

## **Nuclear forensics in law enforcement applications**

**P. M. Grant,\* K. J. Moody, I. D. Hutcheon, D. L. Phinney, R. E. Whipple, J. S. Haas, A. Alcaraz,  
J. E. Andrews, G. L. Klunder, R. E. Russo, T. E. Fickies, G. E. Pelkey, B. D. Andresen,  
D. A. Kruchten, S. Cantlin**

*Forensic Science Center, L-178, Livermore National Laboratory, Livermore, CA 94550, USA*

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Over the past several years, the Livermore Forensic Science Center has conducted analyses of nuclear-related samples in conjunction with domestic and international criminal investigations. Law enforcement officials have sought conventional and nuclear-forensic analyses of questioned specimens that have typically consisted of miscellaneous metal species or actinide salts. The investigated activities have included nuclear smuggling and the proliferation of alleged fissionable materials, nonradioactive hoaxes such as “Red Mercury,” and the interdiction of illegal laboratories engaged in methamphetamine synthesis.

### **Introduction\***

Dismantlement of numerous nuclear weapons by the United States and Russia is resulting in sizable quantities of surplus nuclear materials. Existing controls on some of these materials are being stressed by the sheer magnitude of the effort and by the tumultuous evolution of the former Soviet Union. The increasing availability of highly enriched uranium (HEU) and  $^{239}\text{Pu}$  on the international black market has highlighted the potential for nuclear proliferation by rogue nations, as well as the danger of nuclear terrorism.<sup>1,2</sup> Although illegal nuclear material trafficking and smuggling incidents to date have principally involved hoaxes or sales of nonexplosive radioactive substances,<sup>3</sup> the interdiction of non-weapons-grade nuclear materials by law-enforcement personnel has markedly increased in Europe of late. These species have included nuclides such as depleted- $^{238}\text{U}$ , radioactive substances from smoke-detector and chemical sensors, and other industrial/medical isotopes (such as  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ ).

Nuclear scams that have led to forensic analyses of suspect specimens in our laboratory have included the “Red Mercury” phenomenon, sales of depleted uranium (promoted as HEU), and miscellaneous, misrepresented metals potentially derived from nuclear industry and proffered as weapon components. In addition, thorium compounds can be utilized as catalysts in illegal drug synthesis, and analyses of diverse samples for Th, U, and other actinide contents have occasionally ensued after the seizure of clandestine laboratories by law enforcement. This paper provides some detail in each of these nuclear-forensic effort areas.

### **“Red Mercury” analyses**

A relatively notorious nuclear hoax, first promulgated in 1979 and very active during the 1990s, is “Red Mercury.” This material has been alleged to furnish an incredible component for a nuclear bomb, purportedly allowing ignition of a thermonuclear reaction in tritium without a prerequisite fission trigger. Red Hg has also been variously advertised as an ingredient in stealth technologies, gravity bombs, and missile-guidance systems, and its selling price has been listed at approximately \$500,000 per kg.<sup>4</sup> Black-market sales of Red Hg are often accompanied by extraordinary parameters for physical properties of the substance, and such technical fallacies are regularly encountered in frauds concerning the illicit trafficking of nuclear materials.

In one investigation, two questioned samples declared to be Red Hg specimens were submitted for forensic characterization. The outer sample container was a thick, taped polyethylene bag, within which was a plain brown mailing envelope with no writing or other distinguishing features. Inside the brown envelope were the two questioned specimens, which appeared to be liquid Hg contained in small-gauge, plastic, syringe-needle holders. The Hg contents of the samples were 3.647 and 4.868 g. The needle holders were wrapped in Al foil, and some leakage of the liquid Hg was evident. Also present in the envelope and Al foil were small grey flecks of a flocculent, metallic material, perhaps an amalgam formed by chemical reaction(s) between Hg and Al.

\* E-mail: patgrant@llnl.gov

*Inorganic analysis*

We conducted a qualitative, elemental investigation, spanning the periodic table from  $Z=20$  (Ca) to  $Z=94$  (Pu) and sensitive to  $\mu\text{g/g}$  (ppm) concentration levels, on the two liquid specimens using X-ray fluorescence (XRF) spectroscopy. The results of these inorganic analyses indicated that the two questioned specimens were predominantly Hg, with minor amounts of Cd, Ag, and Zn (and a trace of Pb) also evident. We also analyzed a sample of very pure “bright Hg” for comparison purposes. The XRF signature of this substance was equivalent to those of the Red Hg samples.

*Radioisotopic analysis*

Forensic radiochemistry was employed to screen the Red Hg specimens for general  $\gamma$ -emitting nuclides and key isotopes of the diagnostic heavy elements, Ra, Th, U, and Pu. We performed nondestructive nuclear analyses on both samples. They were counted overnight on high-efficiency, low-background, 4096-channel Ge spectrometer systems for any  $\gamma$  emissions between 50–2000 keV. These analyses gave results that were indistinguishable from the normal background levels of the analyzers.

However, the most sensitive investigation of heavy-element composition was accomplished by radiochemical isolation and subsequent counting of extremely pure chemical fractions. Such separations<sup>5–7</sup> were performed on the two questioned specimens and were followed by  $\alpha$ - and  $\gamma$ -spectrometric analyses. Table 1 gives the results. No evidence of heavy-element signature species was found. In fact, our analytic experience with purported Red Hg samples thus far has been consistent with those specimens being nothing more than elemental Hg, HgO, or HgI<sub>2</sub>.

Table 1. Atoms of heavy element per gram of sample

Nuclide	Red Hg #1	Red Hg #2
<sup>226</sup> Ra	$\leq 7.4 \cdot 10^8$	$\leq 5.8 \cdot 10^8$
<sup>230</sup> Th	$(5.8 \pm 4.4) \cdot 10^9$	$\leq 5.6 \cdot 10^9$
<sup>232</sup> Th	$\leq 1.2 \cdot 10^{15}$	$\leq 5.0 \cdot 10^{14}$
<sup>234</sup> U	$\leq 5.5 \cdot 10^{10}$	$\leq 1.9 \cdot 10^{10}$
<sup>235</sup> U	$\leq 1.2 \cdot 10^{14}$	$\leq 6.6 \cdot 10^{13}$
<sup>238</sup> U	$\leq 1.2 \cdot 10^{15}$	$\leq 3.1 \cdot 10^{14}$
<sup>238</sup> Pu	$\leq 2.8 \cdot 10^6$	$\leq 1.1 \cdot 10^6$
<sup>239,240</sup> Pu	$\leq 5.5 \cdot 10^8$	$\leq 2.5 \cdot 10^8$

**Miscellaneous metal analyses****Case Study #1**

Recently, a somewhat more feasible, nuclear-related sample with an illicit connection was submitted for analysis in support of a federal investigation. The specimen was a small fragment of gray metal that was supposedly representative of a larger quantity of material that could be substituted for HEU in a nuclear weapon. We conducted multidisciplinary and comprehensive forensic analyses, incorporating methods of materials science, radioisotopic chemistry, inorganic chemistry, and organic chemistry.

*Materials and methods*

Nondestructive nuclear and elemental analyses of the sample were conducted initially. These included the use of gas proportional counters to determine the presence of any gross- $\alpha$  or gross- $\beta$  radioactivities, as well as high-performance Ge spectrometry to detect any  $\gamma$  emission between 50–2000 keV. In addition, XRF analyses of the specimen surfaces were effected with a <sup>109</sup>Cd isotopic excitation source, Si(Li) detector, and multichannel analyzer system.

Metallurgy and microprobe analyses were performed with a JEOL-733 automated electron microprobe, implementing backscattered electron imaging and wavelength-dispersive quantitative chemical analyses. An energy of 15 keV and intensity of 10–20 nA were used in this study. Sensitive atomic and isotopic interrogations of the sample were conducted via ion-microprobe mass spectrometry with a Cameca IMS-3f ion microscope.

Following extraction of the specimen with ultrapure 3:1 methylene chloride/isopropanol, nonpolar organic analytes were determined by gas chromatography-mass spectrometry (GC-MS) using a Hewlett-Packard model 5890 GC interfaced to a H-P model 5988 quadrupole MS. Separations were made on a 30-m Alltech SE-30 methyl silicone capillary column. Polar compounds were similarly analyzed after derivatization with N,O-bis(trimethylsilyl)trifluoroacetamide.

Sensitive, but destructive, multielemental analyses over the range  $Z=3$  (Li) to  $Z=94$  (Pu) were conducted by inductively-coupled-plasma mass spectrometry (ICP-MS). A portion of the sample was dissolved in ultrapure HNO<sub>3</sub> under Class 100–1000 cleanroom conditions and was processed using ultrapure reagents and Teflon labware. The ICP-MS was a modified VG/Fisons

*Fig. 1.* Photograph of 9-kg, depleted- $^{238}\text{U}$  questioned specimen

Plasmaquad model operating at an incident power of 1350 w. Ion pulse-counting was performed with an ETP model AF563 system at 2550 V and 20-ns deadtime, and data were collected from 5.5–240 amu in a 4096-channel analyzer. Quantitative results were referenced to a 45-element external standard and three added internal standards (Ga, In, and Tl).

### *Results*

The specimen was determined to consist of a rare, but moderately pure, metal that did, in fact, likely originate from a uranium-refining process. Although no fissionable species or weaponization indicators were detected, the sample did display some very unusual properties. These anomalies included lanthanide fractionation and the presence of long, odd-chain fatty acids. A full report on this study is in press.<sup>8</sup>

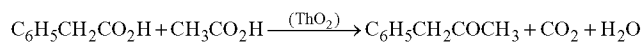
### **Case Study #2**

Another questioned specimen that was submitted for analysis was a massive, radioactive, metal part that a military official had been offered for sale as nuclear weapon material. The sample was consistent with a pervasive swindle, first reported in 1991, and now referred to as the “Southeast Asian Uranium” scam. This piece weighed ~9 kg, had a complex shape, and was evidently intended to attach to another part by means of a bolt (see Fig. 1). Arc-shaped scratches on the surface, surrounding large, circular depressions on opposite sides of the piece, implied that the specimen may have been a component of an assembly that functioned as a hinge (possibly for a shipping container). We performed comprehensive nuclear studies, as well as more conventional forensic analyses, on the sample.

Gamma-spectrometry and XRF analyses revealed that the part was depleted uranium that had been coated with a thin Ni surface layer. The Ni barrier had been penetrated at various locations, and the piece was cast in shape, with no subsequent machining prior to Ni electroplating. The  $^{235}\text{U}$  isotopic content, determined by forensic radiochemistry, was  $\sim 0.3$  wt.%. This value was well below the natural concentration level of 0.71 wt.%, but was too high to be representative of the depleted- $^{238}\text{U}$  modern standard. Radioisotope chronometry placed the date of last purification at  $1961 \pm 3$  years. The density of the part was  $17 \text{ g/cm}^3$ , and electron- and ion-microprobe analyses identified it to be a 10% Mo alloy, with the Ni coating  $>99\%$  pure and  $90 \mu\text{m}$  thick. Utilizing the above forensic information, we determined that the sample was likely a constituent of an aircraft counterweight assembly, possibly of military origin.

### Clandestine laboratory materials analyses

Hot  $\text{ThO}_2$  can be used as a catalyst for the production of phenylacetone (P2P) from phenylacetic and acetic acids.<sup>9</sup>



P2P is an immediate precursor in the synthesis of methamphetamine and is a Schedule II controlled substance.<sup>10</sup> It is legally available only through a Drug Enforcement Administration (DEA) license. Consequently, P2P is manufactured clandestinely in drug laboratories outside of lawful channels. After law-enforcement interdiction of such operations, we have occasionally been requested to analyze suspect materials.

In addition to performing quantitative analyses for Th, we have applied nuclear forensic techniques for other heavy-element isotopes to selected samples as well. For routine DEA applications, such supplementary isotopic data are generally unnecessary. However, for more intricate and intelligence-related investigations, measurements of ancillary radionuclides can provide potentially meaningful information.

For example, the  $^{232}\text{Th}/^{228}\text{Th}$  ratio is an age-dating parameter as it gives the time since the last chemical purification of the specimen. The  $^{230}\text{Th}/^{232}\text{Th}$  ratio provides the concentration of  $^{238}\text{U}/^{232}\text{Th}$  in the parent ore, even if the chemical purity of the Th is such that U is no longer directly measurable in the sample.

In addition, the deviation of certain isotope ratios (e.g.,  $^{226}\text{Ra}/^{227}\text{Ac}$ ) from their natural values can be indicative of the particular chemical process(es) implemented for product decontamination.

Such data could provide collateral information and other indicative leads to law-enforcement investigators. Thus, such intelligence might reveal the source of the Th supply, provide insight into a partial time-history of the material, indicate the likelihood of a common supplier for materials being utilized across multiple illicit laboratories, identify residual "fingerprint" species present at ultralow levels in otherwise pure reagents, and so forth.

We have developed a comprehensive radiochemical procedure for analyses of Th, U, and other potential target analytes of interest in DEA samples. The method accommodates any molecular speciation of the metals (i.e., chemically combined as nitrate, oxide, fluoride, etc.) and any sample matrices that would be possible in confiscated specimens. A report describing these techniques has been submitted for publication.<sup>11</sup>

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