Identifying the Source of Stolen Nuclear Materials

Livermore scientists are analyzing interdicted illicit nuclear and radioactive materials for clues to the materials' origins and routes of transit. **UCLEAR** forensics and attribution are becoming increasingly important tools in the fight against illegal smuggling and trafficking of radiological and nuclear materials. These include materials intended for industrial and medical use (radiological), nuclear materials such as those produced in the nuclear fuel cycle, and much more dangerous weapons-usable nuclear materials—plutonium and highly enriched uranium.

Livermore scientists are among the leaders in nuclear forensics—the chemical, isotopic, and morphological analysis of interdicted illicit nuclear or radioactive materials and any associated materials. (See the box below.) They are also supporting the national effort in nuclear attribution, which is the challenging discipline of combining input from nuclear and conventional forensics to identify the source of nuclear and radiological materials and determine their points of origin and routes of transit.

Nuclear forensics and attribution go beyond determining the physical, chemical, and isotopic characteristics of intercepted nuclear or radiological materials. Simply determining that an interdicted material is, for example, enriched nuclear power plant fuel often tells law-enforcement authorities little about the origin of the material, where it has been, or who is responsible for the theft. "We want to know the exact nature of the interdicted material, who the perpetrators were, and where legitimate control was lost," says David Smith, a geochemist in the Nonproliferation, Homeland and International Security (NHI) Directorate and a leader of the Livermore nuclear forensics team. Knowledge of the pathways involved in transporting the material is also vital to nonproliferation and counterterrorism efforts.

Nuclear forensics and attribution can be difficult because of the large amount of radioactive materials in the industrialized and developing world. For example, the nuclear fuel cycle produces a variety of materials at different processing steps, ranging from processed ore (commonly known as "yellow cake") to uranium oxide fuel enriched in uranium-235. Opportunities exist at many points during the fuel cycle for materials to be diverted outside the channels authorized for their legitimate manufacture, handling, and protection.

Successful nuclear forensics and attribution contribute to the prosecution of those responsible and strengthen national efforts in nuclear nonproliferation and counterterrorism. According to former Livermore physicist Jay Davis, the first director of the federal Defense Threat Reduction Agency (DTRA), "Nuclear attribution, with the accompanying possibility of prosecution and retribution, may be one of our greatest deterrent tools and hence a vital and compelling component of our defense against terrorism."

Livermore and seven other Department of Energy (DOE) national laboratories have been tasked by the Federal Bureau of Investigation (FBI) and the Department of Homeland Security (DHS) with further developing the nation's technical forensics capability for nuclear and radiological materials. Major U.S. government partners in addressing domestic and foreign obligations include DOE, the National Nuclear Security Administration (NNSA), the Department of State, and DTRA. Technical nuclear forensics includes

A Textbook for Nuclear Forensic Scientists

Nuclear Forensic Analysis, written by Livermore scientists Ken Moody, Ian Hutcheon, and Pat Grant, is the first primary reference source for the growing specialty of nuclear forensics. In addition to being a resource for nuclear forensic scientists, the book also serves as a textbook for traditional radiochemistry science courses that increasingly include material on nuclear forensics. The book covers the principles of the chemical, physical, and nuclear characteristics associated with the production and interrogation of a radioactive sample; protocols and procedures; and attribution. The authors discuss principles and techniques used in numerous case studies of nuclear investigations conducted at Livermore.

Hutcheon is deputy director of the Glenn T. Seaborg Institute. Moody, who studied with Nobel Laureate Glenn Seaborg (the discoverer of plutonium), is a radiochemist and co-discoverer of four heavy elements. Grant serves as the deputy director of the Forensic Science Center. "Pat is trained in classical forensics, Ken is a nuclear chemist, and I specialize in instrumentation," says Hutcheon. "Together, we give a balanced viewpoint."

According to Hutcheon, "The book gives one an idea of the analytic techniques we use and the types of material we are asked to investigate. It also describes case studies and the ways we assign attribution." Hutcheon says the book was written to be understandable by nonspecialists. the analysis of conventional evidence that is radiologically contaminated as well as the analysis of the radiological materials themselves.

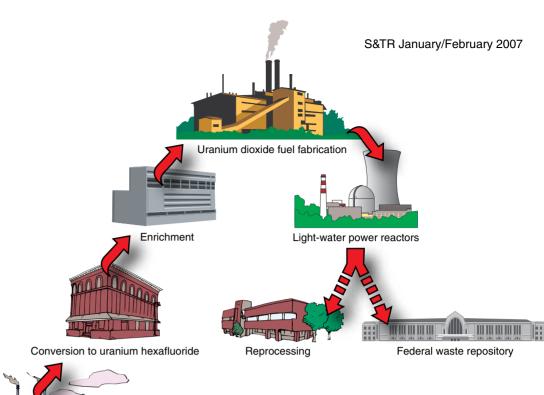
Lawrence Livermore and Savannah River national laboratories serve as a "hub" for nuclear forensics, with the "spokes" of the hub represented by six other national laboratories that provide specialized analyses and serve in supporting roles. A sample identified by one of the hub laboratories could be sent to experts at one or more of the spoke laboratories for complementary analyses and further characterization.

"We're part of a national nuclear forensic and attribution capability that ties nuclear and radiological materials to people, places, and events," says Smith. The effort is also part of a national strategy to counter nuclear terrorism. Other activities at the Laboratory that support the national strategy include developing more capable radiation detectors, designing detection systems for ports and other venues, and working with Russia and nations of the former Soviet Union to secure their nuclear materials.

Focus on Forensics

Nuclear forensics began at the Laboratory under the leadership of scientist Sid Niemeyer in the mid-1990s, and Livermore has maintained a leading role because of a collective group effort. The Laboratory has been involved in nuclear forensics and attribution for more than 15 years, since the collapse of the Soviet Union sparked concerns about the diversion of nuclear materials from former Soviet nuclear laboratories and other sites. Livermore's capabilities in radiochemistry and nuclear physics, originally developed for the nation's underground nuclear testing program, were adapted for use in nuclear forensics and attribution.

The strength of the national capability rests on the ability of Livermore to work in concert with the other laboratories, drawing on a core group of 30 to 50 scientists



Because nuclear material goes through several transformations during the uranium concentration and enrichment cycle, specific signatures may be unique to different stages of the cycle. Scientists want to develop signatures across the entire nuclear fuel cycle.

from diverse backgrounds to work collaboratively on nuclear forensics and attribution cases. This depth of knowledge and breadth of experience are essential to tackling current issues in nuclear forensics.

I Iranium mines and mills

Technical nuclear forensics can be performed on a broad spectrum of possible substances. Some interdicted samples are stolen containers of uranium diverted during one of the mining, milling, conversion, enrichment, or fuel fabrication steps used to convert uranium ore to enriched fuel for nuclear power plants. Alternatively, the interdicted sample could well be a commercial radioactive material such as cesium-137. strontium-90, cobalt-60, or americium-241. These isotopes and others are used in applications such as medical diagnostics, nondestructive analysis, food sterilization, and thermoelectric generators.

"We support investigations with the extraction of evidence that can be used by law-enforcement agencies," says Smith. "Our casework validates and verifies our technical approaches." Livermore and the other national laboratories subject an interdicted sample to a host of extremely sensitive and accurate measurement techniques. Researchers analyze the material's chemical and isotopic composition, which includes measuring the amounts of trace elements as well as the ratio of parent isotopes to daughter isotopes. These measurements help to determine the source location and sample's age. They also examine the material's morphological characteristics such as shape, size, and texture. Together, these characteristics are indicative of the specific processes used to produce the material.

Analytical methods include electron microscopy, x-ray diffraction, and mass spectrometry. (See the box on p. 17.) "As we obtain new tools, we open new frontiers of technical nuclear forensics," says Ian Hutcheon, deputy director of the Glenn T. Seaborg Institute and senior scientist in the Forensic Science Center. For example, Livermore's NanoSIMS, a secondary-ion mass spectrometer, provides a 50-nanometer (a billionth of a meter) spatial resolution. This capability enables researchers to analyze subsamples of less than 1 microgram and perform particle-by-particle characterization to elucidate additional signatures beyond those gained through dissolving the bulk sample. "We are advancing from the micro to the nano level in analyzing samples to obtain forensic clues," says Hutcheon.

Contaminated Evidence

An important element of nuclear forensics is the analysis of nonnuclear materials found with the radioactive material. Livermore scientists have been involved in developing forensics on radiologically contaminated evidence, which enables conventional lawenforcement forensics to be performed on materials that are radioactive. In addition, as a sample is moved from place to place, it picks up clues such as pollen, cloth fibers, and organic compounds. These so-called route materials provide information about who has handled a sample and the path it has traveled.

Nuclear forensics takes advantage of capabilities in Livermore's Forensic

Science Center to tease out fingerprints, paper, hair, fibers, pollens, dust, plant DNA, and chemical explosives from contaminated substrates for the FBI and other government agencies. Established in 1991, the center supports DOE in verifying compliance with international treaties. It has also assisted federal, state, and local law enforcement on a wide range of criminal cases, including the 1993 World Trade Center bombing and the Unabomber investigations. The Forensic Science Center incorporates subject-matter experts from the Laboratory's NHI and Chemistry, Materials, and Life Sciences (CMLS) directorates to build a complete profile of a sample.

Livermore's nuclear forensics program uses laboratories equipped to handle large objects that are contaminated with radioactive materials. These laboratories make it possible, for example, to analyze a contaminated truck axle while still maintaining the evidentiary chain of custody required in a court of law. In addition, the program operates a



Livermore researchers use techniques such as electron microscopy, x-ray diffraction, and mass spectrometry to analyze interdicted radiological and nuclear materials. In this photo, Ian Hutcheon (left), a senior scientist working with Livermore's Forensic Science Center, and analytical chemist Peter Weber use NanoSIMS, a secondary-ion mass spectrometer with nanometer-scale resolution.

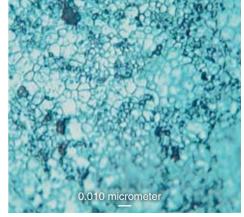
mobile van outfitted to ferry samples around the site or, if necessary, transport material between the Laboratory and offsite locations.

Developing Signatures

A major focus of nuclear forensics is identifying signatures, which are the physical, chemical, and isotopic characteristics that distinguish one nuclear or radiological material from another. Signatures enable researchers to identify the processes used to initially create a material.

Livermore scientists are developing signatures for a variety of nuclear materials. Enriched uranium is of particular concern. The goal is to develop validated signatures across the entire nuclear fuel cycle by experimental measurement or by simulation. Because nuclear material is transformed at different points during uranium concentration and enrichment processing, clues may be unique to different stages.

For example, unirradiated uranium reactor fuel pellets have inherent elemental oxygen content. Because the ratio of naturally occurring isotopes of oxygen-18 to oxygen-16 varies worldwide, these ratios could correlate with the locations of production sites. Similarly, the



The microscopic shape and size of uranium particles offer clues about the material's origin. This optical image of a nuclear fuel pellet shows the morphology of the grains.

variations of lead isotopes could provide clues about where a uranium compound was produced. Age, color, density, trace elements, and surface characteristics of the uranium compound are other important characteristics. A related investigation is studying the effects of different uranium manufacturing processes on the grain size and microstructure of the finished product.

When comparing a sample's signature against known signatures from uranium mines and fabrication plants, researchers can benefit by assembling a library of nuclear materials of known origin from around the world. Livermore scientists, with the help of the Office of Laboratory Counsel, have developed relationships with domestic suppliers of nuclear materials (uranium hexafluoride and uranium oxide reactor fuel) to assemble such a library. Contracts with major U.S. uranium fuel suppliers have provided researchers with samples and manufacturing data. Livermore scientists are using statistics to identify distinguishing signatures in nuclear

Under an agreement signed in 2006, KazAtomProm is providing Livermore scientists with uranium ore samples and data and has identified experts available for consultation on nuclear forensic issues. Front row, left to right: Dave Herr from Livermore's Procurement Office and Sergei Yashin, vice president of KazAtomProm. Back row, left to right: Mike Kristo and David Smith from Livermore and representatives from KazAtomProm. (background) KazAtomProm is the leading uranium mining and fuel production facility in Kazakhstan. materials using samples and data obtained from manufacturers.

Livermore forensic scientists are also seeking to obtain samples of uranium products worldwide to analyze the products' isotopic and trace-element content, grain size, and microstructure. Nations with nuclear capabilities are beginning to share information about their nuclear fuel processes and materials. (See the box on p. 18.) For example, Livermore representatives signed a fiveyear agreement in January 2006 with KazAtomProm, the national atomic energy enterprise in Kazakhstan. KazAtomProm has long provided uranium ores to Russia and other nations of the former Soviet Union. With legal and technical support from NNSA, DHS funded the Laboratory to contract with KazAtomProm to provide Livermore with uranium ore samples and data. In addition, KazAtomProm officials have identified experts available for consultation on nuclear forensic issues.

A similar agreement was signed in

2006 with nuclear power officials in Tajikistan. This agreement provides samples to enable comparison between known and questioned sources. "We want to expand our agreements to all the Central Asian nations that process uranium," Smith says.

The importance of international cooperation was underscored in July 2006, when U.S. President George W. Bush and Russian President Vladimir Putin announced the creation of a Global Initiative to Combat Nuclear Terrorism. The initiative's goal is to strengthen worldwide cooperation in making nuclear materials more secure and preventing terrorist acts that involve nuclear or radioactive substances.

Building a Knowledge Base

The data obtained from U.S. and Central Asian uranium producers are part of a growing technical nuclear forensics knowledge base involving Livermore and seven other DOE national laboratories.



"We are establishing a knowledge management system for nuclear signatures, processes, origins, and pathways," says Livermore nuclear engineer Frank Wong, who is leading the knowledge base effort, which now includes about 30 experts nationwide.

Rapid and credible interpretation of nuclear forensics data is predicated on a knowledge base that accesses and analyzes information derived from measurements or simulation of the full spectrum of the nuclear fuel cycle. Knowledge management allows for the ready comparison of analytical signatures obtained from suspect nuclear samples against known signatures from nuclear production, reprocessing, manufacturing, and storage. It also allows for the analysis of existing knowledge and data with the goal of identifying the most diagnostic nuclear forensic signatures.

The knowledge base is being designed so that it can be accessed by a simple query on a computer. "As an example,



The Lawrence Livermore technical team that addresses the technical nuclear forensics of interdicted materials includes (left to right) in the front row: lan Hutcheon, Erick Ramon, and Michael Kristo; second row: Brett Isselhardt. Giles Graham, and Lars Borg; third row: Ross Williams and Michael Singleton; and fourth row: David Smith and Lee Davisson. Not pictured: Glenn Fox, Pat Grant, Leonard Gray, Steven Kreek, Ken Moody, Sid Niemeyer, Martin Robel, Steve Steward, Louann Tung, Alan Volpe, Philip Wilk, Nathan Wimer, and Frank Wong.

Scientific research in cosmochemistry and isotope geochemistry continues to technically validate Livermore's nuclear forensics capabilities. Cosmochemistry is the study of the origin and development of the elements and their isotopes in the universe and the formation of our solar system. Livermore cosmochemistry projects, funded by the Laboratory Directed Research and Development (LDRD) Program and the National Aeronautics and Space Administration, allow researchers to use the same instruments and procedures that nuclear forensic scientists use when analyzing interdicted nuclear and radiological samples.

One LDRD effort is studying inclusions in Australian zircons more than 4 billion years old to determine when conditions suitable for life first emerged on Earth. The work, led by scientist Ian Hutcheon, involves dating and isotopically and chemically analyzing the zircons

The Cosmic Connection to Nuclear Forensics

and the mineral inclusions in them. The effort focuses on measuring the abundance of trace elements, notably uranium and other actinides in these ancient zircons. This analysis will help researchers to understand the evolution of the atmosphere and hydrosphere during the earliest epoch of Earth's history and to evaluate the evidence for volcanic activity as far back as 4.4 billion years.

Hutcheon says this LDRD effort develops and enhances microanalytical capabilities needed for nuclear forensics. The investigation uses tools that also support Livermore's nuclear forensics work, such as the Laboratory's nanometer-scale secondary-ion mass spectrometer (NanoSIMS) and a new, ultrahigh-resolution scanning electron microscope. "Whether we're measuring oxygen isotopes in Australian zircons or in interdicted uranium yellow cake, the techniques are similar," he says. "Our ancient zircon research is published in peer-reviewed journals, and the suggestions and criticism made in response to these published papers strengthen our nuclear forensics work." Hutcheon was involved in another LDRD cosmochemistry effort headed by John Bradley of Livermore's Institute of Geophysics and Planetary Physics. Using a transmission electron microscope, Laboratory researchers detected a 2,175-angstrom extinction feature (or bump) in interstellar grains embedded within interplanetary dust particles. The Livermore team identified organic carbon and amorphous silica-rich material as responsible for the bump. (See *S&TR*, September 2005, pp. 24–27.)

These measurements may help explain how interstellar organic matter was incorporated into the solar system. Interplanetary dust particles are roughly the same size scales of interdicted radiological and nuclear materials, such as powdered nuclear enriched uranium. By studying these dust particles, scientists gain confidence in using advanced tools to characterize other materials of interest to nuclear forensics work. when key words are entered, the system will find the relevant information," Wong says. He notes that the knowledge base will use a distributed architecture. That is, the information will not be centralized in one location. Instead, information will be distributed across several government sites, such as DHS. FBI, and DOE, with several levels of controlled access. The knowledge base will include a list of subject-matter experts worldwide who could be called upon to assist in specific nuclear forensics casework. Another proposed element is a U.S. evidence archive, where nuclear materials from past cases would be stored as references.

Growing Capabilities

Nuclear forensics and attribution are two important elements among several DHS and NNSA efforts in border protection, radiation monitoring, emergency response, and consequence management to provide the nation with the best protection against nuclear and radiological threats. Nuclear chemist Michael Kristo says that the nuclear forensics and attribution effort has gained strength with the launching of the Global Nuclear Energy Partnership (GNEP) in February 2006.

As part of President Bush's Advanced Energy Initiative, GNEP is a strategy to expand nuclear energy worldwide

Strengthening the Worldwide Effort

The Nuclear Smuggling International Technical Working Group (ITWG) was chartered in 1996 to foster international cooperation in combating illicit trafficking of nuclear materials. "The ITWG was formed with the recognition that nations must work together," says geochemist David Smith of the Nonproliferation, Homeland and International Security Directorate. The ITWG was cofounded by Livermore scientist Sid Niemeyer and has been cochaired by Lawrence Livermore since its inception.

The ITWG works closely with the International Atomic Energy Agency (IAEA) to provide member countries with support for forensic analyses. Priorities include the development of common protocols for the collection of evidence and laboratory investigations, organization of forensic exercises, and technical assistance to requesting nations. Experts from participating nations and organizations meet annually to work on issues concerning illicit trafficking of nuclear materials. The 2006 meeting was sponsored by the European Commission's Institute for Transuranium Elements in Karlsruhe, Germany.

To promote the science of nuclear forensics within the ITWG, the Nuclear Forensics Laboratory Group was organized in 2004. In that year, Livermore scientists wrote a comprehensive description of a model action plan to guide member states in their own nuclear forensic investigations. The plan provides recommendations governing incident response, sampling and distribution of materials, radioactive materials analysis, traditional forensic analysis, and nuclear forensic interpretation of signatures. In 2006, the IAEA published the model action plan as a Nuclear Security Series Technical Document. Participating countries have adopted the plan and used it in their own nuclear forensics investigations.

Exercises are critical to operational readiness within ITWG participants. An international exercise is planned for later this year, when participants will receive identical radiological samples supplied by Pacific Northwest National Laboratory. "It's essential for laboratories to discuss the results from their findings and evaluate their capabilities," says Smith. by demonstrating and deploying new technologies to recycle nuclear fuel, minimize waste, and improve the ability to keep nuclear technologies and materials out of the hands of terrorists. GNEP would build recycling technologies that enhance energy security in a safe and environmentally responsible manner. A basic goal of GNEP is to make it impossible to divert nuclear materials or modify systems without immediate detection. One option is to incorporate tags into nuclear materials at different stages of the fuel cycle so that the materials could be tracked and traced.

Looking to the future, Smith notes that existing Laboratory technical efforts define an emerging Nuclear Forensics Analysis Center funded primarily by DHS. This center would build on Livermore's experience in nuclear assessment, nuclear weapons and materials, and isotope and trace-element science as well as on the capabilities and expertise of the Forensic Science Center and the NHI and CMLS directorates. "Livermore is uniquely positioned," he says. "We have science-based signatures, expertise, and facilities. All the pieces are here for such a center and for making an even greater contribution to the prevention of nuclear materials trafficking and nuclear terrorism."

—Arnie Heller

Key Words: cosmochemistry, Forensic Science Center, Global Nuclear Energy Partnership (GNEP), International Atomic Energy Agency (IAEA), nuclear attribution, nuclear forensics, Nuclear Forensics Analysis Center, nuclear material, nuclear terrorism.

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