. We will discuss our studies to characterize morphology and chemical speciation of a set of uranium trioxide materials, and compare these results with analyses of $\text{U}_3\text{O}_8$ materials.


**Modelling & Simulation**

**Water layers on Actinide Oxide surfaces**

Bengt Tegner

Heriot-Watt University, UK

**Keywords:** Spent Fuel; Surface Chemistry; Storage

My contribution is concerned with the medium term storage of the UK’s civilian stocks of plutonium. Presently, this is stored as PuO$_2$ powder in stainless steel containers at Sellafield, while the government decides its long-term fate. Inside some of these containers, gas has been found to be accumulating. Several different mechanisms have been proposed for this gas build-up, including (i) steam produced by H$_2$O desorption from hygroscopic PuO$_2$ due to self-heating (ii) radiolysis of adsorbed water and (iii) generation of H$_2$ by chemical reaction of PuO$_2$ with H$_2$O, producing a postulated PuO$_{2+x}$ phase. These hypotheses, along with a desire to better understand the chemistry of these systems, have lead me to investigate water interactions with actinide oxide surfaces using plane-wave density functional theory at the PBE+$U$/level. The initial focus of this work was the geometries and energetics of up to a single monolayer of water on the
stoichiometric \{111\}, \{110\} and \{100\} surfaces. This study has now been published \cite{1} and I have since been studying sub-stoichiometric surfaces (oxygen vacancy formation geometries and energies) and water adsorption on them \cite{2}, as well as multiple water layers. Initial results from the multiple water layer study \cite{3} suggest that the second layer is quite strongly bonded to the first, and that with additional layers, the average binding energy rapidly approaches a constant value of about 0.54 eV/water molecule independent of the surface or mode of binding of the first two monolayers.

\cite{3} Bengt E. Tegner and Nikolas Kaltsoyannis, J. Vac. Sci. Technol. A, 2018, 36, 041402, DOI: 10.1116/1.5028210

**Spent fuel modelling for Actinides and fission products in pebble bed high temperature reactors**

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Tsinghua University, China

**Keywords:** pebble bed high temperature reactor; gamma spectroscopy; depletion calculation

For the material accounting of the pebble bed high temperature reactors (PB-HTRs), the most important issue is to determine the amounts of the key material isotopes such as the isotopes of uranium and plutonium from the fuel burnup values obtained via calculations or measurements. The burnup values of spent fuels can be measured by gamma spectroscopy upon the long-lived fission product $^{137}$Cs. In this work, the spent fuel nuclide production and loss are modelled, in which burnup calculations based on the netronics analysis upon the HTR-PM, the demonstration plant to be deployed in Shandong Province, China, are implemented by using the reactor system code package VSOP and the depletion code KORIGEN. A Monte Carlo method is presented to analyze the stochastic behavior of the depletion process of multi-pass fuel recycling, which sampled the radial position of pebble entering the core top randomly and calculated the burnup increment for this pass based on the prior knowledge of the neutron flux and power distribution of the equilibrium core. Correlations between burnup values, fission product amounts (mainly $^{137}$Cs) and actinide amounts in the spent fuels are concluded according to the calculation results. The stochastic behaviors of uranium isotopes and plutonium isotopes are analyzed and the correlation of fission product amounts and actinide amounts is discussed. Thus, the amounts of most concerned nuclear materials in PB-HTR’s spent fuels can be measured by gamma spectroscopy upon the long-lived fission product nuclides.

**Good Day Mr Naxos**

Paul Thompson

AWE, UK

**Keywords:** Evidence; Presentation; Court

The presentation consists of an examination by an experienced legal member of a slightly dubious scientist reporting evidence in court.
Analytical Techniques 2

Material exercises of the International Technical Working Group (ITWG) for Nuclear Forensics

Olivia Marsden¹, and Jon Schwantes²

AWE, UK, Pacific Northwest National Laboratory, USA

The Nuclear Forensics International Technical Working Group (ITWG) is an informal association of nuclear forensic practitioners, which aims to provide a framework for combatting the illicit trafficking of nuclear materials and other radioactive substances by establishing informal communications and cooperation among international experts, including policy makers, scientists, and law enforcement personnel. This effort is supported through the work of various task groups, of which five currently exist: Guidelines, Outreach and Training, Evidence and Testimony, Exercises, and Nuclear Forensic Libraries. The Exercises Task Group (ETG) conducts collaborative materials exercises (CMXs) on behalf of the ITWG for the laboratories which make up the ITWG Nuclear Forensics Laboratories (INFL).

This talk will provide an overview of the ITWG and the ETG, and will discuss the six CMXs that have taken place to date. Key analytical techniques that have been imperative to the exercises will be discussed in the context of nuclear forensics investigations.

Advances in rapid nuclear forensic preparation techniques

David Reading, Ian Croudace, and Phil Warwick

GAU-Radioanalytical

Keywords: Gamma Spectrometry; LA-ICPMS; Preparation Techniques; Rapid; Fusion; Dissolution

The rapid acquisition of accurate and precise radiometric and geochemical data is crucial in nuclear forensic studies where urgent characterisation and geolocation are required. This presentation discusses recent advances made at GAU-Radioanalytical in determining gamma and lanthanide signatures. An historic set of uranium ore concentrate (UOC) reference samples were used to validate the effectiveness of the procedure. The first case study involved the optimisation of a total sample dissolution technique to prepare samples for gamma spectrometry where no geometric assumptions or attenuation corrections are required. The second case study presents a rapid, flux-free fusion technique to produce small homogeneous glass beads suitable for LA-ICPMS measurement. These techniques could readily be applied with analysis completed within a working day from sample receipt. Sample usage can be readily controlled so valuable sample is preserved for archiving or other destructive tests.

Passive, non-intrusive assay of depleted uranium

Helen Parker, Malcolm Joyce, and Jonathan Beaumont

Lancaster University, UK

Keywords: Spontaneous fission; passive; neutron assay; depleted uranium

Uranium-238 is the most abundant, naturally-occurring isotope of uranium and makes up the majority of most uranium-based, nuclear materials. It is not fissile, and decays predominantly via α decay with some associated γ-ray emission. A small amount of spontaneous fission may also be observed, releasing neutrons at an emission rate of \(0.0136\ \text{n.s}^{-1}\ \text{g}^{-1}\). The ability to detect these neutrons, passively and non-intrusively may constitute a suitable method for nuclear forensic applications. This has been achieved with a small modular organic liquid scintillator detector array, which can detect changes in mass of \(^{238}\text{U}\) between 3.69 g and 14.46 g. Four EJ-309 detectors, coupled with a Hybrid Instruments Ltd. MFAx4.3 operated in pulse-
gradient-analysis mode, have been used to assess slices of 0.3% wt. depleted uranium pellets over 18-hour periods. Background-corrected total-neutron-counts were obtained, resulting in a fast neutron count sensitivity per detector of \((2.0 \pm 0.3) \times 10^{-4} \text{ n.g}^{-1}.\text{s}^{-1}\) for masses of depleted uranium. Measurements of \(g\) rays were taken concurrently with neutron counts. As expected, these resulted in a linear dependency with sample-mass, however, large noise interferences distorted the number of counts recorded. The reliability of these measurements is not fully understood and the method may need further research to validate it. Diurnal effects during measurements (including background measurements) were considered, as well as detector cross-talk and cosmically-induced fission. The simplicity and small volume of the depleted uranium source used here negated any investigation into self-attenuation and multiplicity.

**Progress in ultra-low-level \(^{238}\text{Pu}\) measurements by Thermal Ionization Mass Spectrometry**

Jeremy Inglis, Joel Maassen, Azim Kara, Robert Steiner, and Stephen Lamont

Los Alamos National Laboratory, USA

**Keywords:** TIMS; Plutonium; ion-counting

Modern technological developments in mass spectrometry have demonstrated both the attainability and utility of ultra-low-level (i.e. sub-pg) Plutonium (Pu) isotopic measurements, using either thermal ionization mass spectrometry (TIMS) or inductively coupled mass spectrometry (ICP-MS). However, \(^{238}\text{Pu}\) measurements at ultra-low-levels remain extremely challenging because of the problematic presence of \(^{238}\text{U}\) as an isobaric interference. Consequently, an accurate determination of the full isotopic composition of a Pu sample currently requires a time-consuming combination of alpha counting and mass spectrometry techniques. At Los Alamos National Laboratory, advances in ion-counting, sample preparation and loading techniques have been integrated to eliminate the ubiquitous problem of \(^{238}\text{U}\) interference in TIMS analysis of \(^{238}\text{Pu}\). To demonstrate the precision and accuracy that is possible, we report data from the analysis of four commonly used Pu reference materials. Samples were analyzed using a resin bead loading technique optimized for low U content and ionization. An external precision of 3.5% (RSD 2s) was achieved on aliquots approaching 10 fg of \(^{238}\text{Pu}\). In general, the accuracy of the \(^{238}\text{Pu}/^{239}\text{Pu}\) ratio in each CRM analyzed was better than 2.5% and agreed within analytical uncertainty with the declared reference values. The results of this work represent a significant advance in multiple ion counter measurements of Pu by TIMS and illustrate the potential for the complete isotopic characterization of trace amounts of Pu, without the need for alpha spectrometry. LA-UR-19-22399
Posters

P1. Is Thorium truly a “Proliferation Resistant” nuclear fuel: A nuclear forensics perspective
Erin Holland, Alexander Warren, Tomas Martin, and Tom Scott
University of Bristol, UK

Keywords: Thorium; Non-Proliferation; Energy; Forensics

Thorium is a weakly radioactive actinide element approximately as common as lead in the Earth’s crust. It can be found in readily accessible surface deposits such as monazite sands. Unlike uranium, it has a low solubility due to its redox inactivity, and as such it is commonly found in beach sands but not seawater. Thorium has been studied extensively since the very beginning of nuclear power research, but remains far less well understood than uranium.

In recent years, thorium has gained significant global attention as an alternative to uranium as a nuclear fuel. It has often been suggested in the scientific and popular press that the addition of thorium into fuels creates a more “environmentally friendly” and proliferation resistant power source than pure uranium; these views are not a consensus, with some commentators noting that there remains a proliferation risk through the protactinium pathway and the U$^{233}$ component of the spent fuel. Thorium based power forms a central part of the Indian three stage nuclear power programme due to the large natural thorium reserves along its East coast. Further, countries such as China, Canada and Norway are investing heavily in thorium as a potential future fuel source.

To date, there has only been one publicised confiscated thorium sample, however as the thorium fuel cycle becomes more widely used, there is a need to: 1) further characterise the fundamental properties of thorium (e.g. microstructural, mechanical and corrosion properties) and, 2) determine the most effective signatures to identify the origin of any thorium-based material should it be recovered from traffickers, terrorists or other bad actors.

This work will explore the factors which have the potential to generate signatures suitable for use in thorium forensics, and discuss issues influencing the proliferation resistance of thorium based nuclear fuels.

P2. The application of Isochrons to evaluate radiochronometry model age assumptions in UOC
Lauren Harrison, Amy Gaffney, Kerri C Treinan, Eva J Baransky, Ross W Williams, Rachel E Lindvall, Dana L Drew, and Victoria D Genetti
Lawrence Livermore National Laboratory, USA

Radiochronometry is the measurement of parent and daughter isotopes in nuclear materials to determine the model purification age. Radiochronometers can be used to predict a model age within two assumptions: 1) all daughter isotopes were removed upon production; 2) parent or daughter isotopes have not been gained or lost since production. Assessing the degree to which a sample and the resulting model age conform to these assumptions is key to accurately interpreting the forensic history of an unknown sample.

The isochron method uses the natural variation in daughter to parent ratios to define an isochron, the slope of which defines the age and the y-intercept the initial daughter isotopic ratio. This method may therefore test the justification of radiochronometric assumptions for a specific sample and provide a correction of the model age for initially present daughter isotopes. Previous analysis of experimentally produced U compounds showed that this method effectively corrected an anomalously old model age to the actual production age.
Here, we measure and calculate $^{230}$Th-$^{234}$U and $^{231}$Pa-$^{235}$U model ages for commercially produced UO$_4$ and ADU to assess the efficacy of the isochron method in establishing the purification age of early fuel cycle materials. The conventional $^{230}$Th/$^{234}$U and $^{231}$Pa/$^{235}$U ADU model ages are older than the production age, indicating excess initial $^{230}$Th and $^{231}$Pa, but there was not enough Th/U heterogeneity to define a robust isochron. The UO$_4$ isochron age is within error of the conventional model ages, signifying that the in-situ mining technique effectively purifies the UO$_4$ of $^{230}$Th and $^{231}$Pa, a signature not observed in the ADU. These production method signatures identified with the isochron method may be a nuclear forensic signature that should be investigated with more sample analyses. A threshold of sample heterogeneity should be documented and targeted in analytical aliquoting for successful isochron method application.

P3. Assessing High-energy, Laser-driven, X-ray Sources as a Technique to Analyse ILW Containers
Christopher Jones$^1$, Ceri Brenner$^2$, Chris Armstrong$^2$, Satya Kar$^3$, David Neely$^2$, and Tom Scott$^1$

$^1$University of Bristol, UK $^2$Central Laser Facility, UK $^3$Queen’s University, Belfast, UK

**Keywords:** X-ray; ILW; Laser; Uranium

The principle concern regarding intermediate level waste (ILW) containers is the internal volumetric expansion due to the formation of corrosion products. The development of a new technique to identify problematic containers was initiated by radiographic imaging of simulated nuclear waste packages. This was achieved using a 100TW laser pulse to produce a bright burst of high-energy bremsstrahlung radiation for imaging.

The small samples were corroded in oxygen and hydrogen rich environments to simulate long-term storage. These were characterised using electron and ion beam techniques to establish the extent of corrosion and sample morphology. High-resolution x-ray tomography (XRT) was also performed to identify the corrosion state of the material and the effect on the surrounding grout matrix. These data sets were used to ascertain the imaging quality and potential capability of a laser driven inspection system (Figure 1).

A 400mm waste drum was configured inline with a 6mm thick, 50mm diameter, depleted uranium sample and resolved to better than 1mm lateral resolution from a single shot acquisition (Figure 2). Thus, with increased acquisitions and/or higher energy x-ray production, x-ray imaging through full sized ILW containers is proposed.

The study established the non-destructive imaging capability of a high-energy laser driven x-ray source for identifying corrosion products on uranium samples, containment cracking, and for penetrative imaging through large thicknesses of grout. In addition, generation of laser-driven neutrons of thermal energy required for active interrogation of uranium isotopes was demonstrated experimentally thus paving the way for criticality testing of ILW containers.
Figure 1: a) Vulcan laser generated x-ray image of the pre-grouted uranium, corroded by 50%, inducing crack formation. b) XRT radiograph displaying the similar features with 30μm lateral resolution.

Figure 2: Detection achieved through a 400mm barrel in comparison to the dimensions of the full scale, 500 litre, ILW drums.

P4. A compact laser ion source for in situ mass spectrometry for rapid detection of nuclear materials in environmental samples
Ankur Chaudhuri
Canadian Nuclear Laboratories, Canada

Mass spectrometry, a key destructive analysis technique to support nuclear forensics and emergency response, requires extensive sample preparation procedures for solid samples. This is a major obstacle to using a field-portable mass spectrometer for in-situ analysis of solid environmental samples. The sample preparation method typically involves several dissolution and purification steps, performed by trained chemists with specialized supplies, before introduction of the sample into a mass spectrometer for ionization.
and mass analysis. This is further complicated by logistical challenges and significant costs arising from associated waste generation and disposal issues.

A portable laser ion source is proposed, where a focused short-pulsed, high peak-power laser beam is used for both ablation and ionization of a solid sample, to mitigate these in-situ challenges. This laser ionization scheme can be readily mated to Time-of-Flight (TOF) mass spectrometers. A number of factors contribute to multi-passage TOF mass spectrometers as a strong choice for field-portable high-resolution mass spectrometry: advances in TOF mass spectrometer technology have enabled increases in mass resolution while maintaining a relatively compact size of the mass analyzer [1] [2], and TOF mass spectrometer systems are commercially available. Thus, a compact laser ion source coupled with a TOF mass spectrometer can serve well in applications for in-situ nuclear forensics and emergency response. The state of the art of the multi-passage TOF mass spectrometry technique, along with the on-going development of a laser ion source for a field-portable TOF mass spectrometer, will be presented.


P5. Employing a fractional factorial matrix to study the effects of solution processing parameters on morphology of studtite and UO3

Nathan Thompson1, Neil Hyatt1, and Matt Gilbert2
1University of Sheffield, UK 2AWE, UK

Keywords: Studtite; UO3; morphology; matrix

The aim of this project is to investigate how the morphology of uranyl peroxide (UO4·4H2O) powder produced from aqueous reprocessing is affected by the processing route, conditions and impurities present. This will be used to develop a library of characteristic forensic signatures to aid in the discrimination of material from different sources. Variable conditions in this work include reagent concentrations and order of reagent addition (strike order) to produce different agglomerate morphologies.

To date, work in this project has specifically helped elucidate the difference in agglomerate powder morphology between samples of forward and reverse strike addition of reagents. Concentration of reagents may also contribute to this effect. The retention of morphology from precursor to thermal products (uranium trioxide, UO3) has also been observed in this project.

P6. Thorium procurement, policy and disposal

Sam Cross
AWE, UK

Keywords: Thorium; History; United Kingdom

Thorium (232Th) is a naturally occurring fertile material that may be used to produce fissile uranium (233U) via neutron absorption. Making thorium a plausible substitute to natural uranium for nuclear power generation. Since WWII many countries have attempted developing a thorium program. In the UK, interest in thorium peaked in the 1950s, primarily due to worries about access to sufficient uranium supplies. Much of the information relating to the UK thorium programme is spread across several hundred publicly available documents stored in the National Archives, Kew. To capture these unique operations, the records have been
reviewed and consolidated into a format that may be drawn upon to inform provenance assessment during a nuclear forensic investigation. Sources of UK thorium, subsequent processing, distribution and disposal are all discussed in detail. To add context to the UK thorium production program international thorium activity is also visited.

P7. Rapid Radiochemical separations of Americium for nuclear forensics

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1University of Manchester, UK 2AWE, UK

**Keywords:** Radiochemistry, Radiochemical Separations.

This project is sponsored by the Atomic Weapons Establishment (AWE).

AWE is central to the to the UK’s nuclear security programmes and as such is responsible for UK nuclear forensic (NF) capabilities. The UK must maintain, and indeed develop its NF capabilities in order to fulfil its obligation to support international investigations and treaties such as the Nuclear Non-Proliferation Treaty and the Comprehensive Nuclear Test-Ban Treaty[1,2].

In order to fulfil its NF commitments, AWE constantly pursues innovative techniques in the area of radiochemical separations, which are required for the purification of radioisotopes.

Given the physical and chemical similarity of many of the isotopes considered typical of a NF matrix, including but not limited to the fission and activation products of the major actinides, their separation can often be a protracted and laborious process.

Fast separations and subsequent quantification of materials (results in 24-48 hours) are essential in the context of NF given the constraints imposed by legal frame-works, emergency situations and short-lived isotopes.

The separation and quantification of americium in particular is currently a rate-limiting step in the process of obtaining a full data-set on NF samples.

This work describes the development of an extraction chromatography resin based on the use of soft heterocyclic N-donor ligands, particularly CyMe4BTPhen (BTPhen), which have been shown to be efficacious in the rapid separation of americium from lanthanides and actinides[3].

![Figure 1: CyMe4BTPhen Ligand.](image)

P8. Deformation and Strain Concentration in Cast Uranium

Phil Earp¹, John Askew², and James Marrow¹

¹University of Oxford, UK ²AWE, UK

Keywords: Uranium; Crystal Plasticity Finite Element Modelling; Mechanical Properties; Neutron Diffraction

Uranium metal is highly elastically anisotropic, due to its orthorhombic crystal structure, and each grain has a limited number of slip modes; in particular, there are no active slip modes at room temperature to accommodate deformation perpendicular to the (001) plane. Consequently, it deforms extensively by twinning, with (130) and (172) twins commonly observed. Incompatibilities between deforming grains can result in stress concentrations that may initiate fracture. To fully understand local processes such as fracture, there is a desire to develop crystal plasticity finite element (CPFE) models that can be used to predict the strength of engineering components whilst incorporating the internal microstructure of the material. Uranium presents unique challenges to the CPFE modelling community, specifically how to incorporate the effects of twins into a dislocation-based model, and how to experimentally validate these models in coarse-grained microstructures.

Experiments at the University of Oxford, in collaboration with AWE plc, aim to characterise the deformation response of cast uranium across multiple length scales. In this paper, we present a neutron diffraction experiment on this coarse-grained material, the results from which will contribute to the development of crystal plasticity models for orthorhombic metals. In an experiment at the ENGIN-X beamline, ISIS Neutron Source, UK, the development of lattice strain in cast uranium specimens was measured during tensile and compressive deformation up to a total strain of 0.7%.

The lattice strain on the {200} planes saturated with increasing stress, indicating the onset of plastic deformation. Plasticity initiates at lower stress in the coarse-grained material than in a previous study of fine-grained material in the literature. This is attributed to a combination of larger thermal residual stresses in the coarse-grained material and the Hall-Petch effect making twinning easier in large grains. In compression, however, the lattice strain on the {020} planes saturates. The asymmetry between the tensile and compressive response of the material shows that twinning is the dominant plastic deformation mechanism at low strains in the cast material at room temperature. Axial texture changes for the cast uranium were calculated by post processing of the full diffraction spectra. This analysis shows that lattice rotations associated with twinning occurred at the yield point. Interestingly, this lattice rotation disappears after unloading, which indicates that de-twinning occurs in uranium. This emphasises the importance of in-situ studies of deformation and fracture, as post-test analysis may not observe the twins that are responsible for fracture initiation.

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P9. Using a high-performance ICP-QQQ in MS/MS mode for challenging nuclear applications

Raimund Wahlen
Agilent, UK

**Keywords:** ICP-QQQ, radionuclides, interference removal

Thanks to its high sensitivity and capability to differentiate the isotopes of different elements, ICP-MS has become a powerful and widely used tool in the nuclear industry and for radioactivity studies. However, some analytical challenges cannot be solved by Single Quadrupole ICP-MS even High Resolution ICP-MS systems.

Spectral interferences like isobaric overlaps, doubly-charged and polyatomic species and spectral overlaps from peaks at adjacent masses can now be resolved using ICP-QQQ-MS instruments. Controlled reaction chemistry in combination with MS/MS mode can now significantly reduce the impact of all types of spectral interferences in ICP-MS. In addition MS/MS operation provides excellent abundance sensitivity, which allows detection of trace isotopes at m/z ratios adjacent to major matrix peaks - provided the instrument configuration includes two high-performance quadrupole mass spectrometers operating at 1 amu mass resolution.

The poster will highlight the benefits of ICP-QQQ for nuclear applications using uranium isotopic ratios measured free of UH+ interferences using oxygen reaction mode with MS/MS mode. Radionuclides such as Neptunium and Plutonium can also be measured in a uranium matrix whilst avoiding the peak tailing from the matrix.

The perfect control over the masses entering and exiting the cell enables the removal of isobaric interferences that are usually handled through long and complex sample preparations. The example of the measurement of Strontium 90 free of Zirconium 90 will be used to illustrate this capability, significantly reducing the sample preparation and therefore the total analysis time for this application.