

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

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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. 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 two-fold. Several of the ^{252}Cf -based systems which have been established or deployed for waste-related applications will be summarized, in addition to new technologies with potential application to waste characterization. Also, the U.S. Department of Energy (DOE) Californium Industrial Sales/Loan Program for the supply of ^{252}Cf sources will be discussed. While the Sales Program supplies source material for commercial applications, the Loan Program provides sources for 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.

Description

Californium Industrial Loan Program

The DOE inventory of sealed ^{252}Cf sources is stored at the Radiochemical Engineering Development Center of Oak Ridge National Laboratory (ORNL). Californium-252 is produced, purified, and encapsulated at ORNL as a byproduct of DOE's heavy element program. While source material is sold to commercial vendors at a current price of $\$60 \mu\text{g}^{-1}$ of ^{252}Cf , plus encapsulation, packaging, and transportation charges, government researchers and contractors can obtain DOE sources on loan without charge for the radioisotope. 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 for loan and return of a $<7\text{-}\mu\text{g}$ source total $\sim\$11,000$. (This fee is waived under the University Loan Program for university research and teaching applications.) Similar costs for a source in the range of $7 \mu\text{g}$ to 3 mg (neutron intensities ranging up to $7 \times 10^9 \text{ s}^{-1}$) total $\sim\$20,000$, while sources in the 3- to 5-mg range total $\sim\$28,000$. Loan/return costs of pre-existing sources from inventory containing $>5 \text{ mg}$ total $\sim\$32,000$. Sources containing $>8 \text{ mg}$ typically require

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custom fabrication, but a source containing the maximum permitted ^{252}Cf content of 50 mg (neutron intensity $\sim 10^{11} \text{ s}^{-1}$) can be obtained for $\sim \$51,000$. None of these costs include transportation charges. Sources with neutron intensities $< 10^8 \text{ s}^{-1}$ can be obtained at lower cost from commercial vendors, but potential costs for end-of-use source disposal should be evaluated. 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.

Trace Elemental Analysis (INAA)

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 $\sim 6 \text{ mg}$ of ^{252}Cf (neutron flux $> 10^7 \text{ cm}^{-2} \text{ s}^{-1}$) to provide INAA services for environmental and waste management customers, with analyses of organic compounds, sediments, site process solutions, etc.¹ Analytical sensitivities in the parts-per-million range can be achieved for several dozen elements.

Another major NAA system (thermal neutron flux $\sim 10^9 \text{ cm}^{-2} \text{ s}^{-1}$) is currently being established at the Hanford complex for high precision measurements of the vitrification of Hanford fuel reprocessing wastes as well as measurement of trace quantities of fissionable isotopes ($\sim 0.1 \text{ ppm}$ of uranium and plutonium) via delayed neutron counting.²

Principal Components Analysis (PGAA)

Lower-intensity ^{252}Cf sources can be used to induce prompt gamma emission from a sample upon neutron capture, and 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 Idaho National Engineering and Environmental Laboratory and marketed by EG&G ORTEC is used for the nondestructive identification of the contents of munitions, chemical weapons, and general chemical-storage containers.³ PINS systems typically use several micrograms of ^{252}Cf transported in a small shielded container.

A borehole geophysical logging tool developed by Waste Management Technical Services uses several hundred micrograms of ^{252}Cf for subsurface PGAA investigation of a waste disposal site having a significant presence of chlorinated compounds.^{4,5}

Cold Neutron Irradiator (CNI)

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.⁶ One CNI design uses a 2-mg ^{252}Cf source and a moderator at 30 K to deliver a flux $> 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ 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. Clark observed that PGAA provides superior sensitivity by 1 to 5 orders of magnitude for 74 out of 89 elements.⁶ Another advantage of cold neutrons is that they can be transported out of the CNI using neutron wave guides for additional flexibility in analyses and end-use of neutrons. A proposal has been submitted for a joint project between ORNL and Cornell University to build a prototype CNI.

Discussion

The DOE Californium Loan Program provides compact, portable neutron sources for waste characterization by governmental organizations and subcontractors, at total costs ranging from the reasonable to the nominal, depending on source intensity and considering lack of disposal concerns. A few of the better-known ²⁵²Cf-based technologies are discussed above, while the full paper will also address these in more detail as well as other developmental technologies and concepts. These include a hand-held device with a weak ²⁵²Cf source presently used to detect contraband hidden within metal structures⁷; a miniaturized, portable time-of-flight mass spectrometer based on ²⁵²Cf-induced plasma desorption mass spectrometry and used, for example, to analyze solid samples and the decomposition products of high explosives⁸; and ²⁵²Cf-driven subcritical multiplier assemblies used to boost ²⁵²Cf neutron flux by an order of magnitude or more in major nonreactor irradiation facilities.

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