

PREDICTIVE RESEARCH FOR THE DEPARTMENT OF ENERGY'S RADIOLOGICAL TRIAGE PROGRAM

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ABSTRACT

The U.S. Department of Energy's Radiological Triage Program supports first-response teams for the analysis of nuclear data and specializes in interpreting the gamma-ray spectra from portable radioisotope identifiers (RIIDs). Analysts provide rapid assessment of radiological data to ascertain whether a source presents a threat while striving to minimize potential impact on the legitimate movement of people and goods. To serve this mission, it is important that analysts stay knowledgeable about the complex and changing radioisotope industry. New radioisotope products and processes are continually emerging, and each carries a risk of causing innocent alarms and delays. To mitigate this risk, our program proactively researches the production and distribution of these radioisotope products while evaluating RIID performance. This paper discusses some of our recent ventures and findings.

INTRODUCTION

Analysts who work for the Department of Energy's (DOE's) Radiological Triage Program—or Triage—must rapidly and accurately interpret gamma-ray spectra that have been obtained by first responders using hand-held radioisotope identifiers (RIIDs). This requires familiarity with the various RIID instruments, ability to analyze the spectra even when collected under poor conditions, and extensive knowledge about the types of radiological materials that may be encountered in vivo or within a shipment. The majority of radioisotope products encountered are routine and well understood by RIID engineers and emergency responders. However, the scope of radioisotopes being used in the industrial and medical fields is broad and dynamic, and identifying the emerging or less-prevalent radioisotopes often requires assistance from the Triage analysts. Furthermore, evolving production methods of commercial radioisotopes often result in unexpected trace isotopes that can confuse RIIDs and analysts. Familiarity with the uses and radiation signatures of these products is essential to minimizing the impact on the legitimate movement of people or goods. It is imperative to collect high-fidelity data in order to predict what to expect when a particular product is encountered in a field environment.

Predictive research in the Triage program involves collecting information and spectral data for rare or emerging radioisotopes that might be encountered by a first responder, with a focus on those cases that are most likely to be encountered or that could be confused with more serious threats. The Department of Homeland Security (DHS)

Secondary Reachback Program supports different field elements but has technical goals similar to those of the Triage program. Some of the research involves a cooperative effort with DHS Reachback, and both programs are discussed in Ref. 1. Combined results from DOE and DHS efforts are included in the RadAssessor database [2,3], which is updated regularly to include new spectra and RIID results.

COLLECTION OF DATA

Information about emerging radioisotopes can be obtained from literature searches, press releases, trade conferences, observations from radiation monitoring programs, or follow-through with professional contacts. Much of the information is volunteered as a courtesy by partner agencies such as the Federal Bureau of Investigation, U.S. Customs, or the DOE Radiological Assistance Program. If a product is identified as being important for further study, contact is established with an institution that manufactures, distributes, or uses the product. If possible, a visit is scheduled to the institution to collect data from realistic configurations using a variety of RIIDs, such as the ORTEC Detective, SAIC GR-135, and Thermo Electron identiFINDER. Volunteers are sought for in vivo measurements of some medical isotopes, although most of the work with patients is carried out under the DHS Reachback Program. Our goal is to discover information about a radioisotope's applications, production mechanism, trace isotopes, distribution, and prevalence. The following sections describe some of our DOE-supported activities performed at various institutions that invited collaboration in 2005 through 2006.

A. The University of Missouri Research Reactor

The University of Missouri Research Reactor (MURR) is located in Columbia, Missouri, and has the highest neutron flux ($\sim 10^{14}$ n/cm²-s in the core) of any university research reactor in the United States. The MURR facility is a multidisciplinary institution that is involved in radioisotope production and academic research while also providing analytical and irradiation services to a wide array of external customers. Research activities at MURR span several disciplines including the development and testing of radiopharmaceuticals, nutritional epidemiology, archaeometry, materials science, and analytical chemistry. MURR produces both medical and industrial radioisotopes, with an emphasis on the medical applications, and provides irradiation services for topaz coloring, silicon doping, and other commercial and research applications.

MURR is an ideal collaborator for the predictive research program because of its wide variety of research activities and products. On our first visit to MURR, spectra were generated from the following items of interest:

- ¹⁰⁴Ru irradiated target
- ¹⁵³Sm bulk shipment
- ¹⁵³Sm in a single dose vial
- aged ¹⁵³Sm sample
- ¹⁵⁹Gd
- ¹⁶⁶Ho
- ^{166m}Ho
- ¹⁷⁷Lu bulk shipment
- ¹⁸⁸W/¹⁸⁸Re generator
- ¹⁹⁸Au foil
- ²⁰¹Tl
- ⁶⁸Ga/⁶⁸Ge generator

- ^{75}Se
- ^{90}Y
- Al-Si irradiated can
- depleted U shipping container
- irradiated quartz vial
- topaz chips
- irradiated topaz in its shipping drum.

MURR also supplied a sample of concentrated $^{177\text{m}}\text{Lu}$, which is a long-lived component of some medical products that is often mistaken for ^{239}Pu .

Figure 1 shows two gamma spectra acquired from a vial of ^{159}Gd ($T_{1/2} = 18.6$ h), an isotope used for skeletal targeted radiotherapy. The spectrum on the left was acquired using an ORTEC Detective, and the spectrum on the right used an SAIC GR135. Neither of these RIIDs accurately identified the isotope, even though the spectrum signature is distinctive. Analysis of the high-purity germanium (HPGe) spectrum confirms the presence of two trace isotopes: ^{153}Sm ($T_{1/2} = 46.7$ h) and $^{152\text{m}}\text{Eu}$ ($T_{1/2} = 9.32$ h). These trace isotopes are medically benign but are a potential source of confusion because the patient's history may disagree with the RIID isotope report. Coincidentally, ^{153}Sm is also used for skeletal radiotherapy.

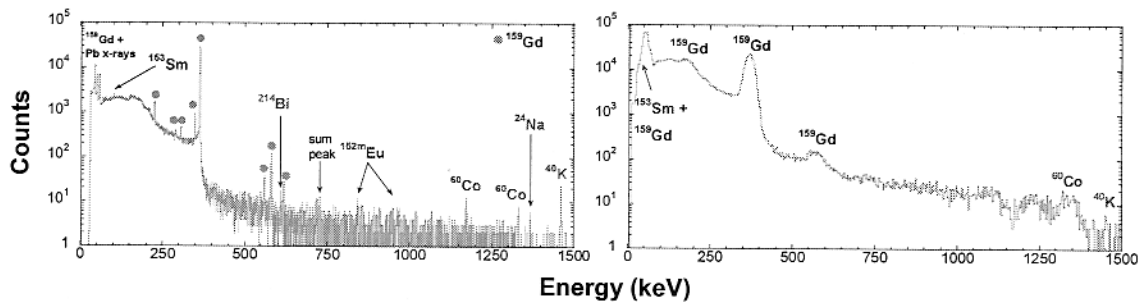


Figure 1. Gamma spectra acquired on a bare ^{159}Gd source using a HPGe ORTEC Detective (left) and a NaI SAIC GR135 (right).

B. MDS Nordion

MDS Nordion is one of the foremost companies in medical and industrial radioisotope production, processing, and distribution. They export radioactive products and related technologies to more than 60 countries around the world. It is the world's largest supplier of medical ^{99}Mo and industrial ^{60}Co sources. They also distribute ^{14}C , ^{57}Co , ^{51}Cr , ^{137}Cs , ^{64}Cu , ^{18}F , ^{67}Ga , ^{111}In , ^{123}I , ^{125}I , ^{131}I , ^{177}Lu , ^{103}Pd , ^{32}P , ^{186}Re , ^{82}Sr , ^{201}Tl , ^{133}Xe , and ^{90}Y , and are active in the research of new products and methods. We visited their Ottawa facility to learn more about their products and collect radiation signature data. A visit to their Vancouver facility is planned.

One of their radiopharmaceutical products, Therasphere, has been developed to treat liver cancer. It consists of microscopic glass beads containing ^{90}Y that are sized to lodge into the hypervascularized regions of a tumor-infected liver. Though ^{90}Y is a pure beta emitter, data collected on site at MDS Nordion on fresh and aged Therasphere doses reveal trace components that produce visible peaks in a gamma spectrum, becoming more visible as the ^{90}Y decays away. These include ^{88}Y , ^{152}Eu , and ^{154}Eu . Later in vivo measurements,

performed by U.S. Customs, confirm the presence of these radioisotopes in patients at very low levels that are medically harmless.

Other products that were measured at the Ottawa facility include Gammacell ^{137}Cs irradiators of various configurations, a 200-kCi ^{60}Co source packed for shipment, a returned ^{192}Ir source in depleted uranium shielding, ^{131}I doses boxed for shipment, and ^{125}I doses boxed for shipment. Trace ^{126}I signatures were notably absent from the latter, which was unlike previous Triage experience with this isotope. We were informed by the MDS Nordion scientists that their irradiation technique is designed to minimize the production of ^{126}I .

C. Theragenics Corporation

Theragenics Corporation, located in Buford, Georgia, is a medical device company founded in 1981. Their products include brachytherapy (seeding) devices primarily used in the treatment of prostate cancer and, through its CP Medical subsidiary, a number of surgical devices including wound closure, brachytherapy, and cardiac-pacing products. The highest-volume product manufactured by Theragenics is the ^{103}Pd TheraSeed, Model 200, shown in Fig. 2. The second most popular product is the ^{125}I I-Seed, Model I25.S06. Theragenics produces the ^{103}Pd for the TheraSeed at their Buford location through a $^{103}\text{Rh}(p,n)^{103}\text{Pd}$ reaction, using eight operational cyclotrons. The ^{125}I used in the I-Seed is imported from a reactor operated by McMaster University in Ontario, Canada.

^{125}I is quite prevalent and has been encountered previously in both patients and shipments interdicted by first responders. Its trace components and gamma spectra, as viewed by common RIIDs, are well-understood. Conversely, despite the sharp increase in ^{103}Pd usage over the past decade, it is still rarely encountered by Triage, and its spectral features have not been investigated extensively. To familiarize ourselves with ^{103}Pd , we visited Theragenics Corporation and collected data on some TheraSeed samples in both bare and shielded configurations. The data will be used in the Triage training program to ensure familiarity with this product.



Figure 2. Photograph of a single TheraSeed brachytherapy source, approximately 4 mm long.

D. The Mallinckrodt Institute of Radiology, Washington University

The Mallinckrodt Institute of Radiology (MIR) at Washington University is one of the premier radiopharmaceutical research institutions in the United States. A primary focus is positron emission tomography (PET) imaging technology. MIR studies a large variety of candidate isotopes. Some are traditional, short-lived, pure positron emitters such as ^{11}C , ^{15}O , and ^{18}F , and others have significantly longer half-lives and emit multiple gamma rays in addition to the 511-keV annihilation radiation. Some of the nontraditional isotopes include ^{76}Br , ^{77}Br , ^{124}I , ^{86}Y , $^{94\text{m}}\text{Tc}$, ^{66}Ga , ^{60}Cu , ^{61}Cu , ^{64}Cu , and ^{82}Rb .

Two isotopes that are of particular interest to our program are ^{64}Cu ($T_{1/2} = 12.7$ h) and ^{76}Br ($T_{1/2} = 16.2$ h), primarily because their long half-lives make it feasible to ship them anywhere within the United States and their use in research is growing. The likelihood of encountering ^{64}Cu is high, as it is currently distributed to more than 40 universities and medical research facilities. One compound, ^{64}Cu -ATSM, is used as a hypoxia imaging agent for PET and has already been approved by the U.S. Food and Drug Administration for use in humans. ^{76}Br is not as prevalent as ^{64}Cu , but its use as a PET imager and surrogate for ^{124}I research is steadily increasing. To become more familiar with both of these two radioisotopes, we visited MIR and collected data using several RIIDs in different measurement configurations.

One of the HPGe spectra acquired during this visit was obtained on a freshly-prepared, unshielded sample of ^{64}Cu and is shown in Fig. 3. There is only one peak in this spectrum that corresponds to the decay of ^{64}Cu , while the rest can be assigned as background peaks or other trace isotopic components in the sample. One of the trace isotopes, ^{55}Co ($T_{1/2} = 17.5$ h), is longer lived than ^{64}Cu and may contribute to confusing RIID results. However, it is doubtful that the trace components ^{61}Co ($T_{1/2} = 99$ min) or ^{60}Cu ($T_{1/2} = 23$ min) would ever be encountered in Triage due to their short half-lives.

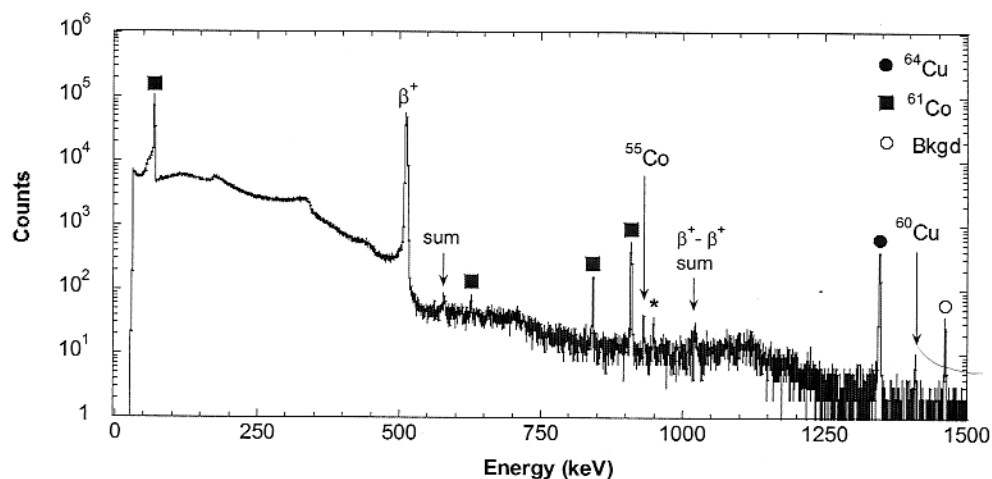


Figure 3. HPGe spectrum acquired with the ORTEC Detective on freshly-prepared ^{64}Cu .

E. Cardinal Health

Cardinal Health has assisted with the procurement of several samples of radionuclides for testing at Los Alamos National Laboratory and has made its Albuquerque facility available for the collection of data. In addition, consultants at Cardinal Health have informed us numerous times about emerging radioisotopes with which we were not familiar. Some of the samples they provided included Mallinckrodt and Bristol-Myers Squibb ^{99m}Tc generators of various ages and shielding designs, waste streams from one of these generators, ^{32}P solutions, ^{111}In , ^{133}Xe with ^{133m}Xe at various ages (Fig. 4), and more common medical isotopes such as ^{99m}Tc , ^{67}Ga , ^{123}I and ^{131}I used at LANL in masking exercises.

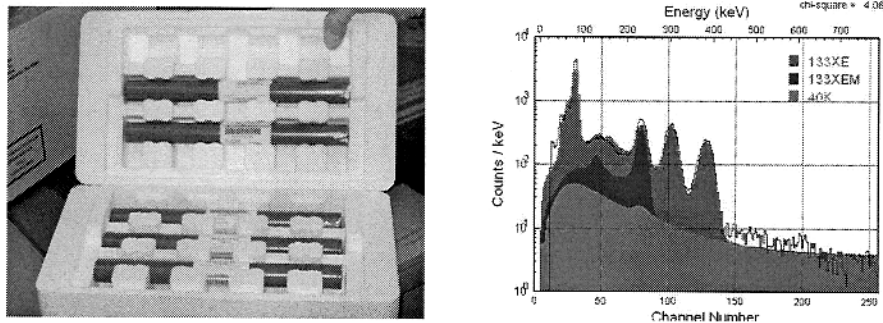


Figure 4. ^{133}Xe in shipping configuration and a typical RIID sodium-iodide spectrum. The metastable component is visible in red; its relative strength varies depending on sample age.

F. Schlumberger

Schlumberger is one of the world's leading oilfield service providers. Their services include well logging using neutron reflectometry and neutron-capture elemental analysis. Pulsed neutron sources are used for some of this work, but large AmBe sources are also used (Fig. 5). These AmBe sources generate numerous portal alarms and potential delays.

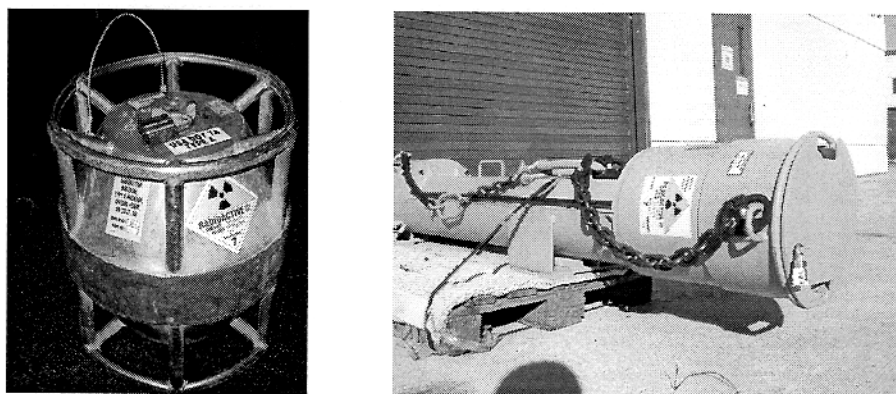


Figure 5. Photographs large neutron sources packaged for shipment, courtesy of Schlumberger.

Schlumberger has worked with us to improve our understanding of their products and to provide valuable training information. Not only does this cooperation reduce the potential for panic that can result from experiencing a large neutron hit, but it also reduces the chance of shipping delays and resulting commercial impact to the company. One interesting feature is that the large neutron sources have a seasonal migration pattern: they are used in Canada when frozen ground conditions allow vehicle access to the oil fields and are shipped to Mexico for use during the rest of the year. This results in numerous international shipments in a predictable pattern along predictable routes. Schlumberger technical advisors have also helped us understand other aspects of the oil industry, such as the use of ^{46}Sc as a tracer in oil fractionating plants.

CONCLUSIONS

Analysts from the DOE Radiological Triage Program are responsible for interpreting gamma-ray spectra quickly and accurately to determine if a radiological event is serious or benign. In the past year, we have collaborated with several institutions that are involved in the production and distribution of a variety of radioisotopes. These institutions have been hospitable and have allowed us access to information that will improve our knowledge and response times in future radiological events.

ACKNOWLEDGEMENTS

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