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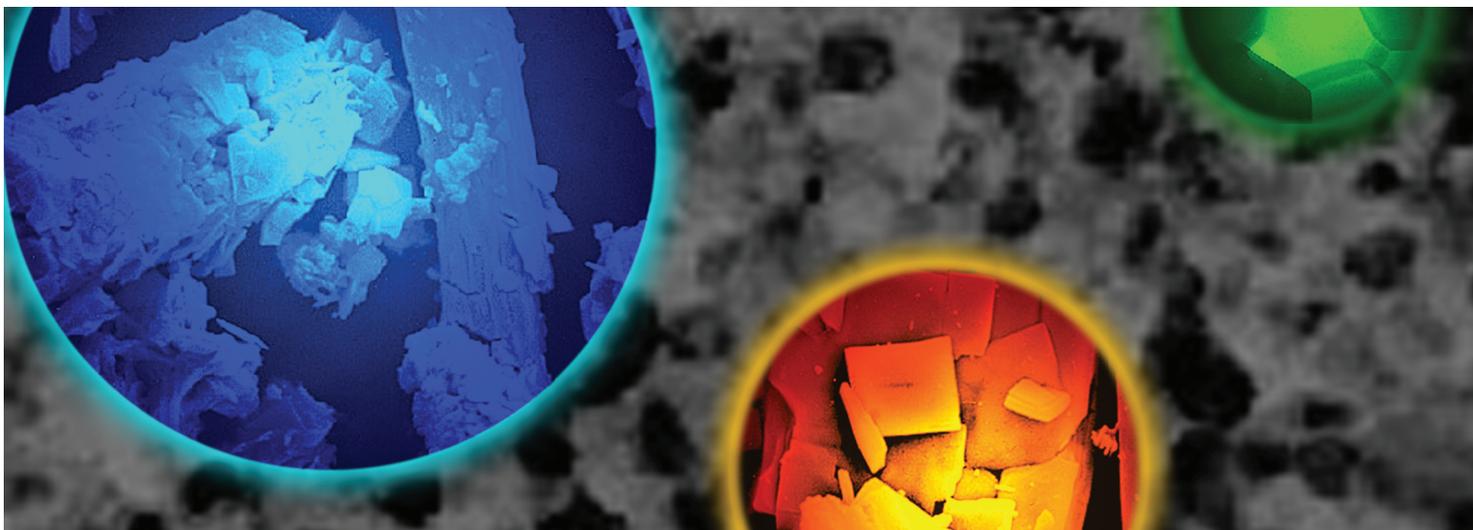
4th Quarter 2007

Nuclear forensics

A new branch of science

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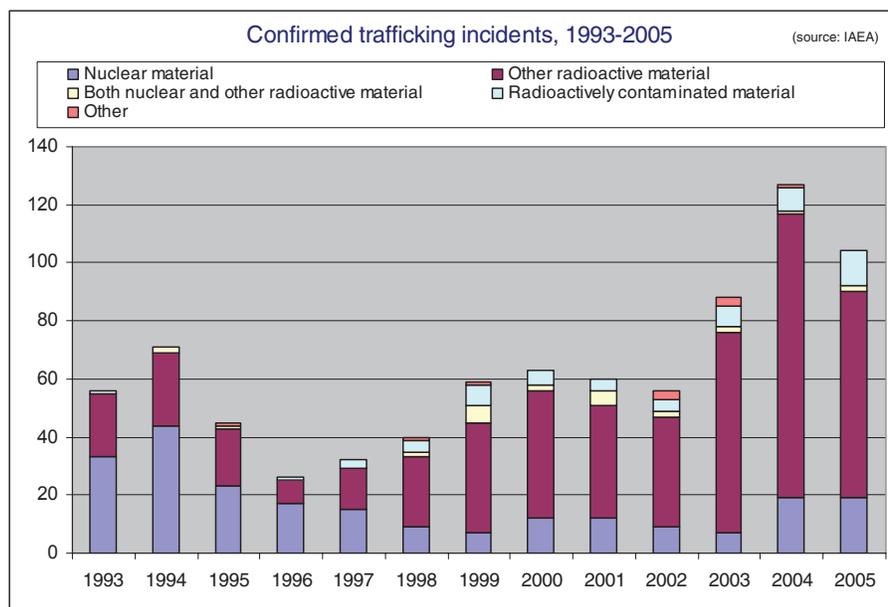
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CSI: KARLSRUHE

NUCLEAR FORENSICS SLEUTHS TRACE THE ORIGIN OF TRAFFICKED MATERIAL

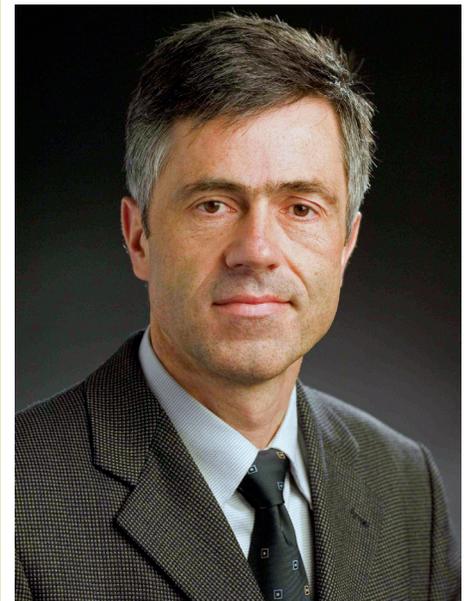
When the former Soviet Union broke up, the new independent states couldn't control their supplies of nuclear material and some of it got "lost." In 1994 alone, 45 confirmed cases of nuclear material trafficking were reported, according to the International Atomic Energy Agency (IAEA). The numbers have decreased to around 10 reported incidents a year, but poorly guarded and easily stolen nuclear materials still pose a serious problem because of the radiological hazards associated with improper transport, handling, and storage. And while the dumping of nuclear material in a landfill or salvage yard is very serious, even more dire consequences could occur if the material ended up in the hands of a terrorist.



When a cache of stolen or dumped nuclear material is intercepted, routine forensics techniques are used to answer the questions of who and how and what. Answering what the material is, where it came from, and what it could be used for is a nuclear whodunit worthy of the "CSI" television series and has resulted in the development of a new branch of science called nuclear forensics.

Since the early 1990s the Institute for Transuranium Elements (ITU) in Karlsruhe, Germany, has been involved in developing the methodology of nuclear forensics to answer the questions of chemical makeup, origin, and use. Tracing where the material came from will help governments improve physical protection of the site of origin and prevent future thefts or illegal disposal.

ARQ thanks Klaus Mayer, Maria Wallenius, Ian Ray, and Klaus-Richard Lützenkirchen of the Institute for Transuranium Elements (ITU) for their contributions to this article.



Klaus-Richard Lützenkirchen

The science is based on analytical techniques related to the nuclear fuel cycle: radiochemistry, nuclear physics, reactor physics, and materials science.

Klaus-Richard Lützenkirchen of ITU's Nuclear Safeguards and Security Unit recently visited Los Alamos and gave a talk on nuclear forensics activities at ITU. The Seaborg Institute sponsored his visit for Transactinium Science. Lützenkirchen discussed typical cases that have been analyzed at ITU and described the various analytical techniques that led to the successful determination of where the materials, specifically plutonium and uranium, came from. Three of the cases are discussed below.

**Information that can be obtained from nuclear* material
*uranium (U), plutonium (Pu)**

Parameter	Information	Analytical technique
Appearance	Material type (e.g., powder, pellet)	Optical microscopy
Dimensions (pellet)	Reactor type	Database
U, Pu content	Chemical composition	Titration, HKED, IDMS
Isotopic composition	Enrichment → intended use; reactor type	HRGS, TIMS, ICP-MS, SIMS
Impurities	Production process; geolocation	ICP-MS, GDMS
Age	Production date	AS, TIMS, ICP-MS
¹⁸ O/ ¹⁶ O ratio	Geolocation	TIMS, SIMS
Surface roughness	Production plant	Profilometry
Microstructure	Production process	SEM, TEM

HKED (hybrid K-edge densitometry), IDMS (isotope dilution mass spectrometry), HRGS (high-resolution gamma spectrometry), TIMS (thermal ionization mass spectrometry), ICP-MS (inductively coupled plasma mass spectrometry), SIMS (secondary ion mass spectrometry), GDMS (glow discharge mass spectrometry), AS (alpha spectrometry), SEM (scanning electron microscopy), TEM (transmission electron microscopy)

One of four uranium-oxide fuel pellets seized in Lithuania. Nuclear forensics sleuths determined that the pellets were manufactured by MZ Electrostal in Moscow for the RBMK-1500 reactor known as Ignalina Unit 2 in Lithuania. The pellets probably came from a fresh fuel assembly that had been stolen from the Ignalina power plant in 1992.



ITU's nuclear forensics methodology takes data from analytical methods from safeguards, material science, and isotope geology to determine the isotopic composition, elemental composition, impurities, macroscopic appearance, microstructure, and age. The data reveal into two types of results: endogenic, or self-explanatory (age, intended use, production mode), and exogenic, which requires reference data (place of production, last legal owner, and smuggling route). Twenty-one seizures analyzed at ITU between 1992 and 1997 included natural uranium, low-enriched uranium fuel pellets, highly enriched uranium, plutonium, and contaminated scrap metal.

First case: uranium pellets

Uranium dioxide (UO₂) pellets are used as fuel in nuclear power reactors. In June 2003, ITU received four uranium pellets from Lithuania. The pellets were analyzed for uranium content and isotopes; chemical impurities, which would point to the source of the raw product; age, which would point to the production time; and microstructure, which would point to the production process.

All of the pellets showed identical geometry; they had a central hole and they were dished. The pellets were weighed and their dimensions measured. The four pellets were measured individually with a high-resolution gamma spectrometer for the first indication of the isotopic composition. The spectra showed gamma lines belonging only to uranium, and analysis showed an average uranium-235 enrichment of 2%. Because the pellets were identical in dimensions as well as in isotopic composition of the uranium, only one of them was dissolved for further analysis.

**Isotopic composition of uranium (U)
by mass spectrometry in mass%**

Technique	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U
TIMS	0.0147 ± 0.0010	2.0005 ± 0.0010	0.0071 ± 0.0067	97.9778 ± 0.0019
MC-ICP-MS	0.0142 ± 0.0002	2.0005 ± 0.0001	0.0071 ± 0.0000	97.9782 ± 0.0010

TIMS (thermal ionization mass spectrometry), MC-ICP-MS (multi-collector inductively coupled plasma mass spectrometry)

The isotopic composition of the uranium was determined by mass spectrometry. Mass spectrometry techniques are able to provide accurate results for the minor abundant isotopes (uranium-234 and -236), which is not the case with gamma spectrometry. The measurement technique routinely used for uranium and plutonium isotope analysis is thermal ionization mass spectrometry (TIMS). An inductively coupled plasma mass spectrometer with multi-collector detection system (MC-ICP-MS) was used to compare the accuracy and precision between these two methods.

The uranium content in solution was determined by three different methods: potentiometric titration, hybrid K-edge densitometry (HKED), and isotope dilution mass spectrometry (IDMS). All three methods determined that the uranium content corresponded to the stoichiometry of uranium dioxide (UO₂) whose theoretical value is 88%. Impurities in the sample were determined after complete dissolution by sector-field ICP-MS using rhodium-103 as an internal standard.

Determining the age of the material, and thus the date when the material was produced, helps identify the production campaign or batch. The radioactive decay of the uranium isotopes provides a unique chronometer that is inherent to the material. This clock is reset to zero each time the decay products (daughter nuclides) are chemically separated from the uranium. The half-life of the uranium isotopes in question is very long, therefore the short periods between the preparation of the uranium fuel and the seizure of the material generated extremely minute amounts of daughter nuclides. Nevertheless, the age could be determined from these parent/daughter ratios. The age of the uranium was calculated using the equation of radioactive decay and its derivatives.

The sample solution was spiked with thorium-228 and uranium-233 before the uranium/thorium separation. The amount of uranium-234 and thorium-230

Uranium content in mass%

Technique	Uranium content
HKED	87.43 ± 0.32
Titration	87.90 ± 0.13
IDMS	87.99 ± 0.24

HKED (hybrid K-edge densitometry), IDMS (isotope dilution mass spectrometry)

**Measured impurities in
μg/g uranium by
SF-ICP-MS**

Element	Concentration
Aluminum (Al)	6.08 ± 0.73
Calcium (Ca)	18.4 ± 2.2
Chromium (Cr)	6.12 ± 0.73
Copper (Cu)	1.80 ± 0.22
Iron (Fe)	91.9 ± 7.4
Potassium (K)	44.7 ± 3.6
Magnesium (Mg)	4.71 ± 0.57
Manganese (Mn)	1.13 ± 0.14
Sodium (Na)	17.9 ± 2.1
Nickel (Ni)	5.14 ± 0.62
Zinc (Zn)	3.40 ± 0.41

SF-ICP-MS (sector-field inductively coupled plasma mass spectrometry)

was determined using the isotope dilution technique, i.e., relative measurements against the (known amount of) spike isotope. The age of the material was determined to be 12.6 years ± 0.8 years. Thus the pellets had been produced at the end of 1990 (remember that the test occurred in 2003). Only the uranium-234/thorium-230 parent/daughter ratio could be used this time for the age determination because of the long half-life of uranium-235 and consequently the very small amounts of built-up daughter nuclides.

At this point, the investigators knew the dimensions of the pellets as well as the isotopic composition and age of the material. For the next step—determining where the pellets came from—the team used a relational database at ITU that contains data from several nuclear fuel manufacturers (including most of Western Europe and Russia). The database contains dimensions of pellets, uranium-235 enrichment, and typical impurities. Besides commercial reactor fuels, the database also contains information on research reactor fuels and information acquired from open literature. Additionally, results of old findings are introduced into the database for a comparison with future cases.

In the case of the four pellets from Lithuania, the database gave a very unambiguous answer. The pellet dimensions and enrichment already were enough to identify them as being made for an RBMK-1500 reactor, which is a Russian-type, water-cooled, graphite-moderated reactor. There are two models of the RBMK reactor: the 1000 and 1500. The 1000 model is older and more widely distributed, while there is only one 1500 model reactor in the world: Ignalina Unit 2 in Lithuania, which started up in August 1987 and is still operational.

Furthermore, there is only one manufacturer for this type of fuel: MZ Electrostal near Moscow. The measured impurities of the pellet material were below the maximum values given in the manufacturer's specifications and they also agreed with the experimental data from earlier findings of the same fuel. The last confirmation parameter was the age, which fit with the production data of the manufacturer (start of fuel production: December 1989). The information contained in the nuclear materials database proved to be essential for the attribution of the material.

ITU's nuclear forensics team was able to further deduce from the absence of uranium-236 that the fuel had been enriched from natural uranium (meaning there was no reprocessed material) and, because the pellets contained no traces of plutonium, that the fuel had never been in a reactor.

The IAEA database on trafficking of nuclear and other radioactive materials and some other open source information reported a case of a fresh fuel assembly being stolen from the Ignalina power plant in 1992. The four pellets under investigation definitively originated from Electrostal, and probably came out of that stolen assembly. This kind of fuel assembly contains about 110 kilograms of uranium. Between 1994 and 1997 more than 100 kilograms of pellets have been confiscated in several seizures; the greater part of the material has been recovered. The material itself is not useable for nuclear weapons because the

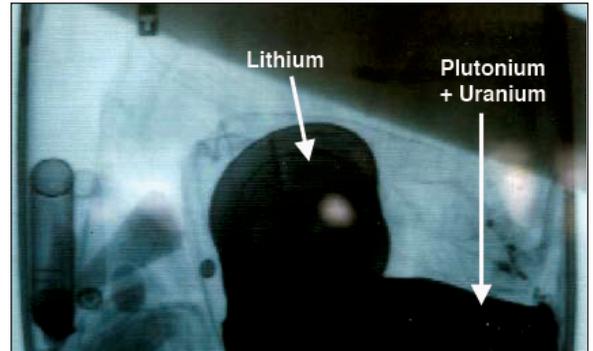
uranium-235 enrichment of 2% is far too low. However, what makes this case spectacular is the amount of the material that was stolen. Efforts have been undertaken to improve the physical protection at nuclear power plants and other storage facilities for nuclear material in the former Soviet Union.

Second case: mixed-oxide (MOX) powder

In August 1994, three men were stopped at the Munich airport carrying a suitcase containing 560 grams of plutonium- and uranium-oxide powder and 210 grams of lithium metal. The powder consisted of 64.9 wt.% of plutonium and 21.7 wt.% of uranium. The plutonium-239 enrichment was about weapons-grade quality, whereas the uranium had a low uranium-235 enrichment. The piece of lithium metal was enriched to 89.4% lithium-6. The MOX powder consisted of three different particle types: plutonium-oxide (PuO_2) platelets, rod-shaped PuO_2 , and hexagonal uranium-oxide (U_3O_8).

Light-water reactors (PWR, BWR, and VVER) were excluded as the origin of the plutonium because the isotopic composition of plutonium after a typical irradiation period of three years in these reactors would have been significantly different. Materials-testing reactors using 36%–90% enriched uranium-235 were also excluded because a higher plutonium-238 abundance would have been expected in this case. Most likely, a reactor type with a softer neutron spectrum (e.g., heavy-water or graphite-moderated) was used for production.

In this case, the nuclear reactor would have operated with an initial fuel enrichment of 1.8% uranium-235 to yield the uranium composition, assuming of course that the uranium and plutonium were from the same reactor. This scenario was also proposed by plutonium isotopic correlation. However, the plutonium-238 and -242 abundances were too high to originate from the low-burn-up spent fuel of an RBMK-1000 reactor. Thus, most likely, the plutonium was a mixture of different spent fuels (e.g., a low-burn-up or weapons-grade plutonium and a high-burn-up fuel) and had no direct connection with the uranium present.

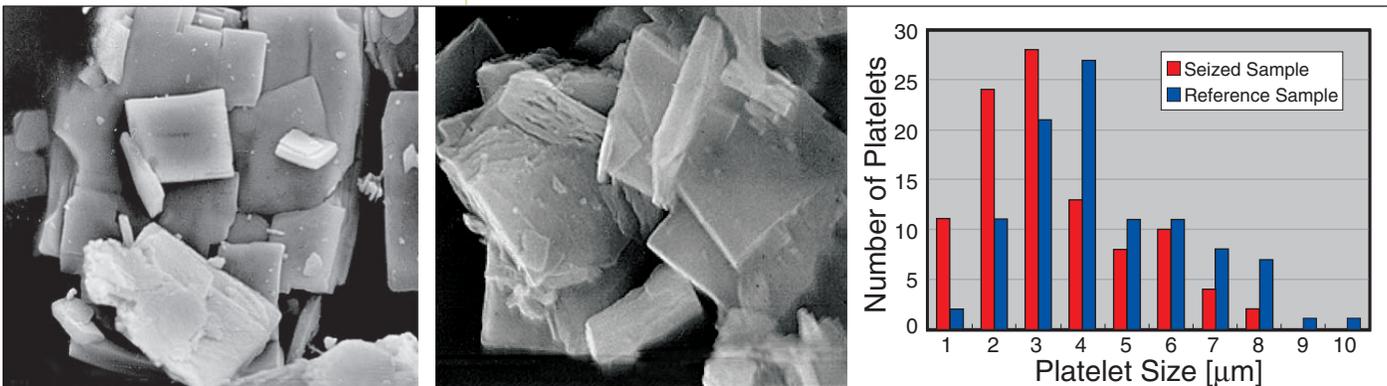


X-ray of a suitcase that contained nuclear material. In August 1994, three men who were expected to be carrying plutonium arrived in Munich from Moscow. Gamma-ray detectors did not respond when their luggage was checked, but X-rays showed a stainless steel container and tin cans.

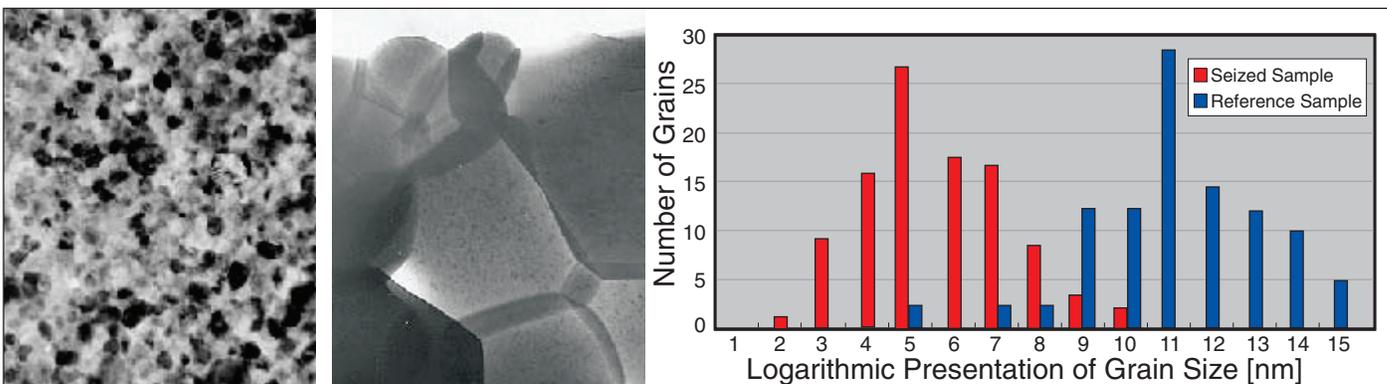
Nuclear material seized at the Munich airport. The suitcase contained 560 grams of plutonium- and uranium-oxide powder (below left) and 210 grams of lithium metal (below center). The structure of the mixed-oxide powder was determined by scanning electron microscopy (SEM). The micrograph (below right) shows the three distinct shapes of the powder: plutonium-oxide platelets, plutonium-oxide rods, and uranium-oxide hexagons.



Because the powder consisted of two different plutonium particle types, individual microparticles were analyzed by SIMS to determine if their isotopic compositions were identical or if the earlier determined isotopic composition for the bulk material was a result of mixing two different compositions. The plutonium-240/-239 ratios in the platelets and the rod-shaped particles were slightly different (0.1159 ± 0.0012 and 0.1245 ± 0.0026 , respectively). However, the difference was much too small to conclude that one plutonium particle type originated from weapons-grade plutonium (plutonium-240/-239 < 0.05) and the other type from high-burn-up fuel (plutonium-240/-239 ~ 0.4 – 0.7). Therefore, the mixing must have taken place before the particles were produced.



Platelet analysis. The plutonium-oxide (PuO₂) platelets in the seized Munich mixed-powder were examined in detail by scanning electron microscopy (SEM), above, to determine platelet-size distribution, and by transmission electron microscopy (TEM), below, to determine grain-size distribution. The SEM analysis shows a reference sample of PuO₂ from a known fabrication plant (above left) and the PuO₂ platelets from the sample seized at the Munich airport in 1994 (above center). The SEM analysis (above right) does not show a significant difference between the two samples. The TEM analysis shows a reference sample of PuO₂ from a known fabrication plant (below left) and the PuO₂ platelets from the sample seized at the Munich airport in 1994 (below center). It is interesting to note that both of the TEM pictures were taken at the same magnification. The TEM analysis (below right) reveals a remarkable difference in grain-size distribution, indicating that a different production process was used for manufacturing the PuO₂.



The age of the plutonium material was determined by gamma spectrometry (bulk sample) and by SIMS (both particles types). The adjacent uranium particles interfered in the SIMS measurements, leading to biased results for the plutonium-238/uranium-234 and the plutonium-239/uranium-235 ratios (isobaric interferences for uranium-238 and plutonium-238, and for uranium-235 from uranium particles and uranium-235 from plutonium-239 decay). Because uranium-236 is a minor isotope in the uranium material, its interference with the grown-in uranium-236 from plutonium-240 decay was negligible. The ages determined for different particle types from the plutonium-240/uranium-236 ratio were similar (within the uncertainties) and they were consistent with the age obtained from the bulk measurement of the plutonium-241/americiu-241 ratio by gamma spectrometry. Both methods gave a production time around the end of 1979 \pm 0.5 years.

Even though the plutonium-239 enrichment is somewhat too low for military purposes, it is not impossible to produce a nuclear device with plutonium of this quality. With regard to the lithium metal, its high enrichment in lithium-6 of 89.4% is noteworthy. One of the possible uses of lithium-6 is to generate energetic tritons via the ${}^6\text{Li}(n,\alpha){}^3\text{T}$ reaction. Such energetic tritons would then be able to initiate deuterium-tritium nuclear fusion in a thermonuclear weapon. Therefore, it may not be a coincidence that plutonium and lithium-6 were found together.

Third case: radioactive waste

In July 2001, plutonium was found in a routine urine analysis of an employee who had been working in a shut-down reprocessing plant under decommissioning in Karlsruhe, Germany. His car and apartment were also found to be contaminated. In addition, his girlfriend and her daughter had incorporated americium and cesium. The employee was arrested and confessed that he had stolen a plastic vial containing a liquid and a swipe cloth. He had managed to get both items out of the reprocessing plant about half a year earlier.

The analytical task was two-fold: first, to confirm that the reprocessing plant in question was really the source of the material; second, to verify whether the two stolen items were the only sources of the contamination and the incorporation. Besides the two stolen items, analyzed samples included vacuum cleaner bags from the contaminated apartments, household gloves used to handle the stolen items, and clothing.

All samples were first measured by gamma spectrometry. The plastic vial contained

Theft of radioactive waste. A plastic vial containing a liquid and a swipe cloth, which were stolen from a decommissioned reprocessing plant in Karlsruhe, Germany, were first analyzed by gamma spectrometry. The vial was found to contain plutonium-238, -239, and -241; americium-241; cesium-134 and -137; and antimony-125. In addition to these elements, europium-154 was also found in the swipe cloth (cellulose fibers from the swipe cloth are shown at bottom left and nylon fibers at right). To quantify the uranium and plutonium isotopes, parts of the samples were dissolved in nitric acid and measured by thermal ionization mass spectrometry (TIMS), inductively coupled plasma mass spectrometry (ICP-MS).





Classical forensics on contaminated items. The nuclear forensics team at the Institute for Transuranium Elements (ITU) in Karlsruhe is helping police departments learn how to perform investigations on radiologically contaminated evidence. The classical technique of taking fingerprints or DNA samples from evidence becomes even more challenging when it has to be performed in a glovebox.

plutonium-238, -239, and -241; americium-241; cesium-134 and -137; and antimony-125. In addition to these elements, europium-154 was also found in the swipe cloth. The other items contained the same nuclides in slightly lower activities. To quantify the uranium and plutonium isotopes, part of the samples was dissolved in nitric acid and measured by TIMS and ICP-MS.

The isotopic compositions of plutonium and uranium were similar in all samples and resembled the spent fuel last reprocessed in the plant before shutdown. The large amount of cesium ingested by the thief's girlfriend was difficult to explain from the activity found in the two stolen items. However, the items were most probably washed before being transferred for the investigations. Because cesium is fairly soluble in water, most of the cesium might have been lost at this stage. The thief was sentenced to prison for breaking the security regulations of the reprocessing plant and for unauthorized possession of radioactive material. Decontaminating the two apartments cost about \$2.5 million.

Current developments

The examples presented here are typical cases analyzed at ITU. A pellet case is often easier to solve than a powder case because information on commercial nuclear fuels is available in ITU's database. Powder is usually not a final product but is an intermediate product or not from a commercial production cycle. To make the origin determination more accurate, researchers are continuously studying samples of known origins.

Existing analytical techniques, as used in material science, nuclear materials safeguards, and environmental analysis, have been adapted to the specific needs of nuclear forensic investigations. Characteristic parameters (e.g., isotopic composition, chemical impurities, and macro- and microstructure) can be combined into a "nuclear fingerprint" pointing at the origin of the material. Further research is being carried out aimed at identifying other useful material characteristics to reduce the ambiguities often remaining in the interpretation of the data and in the source attribution.

The new science of nuclear forensics has also required a change in how police conduct investigations. Using classical forensics techniques on contaminated items must be done in a controlled environment and with proper radiological protection. ITU has helped the police develop procedures for crime-scene management and has set up a dedicated glove box for taking DNA samples and fingerprints from contaminated items.

Response to trafficking of nuclear material

Nuclear forensics is a crucial component of a comprehensive response to nuclear material trafficking. The response measures require a collaborative effort on an international level. ITU, together with several eastern countries of the European Union and countries in the Commonwealth of Independent States

(CIS), has set up projects to increase the efficiency in combating trafficking. A comprehensive approach has been developed that involves all competent authorities in the individual countries.

Assistance is offered to develop a national response plan that is consistent with the Model Action Plan recommended by the Nuclear Smuggling International Technical Working Group (ITWG) on combating nuclear terrorism. The concept of a national response plan has been taken over by the IAEA, put on a broader basis, and promoted for implementation. Training sessions have been offered to law enforcement officers and scientists and demonstration exercises have been carried out in different countries to test the implementation of the Model Action Plan. In a final step, joint analyses of seized samples have been conducted by ITU and requesting countries to demonstrate the preparedness and usefulness of nuclear-forensic analysis.

These efforts are coordinated with other international activities, in particular by the United States and the IAEA, to make efficient use of available resources. On the scientific level, the ITWG serves as a forum for the exchange of experience, advancing nuclear forensics, and interacting with regulatory bodies, law enforcement, and measurement scientists. Nuclear forensics provides an element of sustainability in the fight against trafficking of nuclear material.

FURTHER READING

K. Mayer, M. Wallenius, and I. Ray, "Nuclear forensics—A methodology providing clues on the origin of illicitly trafficked nuclear materials," *The Analyst* **130** (2005).

M. Wallenius, K. Mayer, and I. Ray, "Nuclear forensic investigations: Two case studies," *Forensic Science International* **156**, (2006).

M. Wallenius, et al., "Nuclear forensic investigations with a focus on plutonium," *Journal of Alloys and Compounds* **444-445** (2007).

NNSA SELECTS LOS ALAMOS AS PREFERRED ALTERNATIVE SITE

This article was contributed by David L. Clark, Seaborg Institute, and Joseph Martz, Weapons Program Directorate.

Los Alamos has been selected as the National Nuclear Security Administration's preferred alternative site for plutonium research, development, and manufacturing, along with nuclear weapons design and engineering, and supercomputing. These areas of emphasis for Los Alamos are part of a national plan to transform the nation's weapons complex to be more responsive to emerging threats.

The preferred alternative selection confirms that Los Alamos is first and foremost a science research and development Laboratory. The Laboratory is the nation's choice for materials-centric national security science that relies on effective integration of experiments with exceptional theory, modeling, and high-performance computing. Interdisciplinary excellence in theory, modeling, and simulation with experimental science and nuclear science continue to provide the Laboratory with innovative and responsive solutions to broad national security challenges through the agile, rapid application of key science and technology strengths.

Weapons design and engineering: Los Alamos provides the fundamental science-based understanding of nuclear weapons physics and engineering performance. It is this basic understanding that is the basis for confidence in the nation's nuclear deterrent without the need for further nuclear testing. Los Alamos' design and engineering of both nuclear and nonnuclear weapons components is enabled through small-scale experiments, nonnuclear hydrotests, and subcritical experiments, relying on the full spectrum of scientific excellence across all disciplines, with a focus on materials, high-explosives chemistry, and shock physics.

Plutonium research, development, and manufacturing: Los Alamos has a long and successful history in actinide science and limited plutonium manufacturing that supports a credible, sustainable nuclear deterrent. The Laboratory's expertise in the production, handling, and processing of nuclear and nonnuclear materials makes it the best, most logical site for future limited plutonium manufacturing. The Laboratory is the world leader in actinide science — the exploration of the elements from thorium to lawrencium with particular emphasis on uranium and plutonium, a set of elements on the frontier of scientific inquiry. Los Alamos' scientists publish more than 300 studies a year with a focus on the actinide elements.

In 2007, the Laboratory delivered the first war reserve W88 pit in nearly twenty years with small-scale plutonium experiments, legacy test data, groundbreaking materials science, extensive statistical analysis, adapted computer weapons codes, and a refined manufacturing process that results in increased efficiencies and lower costs.

Los Alamos' Seaborg Institute for Transactinium Science investigates the science that underpins energy security; nuclear power generation; and the production, purification, characterization, analysis, and eventual disposal of actinide elements. The Laboratory also supports actinide research



in physics, chemistry, metallurgy, theory, modeling, and experimental technique development.

New facilities, such as the Chemistry and Metallurgy Research Replacement building, now under construction, along with materials consolidation, means that the nation's special nuclear materials inventory can be protected to meet the security challenges of the 21st century. Additionally, leading-edge new technologies alongside the latest in best practices and procedures will further enhance the Laboratory's already rigorous approach to worker safety, health, and security.

Research-driven supercomputing: Computer modeling and simulation, supported by experimental data and using some of the world's most powerful supercomputers, are central to understanding weapons performance in the absence of nuclear testing. The Laboratory has a suite of supercomputing assets, led by "Roadrunner," slated to be the first computer in the world to operate at sustained petaflop speeds. Phase 3 of Roadrunner is a unique hybrid petascale system, a very large cluster of nodes linked together at high speeds. Each computer node in this cluster consists of two AMD Opteron™ dual-core processors plus four Cell™ processors used as computational accelerators. The Cell processors used in Roadrunner are a special IBM-developed variant of the Cell processor used in the Sony PlayStation 3®. The Laboratory's supercomputing assets also enable research of broader scientific questions related to complex systems like Earth's weather, disease pandemics, and the security of the U.S. electricity grid. Los Alamos will continue to be at the forefront of high-performance computing, exploring advanced architectures, operating systems, and applications.

Broader national security missions: The Laboratory's capabilities in the areas of weapons design, plutonium research, and research-driven supercomputing as outlined above also support a broader set of national security challenges. As the preferred site, Los Alamos would continue its ability to respond quickly to emerging threats and support a broad spectrum of mission objectives in stockpile stewardship, nuclear-energy research, nuclear forensics, nuclear safeguards, and counterterrorism. Large-scale modeling and simulations with broad experimental science capability allow Los Alamos to address challenges such as biothreats, climate change, and infrastructure security. At the same time, world-class nuclear facilities enable waste minimization and environmental cleanup.

Emerging national security challenges also require the Laboratory to advance its scientific user-facility infrastructure and to attract and retain the best talent. Currently in development is a set of facilities called MaRIE, or Material-Radiation Interaction in Extremes. The purpose of MaRIE is to provide tools that will allow the Laboratory to address the critical materials-related scientific questions relevant to a broad spectrum of current and future missions.



HEAT CAPACITY AND THE GOOD OLD DAYS OF URANIUM

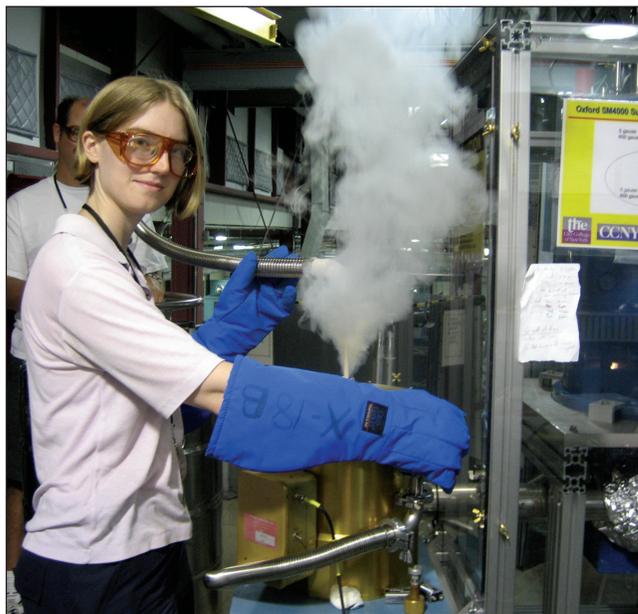
Editor's note: ARQ continues with its series of articles on Seaborg Institute postdocs. This article is authored by Susan Cox, who works in the Materials Physics and Applications Division's National High Magnetic Field Laboratory (MPA-NHMFL). She received her doctorate from the University of Cambridge, Department of Materials Science, in December 2005. Cox immediately began her postdoctoral appointment with her mentor John Singleton at the MPA-NHMFL.

INTERPRETING THE CHARGE-DENSITY WAVE IN LOW-TEMPERATURE ALPHA-URANIUM

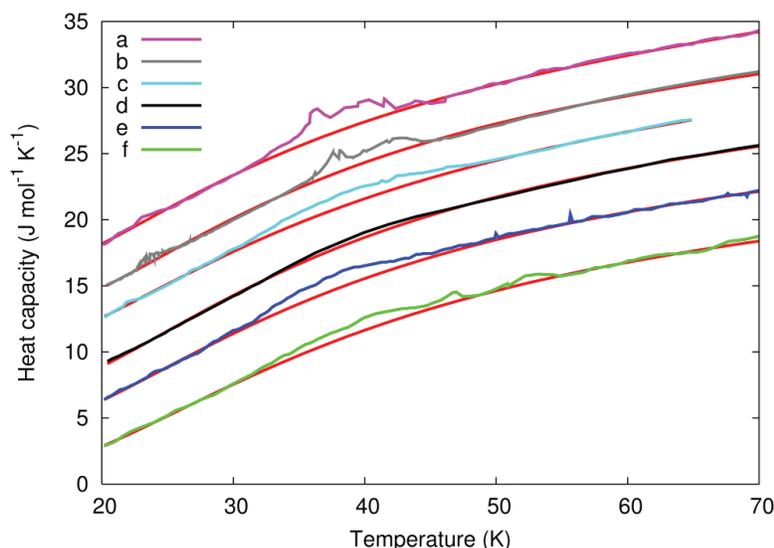
Over the past forty years, neutron, X-ray, electron diffraction, and heat-capacity experiments have indicated that a charge-density wave occurs at low temperatures in alpha-uranium (α -U). This charge-density wave is considered unusual because the wave vector has components in all three dimensions. Despite numerous investigations, there has been a lack of unanimity on how these results should be interpreted. In particular, there has been discussion of the importance of surface effects in transmission electron microscopy measurements and the effects of spatial averaging in X-ray and neutron diffraction. Confusion about the interpretation of heat-capacity data has continued to the present day.

In collaboration with Los Alamos colleagues Edward Rosten, Ross McDonald, and John Singleton, I have shown that the different results can be understood in the context of a single model if peaks in the heat-capacity data are interpreted to arise from “dirty Peierls transitions”: transitions to a charge-density wave state, or changes in the periodicity of that state, in a disordered system. When analyzed in this way, it is also possible to use heat-capacity data as a noninvasive method for bulk microscopy of disorder in the sample. This method is potentially an important step forward in assessing uranium sample quality. Our analysis of historical data indicates that sample quality was highest in the 1950s, showing that those who complain that actinide crystals were better in the “good old days” are actually correct.

Seaborg Institute postdoc Susan Cox fills a bolometer with liquid helium while Mike Klopf of Jefferson Laboratory looks on. The bolometer allows the intensity of light at different wavelengths to be monitored after it has passed through a sample, which sits in a magnet (pictured at far right) that has been installed on an infrared beam line at Brookhaven National Laboratory. The magnet will enable researchers to make magneto-optical measurements to probe how the ground and excited states of actinides and actinide compounds change with magnetic field. The vapor cloud is helium gas, which results from the liquid helium being transferred into a relatively warm metal pot (the bolometer).



Randy Miller



When looking at low-temperature α -U heat-capacity data in the figure above, it is easy to see that there is a smooth background above which three peaks are visible in the single-crystal data and one peak is visible in the polycrystalline data. Such peaks are usually associated with phase transitions, and because it is known that a charge-density wave forms in α -U, one would expect each of the peaks to be attributable to either the appearance of a charge-density wave or a change in the wave vector (periodicity) of the charge-density wave.

The highest-temperature peak is close to 42 kelvin (K), a temperature at which neutron and X-ray experiments show all three components of the charge-density wave appearing with incommensurate wave vectors. The second peak, around 38 K, is close to the point at which the x component of the charge-density wave locks into a value of $q_x = 0.5a^*$. At 22 K the y and z components of the charge-density wave lock into $q_y = 1/6b^*$ and $q_z = 2/11c^*$ (from X-ray measurements) or $q_z = 5/27c^*$ (from neutron measurements). Different combinations of these wave-vector components give rise to two different charge-density wave vectors, $(q_x, +q_y, +q_z)$ and $(q_x, +q_y, -q_z)$.

Transmission electron microscopy studies have discovered that in any area of the sample only one of the wave vectors exists (i.e., the two wave vectors never coexist). However, the results also challenged the simple picture from X-ray and neutron diffraction, which found that the q_x component of the wave-vector never became fully commensurate.

Neutron, X-ray, and electron diffraction all measure the wave vector of the charge-density wave accurately, although neutron and X-ray techniques average over a large volume and transmission electron microscopy only gives information over very short length scales. However, as can be seen from the description of the measured values of the wave vectors, all three techniques produced conflicting results.

Heat capacity of alpha-uranium (α -U) and the modeled background for various samples. Legend: (a) pseudo single crystal U PSC 91 (Crangle and Temporal, 1973), (b) polycrystalline sample (Hall and Mortimer, 1977), (c) single crystal (Mihaila et al., 2006), (d) polycrystalline sample (Mihaila et al., 2006), (e) polycrystalline sample U10 (Crangle and Temporal, 1973), and (f) polycrystalline sample U MM (Crangle and Temporal, 1973). The data are offset from each other by $3 \text{ J mol}^{-1} \text{ K}^{-1}$. When looking at low-temperature α -U heat-capacity data, it is easy to see that there is a smooth background above which three peaks are visible in the single-crystal data and one peak in the polycrystalline data.

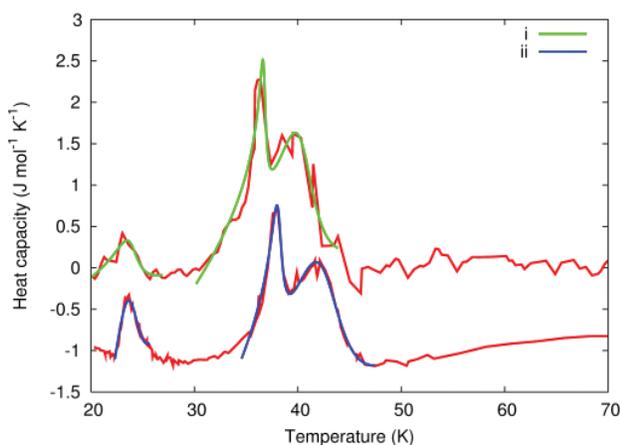
Heat capacity above background and disordered Peierls transitions fits to the data. Legend: (a i) pseudo single crystal U PSC 91 (Crangle and Temporal, 1973), (a ii) polycrystalline sample (Hall and Mortimer, 1977), (b i) single crystal (Mihaila et al., 2006), (b ii) polycrystalline sample (Mihaila et al., 2006), (b iii) polycrystalline sample U10 (Crangle and Temporal, 1973), and (b iv) polycrystalline sample U MM (Crangle and Temporal, 1973). The data are offset from each other by $1 \text{ J mol}^{-1} \text{ K}^{-1}$. Three transitions were found in the single crystals as expected (below left), and in the polycrystalline samples an excess heat capacity was evident in the temperature range where the upper two transitions were observed in the single crystal, with the third transition being absent (below right).

It has long been known that the properties of different uranium samples vary, and it was thought that this probably led to the differing conclusions. But there was no noninvasive method for measuring properties such as the presence of impurities or grain boundaries, which might influence the results. Transmission electron microscopy can be used to measure the properties of a sample, but it damages the sample and observations can be dominated by surface effects. In addition, because of the radioactive nature of uranium, a dedicated microscope is needed, of which there are only a few worldwide.

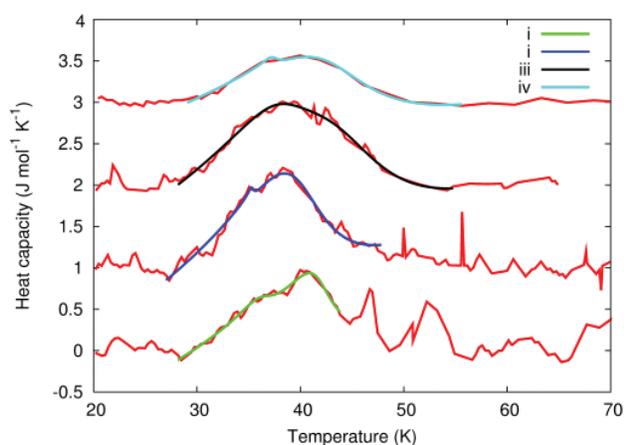
There have been a number of studies of the heat capacity of α -U; the data that we consider here come from studies by Hall and Mortimer (1977), Crangle and Temporal (1973), and Mihaila and others (2006). To make the transitions more visible, a smooth background was removed from the data. The calculation of the high-temperature background requires the Debye temperature, which was obtained by fitting the low-temperature (1.8–10 K) heat-capacity data to a polynomial equation. The high-temperature data were fitted with a Debye model and an Einstein mode, where the Debye temperature obtained from the low-temperature fitting was used.

The data were fitted with iterative reweighted least squares using the Levenberg–Marquardt method. This method has the advantage over the more commonly used least-squares technique that low weights are automatically given to areas that have a poor fit to the model, eliminating the need for sections to be cut out of the data by hand before they are fitted. The heat capacity above background is shown in the figures below. Three transitions were found in the single crystals as expected, and in the polycrystalline samples an excess heat capacity was evident in the temperature range where the upper two transitions were observed in the single crystal, with the third transition being absent.

The heat capacity above background was fitted to a model of a Peierls transition, which is the type of second-order transition that occurs when a charge-density wave is formed. The model for a Peierls transition occurring in a



a



b

disordered material (a dirty Peierls transition) was developed by Chandra (1981) and gives the form of the transition as a function of the level of disorder. The only fit parameters are the disorder length scale and a scale factor. It can be seen that this model fits well in the single-crystal materials. In the polycrystalline materials, the heat-capacity peak can be well modeled as two Peierls transitions with greater disorder than in the single crystals. The third transition observed in the single crystal is entirely absent in the polycrystalline samples. The fits to the transitions give a disorder length scale for each transition and are displayed in the table at right.

The disorder length scale gives an idea of the average distance that separates impurities or physical dislocations in the sample. Therefore, when the sample is better quality and so more ordered, this length scale will be longer; when the sample is poorer quality and so more disordered, this length scale will be shorter. Because all three transitions in one sample are occurring in the same disorder environment, one might expect the disorder parameters to be the same for all three transitions.

However, the disorder parameter indicates the length scale at which the disorder is affecting the transition; the three different transitions will each react differently to the various impurities and grain-boundary dislocations present. Because the same transitions are occurring in the different samples, it is possible to compare how the disorder length scale of a given transition varies from sample to sample. Some caution must be used with the results from polycrystalline samples because the problem is somewhat ill constrained. It is safest to look at the total of the two transitions rather than placing too much weight on the individual results for each length scale.

Because the length scale over which the charge-density wave orders at the third transition is much larger than that for the upper two transitions, it should be the transition most affected by disorder, explaining its disappearance in the polycrystalline samples. This result is consistent with the transmission electron microscopy observation that the charge-density wave is incommensurate at low temperatures (although the study was performed on a thin-film sample, and a considerable amount of disorder is usually introduced during the sample preparation stage).

This work is important because it provides a way of assessing the disorder level in a crystal using a nondestructive technique. It is particularly useful because transmission microscopy is problematic for radioactive materials and can give misleading results. By contrast, it is relatively easy to make probes for heat-capacity measurements that can contain radioactive material to an acceptable standard.

Our analysis method allows different uranium samples to be compared and gives a good idea of the relative quality of different single-crystal samples. From the characteristic disorder length scales given in the table, we can see that the

	L1 (Å)	L2 (Å)	L3 (Å)
U single crystal (U PSC 91, Crangle and Temporal, 1973)	21.0	53.5	21.3
U single crystal (Mihaila et al., 2006)	17.9	40.1	19.0
U polycrystal (U 10, Crangle and Temporal, 1973)	19.4	20.6	absent
U polycrystal (U MM, Crangle and Temporal, 1973)	14.0	53.7	absent
U polycrystal (Hall and Mortimer, 1977)	11.8	13.6	absent
U polycrystal (Mihaila et al., 2006)	12.8	33.0	absent

Disorder length-scale parameters, in angstroms (Å), for different alpha-uranium (α -U) samples. Names and dates refer to the heat-capacity measurements from which the parameters were extracted.

single crystal with the longest disorder length scale, i.e., the least disorder, was the sample U PSC 91, which was grown by E.S. Fisher in 1957. The fact that the highest-quality sample was probably the oldest indicates the difficulty of uranium metallurgy and the little attention that this problem has received in recent decades.

The charge-density wave transitions in both single-crystal and polycrystalline α -U can be modeled as Peierls transitions in systems containing different levels of disorder. The disordered Peierls model links the previously contradictory pictures that arose from transmission electron microscopy studies and X-ray or neutron studies. We have recently found similar effects in another compound, the lanthanum-calcium-manganese-oxygen compound $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$, in which a charge-density wave coexists with a high level of disorder.

In the future, we hope to look at the low-temperature behavior of the heat capacity of α -U because it may be possible to observe the collective mode behavior at low temperatures. In addition, we will further explore the more-general characteristics of charge-density waves in disordered materials in other related systems.



Plutonium Futures "The Science" 2008

*Supporting safe and secure nuclear research
as part of the world energy mix*

Dijon, France 7 - 11 July, 2008



Registration is under way for the Plutonium Futures "The Science" 2008 conference. The 2008 conference, the fifth in the series, will be held in Dijon, France, July 7-11. The deadline for registration is May 15. Complete details on registration, deadlines, and venue can be found on the conference website at www.pu2008.org.

The Plutonium Futures conferences provide an international forum for the presentation and discussion of current research on the physical and chemical properties of plutonium and other actinide elements. Conference topics will include condensed matter physics; materials science; fuel cycle issues; surface, interfaces, colloids, and corrosion; plutonium and actinide chemistry; and detection and speciation analyses.

Former Laboratory Director and Senior Fellow Sig Hecker is an honorary chair. Three Los Alamos researchers are members of committees: David E. Hobart of Actinide Analytical Chemistry (C-AAC), is on the program committee, and David L. Clark, director of the Seaborg Institute, and John Sarrao, leader of the Materials Physics and Applications Division, are members of the international advisory committee.

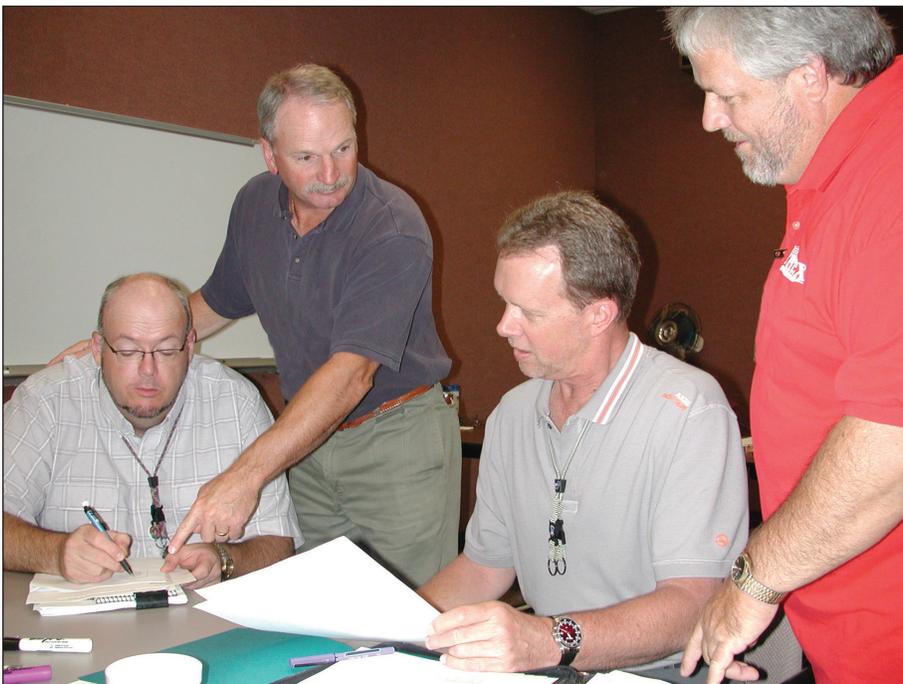
Organizers of the 2008 conference include the Atomic Weapons Establishment (AWE), Commissariat à l'Énergie Atomique (CEA), and the Institute for Transuranium Elements (ITU). Previous conferences have been held in Pacific Grove, Calif. (2006); Albuquerque, New Mexico (2003); and Santa Fe, New Mexico (2000 and 1997).

ATOMICS: CHANGING SAFETY BEHAVIOR IS NO SMALL MATTER

“Do Work Safely.” the DOE’s safety mantra is more than a slogan. It is based on the DOE’s Integrated Safety Management (ISM) guidelines that Los Alamos and other DOE sites must follow. Safety is critical in every activity performed at Los Alamos, but nowhere is it more important than in the special nuclear materials operations performed at facilities within the Stockpile Manufacturing and Support Directorate (SMS).

Integrating science and technology with improved safety practices is critical to the SMS Directorate’s meeting its performance goals as well as meeting its commitments to DOE and NNSA for national security missions in support of nuclear deterrence, energy security, and homeland security. Protection of its workers, the public, and the environment is of paramount importance to SMS. For this reason, managers, technicians, engineers, and support personnel must all view themselves as members of the same team.

SMS has enhanced the behavior-based safety observation process, ATOMICS (Allowing Timely Observations Measures Increased Commitment to Safety), and integrated it with Human Performance Improvement (HPI) principles and Management Observation and Verification (MOV) safety tools. The goal is to reinforce safe behaviors and eliminate at-risk behaviors and conditions for all employees and operations (both programmatic and facility) at TA-55 (where the Plutonium Facility is located), and other sites across the Lab where SMS employees and operations are located.



Jim Kleinsteuber (third from left), ATOMICS facilitator, and Todd Conklin (far left), senior advisor for safety improvement initiatives at Los Alamos, review safety data during a trip to Pantex in Amarillo, Texas, to benchmark Los Alamos’ behavior-based safety observation process. Also pictured are Lonnie Flake (second from left) and Bud Eaton (far right), both of whom are behavior-based safety facilitators at Pantex.

HPI focuses on the aspects of work processes, organizational systems, and the work environment that have significant impact on human performance. It is the foundation of a successful program developed for the nuclear power industry by the Institute of Nuclear Power Operations (INPO) and has been proven highly successful at improving performance, efficiency, and worker safety. MOV is a process by which managers observe workers and work, talk with the workers, and verify that corrective actions are completed. The DOE has integrated HPI principles and tools into its ISM manual, and the SMS Directorate in turn has applied ISM principles to its own safety-observation tool, ATOMICS.

ATOMICS was introduced seven years ago in the Nuclear Materials Technology Division with mixed success. At its inception, ATOMICS was an employee-driven program in which workers focused on behaviors by observing their peers, and managers focused on conditions by conducting walk-arounds. Hundreds of employees were trained as observers who identified thousands of safe and “at-risk” behaviors.

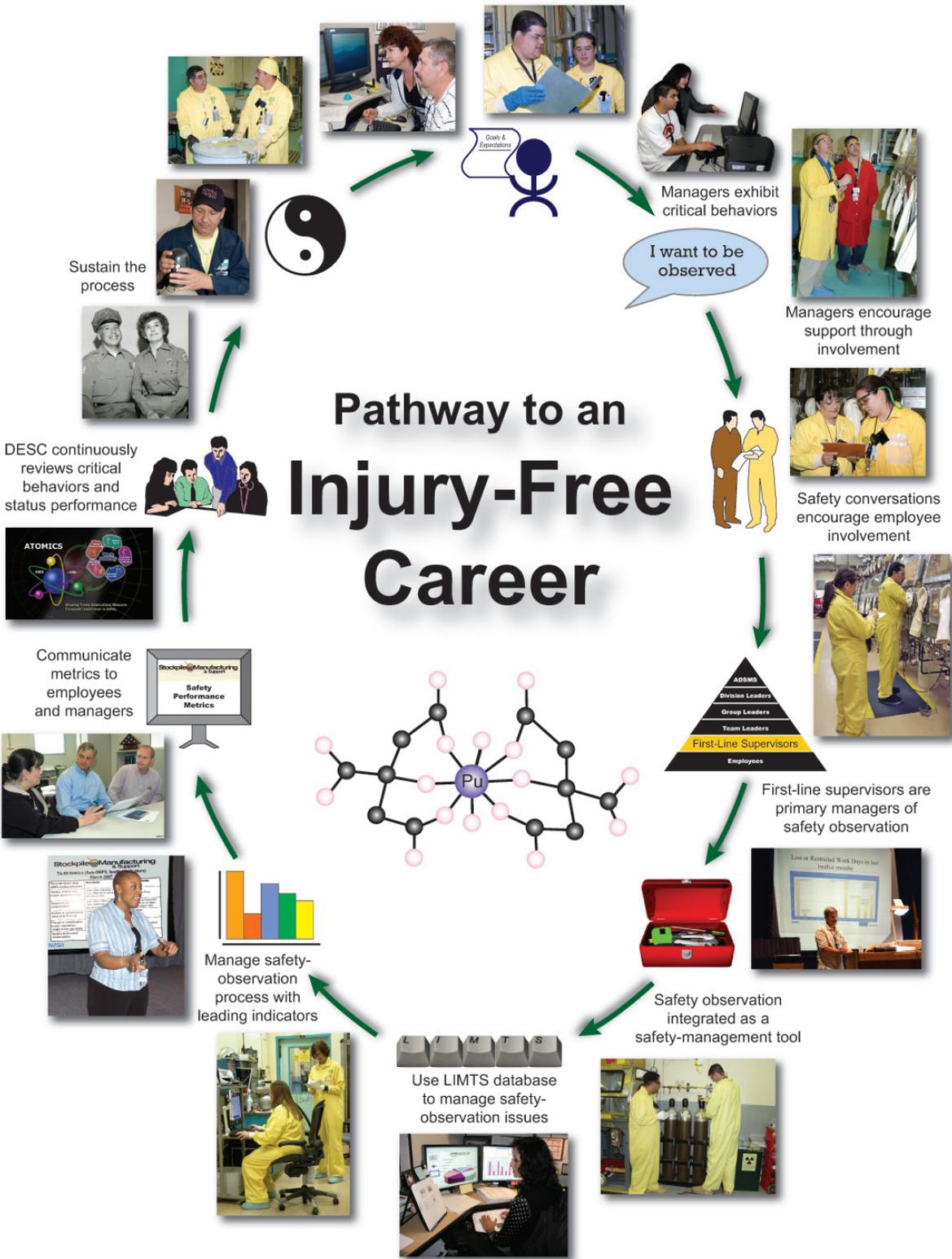
ATOMICS is now a management-driven process owned by all SMS employees. “Full participation is the expectation in SMS,” said Jim Kleinstauber, ATOMICS facilitator. Management will be responsible for process sustainability, and participation may be tracked in a worker’s performance objectives.

The enhanced ATOMICS puts additional emphasis on data analysis and using data to identify root causes for injuries and to track safe work practices. As observations are made, those requiring action that can’t be immediately resolved at a team or group level are placed into the LANL Issue Management and Tracking System (LIMTS) and are tracked until they are resolved.

“ATOMICS is gearing up to collect and analyze data unlike anyone else’s, and a major reason for that is we have recently been established as a pilot observation process for the institution,” said Kleinstauber. “Our hope is to maintain a strong and robust observation program at SMS that will move the directorate toward achieving best in class in safety performance, and at the same time, positively complement the Lab’s quest for Voluntary Protection Program (VPP) recognition in September 2009.”

A recent trip to Pantex in Amarillo, Texas, where a similar program is in place, was used to benchmark the Los Alamos process. “We were told by Pantex facilitators that LANL is a few steps ahead in some areas,” said ATOMICS facilitator Maryrose Montalvo. “LANL is moving faster on merging Human Performance Improvement principles with behavior-based principles, but we need to work on adding conditions. The scope of observation includes the entire job, not just worker behavior. We also need to perform more observations; we’ve done about 9,600 observations versus 65,000 to 70,000 at Pantex.”

Carl Beard, acting SMS associate director, is confident that ATOMICS can be an effective tool for improving safety posture. He and Mike Mallory (former Pantex Plant manager and now Laboratory principal associate director for Operations and Business) are familiar with the success of the program and saw excellent results when they worked at Pantex. Many other DOE sites use



behavior-based safety programs, including Sandia, Strategic Petroleum Reserve, Savannah River, Livermore, Lawrence Berkeley, Oak Ridge, and Argonne.

Safety observation is one strategy that the Laboratory is using to improve processes such as Integrated Work Management (IWM) (IMP300.4), which is undergoing revitalization. Safety performance and processes are being analyzed as part of the implementation of DOE's "Worker Safety and Health Rule 10 CFR Part 851," a new program that establishes management responsibilities, worker rights, safety and health standards, and required training. The new order replaces the Contractor Requirements Document (CRD) of DOE O440.1A "Worker Protection Management for DOE Federal and Contractor Employees."

One enhancement to ATOMICS is the creation of eight observation cards that are hazard and task oriented, versus the single card previously used. Specific cards are used to observe facility conditions, glovebox work activities, good catch/near miss, ergonomics, and outside observations and conditions in all areas of SMS operations. Data from observation cards are entered into a web-based recording and management system called Total Observation Process (TOPs) and are reviewed by the Data Analysis Support Team (DAST), which is made up of employees from throughout SMS. The DAST will recommend improvements on data collection and analysis, evaluate safety management tools, recommend actions to improve communication between managers and workers, and identify and monitor leading (it could happen) behavior and lagging (it's already happened) behavior and job-context indicators.

"Safety is a shared vision between managers and workers alike," said Kleinsteuber. "The goal is to have not just an injury-free workplace, but an injury-free career for everyone." But it must also be understood that while an overall performance objective might be zero injuries or events, the most successful safety programs are built on a foundation of teamwork among all employees and managers.

This shared vision can be summed up in two commitments: management will provide the best possible safety training, processes, and equipment; and all employees will be safe and will participate in initiatives such as safety observation. To meet these commitments, SMS will use a variety of resources to accomplish three objectives: engage management at all levels to improve the safety of the workplace, improve the overall working environment, and increase worker safety awareness and empowerment in a non-threatening and non-disciplinary manner.

The goal is to have all of the directorate's employees actively participating and performing observations and self-reporting by the end of January 2008. "We must work to ensure that the scientific, programmatic and safety cultures all mesh effectively and efficiently," said Beard. For more information on the ATOMICS program, visit the web site at <http://lanl.gov/orgs/adsm/atomics> or contact Kleinsteuber or Montalvo at atomics@lanl.gov.

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