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- The Science of Harold M. Agnew

"To our knowledge, this is the first attempt to use high performance uncertainty quantification to identify key controls at a contaminated site."
Collaboration is a hallmark of actinide research and is a thread that runs through the five articles in this issue of the ARQ.

The cover story is on the new Advanced Simulation Capacity for Environmental Management (ASCEM) program of the US Department of Energy. ASCEM is a multi-laboratory effort joining actinide, environmental, and computing sciences to develop a consistent numerical model to assess and mitigate legacy contamination across all DOE sites. Visualizations shown on the cover are the first results of joint work by scientists from Lawrence Berkeley, Pacific Northwest, Savannah River, and Los Alamos national laboratories at the Savannah River Site to model the fate of 1.8 billion gallons of waste initially placed in unlined basins at the SRS F-area.

The next article opens a window on how advanced computing has been joined with actinide science in modeling existing Los Alamos nuclear facilities and processes to create training exercises that are interactive and realistic. For example, if trainees make bad decisions in the exercises, they are not allowed to move forward. The container training demonstrates which materials would stress each type of container, and again, stops a trainer choosing an unallowed combination. Testing and safety plans are embedded in the programs.

An impressive feat of international cooperation is the recounting of the packaging and repatriation of a collection of highly portable and highly active US-origin radioactive sources that were no longer in service. The sources had been stored near Mumbai and New Delhi, two of the most populous cities in India. The effort to return the materials to the US addressed security, safety, and public health concerns and was spearheaded by the Los Alamos Offsite Source Recovery team. More than seven international and national government agencies were fully engaged with industry in the effort to move and protect these potentially hazardous materials.

Since 2008, a research team at LANL has incorporated synthesis, angle-resolved photoelectron spectroscopy (ARPES), and theory to provide insight into actinide dioxide electronic structure. Los Alamos currently has the only transuranic ARPES capability in the world. As the ARPES team reports in this article, it uses high energy and momentum resolution analyzers to perform studies of uranium and transuranic compounds, with synchrotron radiation or a He lamp as excitation sources. The ARPES measurements have provided new benchmarks for evaluation of theoretical approaches to understanding actinide oxides.

ARQ, as have other Los Alamos publications, offers a tribute to Harold Agnew (1921-2014). Beginning at age 21, he joined scientists of the new Atomic Age and went on to lead and grow the Los Alamos Scientific Laboratory with a steady hand (1970-1979). His scientific contributions, including a PhD done under Enrico Fermi, have sometimes been overshadowed by his legendary management and people skills, but are the focus of this article.

Albert Migliori, Director
Gordon Javvinen, Deputy Director
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ACTINIDE RESEARCH QUARTERLY
is published by Los Alamos National Laboratory and is a publication of the Glenn T. Seaborg Institute for Transactinium Science, a part of the National Security Education Center. ARQ highlights research in actinide sciences, in such areas as process chemistry, metabolism, surface and separation sciences, actinides and molecular sciences, actinide ceramics and nuclear fuels, characterization, spectroscopy, analysis, and manufacturing technologies.

Address correspondence to:
Actinide Research Quarterly
c/o Editor
Mail Stop 8260
Los Alamos National Laboratory
Los Alamos, NM 87545

ARQ can be read online at: www.lanl.gov/arq

If you have questions, comments, suggestions, or contributions, please contact the ARQ staff at: arq@lanl.gov or (505) 667-0392
Seeing Beneath the Surface
Using the ASCEM Open Source Computer Model

Finding the most effective remediation of radioactive contamination in complex geologic systems

Nuclear weapon production during the Cold War has resulted in groundwater contaminations at many production sites in the United States. Low-level radioactive waste solutions, for example, were often disposed into unlined seepage basins with minimal or no engineered barriers. Those locations pose one of the most technically challenging and complex cleanup efforts in the world.1 The US Department of Energy (DOE) Office of Environmental Management (EM) asked the national laboratories to develop numerical tools that could accurately predict the long-term behavior of subsurface radioactive contamination plumes and the engineered materials used for waste disposal, such as glass or cement. The call was for a single-process computational framework to be used throughout the DOE complex in a consistent manner to standardize assessments for environmental cleanup.

In response, computer and environmental scientists from four DOE laboratories created the Advanced Simulation Capability for Environmental Management (ASCEM). ASCEM is a modular open source set of tools that supports standardized performance of computational testing for DOE-EM cleanup and closure and establishes a code base for a growing interdisciplinary community. Team members are from Lawrence Berkeley National Laboratory (LBNL), Pacific Northwest National Laboratory (PNNL), Savannah River National Laboratory (SRNL), and Los Alamos National Laboratory (LANL).

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This article reports on the first application of ASCEM at one challenging site, the F-Area, DOE Savannah River Site (SRS), South Carolina (Fig. 1). The SRS F-Area Seepage Basins (located in the north-central portion of SRS) consisted of three unlined, earthen surface impoundments that received ~7.1 billion liters (1.8 billion gallons) of acidic, low-level waste solutions. Groundwater in F-Area is contaminated with a number of constituents, including uranium isotopes in the relatively mobile (U(VI)) oxidation state, in a plume that extends from the basins to ~600 meters downdgradient where it discharges to a stream. Despite many years of active remediation, the groundwater still remains acidic, and the concentrations of U(VI) and other radionuclides are still significant. Monitored natural attenuation (MNA) is a desired end state for the site. In situ pH manipulation is being used in the downdgradient portion of the plume. It will continue until rainwater eventually neutralizes acidity of the upgradient, stimulating natural immobilization of uranium in the trailing end of the plume. At the pH values in the heart of the plume, 3.2-3.5, carbonate complexes of uranium are insignificant. The reason the uranium is mobile is that the surfaces of aquifer minerals and the dominant aqueous uranium complexes are positively charged at the acid pH, and thus uranium does not sorb well to the aquifer minerals.

The goal of the remediation system is in situ manipulation of the aquifer pH to enhance sorption of the contaminant uranium. Ongoing remediation includes an engineered funnel-and-gate system and periodic injection of a base compound near the gate, intended to raise the pH and immobilize the U(VI). This system is effective, but for life-cycle planning it is necessary to estimate when base injections can be terminated. Monitored natural attenuation (MNA) is a preferred final closure strategy for the site based on the premise that rainwater will eventually neutralize acidity of the groundwater plume, stimulating natural immobilization of uranium at the trailing end of the plume.

At the SRS F-Area, a first step of the flow and reactive transport simulation was a two-dimensional (2D) model developed with Akuna. (The ongoing remediation treatment was not considered; extension to 3D
and consideration of the treatment is ongoing.) The 2D model domain (Fig. 1c) consisted of a 2D vertical cross-section about 2600 m long and 100 m deep, oriented along the plume centerline and passing through the middle of the main basin. The domain includes the sloping topography and hydrostratigraphy with two sandy aquifers (upper and lower) and one clay layer between the aquifers. The variable subsurface was identified using wellbore data collected at the site over many years, as well as more recent seismic data.

Figure 2 shows the 2D evolution of predicted pH and uranium concentration over time. Although the engineering treatments have not yet been included in the model, the simulations provide a good indication of the long-term subsurface behavior. For example, the simulations show that during the waste discharge operation, the low-pH plume front advanced in front of the uranium plume and thus increased the mobility of U(VI). In the postoperational period, the pH values progressively start to rebound as the acidic plume mixes with uncontaminated groundwater, although the pH rebound is impeded by H+ desorption from minerals. The groundwater pH values remain relatively low for a prolonged time period, and the uranium plume migrates towards the creek as the uranium concentration decreases significantly. The simulations also suggest that a significant amount of contamination is left in the vadose zone (the unsaturated region just above the water table) below the basin, due to the capping of the basin, which limits infiltration of fresh water into the system. The elevated U(VI) concentration is also observed in the clay layer due to its low permeability. The simulations suggest that the vadose zone below the basin and the clay layer may act as a long-term contaminant source for the groundwater, if there is significant flow of water through the capped basin. As previously stated, the ASCEM team is improving the model to include other key aspects (such as more detailed variables and engineering treatments), with a goal to provide a ‘living model’ of the site that can be used to explore treatment scenarios and make decisions about closure strategies such as MNA.

Richards’ equation (used to determine the movement of water in unsaturated soils, named for its creator, Lorenzo A. Richards) was solved to simulate unsaturated and saturated flow in the subsurface. A new optimization capability was developed to minimize nonmonotone (increasing and decreasing) behavior due to mesh distortion, and used within the mimetic finite difference (MFD) framework. The flow model was calibrated to obtain the best fit of historical measurements of tritium (3H) concentrations within the F-Area and into the stream. To capture complex geochemical reactions of uranium with clay minerals and also pH dependency of uranium behaviors, the model included various reactions such as equilibrium aqueous complexation, kinetically controlled (different pathways and combinations of chemical produce different products) mineral dissolution and precipitation, and adsorption/desorption. In the current F-Area model, the primary geochemical system consists of 13 reactive chemical components and 8 minerals, the behaviors and parameters of which were determined by the lab experiments. A total time period of about 100 years was simulated, corresponding to a time window from 1955 to 2055.

ASCEM tools simulate plume behavior under different conditions and identify which parameters most influenced plume transport as a function of location and time. To our knowledge, this is the first attempt to use high performance uncertainty quantification (UQ) to identify key controls at a contaminated site. In particular, we evaluated how the uncertainty associated with contaminant source, geochemical reactions, and flow characteristics can influence the predictions of the long-term behavior of pH and U plumes.

Figure 2. Results of 2D simulations of the pH (left column) and uranium concentration (right column) evolution over time at the SRS F-Area.
Figure 3. Monte Carlo analysis results: breakthrough curves and their uncertainty ranges: a) pH at FSB95D, b) pH at FSB110D, c) U(VI) concentration at FSB95D, and d) U(VI) at FSB110D. The black lines are the predicted breakthrough curves, red lines are the mean predicted curves, green lines are the mean ±2 standard deviations, and the magenta dots are observations.

Although some curves predict that the pH could exceed the target value of 5 by the year 2055, the majority of curves predict a slower pH rebound. Figure 3c and 3d show that the breakthrough curves of U(VI) concentrations are the reverse of those for pH. Similar to pH, the plateau of U(VI) concentrations can be seen during the basin operation. After the basin closure, uranium concentrations decrease significantly, although the mean curve does not drop below the maximum contaminant level (MCL) of 1.3E-7mol/L.

Global sensitivity analysis identifies the key controls and the most important parameters for accurate predictions. Such parameter importance ranking will be useful to plan and prioritize the monitoring efforts. Figure 4 illustrates complex patterns of the impact of different parameters on pH and U(VI) concentrations during and following the basin closure. The parameters that most governed system behavior varied as a function of time as well as distance relative to the source and time. Although the geochemical parameters and source concentrations are dominant over the simulated time frame, the hydrological parameters become important in the later time. Table 1 provides a summary of the most influential parameters under both historical (1985) and predicted future (2055) conditions, including aquifer permeability, geochemical and source parameters. For pH, the CEC and source pH have a large impact at both locations during the operation, whereas after the basin closure, the permeability values become important. For the U(VI) concentrations, the sorption site density and the U(VI) source concentration are the dominant parameters during the basin operation, and the permeability of the clay layer becomes important after basin closure. This kind of information is useful for guiding the design of long-term monitoring, sampling, and remediation strategies needed for decision making at the site.

Figure 4. Global sensitivity analysis results using Agni-Amanzi: time profile of sensitivity: a) pH at FSB95D, b) pH at FSB110D, c) U(VI) at FSB95D, and d) U(VI) at FSB110D.

**Table 1. Three most important parameters controlling pH and U plume mobility**

<table>
<thead>
<tr>
<th>Location Close to Basin</th>
<th>Remote Monitoring Well</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>1985</strong></td>
<td><strong>2055</strong></td>
</tr>
<tr>
<td>1 Cation exchange capacity</td>
<td>1 Cation exchange capacity</td>
</tr>
<tr>
<td>2 Source pH</td>
<td>2 Source pH</td>
</tr>
<tr>
<td>3 Geothite specific surface area</td>
<td>3 Geothite specific surface area</td>
</tr>
<tr>
<td>1 Sorption site density</td>
<td>1 Sorption site density</td>
</tr>
<tr>
<td>2 Source U</td>
<td>2 Source U</td>
</tr>
<tr>
<td>3 Clay layer permeability</td>
<td>3 Clay layer permeability</td>
</tr>
</tbody>
</table>

*Geothite is a widespread iron ore mineral*

The parameter symbols are:
- \( k_{ua} \): upper aquifer permeability
- \( k_{lc} \): lower aquifer permeability
- \( p_{ua} \): porosity
- \( m_{ua} \): van Genuchten \( m \_
- \( s_{dens} \): sorption site density
- \( cec \): cation exchange capacity
- \( koh_{ua} \): kaolinite specific surface area
- \( hoh_{ua} \): Geothite specific surface area
- \( se_u \): Geothite specific surface area
- \( recharge \): natural recharge rate
- \( basin_{wt} \): Basin discharge rate
- \( sc_{f} \): source \( H^+ \) concentration
- \( sc_{u} \): source U(VI) concentration
The model prediction and uncertainty quantification presented here are important to develop remediation and closure strategies for the F-Area seepage basins plume because of the insights they provide to the processes controlling pH rebound and how they affect estimates of timeframes for pH rebound. The current in situ remediation is a funnel-and-gate system with periodic pH adjustment within the gates designed to limit flux of contaminants to Fourmile Branch. The duration of its operation depends on pH rebound in the upgradient portion of the plume. Understanding the rate of rebound and the processes controlling it informs future decisions on whether pH rebound in the upgradient portions needs engineered enhancement, and if so, how best to proceed. The model is currently being improved to include more complex features such as the pH manipulation treatments, heterogeneity, and existing/planned monitoring networks. The goal is to provide a living model that can be used by SRS to evaluate the efficacy of remedial actions to assess remediation strategies and predict if and when it will be possible to transition from active to passive cleanup of contaminated groundwater using MNA. The model will also be used to guide the development of monitoring strategies and the interpretation of field monitoring datasets at the F-Area.

Further Reading
compof.2013.04.005.

Safety and Security
Safety and security are very important aspects of these facilities. For the building model, the IMMS team was able to embed each room’s criticality limitations and safety plans, thus giving users more exposure to the requirements of a safe environment and creating greater awareness for safe work habits. Workers were also able to use the visual model as a refresher for the layout of rooms and overall location awareness, which then prevented the need for extended physical stays within facilities that could have had the end result of exposing workers to high dose rates. When workers are comfortable with the layout of a facility, they become more efficient at their work. This increased efficiency leads to better work habits and less exposure time to the material within the facility for the worker.

Virtual Access
Interactive visual models can be used to access controlled, highly radioactive, and specialized facility training areas to save time and effort in preparing paperwork to gain access, as well as to overcome security...
barriers encountered within the facility. These facilities are prime targets for developing visual models because they allow sponsors and emergency response teams to become familiar with a facility without having to physically enter that facility. By using specialized polygons, pictures can replace certain elements, such as signs, to create a sense of realism. Videos and PDF documents also can be placed in specific locations of the model for documentation and training purposes, thus adding to the complexity, usefulness, and uniqueness for each set of users. Embedding relevant documents and visuals increases the experience for the user and makes the model more useful.

**Training and Testing**

The newest and fastest growing applications for the modeling tool have been in training and testing simulations. The development of a user-friendly interface has allowed the tool to reach a new set of users not familiar with modeling. The interface uses features and controls similar to Google Earth. In addition to a user-friendly interface, training procedures can be incorporated into the model and can provide users with step-by-step visual aids as they study their procedure for training. By incorporating procedures, we begin to bridge the gap between the training materials and traditional on-the-job training. For example, preprogramming a key component in a procedure eliminates confusion for users and is essential to successfully bridging that gap.

Incorporating procedures into the model develops it into more than just a visual training aid. Subject matter experts (SMEs) can use the procedure, paired with the visual component, to step through the procedure as a refresher. The model can also be created to test the users on correct steps. If users take a wrong action, the model will not allow them to move forward. This process forces users to truly interact with the procedure while seeing the desired result and ultimately learning the correct process. Supervisors can also receive notice as to when and how often SMEs interact with the model, thus giving the supervisor a greater sense of confidence in the workers’ understanding of training plans. Many procedures used for shipping containers are “use every time,” requiring the user to follow the procedure step by step every time a job is done. To eliminate confusion and enhance efficiency, models can write the use-every-time procedure for the SME in real time while users step through the procedure and fill out the necessary information.

**Glovebox Training**

The IMMS team also designed a glove-box-worker training aid (Figure 2), a virtually simulated model. Because many different techniques are associated with glove changes and bag-out operations, the model and simulations integrated only the main process steps. The virtual glove box is accessible on the Laboratory’s training site as supplemental content.

Another benefit that models provide is the ability to simulate a visual without the time constraints of using traditional engineering tools. A CAD-generated model of technical and engineering drawings displays the materials, dimensions, geometry, and processes. This option is viable when implementing a new design that has been approved and funded; however, a visual model is a fast, inexpensive, and spatially realistic opportunity to demonstrate the conceptualization of an idea. Once stakeholders have a visual representation, they can begin to discuss additional ideas, costs, and benefits of following through to the final desired result. Models can also be developed to target difference phases of a project.

For example, the storage facility is transitioning to the use of SAVY-4000 containers (Figure 3). This transition uses training models to understand the procedure for the containers. The first phase of the model is to integrate the procedure with the visual step-by-step process, thus allowing users to become familiar with how to disassemble the container following the procedure. A very important component of this procedure is to determine the integrity of the seal—a concept that easily can be demonstrated in the visual model. The next phase of the model would then allow users to identify debris...
Packaging and Repatriation of US-Origin Radioactive Sources

Projects in New Delhi and Mumbai
Offsite Source Recovery Project, Los Alamos National Laboratory

The Off-Site Source Recovery Project (OSRP) is a project at Los Alamos National Laboratory (LANL) sponsored by the US Department of Energy (DOE). It falls under the auspices of the National Nuclear Security Administration’s (NNSA) Global Threat Reduction Initiative (GTRI), and is tasked with recovering and repatriating US-origin radioactive sources around the world based on national security considerations. GTRI-OSRP has recovered more than 31,500 sources totaling over 899,000 curies (Ci) in the US; over 2,437 sources totaling 4,133 Ci have been repatriated from 20 countries. Source recovery, packaging, and repatriation efforts were conducted at commercial facilities near New Delhi and Mumbai, the most populous cities in India.

Millions of radioisotope sources are currently used for a wide variety of beneficial civil applications worldwide. These applications include blood and medical supply sterilization, oil exploration, medical treatment, scientific research, food irradiation, and many others. While the number of beneficial uses has multiplied, clear final disposition pathways for sources have decreased, or in many cases, never existed. Large numbers of sources have already or are now reaching the end of their useful working lives due to the decay of the radioactive isotope or fatigue of the encapsulating metal material or sealing welds. This combination of wide beneficial use and the lack of clear final disposition pathways resulted in large numbers of disused sources that could fall or have fallen out of regulatory control (orphan); thus posing a significant security, public health, and safety concern.

Beneficial civilian applications of sources mainly involve radioisotopes of cesium-137 (137Cs), cobalt-60 (60Co), strontium-90 (90Sr), americium-241 (241Am), iridium-192 (192Ir), plutonium-238 (238Pu), plutonium-239 (239Pu), curium-242 (242Cm), radium-226 (226Ra), and californium-252 (252Cf); all identified by the International Atomic Energy Agency (IAEA) as the 10 isotopes of concern.1 Due to their concentrated high activity and portability, any of these materials could be used to make a radiological dispersal device* (RDD), or so-called dirty bomb. The use of an RDD in a populated inner-city could result in economic impacts amounting to billions of dollars and significant social disruption.*

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1. An RDD is a device or mechanism that is intended to detonate conventional explosives laced with radioactive materials or sabotage in place. An RDD is considered a weapon of mass disruption; few deaths would occur, but the radioactive nature of the event would have the potential to cause significant short and long-term economic disruption resultant from public panic, decontamination costs, and denial of access to infrastructure and property for extended periods of time. A radiological exposure device (RED) is a device having the purpose of exposing people to radiation rather than dispersing radioactive material into the air.

* An RDD is a device or mechanism that is intended to detonate conventional explosives laced with radioactive materials or sabotage in place. An RDD is considered a weapon of mass disruption; few deaths would occur, but the radioactive nature of the event would have the potential to cause significant short and long-term economic disruption resultant from public panic, decontamination costs, and denial of access to infrastructure and property for extended periods of time. A radiological exposure device (RED) is a device having the purpose of exposing people to radiation rather than dispersing radioactive material into the air.

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Figure 4. H1700 Packaging and Shipping container screen shot of an integrated packaging and shipping procedure on the left, with each of the actions in the procedure linked to buttons that move the user forward and are represented in the split screen that shows the visual model of the disassembly of the H1700 container.

Figure 5. The H1700 Packaging and Shipping Container.
Over the past decade, terrorist organizations such as al-Qaeda have attempted to acquire radiological materials. According to the 9/11 Commission Report, published in 2004, at that time, over two dozen terrorist groups were attempting to gain such asymmetric weaponry and that number can only be assumed to have increased. The former Director General of the IAEA, Mohamed El Baradei, and current Director General Yukiya Amano have both stated that the continued high amount of theft or loss of source material continues to pose a serious threat of radiological terrorism.

In early 2012, HLS Asia Limited (HLS) made initial contact with GTRI-OSRP by registering an inventory of disused US-origin sources on the OSRP website and formally requesting assistance with their removal. HLS, formerly known as HLS India Limited, in operation since 1987, is a Public Limited Company registered in India. These sources were manufactured in the early 1980s and were consolidated temporarily for GTRI-OSRP, packaging at the HLS headquarters storage facility located in a burgeoning industrial area in Noida, India (~30 km from New Delhi). HLS uses sources for well-logging and other activities associated with the exploration of hydrocarbons. In early 2012, cooperation between GTRI-OSRP and HLS resulted in the packaging and repatriation of 23 sources totaling 61 Ci. This included Category 2 quantities of the isotope 241AmBe (amercurium-beryllium) along with small quantities of 137Cs and 226Ra.

GTRI-OSRP began verification of the origin of the source material and identifying “acceptable knowledge (AK)” that would ensure acceptance of the sources into an authorized waste stream for permanent disposal. HLS submitted a request to the Atomic Energy Regulatory Board (AERB), India’s regulatory authority, for both the export and disposal of the sources in the US. AERB officially approved the request opening up the path for the first Indian source. Source manufacturer documents and other identifying criteria confirmed that the registered sources were of US origin with sufficient AK for disposal. GTRI-OSRP and HLS began preliminary planning for a GTRI-OSRP packaging mission at HLS. The GTRI-OSRP team sent, with HLS customs import assistance, the necessary equipment to perform the recovery. The OSRP field recovery team arrived at HLS in January 2012 (Fig. 1). The GTRI-OSRP team examined each source or device for identifying markings, to ensure they matched the sources that were registered and had already met preliminary AK requirements. This was followed by OSRP and HLS working together to procure, design, and construct an inner box for the three AmBe sources, needed to provide sufficient shielding for transport. HLS provided valuable support by acquiring thick sheets/large quantities of high-density polyethylene beads and by making provision of necessary labor and heavy machinery. HLS also was instrumental in assisting with the export documentation and arrangements required for expedited shipment of the sources to the US. While at HLS, OSRP provided an overview of its project’s mission and a hands-on demonstration of the closure of a field-sealable special form capsule used to package the radioactive source.

In 2012, a major national oil company of India, the Oil and Natural Gas Corporation, Ltd. (ONGC), had ten sources from the Indian state of Agartala slated for disposal that were diverted to the ONGC logging base in Panvel for storage due to a lack of storage space. In May 2012, the ONGC sent the AERB a feasibility study for disposing of these sources in abandoned ONGC wells. Fifteen scientists from the Waste Management Department of BARC and the Radiological Safety Department of AERB, and the Board of Radiation Isotope Technology (BRIT) met to assess the proposal. Although meeting participants agreed to the ONGC proposal conceptually, other concerns such as institutional control of facilities and the structural integrity of sources at higher temperatures merited further study. Participants also advised the ONGC of the existence of GTRI-OSRP as another potential disposal option.

In July 2012, the ONGC contacted GTRI-OSRP and registered all its sources on the GTRI-OSRP website. As wireline logging and perforation activities in both HLS and ONGC are very similar, the sources were nearly identical in type. However, the ONGC possessed far greater numbers and activity of sources than did HLS. This required extensive preliminary research for source characterization and large amount of certified Type A shielded containers. Logging bases throughout India began sending GTRI-OSRP photos and leak tests of the sources. Out of 117 sources originally registered, 80 passed the GTRI-OSRP initial screening (~252 Ci, 137Cs ~16.2 Ci). Reasons for ineligibility varied from being of foreign origin (not US), fell under the Nuclear Regulatory Commission exempt quantity activity levels, or lacked identifying information (isotope or activity). Some of the ineligible sources (mostly 226Ra and 228Th) were accepted by AERB for disposal by the Waste Management Division at BARC. The AERB provided formal approval for the consolidation of the sources in Panvel from many Indian states (ONGC bases: Agartala, Ankleshwar, Ahmedabad, Mehasan, Karaikal, and Nazira) for forwarding on to the US. Senior BARC
and AERB officials visited the ONGC logging based in Panvel to verify the storage and handling facilities.

The packaging mission at ONGC took place exactly a year after the mission at HLS. As before, LANL was responsible for covering the import and export arrangements; with significant in-country support from ONGC on both shipments. The GTRI-OSRP field team arrived at the ONGC Panvel site in mid-January (Fig. 2). A formal meeting was held between GTRI-OSRP and ONGC with scientist observers from AERB, BRIT, and BARC. In addition to direct observation of the source recovery operation (Fig. 3), GTRI-OSRP provided Indian government officials hands-on demonstrations of verification and packaging methodologies employed by the project. Rather than keeping the 241Am sources in their existing shielded containers, the GTRI-OSRP and ONGC teams worked together to remove sources for direct visual verification, encapsulation in special form capsules and placement in 11 Type A containers that provided the necessary shielding (Fig. 4). Field-encapsulation of the sources in special form capsules also ensured that sources with expiring special form certificates would continue to meet special form criteria for transportation. Many sources fell in the 20 Ci range and required extended handling tools, limited exposure time, and maintaining greater physical distance from personnel. A radiological perimeter was established to ensure only essential personnel were exposed. The 137Cs sources remained in their original source holders and were quickly deemed too large to fit in the cavity of the lead insert of the Type A containers. Experts from BRIT and ONGC removed the 137Cs sources from their holders, aided by manufacture diagrams supplied by GTRI-OSRP along with device knowledge from ONGC. These sources could be compliantly packaged and shipped. The ONGC and BRIT/BARC also provided ancillary radiological survey equipment. This cooperative effort was important to GTRI-OSRP in building relationships with key commercial/government entities in India that have the ability to facilitate further future radiological cooperation.

GTRI-OSRP involvement in permanent threat reduction with commercial entities in India such as HLS and ONGC, and with assistance from BRIT demonstrates a shared commitment to the safe and secure disposal of US-origin radiological sources. As a result of this collaborative effort, spanning over a year, HLS and ONGC have agreed to inquire other private entities in India to determine whether there may be more disused US-origin sources located at other commercial sites throughout India. OSRP-GTRI would like to thank the following experts and their teams for essential support and for their efforts, and assistance from BRIT, and with recommendations to the project: BRIT: Dr. AK Kohli, Chief Executive; Mr. KVS Sastrti, Senior General Manager; Mr. Pravin Kumar, Manager, Irradiator Sources; Mr. BN Patil, Manager, RS&TL, HLS-Asia: Mr. Sanjay Gupta, HSE Officer; Mr. Anil Kumar, Head Materials. ONGC: Mr. RK Pandey, General Manager-Chief Logging Services; Mr. JJ Mohanty, Chief Geophysicist; and Mr. PC Chaturvedi, Chief Geophysicist and RSO. Endnotes


2 In January 2003, British officials discovered al-Qaeda training manuals on detonating a dirty bomb along with actual radioisotopes necessary for this at a nuclear laboratory in Herat, Afghanistan. Abu Zubaydah, a detainee at Guantanamo Bay, made statements in 2002 that al-Qaeda already had this capability. In 2005, a significant amount of cesium was stolen from an oil company in La Gloria, Colombia. The material is still missing, and al-Qaeda was suspected of conducting the operation.” Seleem Cesium Still Missing,” El Pais, November 22, 2005.


Photoelectron Spectroscopy of Transuranic Materials

Introduction

Photoelectron spectroscopy is used throughout the world in a wide range of applications to probe the chemical identity, bonding, electronic structure, and surface properties of materials. In the last two decades the spectroscopic capabilities at public synchrotrons for studying most materials has expanded significantly, but regulatory restrictions have greatly limited transuranic research at these facilities. Los Alamos National Laboratory (LANL) has a unique photoelectron spectroscopy capability that allows many of the most important capabilities of photoemission to be used on transuranic materials. The LANL photoelectron spectroscopy system is outlined in general, and the angle-resolved photoemission system (ARPES) is presented in some detail.

Background

Photoelectron spectroscopy (or photoemission) is a photon in–electron out spectroscopy. It is perhaps the most direct experimental probe to discover the electronic structure of materials, which explains the prevalence of the technique. Based on the photoelectric effect reported by Hertz in 1887, characterized by Lennard in 1902, and explained by Einstein in 1905 (for which he won the Nobel prize in Physics in 1921), the modern era of photoemission was ushered in around 1960. At that time the main focus was on fixed-photon-energy light sources both at low energies (gas discharge lamps) with energy up to 50 eV, and high energy x-ray sources ranging from 1200 eV to a few keV. Synchrotron radiation, first observed in 1947, was generally considered a nuisance as it depleted energy from stored electron beams, but a national academy study in the mid-1960s indicated that synchrotron radiation could be beneficial for research as a path towards new capabilities in spectroscopy. Because photoemission measures a relativistic low-energy electron as the output probe, it is very surface sensitive and therefore requires atomically clean surfaces and no barrier between the sample, the photon source, and the electron energy analyzer. The lack of a barrier between the sample and the photon source has basically excluded transuranic photoemission from public synchrotrons as these facilities generally cost hundreds of millions of dollars and the potential for radioactive contamination is unacceptable. There are dozens of synchrotrons around the world now running as public photon sources. The US Department of Energy (DOE) Basic Energy Sciences (BES) operates four synchrotron photon sources in the US. There is no synchrotron facility in the world that allows transuranic photoemission at this time.

In 1992, the BES funded the Laser Plasma Light Source (LPLS) project at LANL. This project was designed to provide tunable photons between 27 and 140 eV for use in transuranic photoemission research. At a storage ring facility (or now a free-electron laser (FEL) facility), such as those run by the DOE, the photons are generated by the path of relativistic electrons being bent passing through a magnetic field (either in an undulator or a bending magnet). The LPLS generates a range of photons by focusing a short pulse of laser light onto a metal target and generating a plasma, as in Figure 1. Over the years we have used many different metal targets ranging from aluminum (Al) to lead (Pb), with mercury (Hg) striking a nice balance between a high-photon flux as a source and low-maintenance upkeep as a target. Since initial funding in 1992, the LPLS has added an x-ray photoelectron spectroscopy (XPS) capability (1500, 3000 eV photons), a high-resolution gas discharge lamp (21, 23, 41, 48 eV photons), and an angle-resolved photoemission capability. In addition to the BES program, the LPLS facility is used for research funded by the Laboratory Directed Research and Development (LDRD) program, Science Campaign 2, US Department of Homeland Security (DHS) nuclear forensics and Life Extension Project (LEP) work at LANL.

Research

Several variations of photoemission are used with the LPLS system, including angle-resolved photoemission (ARPES), resonance photoemission, and the more traditional angle-integrated mode of photoemission (PES). Resonance photoemission takes advantage of tunable photon sources by selecting a specific photon energy to enhance a particular orbital character (s, p, d, or f) in a material by providing multiple initial-state excitation...
paths to the same final-state configuration. While we have previously reported results of resonance photoemission on plutonium (Pu), ARCES results are the focus of this article.

There are limiting cases for the interpretation of PES data depending on the electronic structure of the material under investigation. In a simplistic approach, the two endpoints for PES interpretations would be either an itinerant electron framework where the electrons form bands and move freely throughout the solid or a localized electron framework where the electrons are localized around one atom position such as the core levels in a solid. The properties of Pu materials are defined in large part by the 5f electrons, which are at the boundary between localized and itinerant character. In the series of actinide elements, Pu is the transition point for the 5f electrons moving from bonding character in the lighter actinides (Np and lower) to localized character in the heavier actinides (Am and higher). Moreover, this crossover point in 5f character at Pu in the actinide elements is also found in many Pu compounds. With PES and ARCES we can determine the character of the 5f electrons in any particular Pu material.

For the itinerant electron picture of a solid, PES measures the density of electronic states (DOS) as a function of energy, basically a 1D mapping of occupied states vs. energy. The experimental equivalent of DOS measured with PES is known as an energy distribution curve (EDC) and has typical experimental limitations of energy resolution, photoionization cross-section, and dipole selection rules. In ARCES, there is the added dimension of crystal momentum (k), and the mapping is now 2D, with spectral intensity as a function of energy and k. As an example of ARCES data for a uranium system that shows primarily itinerant electron character, we show valence band ARCES data for the heavy fermion superconductor URu$_2$Si$_2$, in Figure 2. This data was collected at the synchrotron radiation center using a Scienta analyzer on a high-resolution undulator beamline. It is clear from this data that the electronic structure is dominated by itinerant electron character as the full symmetry of the lattice is evident, and the large-scale (eV) electronic structure agrees well with the band structure calculation. If the electronic structure were dominated by localized states, then there would be no momentum (angle) dependence to the electronic structure and the states would be flat, horizontal lines in the ARCES data.

In the case of Pu materials, the 5f electrons can range from fully localized to rather delocalized (or itinerant) depending on the compound, alloy, or phase of Pu. We will show ARCES data for two plutonium compounds, PuO$_2$ and PuCoGa$_5$. These materials both show crystal momentum dependence in the ARCES data, indicating that the treatment of the 5f electrons in these materials requires at least some consideration of delocalized 5f character. Figure 3 is ARCES data for PuCoGa$_5$, which is a 18.5 K superconductor discovered at Los Alamos in 2002. We published the original PES data on this material in 2003 before we had an ARCES capability at the LPLS. Now, with the addition of ARCES, we can see details in the electronic structure that reveal the 5f electrons hybridizing with conduction band states near the Fermi energy (zero binding energy). By integrating the 2D energy—momentum map we recover a limited EDC, shown in the left frame, and the full 2D map in the right frame.

The EDC in Figure 3 shows two peaks, one at the Fermi energy and one at a binding energy of ~1eV. The data is taken at a photon energy of 40.8 eV, which has a large cross-section for the Pu 5f states and a somewhat smaller cross-section for the Co 3d states. These two peaks are associated with a hybridized (or itinerant) 5f character and a localized 5f character. The right side of the figure shows the variation in the 5f intensity at the Fermi energy with high (blue) intensity at the 0 and +/- 8 degree points and lower intensity (green) in between these points. Together, this data set indicates the dual nature of the 5f electron character in PuCoGa$_5$. In addition to the full valence band data on PuCoGa$_5$, we can look with greater detail at the electronic structure near the Fermi level. In Figure 4 we show ARCES data for PuCoGa$_5$ at a lower photon energy, which enables visualization of the 5f and 6d electron states. This data is over an energy interval which is an order of magnitude smaller than shown in the previous figure, and details of the Fermi surface are evident from such data. The top
The frame of Figure 4 is the energy-momentum map for PuCoGa$_5$, at an energy of 21.2 eV. In the bottom frame, are EDCs taken at a specific angle, here represented as a horizontal line taken across the 2D map, which shows a peak in the electronic structure at 8 deg that sequentially moves toward and through the Fermi energy (zero) as the angle moves from 8 to -8 deg.

The amount of information that is available from the LPLS system running in ARPES mode is large as it is composed of a significant number of 2D maps building the full 3D electronic structure of a material, where the three dimensions are crystal momentum, binding energy, and measured intensity. While PES and ARPES can provide extremely valuable information on electronic structure, the technique depends critically on high quality samples and high quality, in-situ surface preparation. Fortunately, LANL has experts in transuranic single crystal and polycrystalline sample growth. Our samples are provided by MPA-CMMS, MST-16, and MPA-11. We use primarily two methods for sample surface preparation: in-situ cleaving and laser ablation.

Figure 5 shows the laser ablation setup for cleaning samples. We currently have two lasers at the LPLS, one for generating the plasma from the Hg source for tunable photoemission (Figure 1) and the other for cleaning samples for PES and ARPES. In principle, either laser would suffice to complete either one of the tasks. The larger laser is an established YAG technology operating at 50 Hz, 950 mJ/pulse and a pulse width of 10 nanoseconds (ns) at 1064 nanometer (nm) wavelength. We currently use this laser for generating the Hg plasma for the tunable photon source. Our new laser is a picosecond (ps) class laser that operates at a full power of 160 μJ/pulse, 20 kHz, and 6 ps pulse width. The ps laser has the advantage that it can ablate material from the sample surface without significantly heating the sample as the pulse width is of the same time scale as the phonon coupling time, whereas the ns laser operating in the infrared range definitely drives the sample long enough to create substantial sample heating.

When we are cleaning actinide materials, the sample is inserted in the tube to the right to contain the ablated material in a confined region and minimize the contamination of the main measurement chamber. The laser ablation cleaning method has proven very effective for actinide materials. We focus the laser beam for both cleaning and plasma generation. When we are generating a plasma on the Hg target, we focus the beam to 25 microns and generate a power density of 1012 Watt/cm$^2$. Depending on the material, cleaning can be at power density levels orders of magnitude below levels used for plasma generation with a spot size of 200 to 1000 microns rastered across the sample surface.

In addition to strongly correlated intermetallic samples investigated in Figures 2–4 we also look at Mott insulators and PuO$_2$, where large values of Coulomb U lead to interesting physics and an electronic structure rich in features. In this research effort on the actinide dioxides, the theoretical predictions preceded the experimental work. Rich Martin (T-1) and collaborators developed a hybrid functional theory that works particularly well for actinide oxides. They predicted that even though the radial wavefunction of the actinide 5$f$ electrons is contracting as one moves across the actinide series from Th to Pu, UO$_2$ would have very little hybridization between the O 2$p$ levels and the U 5$f$ orbitals, while PuO$_2$ would have substantially more hybridization due to an overlap in the 2$p$-5$f$ orbitals at Pu. While we were able to confirm this general trend with PES, the direct experimental evidence of the hybridization from ARPES required single crystals of multi-millimeter dimension that did not exist. Bulk single crystals
of PuO$_2$ were not available, but the LANL-developed polymer assisted deposition (PAD) thin-film growth technique worked very well on both UO$_2$ and PuO$_2$. These samples were synthesized and then measured with the LPLS system. In Figure 6 we show the ARPES data for PuO$_2$ taken at a photon energy of 40.8 eV to enhance the Pu 5f character. In Figure 6, the dashed line traces the extent of the dispersion observed in the ARPES data, which directly equates with the amount of hybridization-driven dispersion found in calculations. Moreover, the black bar in this data map represents the extent of the dispersion observed in the UO$_2$ ARPES data. There was over an order of magnitude increase in ARPES dispersion and thus hybridization going from UO$_2$ to PuO$_2$, just as theory had predicted. This joint actinide effort at LANL worked exceptionally well between synthesis, spectroscopy, and theory to provide a new insight into actinide dioxide electronic structure.

Over the years, the LPLS project has provided a source of unique data for actinide and transuranic photoemission. While daily operations closely resemble those for any other photoemission system in terms of functionality and procedure, access to the contaminated vacuum system requires substantially more thought, planning, and preparation. In Figure 7 we show a 2012 hotjob to replace internal components on the system to keep the LPLS system competitive with conventional PES and ARPES capabilities. During nearly 20 years of Pu and Np operations, the system components have all been upgraded and replaced, often multiple times. The current incarnation of the LPLS system has the broadest range of capability and the most impressive functionality and procedure, access to the contaminated vacuum system closely resemble those for any other photoemission system in terms of daily operations.

### References
10. LANL transuranic single crystal samples: the PuCuO$_2$ samples were prepared using the flux growth by Eric Bauer, Jeremy Mitchell, and Paul Tobash. The PuCuO$_2$ samples were prepared using polymer assisted deposition by Eve Bauer, Brian Scott, Mark McGloley, and Quanxi Jia.
The Science of Harold M. Agnew

Harold Melvin Agnew (March 28, 1921–September 29, 2013) did not know that he would be a Forrest Gump of the new nuclear age—a participant in many of its most important events. It all began in 1942. He was 21 years old, a recent graduate of the University of Denver with a degree in chemistry, and Phi Beta Kappa. Agnew briefly worked at the Bureau of Standards as a science aide, but soon decided to try and join the United States Army Air Corps with his girlfriend, Beverly. Instead, the head of the physics department at the University of Denver, Joyce C. Stearns, invited Agnew to come with him to the University of Chicago, where Stearns became the deputy head of the Metallurgical Laboratory, or “Met Lab,” of the Manhattan Project. Harold and Beverly married on May 2 and moved to Chicago.

1942 was a fateful year for the Manhattan Project. Only a few weeks after Los Alamos was selected as the location for the project’s weapons design laboratory, Nobel Laureate Enrico Fermi’s team at the University of Chicago produced the world’s first self-sustaining chain reaction. As a member of Fermi’s team, Agnew worked on the world’s first reactor and helped conduct the famous chain reaction experiment of December 2 underneath Stagg Field. Fermi’s chain reaction essentially verified that an atomic bomb was possible, making it one of history’s most significant experiments. Many years later, Agnew recalled: “I just thought...this is just another one of Fermi’s experiments and it worked. I had no reason to doubt that it was going to work and I just went back to work. I didn’t really appreciate the significance. Some people will tell you, ‘Oh yes, they thought how important to the world...’ a bunch of bologna.”

With their work at Chicago complete, the Agnews and Fermi joined the staff at Los Alamos in early 1943. Fermi eventually became a division leader and one of the Laboratory’s two associate directors. Meanwhile, Agnew attempted to find a suitable material for the bomb’s tamper, a component designed to contain the neutrons produced during the early stages of the atomic detonation. This work remained a priority because the containment of neutrons would help ensure an efficient implosion.

By April 1945, the weapons design work was largely complete. The emphasis shifted to preparing the weapons for testing and combat, thus leaving many skilled young scientists, including Agnew, available for new assignments. Luis Alvarez, a future Nobel Laureate who had been heavily involved in detonator development, was tasked with developing a method for accurately measuring the yield of nuclear weapons. Alvarez enlisted the help of three young scientists: Agnew, Bernard Waldman, and Larry Johnston.
This diverse group; a chemist, an electrical engineer, and a physicist; started immediately. After consulting with Bill Penney, an eminent British scientist on the Los Alamos staff, the team decided to try and derive the yield by measuring blast pressure.

The Alvarez team decided to use a modified Firing Error Indicator developed by Jesse Dumond and Pief Panofsky from Cal Tech. The instrument was mounted in an aluminum canister that also included a microphone connected to a battery powered FM transmitter, which would broadcast a signal proportional to the blast wave pressure from the detonation. The canisters would be deployed by parachute during the combat missions.

Agnew and his teammates quickly ordered the first set of canisters and left Los Alamos for Wendover Airfield in Utah, home of the 509th Composite Group, to practice using them. During the practice missions, the team would need to master synchronizing the deployment of three canisters with an atomic bomb, checking the calibration in flight, and maintaining communication throughout the bombing run. This proved no simple task. In Japan, the difficulty of measuring the yield would be further compounded by flying over enemy territory and possibly encountering antiaircraft fire. Additionally, the plane would be performing a high speed, 150 deg. diving turn to escape the blast while Agnew and his colleagues were attempting to take measurements.

In June 1945, weeks before “Trinity,” the initial test of the bomb, Agnew travelled to the island of Tinian to help prepare facilities for the atomic strikes against Japan. The rest of the Alvarez team arrived August 1 with orders to install equipment aboard The Great Artiste, the B-29 chosen to serve as the diagnostic plane for both combat missions. Early in the morning of August 6 Alvarez, Johnston, and Agnew boarded The Great Artiste for the trip to Hiroshima and successfully collected yield data during the attack. Agnew also took movies during the flight. Upon their return to Tinian, the team analyzed the data recorded by Johnston and Agnew and calculated Little Boy’s yield.

After the war, Agnew returned to the University of Chicago to complete his PhD work under Fermi. During his time at Chicago, Agnew performed research on atomic interactions of matter with high-energy particles. The development of his thesis, “The beta spectra of Cs137, Y91, Chlorine147, Ru106, Sm151, P32, Tm170,” provided Agnew with the strong experimental and theoretical background he would need upon returning to Los Alamos.

In 1949, Agnew returned to Los Alamos and soon joined the weapons program. He explored the feasibility of constructing a thermonuclear bomb, a weapon many times more powerful than the bombs used to help end World War II. The first major breakthrough occurred in 1951, during Operation Greenhouse, when thermonuclear burn was achieved by a fission device. At the end of 1952, a full-scale undeliverable thermonuclear device was tested as part of Operation Ivy. Code-named Ivy-Mike, the test achieved a yield of 10.4 megatons. In the following years, Agnew would play a pivotal role in creating deliverable, lightweight, solid-fueled thermonuclear weapons.
At this juncture, Agnew’s experience studying atomic interactions at the University of Chicago began to pay significant dividends. He explored the possibility of using lithium hydrides in a more compact, lightweight configuration. In the spring of 1954, as part of Operation Castle, Agnew’s concept for using solid rather than liquid fuel was successfully demonstrated. This innovation soon yielded the first aircraft-deliverable thermonuclear weapons, thus helping the United States maintain its lead in deterrence technology.

Agnew continued his work in the weapons program as the Cold War escalated. Demand for lighter, more compact devices grew significantly in the late 1950s as advanced missile technology emerged. One means to reduce the weight of nuclear explosive devices is to increase their efficiency. From 1956 through 1958, Agnew focused his research on “boosting” to achieve that end. Boosting is the technique of fusing deuterium and tritium to increase yield. Between 1959 and 1961, nearly 20,000 new weapons were added to the stockpile. Agnew’s contributions helped make the stockpile of the late 1950s and early 60s lighter, more efficient, and more versatile. Next, he would help make the stockpile safer.

With thousands of new warheads being deployed worldwide, the need for safety became paramount. After a trip overseas to inspect nuclear...
Agnew's career in management started in 1955, when he was appointed an assistant division leader in the Theoretical Division. Less than a decade later, Agnew became the leader of W Division (Weapons Nuclear Engineering), a post he held from 1964 until he became director. As the 1960s progressed, Agnew dedicated an increasing amount of his time to management activities. He transitioned from being a scientific innovator, to cultivating Los Alamos as a center of excellence for innovators in virtually every major scientific field.

From 1970 to 1979, Harold Melvin Agnew served as third director of Los Alamos Scientific Laboratory (its name changed to “Los Alamos National Laboratory” in 1981, with a broader, multidisciplinary scope, largely as a result of Agnew’s vision). Up until Agnew became director, every program at the Laboratory had some connection back to the weapons program. During Agnew’s tenure, the Laboratory created research programs in a variety of fields, many of them with absolutely no connection to weapons. In so many ways, Agnew invented the modern, multidisciplinary national laboratory. Agnew’s career in management started in 1955, when he was appointed an assistant division leader in the Theoretical Division. Less than a decade later, Agnew became the leader of W Division (Weapons Nuclear Engineering), a post he held from 1964 until he became director. As the 1960s progressed, Agnew dedicated an increasing amount of his time to management activities. He transitioned from being a scientific innovator, to cultivating Los Alamos as a center of excellence for innovators in virtually every major scientific field.

During his years as director, Agnew fought to preserve the scientific integrity of Los Alamos. He called for higher wages, higher funding levels for scientific programs, and fought against the bureaucratization of science. In 1976, during his testimony before the National Science Board, Agnew boldly said, “Bureaucratic regulations and requirements for conformity will stifle basic research. Bureaucracy will eradicate creative endeavor and innovation in the long run.” When Agnew retired as director, in 1979, he left the Laboratory with more staff members and more scientific programs than it had ever had. Harold Agnew was himself a scientific innovator, but his greatest innovation was reimagining the concept of a national laboratory.

Selected publications of Harold Agnew:

Tokamak Applications

Gas-Cooled Nuclear Power Reactors

The Beta-Spectra Of Cs, Y, Pm, Ru, Sm, P, And Tm

Evidence For An Excited State Of He

Double Magnetic Lens Nuclear Spectrometer
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