The U.S. Department of Energy Californium-252 Program

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Soon after the discovery of the ^{252}Cf radioisotope in 1952, researchers realized its potential as a spontaneous source of neutrons. Under the guidance of the Atomic Energy Commission, the Savannah River Laboratory (SRL) undertook an ambitious market evaluation and source fabrication program in the 1960s. Routine production of ^{252}Cf was initiated at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL), initially from plutonium oxide target rods and ultimately from more productive recycle curium oxide target rods. The ^{252}Cf radioisotope was then shipped to SRL for incorporation into a variety of SRL-designed sources for industrial, medical, and research applications. ORNL independently developed another line of sources, primarily for research applications. Industrial source fabrication operations were consolidated at ORNL in 1986. In 1992, SRL fabricated its last ^{252}Cf sources. Today most of the world's supply of ^{252}Cf is processed, purified, and encapsulated for shipment at the Radiochemical Engineering Development Center (REDC) of ORNL.

DESIGN AND FABRICATION OF CALIFORNIUM-252 SOURCES

Californium-252 is conveniently packaged into small sealed source capsules with a wide range of intensities.[1] The fission neutron energy spectrum of ^{252}Cf is analogous to that of a reactor, with an average neutron energy of 2.1 MeV and a most probable energy of 0.7 MeV. A cylindrical source as small as 5 cm long by 1 cm in outer diameter (OD) is licensed to contain a maximum of 50 mg of ^{252}Cf (>10^{11} neutrons/s, at 2.314×10^{9} neutrons$\cdot$s$^{-1}\cdot$mg^{-1}). Other

specialized products are a physically small source used as a calibration point source and a 3-mm-OD medical brachytherapy source used for cancer treatments. Most sources are double encapsulated. The outer capsules are typically fabricated from 304L stainless steel or, for reduced gamma emission by neutron activation, Zircaloy-2. The inner capsules are made from the same alloys for the ORNL series of sources, or from Pt–10 wt % Rh alloy for the SRL series. An exception is the medical sources, which have both inner and outer capsules fabricated from Pt–10 wt % Ir alloy. Special-order ^{252}Cf electroplates are also available with specific activities potentially >300 μg/cm^2 on platinum or stainless steel substrates.

The chemical form of ^{252}Cf in the ORNL-type sources is californium oxysulfate, $Cf_2O_2SO_4$, contained in microspheres resulting from the pyrolysis of cation-exchange resin loaded with californium. These microspheres are compacted inside aluminum powder or tubing into a small pellet, followed by encapsulation. SRL-type sources contain californium oxide, Cf_2O_3, obtained by filtering a californium oxalate precipitate within an inner capsule followed by calcination to the oxide prior to capsule closure. The primary commercial product is a cermet wire containing Cf_2O_3 dispersed throughout a palladium matrix, with a wire diameter of ~1.3 mm and specific activities typically ranging from 2 to 200 μg of ^{252}Cf per cm of wire.

In over 20 years of fabrication at ORNL (involving hundreds of sources) only one source, single encapsulated, was supplied to a customer and subsequently evidenced a barely detectable pinhole flaw during routine usage. These small sources are surprisingly rugged: one ^{252}Cf source capsule (without the ^{252}Cf) was subjected to the impact of 4.5 kg of plastic explosive without breaching capsule integrity.[2]

PRODUCTION AND DISTRIBUTION OF CALIFORNIUM-252

A heavy element production "campaign" is conducted at the REDC approximately every 2 years, typically producing 350 to 400 mg of ^{252}Cf (along with lesser amounts of berkelium, einsteinium, and fermium radioisotopes for research) from $\leq$100 g of curium in approximately ten target rods. Although the HFIR is scheduled for an extended maintenance outage beginning in late 2000, the 2-year campaign cycle will be scheduled around the outage to ensure a reliable ^{252}Cf supply. During an unplanned HFIR outage of ~4 years in the late 1980s, sufficient ^{252}Cf inventory existed to continuously meet ongoing demand. The HFIR has a projected lifetime of several decades. Curium feedstocks at ORNL are adequate for ~10 years of ^{252}Cf production. Curium feedstocks from elsewhere within the DOE complex are sufficient for ^{252}Cf production for several decades at current rates, assuming those feedstocks are transferred to ORNL.

With ~200 mg of ^{252}Cf produced annually at ORNL, the largest fraction (~25%) is distributed for neutron radiography, followed by commercial sales. A significant fraction is lost by decay prior to distribution. These undistributed inventories can now be accessed by researchers via the Californium User Facility for Neutron Science at ORNL.[3]

Californium-252 is supplied via several programmatic mechanisms.[4] In the case of commercial applications, DOE will not supply a source directly to the end user if a suitable source is available from a commercial vendor. Instead, DOE sells the ^{252}Cf radioisotope to vendors under the Californium Industrial Sales Program at the current price of \$60/μg for subsequent distribution. For noncommercial government-funded and educational applications and for university/medical research, the Californium Industrial/University Loan Program will loan sources from the prefabricated source inventory at the REDC *without charge* for the radioisotope; the

loanee only incurs the cost for the technical services required to provide the source (plus transportation). If required, a source can be fabricated to specification under this program at additional cost. The University Loan Program is unique in that existing small sources (typically a few micrograms of ^{252}Cf) are loaned to academic institutions virtually without cost (except for transportation) for research and student use.

1. J. B. Knauer and R. C. Martin, "Californium-252 Production and Neutron Source Fabrication," in *Californium-252: Isotope for 21st Century Radiotherapy*, J. G. Wierzbicki, Ed., Kluwer Academic Publishers, The Netherlands, 1997, pp. 7–24.
2. *Environmental Assessment of the Thermal Neutron Activation Explosive Detection System for Concourse Use at U.S. Airports*, U.S. Nuclear Regulatory Commission Report NUREG-1396, 1990, p. 42.
3. R. C. Martin, T. E. Byrne, and L. F. Miller, "Biomedical Neutron Research at the Californium User Facility for Neutron Science," *J. Radioanal. Nucl. Chem.* **236**(1–2), 5–10 (1998).
4. "Representative Costs for ^{252}Cf Source Loans," in *Californium-252 Newsletter*, Oak Ridge National Laboratory, Vol. 3, No. 2, October 1999 (available from the author).