Applications and Availability of Californium-252 Neutron Sources for Waste Characterization

Rodger C. Martin* and Steven E. Kos†

*Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6385 USA;
E-mail *martinrc@ornl.gov*
†Waste Management Federal Services, Inc., 345 Hills Street, Richland, Washington 99352 USA;
E-mail *Steve_Kos@gtsduratek.com*

Abstract

This paper reviews some existing and potential applications of compact, portable ^{252}Cf neutron sources to waste and nuclear materials characterization. For example, (1) Fluor Hanford uses a neutron activation analysis facility to assay the sodium and actinide content of vitrified waste glass; (2) neutrons from ^{252}Cf ionization chambers are used to probe fissile material and deposits to estimate subcriticality and plan for removal; and (3) a handheld device can noninvasively detect the presence of hydrogen or moisture via neutron backscattering. This paper emphasizes a geophysical logging system developed by Waste Management Federal Services, Inc., and deployed to Pit 9 at the Idaho National Engineering and Environmental Laboratory, which successfully characterized the presence of chlorine, plutonium, and other radioactive species. The U.S. Department of Energy Californium Industrial Loan Program provides single sources emitting up to ~10^{11} neutrons/s for ~$50,000 (or less for lower-intensity sources) to qualified government subcontractors.

Introduction

The ^{252}Cf radioisotope is an intense neutron emitter that is readily encapsulated in compact sealed sources. Several technologies ranging from the routine to the conceptual employ ^{252}Cf in the characterization of waste and nuclear materials. Common applications include gamma spectroscopy via instrumental neutron activation analysis (INAA) for trace elemental analysis, prompt gamma activation analysis (PGAA) for elemental determination of the principal components of a sample, and passive-active neutron shufflers for determination of fissile content.

The purpose of this presentation is threefold. Several of the ^{252}Cf-based systems that have been established or deployed for waste-related applications are summarized, in addition to new technologies with potential application to waste characterization. A geophysical logging system used to quantify down-hole chlorine, plutonium, and other contaminants at Pit 9 of the Idaho National Engineering and Environmental Laboratory (INEEL) is detailed. Finally, the U.S. Department of Energy (DOE) Californium Industrial Sales/Loan Program for the supply of ^{252}Cf sources is discussed. While the Sales Program supplies source material to ^{252}Cf reencapsulators for commercial applications, the Loan Program provides sources for temporary or extended use by U.S. governmental agencies and subcontractors and university researchers. The Loan

Program can often provide intense neutron sources at significant discounts to qualified users as well as eliminate source disposal issues after task completion.

Review of Waste and Nuclear Materials Characterization Applications

In addition to the common use of ^{252}Cf sources as neutron calibration standards and in neutron shufflers for determination of fissile content of waste containers, a variety of specialized applications address problems in waste and nuclear materials characterization.

Characterization of Fissile Material

Researchers at Oak Ridge National Laboratory (ORNL) have used low-intensity (~1-μg) ^{252}Cf electroplates within small ionization chambers to probe and characterize fissile material. The response of the material as measured by a detector array is analyzed using standard time-correlation and/or frequency-analysis techniques. This methodology has been applied to estimate the spatial distribution, mass, and hydration of deposits of $UO_2F_2 \cdot nH_2O$ in process piping to estimate the subcriticality of the deposit and plan for its removal.[1] Other applications are the identification and inventory of nuclear weapons components and other uranium items for nuclear materials control and accountability, the determination of k_{eff} for optimization in storage and shipping of fuel elements, and measurement of mass flow rate of ^{235}U in a UF_6 gas stream for downblending of Russian weapons-grade uranium. Several of these cart-portable Nuclear Materials Identification Systems are reported in use at the Oak Ridge Y-12 Plant, as well as a more portable battery-operated system.[2]

Trace Elemental Analysis (INAA) of Waste and Environmental Samples

A significant thermal neutron flux is required for conventional INAA. Typically this requires an operating nuclear reactor, although several INAA facilities have been demonstrated using intense ^{252}Cf sources. A facility at the Savannah River Technology Center currently uses ~6 mg of ^{252}Cf (neutron flux $>10^7$ cm^{-2} s^{-1}) to provide INAA services for environmental and waste management customers, with analyses of organic compounds, metal alloys, sediments, site process solutions, etc.[3,4] Analytical sensitivities in the parts-per-million range can be achieved for several dozen elements. The facility is also used to produce short-lived radioactive tracers for radiochemical analyses and separation methods testing. Advantages of INAA with respect to other trace element analytical techniques are that bulk samples can be analyzed without digestion, avoiding analytical uncertainties introduced by the digestion process, and that INAA is more cost-effective for bulk samples in situations where INAA sensitivity is adequate.

Another major INAA system (thermal neutron flux ~10^9 cm^{-2} s^{-1}) has recently been established at the Hanford complex for high-precision measurements of elemental sodium content within vitrified fuel reprocessing wastes.[5] This approach circumvents the problems of standard analytical measurements on high-dose-rate radioactive samples (i.e., sample preparation via dilution of highly radioactive waste with attendant radiological and analytical disadvantages).

This system has the additional capability of quantifying trace residues of fissionable isotopes (~0.1 ppm of uranium and plutonium) via delayed neutron counting.

Principal Components Analysis (PGAA) for Military Base Cleanup

Lower-intensity ^{252}Cf sources can be used to induce prompt gamma emission from a sample upon neutron capture. Coupling with gamma spectroscopy provides analysis of most elements with sensitivities in the percent range or better with minimal sample activation. The commercial Portable Isotopic Neutron-Spectroscopy (PINS) Chemical Assay System developed at the INEEL and marketed by PerkinElmer Instruments (formerly EG&G ORTEC) is used for the nondestructive identification of the contents of munitions, chemical weapons, and general chemical-storage containers.[6] PINS systems typically use several micrograms of ^{252}Cf transported in a small shielded container.

Handheld Hydrogen and Moisture Detector

A handheld instrument containing a low-intensity ^{252}Cf source has been developed by Nova R&D, Inc., and deployed with the U.S. Coast Guard to detect contraband hidden in compartments behind metal and other structures.[7] Fast neutrons from the device, called a Compact Integrated Narcotics Detection Instrument (CINDI), penetrate the barrier material and are backscattered by any hydrogen-rich materials present. Unexpected detection of backscattered neutrons suggests the potential presence of contraband such as narcotics. Panels made of steel, wood, fiberglass, or lead-lined materials can be probed. A recent upgrade makes possible simultaneous detection of both neutron and gamma backscattering. Although this technology has not been applied to waste characterization issues, the convenience of this existing handheld sensor for noninvasive determinations of hydrogen and/or moisture content in, for example, waste drums is promising.

Cold Neutron Irradiator (CNI) for Enhanced Neutron Activation Analysis Sensitivity

A concept developed by D. D. Clark of Cornell University employs a cryogenic moderator to enhance sample neutron capture rates for greater elemental sensitivity using ^{252}Cf-based PGAA.[8] One CNI design uses a 2-mg ^{252}Cf source and a moderator at 30 K to deliver a flux $>10^8$ cm^{-2} s^{-1} at the sample. Cold neutrons increase sensitivity by a factor >2 for a given flux. Alternatively, half the ^{252}Cf mass can be used to provide the same sensitivity, an important cost consideration if commercial sources are purchased. The same concept can boost the sensitivity of INAA using more intense sources, but, for the same flux, PGAA provides a significantly higher count rate per milligram of sample for most elements. For comparable flux, Clark observed that PGAA provides sensitivity superior to INAA by 1 to 5 orders of magnitude for 74 out of 89 elements.[8] Another advantage of cold neutrons is that they can be transported out of the CNI using neutron waveguides for additional flexibility in analyses (e.g., low background measurements) and end use of neutrons. A proposal has been submitted for a joint project between ORNL and Cornell University to build a prototype CNI.

Geophysical Logging at Pit 9

A borehole geophysical logging tool developed by Waste Management Technical Services, Inc. (WMTS), uses ~5 μg of ^{252}Cf for subsurface PGAA investigation of a waste disposal site having a significant presence of chlorinated compounds and transuranic elements.[9] Rocky Flats Plant waste was successfully detected within Pit 9 of the Radioactive Waste Management Complex at the INEEL and measurements made of elemental chlorine (from chlorinated solvents, salts, and organic compounds) using ^{252}Cf PGAA and of ^{239}Pu, ^{238}U, ^{235}U, ^{241}Am, and ^{137}Cs using several other detectors. PGAA measurements were obtained using a high-purity germanium (HPGe) gamma detector. Other detectors were used to measure the ambient photon spectrum (high-resolution HPGe detector), moisture profile (50-mCi americium-beryllium source and ^{3}He detector), passive thermal neutron background (i.e., fission rate of transuranic radionuclides, measured with ^{3}He detector), and azimuth (directional) HPGe log surveys to determine directional distributions of Pit 9 waste.

The logging system is self-contained in a van body mounted on a heavy-duty truck chassis. The truck also contains a winch system for communicating with and positioning the logging tools. These borehole probes are contained in cylindrical housings. The passive-neutron and neutron-moisture logging tools are operated with the same cable and instrumentation and retrieval and data acquisition systems as the spectral-gamma tools. The passive-neutron and spectral-gamma detectors are configured to acquire measurements within 15 cm of the borehole bottom.

The PGAA probe is the only logging tool within the system that can measure nonradioactive elements. This tool employs a HPGe detector with 20% efficiency relative to a standard 7.6- × 7.6-cm NaI detector and has dimensions ~9-cm diameter by 1.8-m length. The source-to-detector distance is ~40 cm; thus, measurements could not be obtained closer than ~46 cm from the bottom of the borehole. The detector is shielded from ^{252}Cf source neutrons and gamma rays by tungsten metal. The logging tool also contains the high-voltage power supply, preamplifier, and a liquid nitrogen dewar and cryostat assembly. The source contained ~5.3 μg of ^{252}Cf at the time of logging and was stored in a small shielded container when not in use. Transfer of the source to the lower end of the logging tool is accomplished using a long source-handling tool to minimize personnel exposure.

Movement of the logging tools is controlled by a surface system computer via a servo-controlled hydraulic winch. For the PGAA logging tool, the data acquisition time is 300 s for each 15-cm incremental location (i.e., in a move-stop-acquire mode). In addition to the computer controller, the surface electronics include a high-count-rate nuclear spectroscopy amplifier coupled to a computer-controlled multichannel analyzer.

Measurements were made during January and February 2000, within 20 steel-cased boreholes drilled in a pattern within the 12- × 12-m area of investigation at Pit 9. Depth of the borehole casings was typically 4±1 m. The waste seam was determined by borehole measurements of moisture content to be approximately 1.2 to 2.6 m thick, buried beneath 1.2 to 1.8 m of overburden soil. Although PGAA measurements of hydrogen, silicon, calcium, iron, and aluminum as well as chlorine were obtained, only the chlorine measurements were calibrated, providing a minimum detection limit of 300 ppm. Chlorine was detected in all 20 of

the boreholes, at concentrations ranging from 1000 to 30,000 ppm. Plutonium-239 was detected via gamma spectroscopy at concentrations from 18 to 124,000 nCi/g and ^{238}U from 24 to 2500 pCi/g in approximately half the boreholes, and ^{235}U from 4 to 33 pCi/g in four boreholes. Americium-241 was also detected at low levels. Cesium-137 concentrations were measured at less than 1 pCi/g in those boreholes that did not exhibit spectral interferences from ^{241}Am.

The WMTS report[9] provides detailed information on the operation and calibration of the system as well as recommendations for more detailed interpretation of the spectral data and for further logging tool development.

Californium-252 Source Availability

Californium Industrial Loan Program

The DOE inventory of sealed ^{252}Cf sources is stored at the Radiochemical Engineering Development Center of ORNL. Californium-252 is produced, purified, and encapsulated at ORNL as a by-product of DOE's heavy element program. While source material is sold to commercial vendors at a current price of $60 per μg of ^{252}Cf, plus encapsulation, packaging, and transportation charges, government researchers and contractors can obtain DOE sources on loan without charge for the radioisotope.[10] If an appropriate source is available from the DOE source inventory at ORNL, the loanee pays only the technical service charges incurred for source preparation, shipment, and return. As part of the loan agreement, DOE requires source return to ORNL after use, eliminating source disposal concerns and costs to the user.

One microgram of ^{252}Cf emits 2.314×10^6 fast neutrons/s with a 2.645-year half-life. Typical costs[11] for loan and end-of-loan return of a <7-μg source total ~$11,000. (This fee is waived under the University Loan Program for university research and teaching applications.) Similar costs for a source in the range of 7 μg to 3 mg (neutron intensities ranging up to 7×10^9 s^{-1}) total ~$20,000, while sources in the 3- to 5-mg range total ~$28,000. Loan/return costs of preexisting sources from inventory containing >5 mg total ~$32,000. Sources containing >8 mg typically require custom fabrication, but a source containing the maximum permitted ^{252}Cf content of 50 mg (neutron intensity ~10^{11} s^{-1}) can be obtained for ~$50,000. None of these costs include transportation charges. Loan costs for high-intensity sources compare very favorably with procurement costs for electronic neutron generators and accelerators with comparable intensities. The choice of radioisotopic vs electronic neutron sources is dependent on the specific application and on a variety of practical factors. The standard for pulsed applications is the neutron generator, but 14-MeV neutrons require more moderation and shielding for thermal neutron applications compared with ^{252}Cf (2.1-MeV average energy). Use of a neutron-absorbing shutter for timed bursts of neutrons from ^{252}Cf has been reported.[1]

Californium Industrial Sales Program

Californium-252 sources for commercial applications typically must be obtained from one of several commercial ^{252}Cf vendors rather than directly from ORNL. An exception is when a custom source request cannot be supplied by a commercial vendor. Because the cost of the

radioisotope is not a significant factor for sources under several tens of micrograms, low-intensity sources can often be purchased from a commercial vendor at lower cost than a loaned source. However, end-of-useful-life disposal of a source becomes the user's responsibility unless a return agreement exists with the vendor. A list of commercial vendors is available on request from the first author of this paper.

Summary

The DOE Californium Loan Program provides compact, portable neutron sources for waste and nuclear materials characterization by governmental organizations and subcontractors, at total costs ranging from the reasonable to the nominal, considering source intensity and lack of disposal concerns. A sampling of ^{252}Cf-based technologies relevant for waste and nuclear materials characterization is discussed above. Names of contacts for further information on any of these technologies are available from the first author. Information on the down-hole logging system is available from author S. E. Kos. Further information and contacts are available from the web site *http://www.ornl.gov/divisions/ctd/cuf.htm.*

Acknowledgments

Oak Ridge National Laboratory is managed by UT-Battelle, LLC, under contract DE-AC05-00OR22725 for the U.S. Department of Energy.

References

1. J. K. MATTINGLY, J. T. MIHALCZO, T. E. VALENTINE, J. A. MULLENS, J. MARCH-LEUBA, T. UCKAN, "Correlation Measurements with ^{252}Cf to Characterize Fissile Material," *Trans. Am. Nucl. Soc.*, **82**, 95 (2000).
2. J. T. MIHALCZO et al., "Field Use of NMIS at Oak Ridge," Oak Ridge Y-12 Plant Report Y/LB-16,019 (1999).
3. D. P. DiPRETE, R. A. SIGG, "Neutron Activation Analysis Detection Limits Using ^{252}Cf Sources," *Trans. Am. Nucl. Soc.*, **82**, 97 (2000).
4. D. P. DiPRETE, S. F. PETERSON, R. A. SIGG, "Low-Flux NAA Applications at the Savannah River Site," *J. Radioanal. Nucl. Chem.*, **244**, 343 (2000).
5. G. L. TROYER, M. A. PURCELL, "^{252}Cf Neutron Activation Analysis of High-Level Processed Nuclear Tank Waste," *Trans. Am. Nucl. Soc.*, **82**, 98 (2000).
6. A. J. CAFFREY, J. D. COLE, R. J. GEHRKE, R. C. GREENWOOD, "Chemical Warfare Agent and High Explosive Identification by Spectroscopy of Neutron-Induced Gamma Rays," *IEEE Trans. Nucl. Sci.* **39**, 1422 (1992).
7. T. O. TÜMER, L. DOAN, C. W. SU, J. BARITELLE, B. RHOTON, "A Sensitive, Selective, and Portable Detector for Contraband: The Compact Integrated Narcotics Detection Instrument," *Trans. Am. Nucl. Soc.*, **82**, 96 (2000).
8. D. D. CLARK, "Production and Utilization of Cold Neutrons for Elemental Analysis," *Trans. Am. Nucl. Soc.* **71**, 162 (1994).

9. S. E. KOS, R. K. PRICE, R. R. RANDALL, “Geophysical Logging at Pit 9, INEEL,” Waste Management Federal Services, Inc., report (2000).
10. R. C. MARTIN, J. B. KNAUER, P. A. BALO, “Production, Distribution, and Applications of Californium-252 Neutron Sources,” *Appl. Radiat. Isot.*, **53**, 785 (2000).
11. R. C. MARTIN, “Representative Costs for ^{252}Cf Source Loans,” *Californium-252 Newsletter*, **3**(1), 2 (1999); available from the first author.