Production, Distribution, and Applications of Californium-252 Neutron Sources

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The radioisotope ^{252}Cf is routinely encapsulated into compact, portable, intense neutron sources with a 2.6-year half-life. A source the size of a person's little finger can emit up to 10^{11} neutrons/s. Californium-252 is used commercially as a reliable, cost-effective neutron source for prompt gamma neutron activation analysis (PGNAA) of coal, cement, and minerals, as well as for detection and identification of explosives, land mines, and unexploded military ordnance. Other uses are neutron radiography, nuclear waste assays, reactor start-up sources, calibration standards, and cancer therapy. The inherent safety of source encapsulations is demonstrated by 30 years of experience and by U.S. Bureau of Mines tests of source survivability during explosions. The production and distribution center for the U.S. Department of Energy (DOE) Californium Program is the Radiochemical Engineering Development Center (REDC) at Oak Ridge National Laboratory (ORNL). DOE sells ^{252}Cf to commercial reencapsulators domestically and internationally. Sealed ^{252}Cf sources are also available for loan to agencies and subcontractors of the U.S. government and to universities for educational, research, and medical applications. The REDC has established the Californium User Facility (CUF) for Neutron Science to make its large inventory of ^{252}Cf sources available to researchers for irradiations inside uncontaminated hot cells. Experiments at the CUF include a land mine detection system, neutron damage testing of solid-state detectors, irradiation of human cancer cells for boron neutron capture therapy experiments, and irradiation of rice to induce genetic mutations.

Keywords: californium, ^{252}Cf, neutron, irradiation

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Introduction

The radioisotope ^{252}Cf is an intense neutron emitter that is routinely encapsulated in compact, cylindrical source capsules. Decay by alpha emission (96.91% probability) and spontaneous fission (3.09% probability) results in an overall half-life of 2.645 years and neutron emission of 2.314×10^6 s^{-1} μg^{-1}, with a specific activity of 0.536 mCi/μg. The neutron energy spectrum is similar to a fission reactor, with most probable energy of 0.7 MeV and an average energy of 2.1 MeV. Portable ^{252}Cf neutron sources can provide an ideal nonreactor source of neutrons for lower-flux applications. Large masses of ^{252}Cf (>100 mg) can approach reactor capabilities for applications such as neutron radiography with reduced design, regulatory, and staffing costs relative to those for a reactor. Procuring large masses of ^{252}Cf is either very expensive or very inexpensive, depending on whether the radioisotope is purchased or loaned.

As background, a single source slightly smaller than a person's little finger (i.e., 5 cm long by 1-cm diameter) is licensed to contain up to 50 mg of ^{252}Cf (neutron intensity $>10^{11}$ s^{-1}), although similar sources are available with intensities as low as 10^4 s^{-1}. The dose equivalent rate from 1 μg of ^{252}Cf at 1 m in air is 0.0221 mSv/h (2.21 mrem/h) from fast neutrons plus 0.0019 mSv/h from gamma rays. As a rule of thumb, the monthly decay of ^{252}Cf is slightly more than 2% of the initial mass.

ORNL has an ongoing loan program to supply sealed ^{252}Cf sources to qualified organizations at a significantly discounted cost. With the recent establishment of the CUF, individuals and organizations involved in developmental neutron R&D can perform experiments within uncontaminated hot cells using a wide range of available ^{252}Cf source sizes without the regulatory, radiological, and infrastructural concerns of source possession.

Applications of ^{252}Cf Neutron Sources

The largest masses of ^{252}Cf are used for neutron radiography of weapons components (Barton et al., 1999). Californium-252 is commonly applied to PGNAA of samples such as coal, cement, explosives, and chemical munitions (Osborne-Lee and Alexander, 1995). PGNAA provides rapid nondestructive elemental assay of the principal components of a sample. Other commercial applications include fissile material and transuranic waste analyses via delayed neutron counting, fuel rod scanning (to determine enrichment and uniformity of fissile material within the fuel rod), and start-up sources to initiate the fission process within nuclear reactors. Specialized applications include conventional neutron activation analysis (NAA) followed by gamma spectroscopy for trace multielemental analysis (MacMurdo and Bowman, 1978), cancer brachytherapy (insertion of ^{252}Cf sources into or around the region of the tumor) (Tacev et al., 1998), confirmation of the presence of land mines (Clifford et al., 1999), and calibration and check sources for neutron detection instruments and monitors. A literature survey was recently conducted on publications involving either use of ^{252}Cf sources or calculation of ^{252}Cf properties. Over 100 publications were cited from May 1, 1998, through September 30, 1999. A listing of these references is available upon request from the authors.

Source Integrity During Use

Californium-252 sources fabricated during the earliest years of the DOE Californium Program (nearly 30 years ago) are still in use without problems. In over 20 years of ^{252}Cf source fabrication at ORNL, including hundreds of sources, customers have been inconvenienced only twice by minor problems with source capsule integrity, both involving single-encapsulated sources. In the 1980s, one source had a pinhole flaw, detectable only under rigorous scrutiny. Another source was not removed from a corrosive bath by a customer for several months, degrading the encapsulation. Neither source posed a serious operational concern, and both were returned to ORNL without incident. Relatively few sources are single encapsulated. No double-encapsulated sources have been returned to ORNL as a result of encapsulation concerns. Inspection of sources at ORNL is rigorous, and sources are permanently removed from circulation at any hint of an encapsulation anomaly.

Californium-252 sources are used in explosives detection systems. Concerns about blowing up a ^{252}Cf source with potential dispersion of radioactive material have been addressed by U.S. Bureau of Mines tests on sealed nonradioactive (dummy) ^{252}Cf source capsules to be used in an airport luggage inspection system (U.S. Nuclear Regulatory Commission, 1990). These capsules were found to be remarkably durable under the impact of 4.5 kg of plastic explosive. Although the experimental mock-up was destroyed, the source appeared visually to be in good condition and was confirmed to be intact in subsequent leak tests. Other reports of capsule integrity include aviation fuel and oil well fires and firing from a 50-caliber machine gun into a concrete wall without breaching the capsules (Rivard, 1998).

Heavy Element Production in the United States

Californium-252 was discovered in 1952 in thermonuclear test debris. Its potential as a spontaneous source of neutrons was soon realized. Demand for larger masses of ^{252}Cf rapidly increased after the first micrograms were produced in research reactors. A market evaluation and source fabrication program was initiated at Savannah River Laboratory (SRL) in the 1960s. Transfer of source fabrication operations and program responsibilities from SRL to ORNL was completed in 1986 (Knauer and Martin, 1997). Today most of the world's supply of ^{252}Cf is processed, purified, and encapsulated for shipment at the REDC.

Over 70 heavy element production "campaigns" have been conducted at the REDC. These campaigns consist of the fabrication of actinide oxide target rods (nominally ten rods containing ~100 g of actinide), followed by irradiation in the flux trap (thermal neutron flux $>10^{14}$ cm^{-2} s^{-1}) of the neighboring High Flux Isotope Reactor (HFIR). After irradiation, the targets are transferred back to the REDC, where they are dissolved, their heavy element components separated and purified, and the californium fraction transferred to the Californium Facility for further purification and source fabrication. The actinide feed material (americium and curium) is recycled into subsequent batches of target rods. The first campaigns irradiated plutonium oxide for nearly 2 years. After repeated recycling, the average isotopic mass of the actinide feed material increased to the point that ^{244}Cm is now the primary actinide, with reduced americium content but significant ^{246}Cm content. This heavy feed material reduces the required irradiation times to less than 1 year.

Current ^{252}Cf production averages ~250 mg per year. Recent production history includes processing campaigns that began in September 1994 (>450 mg of ^{252}Cf), March 1996 (>400 mg), and September 1998 (~370 mg). Ten curium oxide targets were fabricated and began irradiation in HFIR in early June 1999, with another campaign planned to coincide with an extended HFIR maintenance shutdown in mid-2000. Another batch of targets will be fabricated and ready for irradiation when the HFIR returns to operation. This sequence of campaigns will ensure an uninterrupted supply of ^{252}Cf to users during and after the HFIR shutdown. The only recent interruption in ^{252}Cf production was an unplanned shutdown of the HFIR between 1987 and 1989. The inventory of ^{252}Cf was sufficient to supply orders throughout this period. Inventories of ^{252}Cf have been sufficient to meet demand since the earliest days of the Californium Program. Other heavy element production is typically ~10 mg of ^{249}Cf, tens of mg of ^{249}Bk, ~1 mg of ^{253}Es, ~0.1 mg of "milked" ^{253}Es (from ^{253}Cf decay), a few micrograms of ^{254}Es, a few nanograms of ^{255}Fm, and <1 pg of ^{257}Fm. These isotopes are distributed to DOE-supported laboratories for research by interested parties without charge for the isotope.

Standard REDC source designs have been documented elsewhere, including specialized medical source designs (Knauer and Martin, 1997). The REDC distributes ^{252}Cf in two physical forms. For commercial reencapsulation, a cermet of californium oxide, Cf_2O_3, in a palladium matrix is shaped into thin wires (>1-mm diameter, with 200-μg cm^{-1} specific activity) or melted into pellets. For industrial and research sources, the ^{252}Cf is converted into either an oxide powder (by heating an oxalate slurry inside a source capsule) or oxysulfate microspheres (after firing of the ion-exchange resin, the microspheres are pressed into a pellet with aluminum powder). A few sources are single encapsulated (most notably, a point calibration source for the National Institute of Standards and Technology), but the vast majority are double encapsulated for safety.

Source Distribution Programs

Californium-252 is distributed to users under the auspices of the DOE Californium Sales/Loan Program. For commercial applications, ^{252}Cf is supplied to users by way of domestic and foreign commercial vendors. These reencapsulators typically purchase ^{252}Cf in bulk, usually as cermet wires, and fabricate sources to customers' specifications. For commercial applications, the Californium Sales/Loan Program may not compete with commercial vendors unless ORNL is uniquely qualified to meet the source requirements. A list of commercial vendors will be provided by the authors upon request.

To benefit government programs, government-funded research, or university research and training, the Californium Industrial/University Loan Program was established in the 1970s to provide low-cost access to ^{252}Cf sources. The Medical Loan Program addresses medical applications as part of the University Loan Program. Government agencies, government subcontractors, government-funded researchers, universities, and university-affiliated medical institutions are all eligible for loan of ^{252}Cf sources from ORNL. DOE has approved some international loans of ^{252}Cf sources. For example, between 1973 and 1975, ^{252}Cf sources for medical research were provided under the auspices of the International Atomic Energy Agency (Hall and Rossi, 1974).

DOE loans provide the ^{252}Cf radioisotope free of charge. If a preexisting source from the REDC inventory can meet the user's needs, then no source fabrication charges are incurred. Other charges include the loan fee for the source, source and container handling fees, and standard radioactive materials container packaging (RAMSPAC) fees. The loan fee includes a prepayment of the source handling charges incurred upon return of the source to ORNL to close out the loan. The only fee charged by ORNL at the time of source return is the container handling fee (and RAMSPAC fee for ORNL-supplied containers). Transportation charges for the source and container are paid directly to the shipper by the user and are based on per-mile charges (typically $1.25 to $1.50 per mile to and from ORNL) for large Type B shipments (see the discussion that follows); Type A shipments are usually much less. ORNL-owned shipping containers are commonly used, although customer-owned or customer-supplied containers can also be used.

University/Medical Loan Program

A university or medical loan is typically limited to two sources, each $\leq 5\ \mu$g of ^{252}Cf, although exceptions for larger sources can be made with adequate justification. To promote educational and research use of these relatively low-intensity sources, all standard source and container handling fees are waived, with the educational institution required to cover only the cost of transportation of the container.

Industrial Loan Program

Qualified nonuniversity (or high-intensity-source) users can obtain sources under the Industrial Loan Program. Available sources range from submicrogram quantities to tens of milligrams of ^{252}Cf. Preexisting sources containing 8 mg or more of ^{252}Cf are often available, thus avoiding source fabrication fees; however, larger sources must be fabricated on demand. To estimate the total cost for a loaned source, recent cost estimates provided by the ORNL Isotope Distribution Office are summarized in Table 1; some normalization of estimates was performed to provide a comparable cost basis. Estimates will change along with changes in hourly manpower costs, and estimates vary for nonroutine requests. A formal price estimate should be requested for precise planning. The typical range for each type of fee (as of September 1999) is summarized in Table 2. The total cost estimate for a loan (Table 1) is obtained by combining the requisite fees listed in Table 2.

Because the radioisotope is free of charge, the primary cost driver for most loaned sources is the size of the requisite shipping container and its associated handling charges. For transportation purposes, the most common classifications for radioactive source shipments are Type A (<5 mg of ^{252}Cf) and Type B (>5 mg). Type B containers tend to be larger and heavier and require dedicated transportation, thus the sharp increase in cost between 300-μg and 8-mg sources in Table 1. Another significant cost increase is evident when source fabrication is required. Multiple sources can be shipped inside a shipping container as long as radiation readings on the outside of the container do not exceed regulatory limits. The last entry in Table 1 requires two shipments because no Type B container is licensed to transport more than 80 mg of ^{252}Cf. For small ^{252}Cf masses (<50 μg), commercially available sources cost significantly less than those available from the ^{252}Cf Industrial Loan Program. However, ORNL is obligated to accept return of loaned sources. Purchase from a ^{252}Cf vendor may ultimately incur waste disposal costs unless nondisposal options are available.

Isotope costs

The current cost of ^{252}Cf purchased from DOE under the ^{252}Cf Sales Program is $60 per microgram ($60,000 per milligram). Historically, in the early days of the Californium Program (early 1970s), the price of ^{252}Cf was set at $10 per microgram. That price increased to $27 μg^{-1} in 1987, $50 μg^{-1} in 1989, $55 μg^{-1} in 1994, $56 μg^{-1} in 1998, and $60 μg^{-1} in 1999. No further price increase is projected, at least through September 2000. The cost of the radioisotope does not include the technical service fees required for encapsulation of the ^{252}Cf and preparation for shipment. Technical service fees for a multi-milligram shipment typically range from $20,000 to $25,000. The customer also pays all transportation charges.

^{252}Cf Shipping Containers

Table 3 lists the range of shipping containers commonly used by the REDC for ^{252}Cf shipments. Some are owned by ORNL; some are owned by commercial ^{252}Cf vendors or container manufacturers. This table can also be used for qualitative estimates of shielding requirements for source storage. Containers are typically cylindrical, except for the semispherical Type B containers.

The only licensed ^{252}Cf Type B shipping containers in the United States are owned by ORNL. Because these containers were fabricated in the 1970s, under regulatory requirements for documentation and testing different than those in place today, a project to build a new ^{252}Cf Type B shipping container has been initiated by ORNL and the DOE. The new cask will hold up to 60 mg of ^{252}Cf and will be licensed for all modes of domestic and international transport (ground, sea, and air). The project is currently progressing, with container delivery from the vendor currently estimated in late 2001.

The Californium User Facility for Neutron Science

The Californium Facility (CF) at ORNL stores the national inventory of ^{252}Cf neutron sources for the loan program. The CUF was established in 1996 to make these sources and the CF source-handling infrastructure available to outside researchers and industry and to provide a cost-effective option for ^{252}Cf-based studies without the capital outlay or regulatory issues typically involved. The CUF provides a unique resource for radiation effects testing and is ideal for experiments requiring moderate neutron fluxes such as radiation effects on electronic or biological systems, versatile irradiation geometries, or large samples.

The CUF encompasses two large uncontaminated walk-in hot cells for experimental set-up and irradiation, the water-filled ^{252}Cf storage pool, available neutron sources, pneumatic transfer capabilities, and associated infrastructure and technical support. Sealed sources containing submicrogram quantities to tens of milligrams of ^{252}Cf are routinely handled. A maximum of 60 mg may be used in-cell, capable of providing thermal and fast neutron fluxes exceeding 10^8 cm^{-2} s^{-1}. Approval to increase this 60-mg limit to 100 mg has been requested. Total fluxes of $>10^9$ cm^{-2} s^{-1} are available for irradiations within the water-filled ^{252}Cf storage pool. Sources totaling <175 μg (up to 4×10^8 neutrons/s) may be used in out-of-cell tests within the CUF. The REDC has several shielded shipping containers that could be used to shield sources during out-of-cell testing.

After hands-on experimental setup inside the uncontaminated hot cell, all personnel are evacuated, the cell is closed, and the ^{252}Cf sources are pneumatically transferred into the cell and positioned remotely using manipulators. Irradiation samples up to 5.7 cm in diameter can be cycled through the closed hot cell via sample ports. For ^{252}Cf storage pool irradiations, a sample can either be pneumatically transferred into the storage lattice or manually lowered into the pool, and the neutron field tailored by rearranging sources. For an estimated cost of ~$15,000, a low-tech manually operated irradiation station could be installed to access up to several hundred milligrams of ^{252}Cf within the pool. REDC staff can provide additional assistance in gamma spectroscopy and neutron and photon transport calculations for experimental design.

Irradiation testing of semiconductor detectors

A Northeastern University – University of Minnesota consortium used up to 59 mg of ^{252}Cf in a series of irradiations to test the neutron hardness of avalanche photodiode detectors for use in high-energy physics experiments at the CERN accelerator (Reucroft et al., 1997). Real-time data acquisition and computer control were provided by cables connecting the experiment to out-of-cell hardware. The experiments simulated the expected in-service fast neutron flux of 2×10^5 cm^{-2} s^{-1}, and accelerated irradiations provided the predicted lifetime fast fluence of 1.2×10^{13} cm^{-2} in 335 hours of irradiation.

Confirmation of non-metallic land mines by thermal neutron activation

To support peacekeeping activities, the Canadian Department of National Defence has designed a mobile multisensor system to detect buried land mines. The thermal neutron activation (TNA) sensor is designed to confirm a mine's presence by detection of nitrogen within the explosive via PGNAA. A prototype TNA system based on 40 to 80 μg of ^{252}Cf has been demonstrated to confirm the presence of most antitank mines and many antipersonnel mines within a few minutes of analysis. The high-rate-counting electronics were tested at the CUF with a range of ^{252}Cf source intensities, and acceptable performance at count rates up to 1.5×10^6 counts per second was demonstrated using 300 μg of ^{252}Cf (Clifford et al., 1999). The system was thus demonstrated to be capable of 4 times faster operation than in previous tests.

Irradiation of cancer cells

Boron neutron capture therapy (BNCT) experiments on the relative effectiveness of prototype boron-containing compounds for cancer treatments have been performed at the CUF. Living lung cancer cells impregnated with the boron compounds were exposed to ^{252}Cf neutrons. The relative cell survival fractions after irradiation indicated the efficacy of each compound for use in BNCT. Four ^{252}Cf sources (28 mg total) surrounding the test tube sample provided a thermal neutron flux of 2×10^8 cm^{-2} s^{-1} during the 0.5- to 4.0-min irradiations.

Neutron-induced genetic mutation of rice

After a 20-year lull, interest in radiation-induced genetic mutations in plants is increasing due to new techniques for DNA sequencing. Researchers at the U.S. Department of Agriculture, Agricultural Research Service, Rice Research Unit, located in Beaumont, Texas, are interested in inducing genetic mutations in rice by fast neutron irradiation, with a goal of identifying and cloning genes of agronomic interest. Neutron-induced deletion of a necessary gene or regulator will degrade fertility, nutritional quality, or some other desirable property of the plant. By comparing the DNA of the damaged plants with that of the unirradiated plants, one can conclude that any missing DNA base sequences in the damaged plants correspond to the gene or piece of DNA that provides desirable plant properties, thus facilitating cloning of genes from plant varieties with desirable traits.

In order to reproduce earlier reactor beam irradiations with low gamma contamination, REDC staff used the MCNP-4B radiation transport code to design a simple irradiation experiment. Unshielded ^{252}Cf has ~10% gamma contribution to total dose. By placing 5 cm of lead (i.e., a standard lead brick) between the

^{252}Cf sources and rice sample, the gamma contribution to dose was reduced to 2% of total. By sandwiching a 2.5-cm-thick rice sample between two lead bricks and placing 25 mg of ^{252}Cf on each side of the "sandwich," an average dose rate to the rice of 19.5 Gy/h was calculated. This simple in-cell experiment provided a fast neutron dose that was 55% that of a reactor beam line (Pinson, 1999). Rice samples were irradiated to doses ranging from 20 to 120 Gy. Germination rates have not yet been reported.

Summary

The reliability and wide range of applications of ^{252}Cf sources have been demonstrated during 30 years of use. Sources are available from commercial vendors for commercial applications, while the Industrial Loan Program provides high-intensity sources at considerable savings for government and subcontractor use. The University/Medical Loan Program encourages educational and medical research and training by providing sources at negligible cost. The value and convenience of the CUF to the neutron research community has been demonstrated in several noteworthy experiments. The ability of the CUF to provide neutron dose rates approaching those of a reactor beam was computationally demonstrated.

For information on the availability of ^{252}Cf sources, the primary contact is Paul A. Balo, telephone (423) 574-1948, e-mail *balopa@ornl.gov*. For information on ^{252}Cf sources and applications, accessing the Californium User Facility, or obtaining copies of the *Californium-252 Newsletter*, the primary contact is Rodger C. Martin, telephone (423) 576-2280, e-mail *martinrc@ornl.gov*. More detailed information on previous experiments performed at the CUF, CUF capabilities, and general ^{252}Cf applications is available at the web site *http://www.ornl.gov/divisions/ctd/cuf.htm*. The site *http://redc.ct.ornl.gov* provides information on heavy element campaign schedules and on general REDC operations. Information on the availability of heavy elements other than ^{252}Cf and of other isotopes can be obtained from the web site *http://www.ornl.gov/isotopes/catalog.htm*.

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Table 1. Recent Cost Estimates for Industrial Loans

Date	Number of sources	Requested ^{252}Cf mass per source (μg)	Estimated cost	Source fabrication required?	^{252}Cf value @ $60/$\mu$g
12-98	1	0.2	$7,400	no	$12
8-98	1	6	$9,200	no	$360
3-99	1	10	$10,600	no	$600
5-99	1	50	$13,600	no	$3,000
11-98	1	300	$14,200	no	$18,000
8-98	1	8,000	$23,000	no	$480,000
1-99	2	27,000	$51,700 (single source @ $34,800)	yes	$3,240,000
7-99	2	50,0000	$80,800 (single source @ $44,500)	yes	$6,000,000

Table 2. Fee Breakdown for ^{252}Cf Source Loans[a]

Single-source handling fees	$2000 to 2500
RAMSPAC packaging fees (shipments to customer)	$1045
Container handling fees (source shipment to customer)	
<5000-lb container (Type A)	$6500 to 7000
Type B container	$8500 to 10,500
Container handling fees (source return to ORNL)	
Type A	$1000 to 2000
Type B	~$4000
Single-source loan fees (assumes 5-year loan period)	
≤7 μg	$3805
>7 μg to ≤3.7 mg	$6970
>3.7 mg	$9975
Each additional source (waived for sources <1 μg)	$1850
Single-source loan extension fees (assumes 5-year extension)	
≤7 μg	$825
>7 μg to ≤3.7 mg	$1510
>3.7 mg	$2160
Each additional source (waived for sources <1 μg)	$400

[a]Representative fees as of September 1999. Additional fees are incurred for nonroutine orders, services, or source fabrication.

Table 3. ^{252}Cf Shipping Containers Used by ORNL

Container type	Weight (lb)	Height x width (in.)	Nominal maximum ^{252}Cf content[a]
10-gal 6M Type B drum	160	17 x 14	6 μg
55-gal drum	650	36 x 24	100 μg
Type A	300	20 x 18	15 μg
Type A	680	28 x 24	50 μg
Type A	1220	34 x 30	200 μg
Type A	2100	38 x 30	500 μg
Type A	3180	46 x 42	1.5 mg
Type A	3800	48 x 44	3.0 mg
Type A[b]	4600	50 x 50	3.7 mg
Type A[b]	7400	60.5 x 58	5.0 mg
Type B	9550	75 x 62	80 mg
Type B	23,500	80 x 68	60 mg

[a] ^{252}Cf contents that generate maximum permissible external dose-equivalent rates for routine transportation; that is, 2 mSv/h (neutron plus gamma) at the container surface and 0.1 mSv/h 1 m from the surface.

[b] Vendor-owned container.