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CALIFORNIUM-252: A Remarkable Versatile Radioisotope

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MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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Manuscript Completed: October 10, 1995

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for the
U. S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400

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ACRONYMS

AEC	Atomic Energy Commission
ANS	American Nuclear Society
BWR	Boiling Water Reactor
CCNS	Californium Cold Neutron Source
CONAC	Continuous ONline Analyzer of Coal
DOE	U. S. Department of Energy
ELAN	ELemental ANalyzer
ERDA	Energy Research and Development Administration
FDA	Food and Drug Administration
GE	General Electric
HFIR	High Flux Isotope Reactor
INAA	Instrumental Neutron Activation Analysis
INEL	Idaho National Engineering Laboratory
LANL	Los Alamos National Laboratory
MAPS	Magnetic And Passive Scanning device
MCI	Mineral Control Instrumentation
NAA	Neutron Activation Analysis
NR	Neutron Radiography
NRC	U. S. Nuclear Regulatory Commission
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory
PAN	Passive/Active Neutron (shuffler)
PDMS	Plasma Desorption Mass Spectrometry
PGNAA	Prompt Gamma Neutron Activation Analysis
PINS	Portable Isotopic Neutron Spectrometry
ppm	parts per million
PWRs	Pressurized Water Reactor
REDC	Radiochemical Engineering Development Center
SJSU	San Jose State University
SRL	Savannah River Laboratory
SRTC	Savannah River Technology Center

CHEMICAL SYMBOLS

Al	aluminum
^{241}Am	americium-241
Am:Be	americium combined with beryllium
^{10}B	boron-10
^{133}Ba	barium-133
^{249}Bk	berkelium-249
^{250}Bk	berkelium-250
C	carbon
Ca	calcium
Cl	chlorine
^{249}Cf	californium-249
^{250}Cf	californium-250
^{251}Cf	californium-251
^{252}Cf	californium-252
^{244}Cm	curium-244
^{245}Cm	curium-245
^{246}Cm	curium-246
^{247}Cm	curium-247
^{248}Cm	curium-248
^{249}Cm	curium-249
^{250}Cm	curium-250
^{60}Co	cobalt-60
^{137}Cs	cesium-137
^{253}Es	einsteinium-253
^{254}Es	einsteinium-254
Fe	iron
^{257}Fm	fermium-257
Gd	gadolinium
Ge	germanium
H	hydrogen
Li	lithium
N	nitrogen
(n,α)	nuclear reaction in which a nucleus emits an alpha particle following neutron absorption
Po:Be	polonium combined with beryllium
^{238}Pu	plutonium-238
^{238}Pu:Be	plutonium-238 combined with beryllium
S	sulphur
Si	silicon
^{124}Sb:Be	antimony-124 combined with beryllium
^{235}U	uranium-235
U_3O_8Al	uranium oxide-aluminum cermet

EXECUTIVE SUMMARY

Californium-252 (^{252}Cf), a radioactive isotope, is a material of major importance. First isolated from the debris of the Mike thermonuclear test at Enewetak in November 1952, ^{252}Cf has found many applications in medicine, scientific research, industry, and nuclear science education. Californium-252 is a unique source of neutrons because it yields a highly concentrated and reliable neutron spectrum from a very small assembly. During the past 40 years, ^{252}Cf has been applied with great success to cancer therapy; neutron radiography of objects ranging from flowers to entire aircraft; startup sources for nuclear reactors; elemental analysis of coal, nuclear fuel, explosives, and the human body; and many other beneficial uses. For many of these applications, ^{252}Cf is unique and irreplaceable.

Many of the numerous applications of ^{252}Cf are summarized in this report. The Californium use tree (Fig. 1.2) emphasizes the large number and broad range of uses of this isotope and underscores the fact that this is made possible through the sponsorship of the U.S. Department Of Energy (DOE). Many of these applications, which were only ideas when the tree was conceived, have come to fruition and now save lives, time, and money. Some of these benefits could not be realized without ^{252}Cf. A few examples of these benefits are briefly described in the following:

- Use of ^{252}Cf in brachytherapy has proven very effective in the treatment of certain cancers that are otherwise incurable (e.g., certain cervical and brain tumors); consequently, hundreds of lives have been saved with ^{252}Cf. Medical researchers are pursuing other potential uses (e.g., treatment of eye and brain cancers).

- Reactor startup is one of the most critical applications of ^{252}Cf. When a nuclear reactor is started up for the first time, a stable source of background neutrons is needed to ensure a properly controlled approach to criticality. A similar situation exists during fuel reloading, particularly if the reactor has been down for an extended period of time (e.g., for maintenance). Today, most existing and virtually all new reactors in the United States and many reactors in other countries are designed to take full advantage of ^{252}Cf startup sources produced in the United States.

- The nondestructive aircraft examination facility at McClellan Air Force Base in California uses neutrons produced by a ^{252}Cf source. The "n-ray" examination (similar to an x-ray, which is also used) reduces the probability that aircraft will be lost because of in-flight structural failure or fuel leakage through corroded aluminum honeycomb in the wings. The facility also reduces the time required for routine maintenance inspections, thus resulting in fewer planes out of service at any time. This benefit can lead to smaller fleets and subsequent cost reductions.

Applications such as these, which require a very intense neutron source that is also mobile and compact, must use ^{252}Cf—there is no substitute. Furthermore, multi-million-dollar industries have grown out of the availability of ^{252}Cf, and are projected to continue to grow over the next decade. These industries include on-line bulk stream analysis and active-fuel-rod-scanning industries, for which ^{252}Cf is *the* essential component. Other industries—such as the nuclear power industry, which has designed its reactors around the compact ^{252}Cf startup source—have become dependent on ^{252}Cf to the point that switching to an alternative neutron source would represent an enormous new investment.

It is clear that ^{252}Cf is an important material that is having a worldwide impact. Yet, it is produced in only two places in the world: the United States and Russia. Continued production of this material is strongly supported by many people, nuclear facilities, academic institutions, and businesses for which its availability is essential and critical.

1. INTRODUCTION

Californium-252 (^{252}Cf) is a radioactive isotope of one of over 100 known chemical elements. A product of the nuclear age, ^{252}Cf has found many applications in medicine, scientific research, industry, and nuclear science education. Californium-252 is unique as a neutron source in that it provides a highly concentrated flux and extremely reliable neutron spectrum from a very small assembly. During the past 40 years, ^{252}Cf has been applied with great success to cancer therapy, neutron radiography of objects ranging from flowers to entire aircraft, startup sources for nuclear reactors, fission activation for quality analysis of all commercial nuclear fuel, and many other beneficial uses, some of which are now ready for further growth. Californium-252 is produced in the High Flux Isotope Reactor (HFIR) and processed in the Radiochemical Engineering Development Center (REDC), both of which are located at the Oak Ridge National Laboratory (ORNL) in Oak Ridge, Tennessee. The REDC/HFIR facility is virtually the sole supplier of ^{252}Cf in the western world and is the major supplier worldwide.

1.1 BRIEF HISTORY OF ^{252}Cf

Extensive exploitation of this product was made possible through the ^{252}Cf Market Evaluation Program, sponsored by the United States Department of Energy (DOE) [then the Atomic Energy Commission (AEC) and later the Energy Research and Development Administration (ERDA)]. This program included training series, demonstration centers, seminars, and a liberal loan policy for fabricated sources.

The Market Evaluation Program was instituted, in part, to determine if large-quantity production capability was required at the Savannah River Laboratory (SRL). Because of the nature of the product and the means by which it is produced, ^{252}Cf can be produced only in government-owned facilities. It is evident at this time that the Oak Ridge research facility can meet present and projected near-term requirements.

Californium-252 was first isolated from the debris of the Mike thermonuclear test at Enewetak in November, 1952. This isotope is characterized by spontaneous fission and the attendant strong neutron emissions and by a 2.64-year half-life. The unique potential of this new isotope to perform as a miniature nuclear reactor—providing a highly concentrated and reliable source of neutrons—became evident when increasing amounts became available from reactor irradiations in 1967. Early programs to develop industrial and medical applications of ^{252}Cf used small amounts of material loaned by the government. Then, the AEC announced in November 1970, that ^{252}Cf would be made available for sale at \$10/μg. The first sale of 1 mg took place in early 1971 to the University of Kentucky Chemistry Department to perform activation analyses on specimens retrieved from the moon. Other sales quickly followed, including sales to a half-dozen countries.

The production, shipment, and sales history of ^{252}Cf from ORNL is summarized in Table 1.1. From 1971 to 1985, sales rose sharply to a peak of nearly 60 mg/year. Cumulative annual production and sales of ^{252}Cf are shown in Fig. 1.1. Since 1985, the trend in sales has been fluctuating with current sales near 30 mg/year. The loan program to DOE facilities and others maintained a high level of participation after the initial flurry of interest and as experimental and test programs were replaced by established applications (see Table 1.1 and Fig. 1.1*d*). Sales in 1983 and 1984 totaled 98 mg, bringing \$1 million in revenues to the government. Since 1990, sales have averaged nearly 30 mg/year, and revenues have averaged about \$1.5 million annually at the current price of \$50/μg.

Table 1.1 History of the production, shipment and sales of ^{252}Cf

Fiscal year	Amount produced (mg)	Amount shipped (mg) [a]	Amount sold (mg)
1967	5.6	0.2	0
1968	0.5	3.0	0
1969	15	3.1	0
1970	52	33.7	0
1971	284	197.5	.61
1972	513	273.6	13.2
1973	428	194.7	13.6
1974	386	53.3	12.6
1975	717	225.0	19.8
1976	277	111.9	29.3
1977	499	286.7	26.3
1978	632	108.0	29.0
1979	322	127.2	26.6
1980	364	232.6	31.6
1981	899	36.5	36.5
1982	370	185.0	21.5
1983	291	163.8	51.0
1984	367	169.6	47.5
1985	450	127.4	55.5
1986	416	176.9	*b*
1987	429	273.7	<0.1
1988	0	131.2	19.3
1989	0	48.5	44.5
1990	0	150.9	39.4
1991	0	67.7	17.7
1992	375	85.2	33.1
1993	0	134.3	28.3
Total	8,092	3,601	597

[a] Including sales.

[b] Not available at press time.

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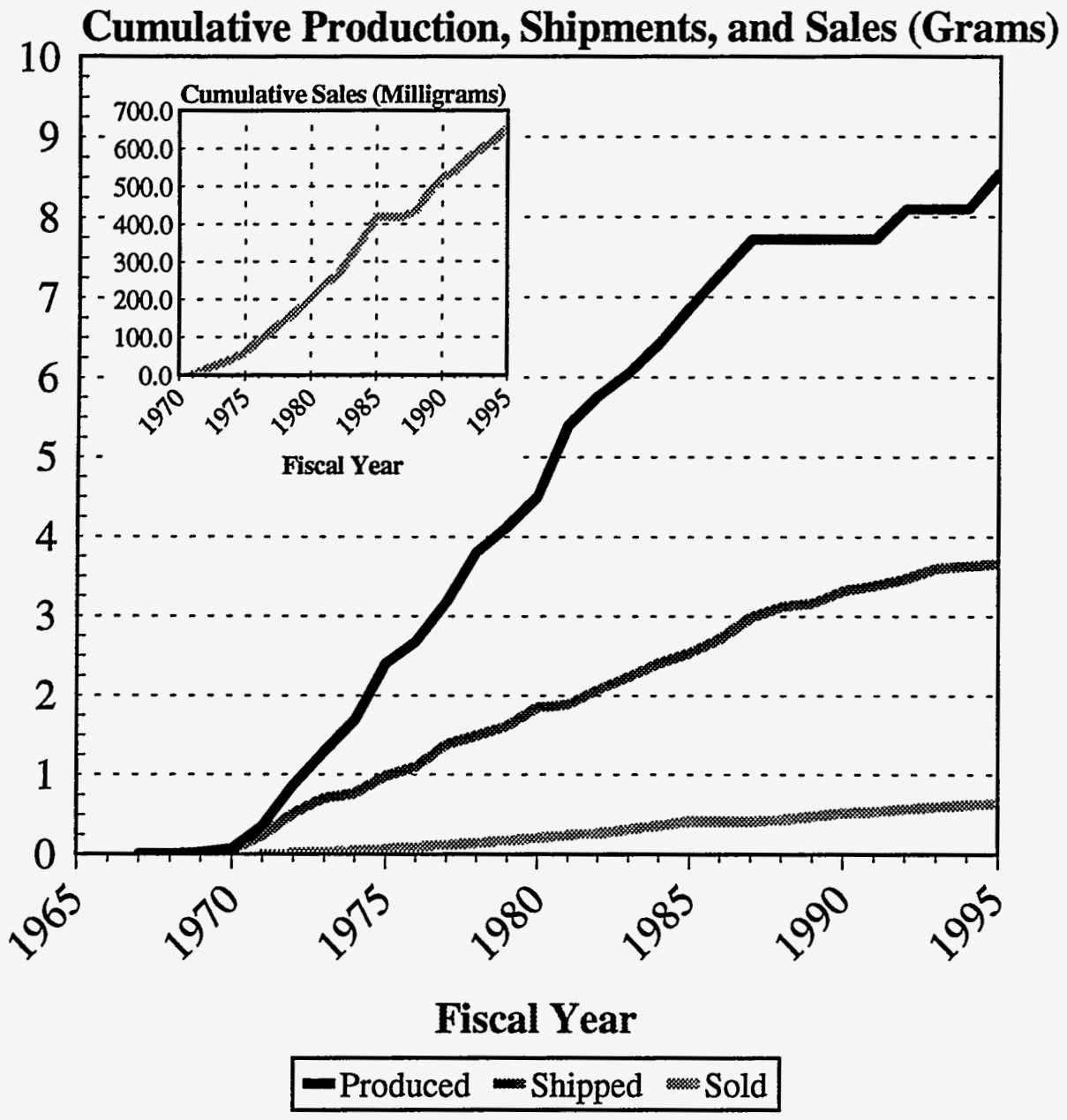

Fig. 1.1 Cumulative annual production, shipments, and sales of ^{252}Cf (from data in Table 1.1).

1.2 CALIFORNIUM-252 (^{252}Cf) WORKSHOPS

Since the major isotope production programs made ^{252}Cf one of the most available and the least expensive isotopic neutron source over 20 years ago, there have been development efforts around the world to take advantage of its unique properties. Corporate, medical, academic, and government researchers have come together periodically to share their discoveries and applications and to plan new uses of ^{252}Cf.

The first conferences on ^{252}Cf were sponsored by the American Nuclear Society (ANS), including an October, 1968 symposium on ^{252}Cf which was sponsored by the New York Metropolitan Section of the ANS,[1] a topical meeting on neutron sources and their applications,[1] and a topical meeting held in Washington, D.C. in 1972.[2] At that time, the focus was on the development of ^{252}Cf source fabrication and shielding methods; the design of neutron source facilities for a variety of purposes such as neutron activation analysis, neutron radiography, and education; and research to evaluate ^{252}Cf for a range of potential applications, such as:

- medical applications
- industrial process control
- elemental analysis of sample and bulk streams
- oceanographic exploration and studies, and
- materials inspection, including radiography and fuel rod scanning

The range of applications then developing or envisioned is perhaps best summarized by the "Californium Use Tree," (see Fig. 1.2) a figure first introduced by J. L. Crandall in 1968[2] and updated for the 1972 meeting.

In 1988, after assuming responsibility for the DOE ^{252}Cf Industrial Sales/Loan Program, ORNL hosted the ^{252}Cf Workshop in Oak Ridge, Tennessee. This workshop drew participants from around the world; it featured reports of numerous established applications—many of them commercial—and revealed an increasing demand for ^{252}Cf. Among the applications reported at the workshop were:

- cancer therapy trials, which showed that ^{252}Cf treatments can saves lives in certain otherwise incurable and terminal tumor cases
- neutron inspection of bulk materials to detect explosives in baggage, which was reported to be practical and commercially viable
- investment in neutron radiography—such as the major facility at McClellan Air Force Base—to improve aircraft safety
- numerous proven and promising applications for neutron activation analysis
- uses of ^{252}Cf in instrument calibration and as a neutron field standard for materials and personnel dosimetry, and
- ^{252}Cf-based instruction and research at colleges and universities where nuclear reactor facilities were not available

The 1993 World Congress on Neutron Radiography (NR),[3] updated the status of ^{252}Cf radiography applications at the McClellan Air Force Base and the Pantex and Mound facilities. Included are several reports (a) on ^{252}Cf- NR experience at the McClellan Air Force Base and (b) a feasibility study on the use of ^{252}Cf-NR for space shuttle inspection.

The ANS 1993 Summer Meeting featured a symposium on Neutron Activation Analysis Using ^{252}Cf. Researchers from the Savannah River Technology Center (SRTC), Cornell University, and Idaho National

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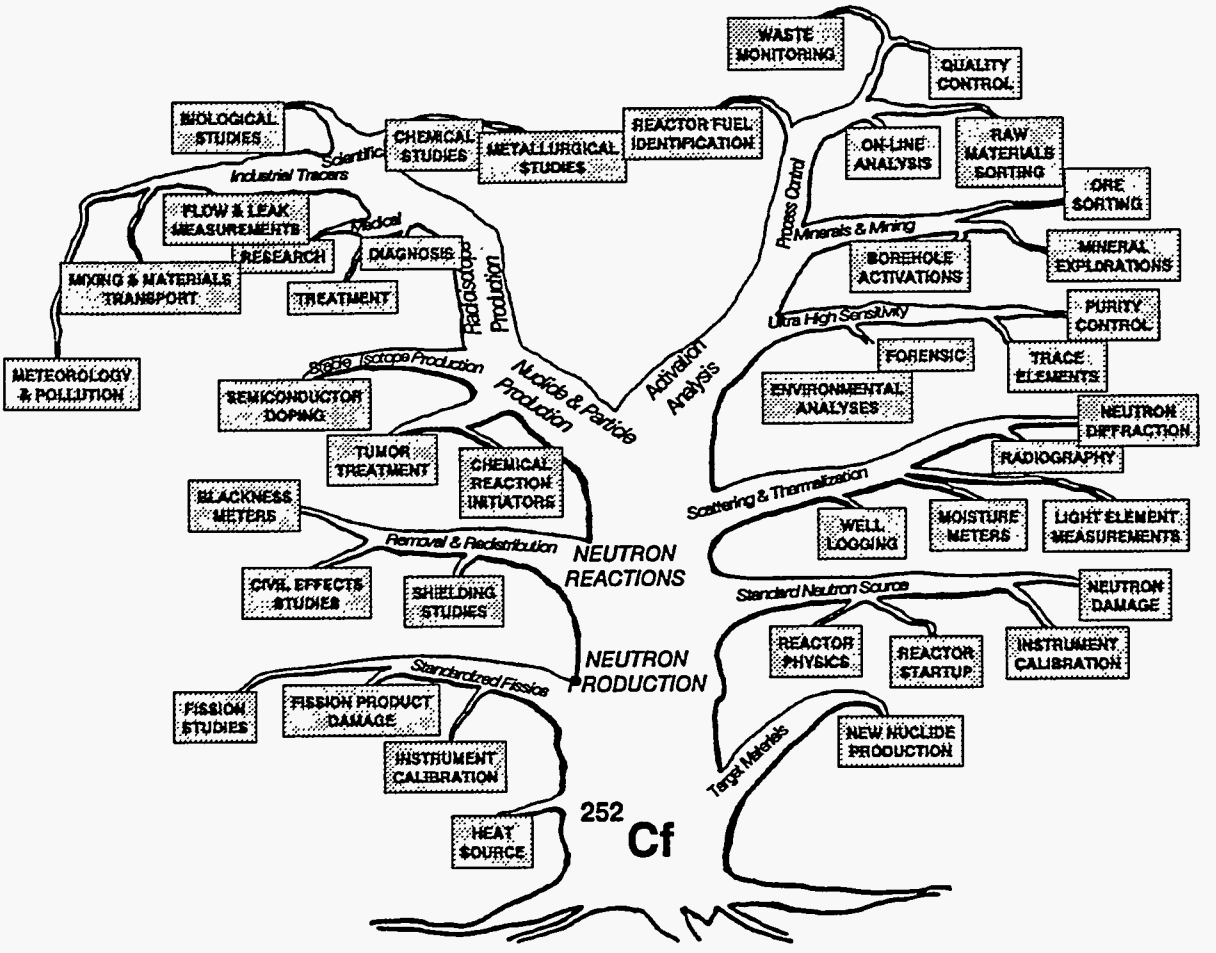

Fig. 1.2 Californium use tree [modified with permission from the original source, (reference 3)]

Engineering Laboratory reported on contemporary and new methods in activation analysis, including a proposed cold neutron irradiator and a field-demonstrated portable prompt gamma spectrometer.[4]

The latest conference, hosted by ORNL, was held in Oak Ridge in April of 1995†. Conferences such as these, in addition to providing a forum for information exchange, have served to highlight the many uses of ^{252}Cf and underscore its growing importance to humanity.

1.3 CURRENT STATUS

1.3.1 ^{252}Cf Production

Californium-252 is currently being produced at a rate of about 400-500 mg/year (Fig. 1.1) at the HFIR and is processed in the REDC at the ORNL. These facilities were constructed to produce special transplutonium isotopes for research purposes. These products include ^{248}Cm, ^{249}Bk, ^{249}Cf, ^{253}Es, ^{254}Es, and ^{257}Fm, as well as ^{252}Cf, which may be sold if in excess to research needs. After its production, the ^{252}Cf decays into ^{248}Cm, a valuable research isotope with a long half-life. The present ^{252}Cf production program, which is supplying the entire western world, can therefore be regarded as a spin-off from DOE's research effort. The ^{252}Cf must decay for several years before the ^{248}Cm can be recovered. During that time, each milligram of ^{252}Cf emits neutrons at a rate of 2.3×10^9/s. To gain benefit from neutron emissions that occur while waiting for the ^{252}Cf to decay to ^{248}Cm, DOE makes a large number of neutron sources available on a loan basis to various institutions, including

- DOE sites,
- medical institutions,
- educational institutions,
- other federal government agencies, and
- industrial organizations performing research or development contracts for the federal government,

Thus, a much larger quantity of ^{252}Cf finds its way into industrial, academic or commercial uses than just the amount indicated by sales. Table 1.1 summarizes the quantities of ^{252}Cf produced, sold, and shipped since the inception of these programs. This same information is shown in Fig. 1.1. In 1984, 170 mg were shipped from the REDC for both loans and sales (see Table 1.1). From 1970 to 1990, shipments averaged 150 mg/year. Since 1990, the amount shipped has averaged approximately 110 mg/year.

Because of the unique and unexcelled properties of ^{252}Cf as a portable, compact neutron source, it has become of key importance to many applications, some of them of far-reaching consequence. Established applications of ^{252}Cf are growing steadily, and several are now poised for even greater growth. The key properties of ^{252}Cf, which make it so invaluable as a neutron source, include:

- it is a 100% reliable source;
- it can be packaged into a small and simple unit;
- it can be fabricated into sources suitable for medical applications;
- it can provide a highly intense source of neutrons;
- its neutrons span a very useful energy range;

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†Information from the April, 1995 conference was not received in time for inclusion in this report.

- its side reactions are minimal or well known (e.g., extraneous radiation, heat evolution, and gas generation); and
- its half-life is in a very usable range— being neither too long nor too short.

1.3.2 HFIR Transmutation Chain

The HFIR produces transplutonium elements through ^{257}Fm by transmutation from curium oxide targets. The transmutation involves two processes: (1) neutron absorption, which increases the nuclear mass by 1, and (2) beta decay, which increases the atomic number (proton number) by 1. For example, ^{244}Cm absorbs a neutron to become ^{245}Cm. Then, ^{245}Cm absorbs a neutron to become ^{246}Cm and so on up to ^{249}Cm. Neutron absorption at this point in the chain is largely replaced by beta decay because the half-life of ^{249}Cm is short enough (64.2 min) to allow most of these nuclei to eject a beta particle before they can capture a neutron and become ^{250}Cm. The ^{249}Bk formed by the beta decay captures a neutron and becomes ^{250}Bk, which has a half-life for beta decay of 3.2 h. The decay of ^{250}Bk leads to the formation of ^{250}Cf and so on (see Fig. 1.3).

1.3.3 HFIR Facility

HFIR is a light water, pressurized research reactor that uses light water to cool the nuclear core and to slow down, or moderate, the neutrons, which are reflected back into the core by a beryllium reflector. The pressure vessel is located in a pool filled with light water. The fuel consists of a U_3O_8-Al cermet containing enriched ^{235}U and clad in aluminum plates. The HFIR produces 85,000 kW of thermal energy. Its continuous working thermal flux level averages 1.8×10^{15} neutrons/(cm^2s). The reactor core, which consists of an annular fuel element, holds 30 target rods. Some additional characteristics of the HFIR are given in Table 1.2.

1.3.4 Radiochemical Engineering Development Center

The REDC, which is located adjacent to the HFIR, fabricates the target rods of curium oxide clad in aluminum for irradiation in the HFIR. After the target rods are irradiated in the HFIR's high neutron flux, the targets are processed in the REDC at ORNL. The transplutonium elements produced are chemically separated and purified in a highly sophisticated, remotely operated process in equipment located in the REDC. The REDC also packages products of these transplutonium elements and ships them to research communities, source fabricators, and other users.

The elements separated from the HFIR targets at the REDC include actinides, lanthanides, and other fission products and the aluminum cladding. The separations performed at the REDC take advantage of the small differences in the chemical behaviors of the lanthanide and actinide elements, multiplied by the ion-exchange and solvent-exchange procedures that are used. Some of the processes used include both caustic and acid dissolution of the targets, solvent extraction using both the CLEANEX[5] and TRAMEX[6] processes, and both anion- and cation-exchange chromatography (see Fig. 1.4). Because the isotopes processed are highly radioactive, most chemical manipulations must be carried out in hot cells. The REDC is the only facility in the world capable of handling the complex chemical procedures required to separate and purify the transplutonium elements on a scale of 1 g of ^{252}Cf. Mainline production from the REDC/HFIR facility is summarized in Table 1.3.

1.3.5 Fields of Application

^{252}Cf is the sole commercially available isotope capable of fabrication into physically small sources which emit neutrons intensely over a practical period of time. This valuable isotope became available for commercial source fabrication in 1971, and since that time, it has found numerous applications (see Table 1.4). In 1982, Janzow[7]

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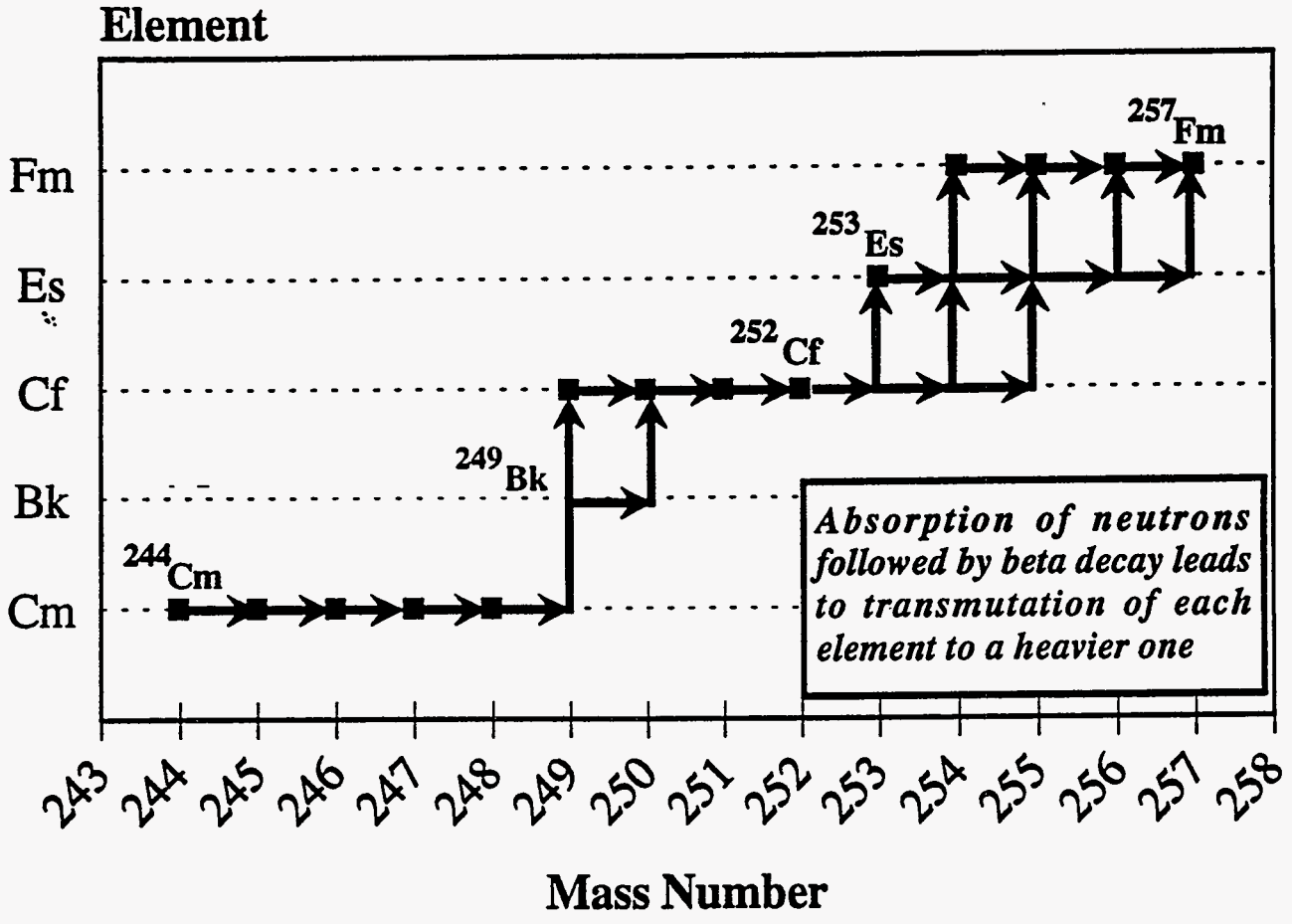

Fig. 1.3 Path of element transmutation in the High Flux Isotope Reactor (based on figure p. 55, ORNL Review, No. 3, 1985).

Table 1.2 History and characteristics of the High Flux Isotope Reactor (HFIR)

Date of first operation	1965
Date of first operation at full power	1966
Power	Initially 100,000 kW; in 1989, power limited to 85,000 kW (thermal) to improve safety margins for the reactor vessel.
Fuel	U_3O_8-Al; Al-clad, U-Al cermet
Fuel load	9.4 kg of ^{235}U
Fuel element	Two concentric annular elements each about 60 cm high; inner element: inside diameter 13 cm, outside diameter 27 cm; outer element: inside diameter 28 cm, outside diameter 45 cm. Thickness of (curved) fuel plates 1.3 mm
Reactor vessel	Stainless-clad carbon steel, 6.1 m high, 2.4 m-diameter top located 5.2 m below surface of the reactor pool
Control rods	Concentric cylinders; black zone: europium oxide-aluminum, grey zone: tantalum-aluminum, white zone: aluminum
Moderator	Light water
Reflector	Beryllium 30 cm thick surrounded by water
Coolant	Water flow 65,000 L/min through vessel
Average power density in core	1.7 MW/L
Peak thermal neutron flux (unperturbed)	3×10^{15} neutrons/(cm^2s)
Thermal neutron flux (average with targets)	1.8×10^{15} neutrons/(cm^2s)

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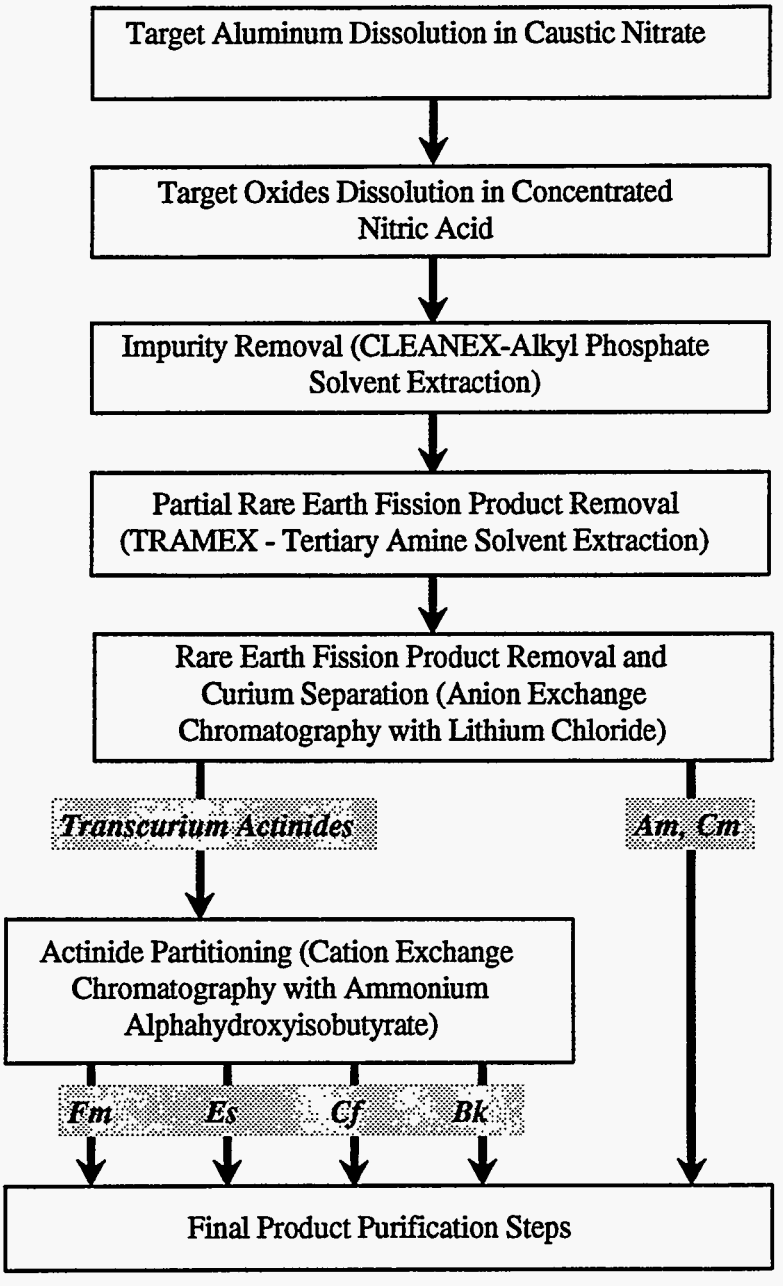

Fig. 1.4 Cell processing steps at the Radiochemical Engineering Development Center (based on figure p. 58, ORNL Review, No. 3, 1985).

Table 1.3 Production of elements 96–100 at the High Flux Isotope Reactor and the Radiochemical Engineering Development Center.

Isotope	Half-life	Principal decay mode	Nominal production (amount/year)
Curium-248	3.397×10^5 year	alpha	150 mg
Berkelium-249	320 d	beta	50 mg
Californium-249	350.6 year	alpha	From ^{249}Bk decay
Californium-252	2.645 year	alpha	500 mg
Einsteinium-253	20.4 d	alpha	2 mg
Einsteinium-254	275.7 d	alpha	4 μg
Fermium-257	100.5 d	alpha	1–2 pg

reported estimates of the distribution of ^{252}Cf use in several major application categories, both by the number of sources used and by ^{252}Cf mass. As shown in Fig. 1.5, within a few years after this unique isotope became available to commercial source fabricators, several major uses of ^{252}Cf had been established, including, in order of the amount of ^{252}Cf used, (1) reactor startup, (2) fuel rod scanning, (3) activation analysis, (4) education and research, (5) medical research, and (6) calibration and dosimetry.

Since 1982, the use of ^{252}Cf has spread greatly (see Table 1.4). Over a decade later the distribution of ^{252}Cf usage has changed significantly,[8] as shown in Fig. 1.5. Reactor startup and activation analysis applications are still very important areas, while new uses have appeared, (e.g., in aircraft inspection and luggage scanning) and others have grown (e.g., medical treatment and research, calibration and dosimetry). More recently, on-line coal-analysis instruments have appeared that are designed to use and to be completely dependent upon ^{252}Cf. Previously established use areas have broadened. Medical uses of ^{252}Cf, for example, have expanded beyond treatment of cervical cancer to include research in (a) the treatment of head and neck cancers, malignant gliomas, skin cancer, and other cancers and (b) in studies of the effects on AIDS. Other use areas have grown more dependent on ^{252}Cf. Reactor startup, for example, is an application area that has come to use ^{252}Cf extensively.

Clearly, ^{252}Cf has become a material of worldwide importance. The availability of this isotope greatly impacts the lives of many people and the well-being of many industries.

Table 1.4 Usage of ^{252}Cf sources after one and two decades of commercial availability

Use Area	1982		1994		Comment
	% of sources	% of ^{252}Cf	% of sources	% of ^{252}Cf	
Reactor startup	55.3	48.3	14	6.9	Large financial impact
Fuel rod scanning	8.2	25.3	5	12.1	Important to fuel fabrication
Activation analysis	18.9	19.4	20	0.69	Wide applications in many fields
Education and research	5	2.4	14	0.41	Enables education and research not otherwise possible
Medical research and treatment	3.1	0.7	2	0.62	Saves lives
Calibration and dosimetry	3.1	0.1	39	0.14	Versatile and important tool
Neutron Radiography (NR)			2	77.4	Versatile and important tool
Bulk materials analysis			3	1.7	Commercial application dependent on ^{252}Cf

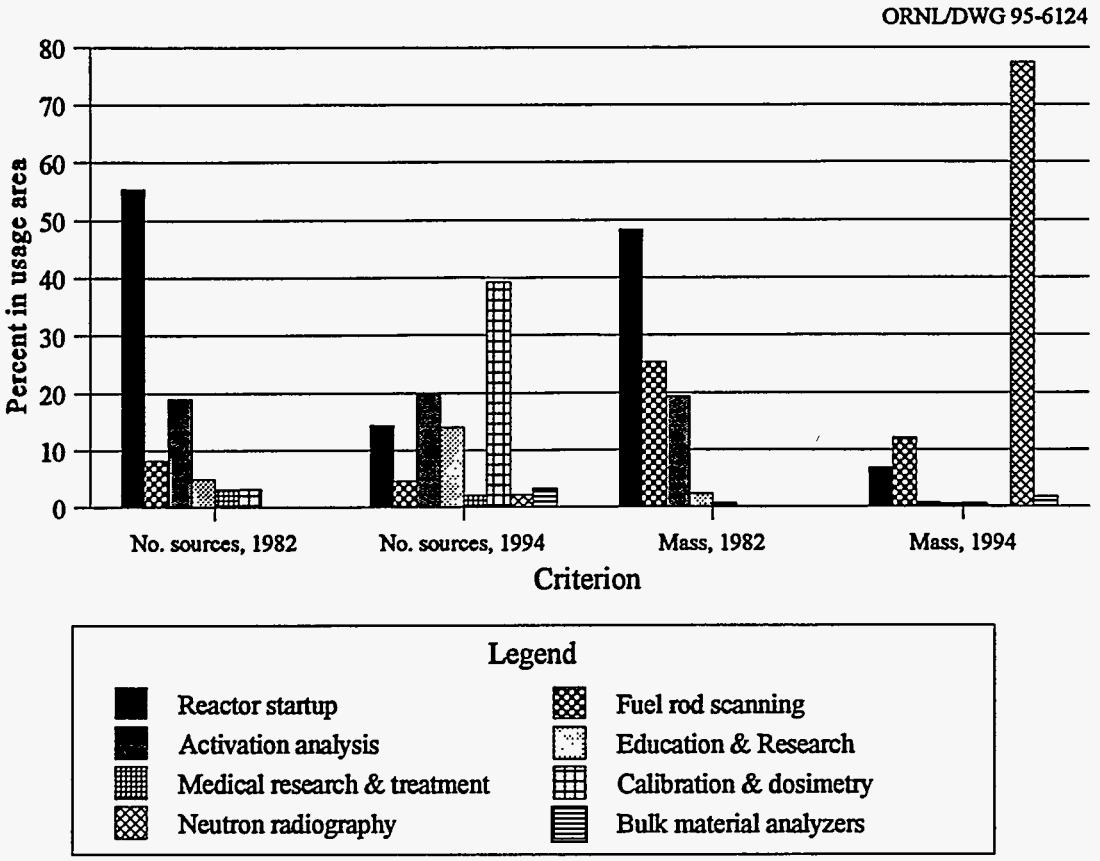

Fig. 1.5 Distribution of ^{252}Cf use by application area, based on number of sources and mass of ^{252}Cf used, after one and two decades of commercial availability (from data in Table 1.4).

2. MAJOR AREAS OF USE AND USERS

The many applications of ^{252}Cf are summarized in Fig. 1.2. The Californium use tree depicts the large number and broad range of uses to which this vital isotope is being put and aptly represents the fact that these important advances were made possible through the sponsorship of the DOE. Many of these applications save lives, time, and money. Some could not be accomplished without ^{252}Cf. There are some areas of use which are still growing. For many of these applications, ^{252}Cf is irreplaceable. Some of these applications are described in this section.

2.1 REACTOR STARTUP WITH ^{252}Cf SOURCES

Reactor startup is one of the most critical applications of ^{252}Cf use. When a nuclear reactor is started up, a stable source of background neutrons is needed to ensure adequate instrumental response to provide a properly controlled approach to criticality. A similar situation exists during fuel reloading, particularly if the reactor has been down for an extended period of time (e.g., for maintenance or modification).

Californium-252 was first used for the startup of a commercial reactor in 1973.[9] Since then, ^{252}Cf has become the industry standard for pressurized water reactors (PWRs), with virtually all PWRs in the United States and many abroad now using ^{252}Cf startup sources. This growth in ^{252}Cf is due to the greatly superior properties of ^{252}Cf sources as compared to alternative sources (Table 2.1). At one time, about half of the ^{252}Cf sold and about half of the capsules fabricated commercially went into this one application. These sources are part of the fuel assemblies and remain in place after the reactor has started up; thus, the material is consumed and cannot be returned.

Po:Be sources were used for early reactor startup, but the short half-life of polonium was a drawback. ^{238}Pu:Be sources were used later. The half-lives of these sources were better, but gas generation and heating were problems. The size of the source required was also a disadvantage. Today, the nature of plutonium waste—transuranic, strongly alpha emitting, highly radioactive waste, and with no currently acceptable disposal alternative—is a strong deterrent to the use of ^{238}Pu:Be. Since the acceptance of ^{252}Cf for PWR startup sources, the production of ^{238}Pu:Be sources gradually ceased. Currently, no United States manufacturer is licensed to produce ^{238}Pu:Be sources.

The ^{124}Sb:Be sources are fabricated using natural antimony (42.7% ^{123}Sb), which is activated by exposure to neutrons in an operating reactor before being used for startup of a new reactor. These sources have been manufactured by General Electric Company (GE) for many years.[10] Because of the relatively short half-life of ^{124}Sb, the shelf life of an activated ^{124}Sb:Be source is limited to about 6 months. Also, the Sb:Be must be activated initially to be useful as a startup source, which means that shipment to an operating reactor for irradiation is necessary. Shipments of these sources must be made using Type B containers, which are much less readily available. Both the short half-life and stringent packaging requirements have made the use of ^{124}Sb:Be sources impractical for new reactor startup, especially since ^{252}Cf sources can be shipped in Type A packages and are already "activated."

^{124}Sb:Be sources are continuously reactivated once in place in an operating reactor. Hence, they are useful for reloading or restarting the reactor for a decade or longer. On the other hand, ^{252}Cf sources are burned out in about 3 years. For these reasons, a hybrid source was developed, which contains ^{252}Cf for the initial startup and ^{124}Sb:Be for restarts. Hybrid ^{252}Cf/^{124}Sb:Be sources are now widely used for restarts after brief shutdown periods. However, even with hybrid sources, ^{252}Cf is required for restarts after extended shutdowns (e.g., six months or longer).

Table 2.1 Some properties that make ^{252}Cf sources superior for reactor startup applications[a]

Property, units	^{252}Cf	^{238}Pu:Be[b]	^{124}Sb:Be[c]
Volume, cm^3	<1	50	100
Heat, W	0.03	300	60,000
Temperature limit, °F	1,500	850	2,000
Swelling problem	No	Yes	Yes
Radioactivity, Ci	<1	400	2,500
Gas Generation (cumulative atoms):			
After 10 years	1×10^{18}	1×10^{22}	1×10^{24}
After 20 years	1×10^{18}	2×10^{22}	2×10^{24}
Half-life, y)	2.645	89	0.16 (60 days)
Shipping cask requirement	Type A	Type B	Type B
Waste disposal concerns	Typical	High	Typical

[a] Extensive data are for a source of 2×10^9 neutrons/s, suitable for large reactors of about 1,000 MW(e).

[b] These are no longer made.

[c] GE still makes these for their BWRs.

Table 2.2 lists manufacturers of nuclear power reactors for reactors located in the United States and throughout the world. All of the manufacturers of United States reactors design for reactor startup with ^{252}Cf sources. Californium startup source manufacturers are listed in Table 2.3. A GE facility, located in Vallecitos, California, supplies most of the commercial nuclear power reactors in the United States and all of PWRs and boiling-water reactors (BWRs) in Japan.[10] Reactors in Europe are supplied with ^{252}Cf-based startup sources of United States origin by Amersham International, which is located in the United Kingdom, although Frontier Technology supplied three startup sources for the German "Konvoi" reactors.

Californium sources used in reactor startup applications use about 3 mg for BWRs, consisting of about 6 sources at about 0.5 mg/source. PWR startups require about 0.5 mg, using sources in the 150–250 μg range. In the United States, there are 116 commercial nuclear power plants (38 BWR + 78 PWR) existing or under construction; of these, 108 are operational.[11] Worldwide, there are 504 nuclear power reactors existing or under construction; of these, 412 are in operation. Foreseeable reactor startups, including both new starts and restarts, number 8 in the United States and nearly 100 worldwide. These startups will require ^{252}Cf sources amounting to on the order of 6.5 mg for BWRs and PWRs in the United States and a total of 62 mg for BWRs and PWRs worldwide.

Sources for reactor startup require extensive design and analysis to ensure proper and safe functioning within the reactor. Similar to fuel rod design, there are many areas of concern in startup source design, including:

- heat generation and removal,
- gas generation and internal pressure buildup,
- fuel swelling,
- materials stress and compatibility, and
- radiation effects.

Further, reactor designs are dependent on the nature of the startup source. Reactors designed to take full advantage of the compactness of ^{252}Cf startup sources would require significant modification to make use of an alternate source. In addition, there would be licensing complications associated with changing to alternate sources.

2.2 NEUTRON ACTIVATION ANALYSIS (NAA) APPLICATIONS WITH ^{252}Cf SOURCES

One way in which neutrons may interact with matter is by rendering it slightly radioactive. When a material is exposed to a neutron flux, neutron capture can occur. When this happens, atoms that may have been previously stable (nonradioactive) are transmuted into heavier isotopes, which are frequently emitters of beta and gamma radiation. The spectrum of energies emitted by each isotope gives a unique "signature" for that isotope. The energies emitted by an activated material can be detected and the energy spectrum then sorted out by sensitive radiation detection and analytical devices. The process of irradiating a material with neutrons to produce transmuted isotopes followed by detection and analysis of the gamma spectrum to determine the elements present in the material is known as neutron activation analysis, or NAA.

NAA is very sensitive, making it useful in measuring trace quantities of many elements. Most elements are detectable at concentrations in the ppm range with suitable equipment. The technique can be highly automated, making it possible to analyze a large number of samples for many elements on a round-the-clock basis. Because of the compact, rugged, high flux, and reliable nature of ^{252}Cf sources and their relatively low cost, they are unexcelled for use in nonreactor NAA applications. Hence, sources used in nonreactor NAA are typically ^{252}Cf

Table 2.2 Manufacturers of nuclear power reactors in the United States and the rest of the world.

Abbreviation (if any)	Reactor manufacturer	Country
	Builders of reactors located in the United States	
W	Westinghouse Electric Co.	USA
B&W	Babcock & Wilcox Co.	USA
C-E	Combustion Engineering, Inc	USA
GE	General Electric Co.	USA
	Builders of reactors located outside the United States	
AEG	Allegemeine Elektricitaets-Gesellschaft, AEG Telefunken	Germany
AA	ASEA-Atom	Sweden
AEE	Atomenergoexport	USSR
AECL	Atomic Energy of Canada Ltd.	Canada
APC	Atomic Power Construction, Ltd.	UK
B&W	Babcock & Wilcox Co.	Sweden
BBR	Babcock-Brown Boveri Reaktor Gmbh	Germany
CNNC	China National Nuclear Corporation	China
	Framatome	France
GETSCO	General Electric Technical Services Co.	USA
GE Can	GE Canada	Canada
	Hitachi	Japan
KWU	Kraftwerk Union AG	Germany
L&T	Larson & Toubro	India
MTM	Mintyazhmash	Russia
MHI	Mitsubishi Heavy Industries	Japan
NNC	National Nuclear Corporation	UK
FECNE	Nuclear Power Plant Equipment Factory	Romania
	Skoda	Slovakia
TNPG	The Nuclear Power Group	UK

Table 2.3 Manufacturers of ^{252}Cf nuclear power reactors startup sources

Reactor startup source manufacturer	Location
Amersham International	Buckinghamshire, UK
CEN	France
Frontier Technology	Xenia, Ohio
General Electric Co.	Vallecitos, California

sources, with outputs ranging from 10^9–10^{10} neutrons per second. For many applications ^{252}Cf sources are the only practical source of neutrons.

Prompt gamma neutron activation analysis (PGNAA) comprises those NAA techniques that analyze γ-rays emitted essentially instantaneously following neutron capture. The γ-rays emitted during the first 10^{-12}s of activation are classed as *prompt*. Some activated elements emit *delayed*, or decay, γ-rays, in addition to the prompt γ-rays, for a long time following neutron capture. Analysis of the decay γ-rays is NAA, as described earlier, which is best performed after neutron irradiation has stopped and at a location away from the neutron source. Detection of the prompt, induced radiation is performed simultaneously with the neutron irradiation.

PGNAA has been used for more than two decades for chemical analysis in process streams and laboratories. The use of neutron capture prompt γ-rays in multielement sample analysis has greatly increased during the past few years. Extensive experimental work has been performed in bulk-coal analysis, mineral exploration, and in several other applications. In all of these applications, the detected γ-ray intensities from the elements present are used to estimate their concentrations. The γ-ray intensities from a specific element are a function of the concentrations of all the elements present and of several other sample matrix parameters such as bulk density, porosity, moisture content, and temperature.

NAA, including PGNAA, is a very active area for ^{252}Cf use and consists of a broad range of applications. There is extensive literature on NAA.[9, 12, 13, 14] Some of the more established uses of ^{252}Cf are:

- multielemental analysis for such purposes as clinical research, forensic work, dating of metallic objects and the measurement of the wear of materials,[15]
- fuel rod scanning,
- uranium borehole logging,
- oil exploration, and
- gold exploration

Recent and emerging applications of NAA include methods for:

- on-line analysis of ores, coal, and sludges,
- field-portable analysis,
- cold-neutron analysis,
- prompt γ-ray analysis,
- NAA-based determination of sodium content in food, and

- luggage analyzers to detect the presence of explosives or pharmaceuticals

At the recent meeting of the American Nuclear Society (ANS), in San Diego, as in previous conferences on ^{252}Cf (see Sect. 1.2), there were numerous reports of work that makes use of portable ^{252}Cf sources for NAA. This contemporary work, which capitalizes on a range of the advantageous properties of ^{252}Cf, is described briefly in Sect 2.2.1. The remainder of this subsection describes more specific applications of NAA using ^{252}Cf that are beneficially impacting our society.

2.2.1 Multielemental Analysis with ^{252}Cf Sources

One of the first applications of NAA was in the study of moon rocks for the National Aeronautics and Space Administration (NASA), which was conducted at the University of Kentucky. The SRTC, located at the SRL, has an NAA facility that makes use of ^{252}Cf neutron sources.[16] The SRTC has used a ^{252}Cf NAA facility since 1976, and it has since developed methods for a more extensive NAA system. The ^{252}Cf facility at SRTC provides multielement analysis of solid and liquid samples for applications that include alloys, sediments, rocks, inks, and site process solutions. The current NAA facility uses a ^{252}Cf mass of about 100 mg to achieve a peak thermal neutron flux of about 2×10^9 n/(cm^{2}s). The system is particularly advantageous for analysis of bulk samples without chemical dissolution. For example, materials that may come in contact with stainless steels are frequently tested to ensure that chloride concentrations are below values of concern.

Two 20-mg sources are used to perform NAA on environmental samples at the Oak Ridge Associated Universities (ORAU) facility in Oak Ridge, Tennessee. Gleason applied NAA to the determination of contaminant concentrations in soils, sediments, slags, and sludges required in an extensive environmental survey.[17] Elements such as uranium, thorium, chromium, mercury, silver, and arsenic were determined at the parts-per-million (ppm) level at the ORAU facility. The use of ^{252}Cf for environmental sample analysis seems destined to grow in importance.

2.2.2 Fuel Rod Scanning

Nuclear reactor fuel rod scanners use a ^{252}Cf source plus γ-ray detection to determine the level of enrichment as a function of position along the fuel rod. Neutrons from ^{252}Cf induce fission in the ^{235}U producing neutrons, delayed neutrons, and prompt fission gamma radiation. Analysis of the delayed gamma radiation is most often used in fuel rod scanning. In addition to giving a measure of the total fissile material content, this analysis reveals the degree of uniformity of fissile material loading over the length of fuel rods. Accurate knowledge of the amount of fissile material loading combined with accurate measurement of the degree of uniformity of loading permits optimal performance for nuclear power generation.

The use of neutron sources produces stronger gamma spectra from the fuel rod being scanned, thus, providing for much improved quality control. This type of scanning, called *active scanning*, is used by every commercial nuclear facility. Nearly all of these facilities[18] are ^{252}Cf-based. For example, GE has been doing active scanning since early 1970s. Nuclear facilities managers demanded scanning and used it as an integral part of their operations to ensure:

- fuel rod quality (e.g., integrity and freedom from defects),
- axial distribution of fuel (e.g., ^{235}U enrichment),
- accountability of nuclear material, and
- optimal billing of customer by quality of product.

Fuel rods have become more diverse, with zoned rods having as many as five zones with five different levels of enrichments. Better knowledge of the fuel allows tighter engineering margins, which affect design and reduces fuel costs. In other words, fuel rod scanning lowers the costs of nuclear power.

The GE facility processes about 3,000 assemblies per year; mostly 9×9 arrays of 72 rods per assembly; thus, hundreds of thousands of rods per year are processed at GE alone. Of these, the vast majority, about 90%, are low-enriched uranium fuel rods, which are processed using active scanning. The remaining 10% are gadolinium (Gd) rods. Because Gd is a neutron poison, such rods cannot be processed by active scanning; consequently, a passive scanning system is used as described below.

Californium-252 sources are typically 3 mg/source and are useful for fuel rod scanning for about two half-lives, or until about 1 mg of ^{252}Cf remains. Beyond that point, the high signal-to-noise ratio is lost along with the exceptional performance (0.5% precision for a zone of enrichment) that is characteristic of active scanners.

Finding an alternative to ^{252}Cf would be difficult and would pose a serious problem. Existing fuel rod scanning equipment would be obsolete. It is not likely that scanning could be given up because customers now demand it.

MAPS, the magnetic and passive scanning device (a magnetometer, super-conducting device) used by GE for its Gd-rod scanning, could provide the function but not the performance of active scanners. Because there is less radiation produced to scan for (about 1/20th), about 6 times as many detectors would be needed per station. In addition, there would be a loss in both throughput and precision. The scan rate would drop by about an order of magnitude, while precision would decrease by a factor of 1.5–2.[19] The reduction in throughput would be costly, requiring perhaps 10 times as many stations as before, at a cost of about $1 million/station. The loss in precision, which could not be recovered, would result in decreased fuel use efficiencies throughout the nuclear power industry.

National Nuclear Corporation of Sunnyvale, California, builds passive and active scanners.[19] So far, there are 10 active scanners in the fields; these have been in business fewer than 2 years. A scanner typically costs about $1 million. Currently, the active scanner industry is wholly dependent upon the availability of ^{252}Cf sources.

2.2.3 Field-Portable NAA Devices

Idaho National Engineering Laboratory (INEL) developed the portable isotopic neutron spectrometry (PINS) of gamma rays for in-field inspection and verification of chemical weapon inventories.[20] PINS provides a system that can be carried into an area inaccessible by wheeled transport because of rough terrain or confined spaces, and it is capable of battery-powered operation. The portable system consists of a ^{252}Cf source of about 5μg; a germanium detector with cryostat mounted on a satellite dewar providing a 24-h supply of liquid nitrogen; a portable, battery-powered, multichannel analyzer and amplifier; and a notebook-sized personal computer interfaced to the multichannel analyzer. The system can perform spectral peak analysis, elemental identification and, under certain conditions, chemical identification. The system has been successfully used to assay military munitions, and it is being considered for the in-field assay of low-level radioactive waste. The potential savings that could result from the availability of rapid, in-field, bulk chemical assays may be expected to increase the demand for ^{252}Cf sources.

2.2.4 Enhanced Analytical Methods Using Cold Neutrons from ^{252}Cf Sources

Currently, there are limited facilities in the United States and worldwide where research involving cold neutrons is performed. Sources for existing cold neutron facilities are either at nuclear research reactors or spallation accelerators. These facilities are in high demand.[21] A nonreactor-, nonaccelerator-based cold neutron source is of interest because of the difficulties associated with reactor- and accelerator-based neutron sources. Because of the high radiation fields generated by reactors and accelerators, neutrons often must travel distances up to 50 m beyond the cold source before they are isolated enough from gamma radiation to be useful.

In 1989, Robinson and co-workers,[22] at ORNL, proposed a ^{252}Cf cold neutron source. The Californium Cold Neutron Source (CCNS), which was proposed by the ORNL group, would make use of several hundred milligrams of ^{252}Cf. It is thought that the CCNS would increase the opportunities for scientists to conduct research in areas such as neutron tomography, localized irradiations in cancer treatment, neutron optics (neutron focusing), and low-temperature moderator development.

Cornell University is undertaking optimization studies of a novel design that exploits the combination of the superior analytical properties of cold neutrons with the simplicity and portability of isotopic neutron sources such as ^{252}Cf.[23] The device, termed a *cold-neutron irradiator*, is suitable for both NAA and PGNAA. The advantage of the cold irradiator, which uses cold neutrons corresponding to the temperature of liquid helium rather than to room temperature, is that the neutron reaction cross sections (proportional to $1/v$, where v is the neutron velocity) are greater by more than a factor of 8. Because of the greatly enhanced reaction rates possible, the cold neutron irradiator is an exciting prospect. The cold neutron irradiator would use either a ^{252}Cf or an Am-Be source.

2.2.5 NAA Facility for the Food and Drug Administration (FDA) Food Examination

The staff of the Nuclear Science Facility at San Jose State University (SJSU) is developing a ^{252}Cf-based neutron activation analysis program that will permit analysis for trace concentrations of environmental, nutritional, and high-purity manufacturing interest.[24] The program will enhance the quality of the nuclear science summer school at SJSU (which is sponsored by DOE) and will also support other federal, state, and commercial programmatic activities. The FDA, in particular, has an interest in the program.[25] The FDA intends to develop a program to measure the level of sodium content in processed foods to ensure that food packages are accurately labeled. The FDA has an agreement with SJSU to provide funds to support the development of the SJSU NAA program for this purpose. The FDA has also contracted with ORNL to have the necessary ^{252}Cf sources fabricated and shipped.

The State of California is also interested in measuring trace concentrations of elements as part of its environmental monitoring and restoration programs.[25] Located at the headwaters of the "Silicon Valley," in California, San Jose State University is near many companies that manufacture computer chips, esoteric alloys, and other materials that require absolutely pure feed stock. Such companies may be interested in having an inexpensive, quick turnaround trace-contamination-measurement program available.

The NAA program at San Jose State University is based upon a maximum of four 50-mg ^{252}Cf sources, which, altogether, will provide a source strength of over 10^{11} neutron/s. This source strength would permit analysis of samples of sodium, magnesium, aluminum, sulfur, chlorine, potassium, calcium, and other elements of importance. The current program at San Jose State University, which is based upon a neutron "howitzer" with a 10-Ci ^{238}Pu:Be source, is limited by the low rate of emission of neutrons (10^6 neutron/s).

The NAA program that is being planned for San Jose State University has several advantages over one located at a reactor. Many reactors have extensive, unscheduled downtime; a regulatory compliance program cannot be effectively managed if measurement services are not routinely available. Also, many food samples may not be compatible with the heat associated with the environment of a reactor. Further, changes in spectrum and flux caused by nearby experiments could adversely affect the quality of NAA measurements. Finally, development of the license application for a dedicated reactor under the U.S. Nuclear Regulatory Commission's (NRCs) user fee program could be prohibitively expensive—and there is no assurance of success—whereas amendment of an existing broad-scope university radioactive materials license is comparatively straightforward.

2.2.6 In Vivo Neutron Activation Analysis

The technique of in vivo neutron activation analysis has been widely used in body composition studies[26, 27, 28, 29] with a variety of neutron sources and spectra. Various laboratories have tried to obtain the best compromise between achieving uniformity of activation and minimizing the body dose per incident neutron, considering also cost and ease of operation. Previously, ^{238}Pu:Be sources were favored, and systems for total-body and partial-body analysis have been successful using these sources as well as neutron generators. Californium, however, has emerged as an alternative for partial body in vivo NAA with advantages over the other alternatives—namely:

- A ^{252}Cf neutron has approximately 40% greater peak thermal fluence generated within the body (human or animal) per unit dose delivered relative to ^{238}Pu:Be. This benefits element detection limits,
- The lower-energy neutrons of ^{252}Cf are less damaging to the Ge semiconductor detectors,
- The smaller physical size of ^{252}Cf sources permits more effective collimation of the neutron beam,
- Transporting of ^{252}Cf sources is easier than for alternative isotopic sources.

2.2.7 On-Line Analysis Systems

The most widely applied on-line elemental analysis technique is PGNAA, which uses ^{252}Cf as a source of neutrons. The prompt γ-rays are most useful for on-line bulk stream analysis of materials such as coal. Important constituents of coal (e.g., for instance, such as hydrogen and carbon) would be missed without PGNAA because these elements don't produce delayed γ-rays.

On-stream bulk material analysis devices use neutrons from ^{252}Cf sources along with prompt gamma analysis to continuously measure elemental concentrations at industrially useful rates. Analyzers are used to measure sulfur and ash content and other elemental components in bulk coal or cement at rates of up to 1,000 ton/h. By comparison with the other techniques used for on-line bulk material analysis, PGNAA devices using ^{252}Cf provide much more information and at a greater rate of throughput. Although it is the most costly of the major on-line techniques, the ^{252}Cf-based elemental analyzers are the preferred and most widely used technique in several industries, including the mining and electric power industries. Furthermore, the use of ^{252}Cf-based elemental analyzers is growing rapidly in the cement industry.[16, 30]

Commercial analyzers were developed in the late 1970s by the Science Applications International Company (SAIC) using funding from the Electric Power Research Institute (EPRI), and MDH-Motherwell, which in turn was funded by the Pennsylvania Electric Company and the New York State Electric and Gas Corporation. The first analyzers were CONAC (Continuous ONline Analyzer of Coal), developed by Scientific Applications, and ELAN (ELemental ANalyzer), developed by MDH-Motherwell. The first application of a PGNAA-based bulk material analyzer to cement was by Texas Nuclear in 1984. Currently, there are three manufacturers of commercial PGNAA machines: GAMMA-METRICS, Mineral Control Instrumentation, Ltd. (MCI), and Energy Technologies, Inc. (ETI), formerly a division of SAIC. A discussion of these three analyzers follows.

GAMMA-METRICS Coal Analyzer, Model 3612. This analyzer was introduced by GAMMA-METRICS (San Diego, California) in 1984 with the capacity to handle up to 500 ton/h. A smaller, 100-ton/h-capacity version (MODEL 1812C) was introduced in 1986. The 3612L handles up to 1,000 ton/h. In 1991, a total of 30 GAMMA-METRICS coal analyzer units were in operation at coal installations.[15] As of 1993, GAMMA-METRICS had over 100 ^{252}Cf-based analyzers in the field including coal, cement, and laboratory-sample analyzers.[31] GAMMA-METRICS also has an automated blending program called COBOS (COal Blending Optimization System), which estimates the proportions of different sources of coal needed to achieve a desired blend quality using the analysis of the blended coal.

MCI COALSCAN 9000: Developed by MCI in Unley, South Australia, this analyzer uses the dual energy γ-ray transmission technique in addition to the standard PGNAA arrangement to provide a separate and an independent ash gage. It also includes an integral microwave moisture meter. As of 1991, there were six installations of the COALSCAN 9000 analyzer worldwide.

ETI Model 200 Sulfur Meter: This analyzer, originally developed by SAIC at its Oak Ridge, Tennessee facilities, is now available from ETI, in Knoxville, Tennessee. ETI's sulfur meter provides direct analyses of sulfur, ash, and moisture. This analyzer determines sulfur by PGNAA, ash by γ-ray transmission, and moisture by microwave meter. The Model 200 was newly released in 1991.

There were 44 units installed or on order worldwide by 1991. Now there are over a 100. Currently, GAMMA-METRICS dominates the market, supplying about 80% of the units in use.[31] On-line elemental analyzers are being installed at an increasing number of facilities worldwide[16] for coal-quality monitoring and to provide information that is needed to improve key process operations. Current applications include:

- *run-of-mine/mine grade monitoring*—On-line analysis is used to alert mining operators to contamination in the output (e.g., high ash material in coal mining) so that operations can be adjusted,

- *sorting*—On-line analysis is used to identify high-ash and low-ash sections of a crushed coal stream, allowing segregation and the production of stockpiles sorted by quality,

- *blending*—On-line analysis is used to monitor and control the blending of two or more bulk streams to achieve a certain product specification by varying the blending ratio continuously to compensate for fluctuations in the qualities of the blend components,

- *preparation-plant monitoring/control*—The operator of a preparation plant is provided with instantaneous information allowing the achievement of a product with a specified composition in this, the largest application for on-line analysis,

- *consignment monitoring/control*—On-line analysis is used to monitor and control the quality of the bulk product prior to transport to customers, including shipments to steel plants, at train loading and shipping stations, and mine production,

- *power-station-feed monitoring*—On-line analysis is used to monitor the coal feed entering the plant with this "feed-forward" information being used manually or in a control system to optimize plant operation, including emissions control equipment.

In the coal industry, PGNAA with ^{252}Cf sources has been universally adopted as the system which can provide a nearly complete elemental analysis of coal, together with its bulk ash content and heating value, with good

correlation accuracies (see Table 2.4). The basic price of analyzers of this type is typically less than $500,000, with the total cost depending on the particular additional equipment needs of the customer, such as moisture meters and sampling systems. Nevertheless, customers have reported such improvements in plant performance that short payback times were achieved.[15]

2.2.8 Explosives Detection with ^{252}Cf

Scientists have long searched for ways to identify concealed weapons that can be used in terrorist attacks, due to the continued threat that terrorism poses. Explosives, which can be shaped to look innocuous, are hard to detect. Explosives are also a concern for treaty verification purposes, due to the hazards of direct sampling of chemical and explosive munitions. A nondestructive evaluation method is needed in each of these instances.

Currently, PGNAA is under development for explosives and illicit contraband detection, both as a stand-alone application[32] and in combination with two other methods: X-ray imaging and vapor detection. Vourvopolos recently reported[33] on the use of x-ray imaging, combining high and low energy x-ray machines; vapor detection using a "sniffer" to collect vapor samples with analysis of the vapor by gas chromatography, chemiluminescence and mass spectroscopy; and PGNAA. PGNAA shows promise in this application area, since determinations must be carried out at a speed to minimize disruption and delay at airport and other security stations.

The PGNAA method uses neutrons from a ^{252}Cf source to induce capture and inelastic reactions in munitions or contraband, and the resulting γ-rays are measured with a high-purity germanium detector. Caffrey and coworkers[34] have developed a portable, battery-operated assay system for field verification that performs automatic spectrum analysis and agent identification in near-real time. The field portable unit has reliably identified nerve agents, blister agents, white phosphorous, and high explosives.

Though yet developing, explosives and contraband detection using PGNAA is clearly an application of ^{252}Cf with far-reaching potential. Successful deployment of detection devices which have already been successfully tested can help to prevent the loss of lives and property around the world.

2.3 NEUTRON RADIOGRAPHY (NR) WITH ^{252}Cf SOURCES

Neutron Radiography (NR) is a technique used for detecting flaws or voids in metals, especially in castings, forgings, and weldments. Similar in principle to x-ray or gamma radiography, NR differs in that it is primarily useful for detecting and imaging low-atomic-weight materials, such as plastics and explosives. Sources suitable for NR must be physically small in size and have outputs on the order of 10^9 neutron/s. For this reason, ^{252}Cf is the preferred isotope for most non-reactor applications. Neutron radiography has uses ranging from small, delicate materials, such as flowers, to very massive materials. These varied applications demonstrate the usefulness of NR.

2.3.1 Nondestructive Aircraft Examination

For applications requiring a very intense and highly concentrated neutron source that is also compact, there is no substitute for ^{252}Cf. The nondestructive inspection facility at McClellan Air Force Base near Sacramento, California, is one important example. This facility, equipped with two bays (one for x-ray and one for n-ray), provides for examination of complete aircraft. The n-ray examination at the McClellan facility reduces the probability that aircraft will be lost due to in-flight structural failure because of corroded or debonded honeycomb in the wings. Possible sources for the corrosion are fuel leakage or water intrusion. Since being installed, the

Table 2.4 Comparison of on-line analysis techniques for bulk coal

Technique	PGNAA	X-ray/γ-ray backscatter	Dual energy γ-ray transmission	Natural gamma	Microwave	Compacitance
Excitation source	^{252}Cf	^{238}Pu, ^{241}Am	^{241}Am, ^{137}Cs, ^{133}Ba	Naturally occuring isotopes	Microwave transmitter	Charged plates
Measured quantities	S, C, H, N, Fe, Al, Si, Ca, Cl	Mean atomic number	Mean atomic number	Quantity of radioactive elements	Moisture	Moisture
Instrument displays	Ash, sulphur, other elements, heat value	Ash	Ash	Ash	Moisture	Moisture
Typical correlation accuracies to within wt %	0.5 ash 0.1 sulphur 400 kJ/kg (or) 150 Btu/lb)	1–2	0.3–1.5	0.4–2	0.5	0.5
Approximate measurement time	1 min	10 min	2 min	2 min	<1 s	2 min
Sample size	5–500 tons/hour (Could be full flow)	laboratory/ conveyor belt	conveyor belt	conveyor belt	conveyor belt	conveyor belt
Approximate cost, $K	<$500	$80–120	$100–130	$60–80	$50–70	$30-50

A. T. Kirchner, "On-Line Analysis of Coal," ISBN 92-9029-193-1, IAEA Coal Research: London, United Kingdom, 1991.

existing n-ray examination facility has reduced the time required for routine maintenance inspections, which in turn reduces the number of planes out of service, reducing the need for and cost of larger fleets.

The primary applications of neutron radiography in aircraft maintenance[35, 36, 37, 38] are the detection of corrosion and entrapped moisture (and liquids such as water, fuel) in aircraft structures (see Table 2.5). Surface, exfoliation and galvanic lap corrosion in an early stage are readily detectable by NR, as well as intergranular, stress and pitting corrosion in advanced stages. Conventional non-destructive methods detect such corrosion only after substantial structural damage has been done. Also, NR detects moisture and liquids that lead to corrosion of metal honeycomb structures and to debonding in carbon-fiber composite structures in layers with a thickness of only a few tenths of a millimeter—far better than the only competing nondestructive testing method, X-ray radiography, which identifies liquid layers of not less than several millimeters in thickness.

NR is also being considered for space shuttle inspection, because it can show entrapped moisture and naturally occurring aluminum corrosion, even in small amounts. A facility dedicated to NASA operations has been conceptualized.[39] A trial has been made at the McClellan Air Force Base NR facility, and further trials using a mobile NR system have been recommended.

2.3.2 Nondestructive Examination of Weapons Components

Nondestructive examination of weapons components has been an important use of ^{252}Cf in U.S. defense programs for over a decade. NR was used at the EG&G Mound Applied Technologies, Inc., facility to examine weapons and explosives components for more than ten years.[40] ^{252}Cf was chosen over accelerators because it was a more reliable and less costly alternative. Other isotopic sources, such as Pu:Be, require excessive exposure times with unacceptably poor resolution. Reactor-based examination was not acceptable due to risk and classification concerns. Although the Mound facility no longer uses NR, ^{252}Cf-based NR is actively used for research, development, and production applications at other defense facilities.

2.3.2.1 Pantex Neutron Facility

Examination of weapons components, including high explosives, is an important application area for NR. An example of this type of application is the neutron facility planned for use at the Pantex Plant, in Amarillo, Texas.[41] This facility, which is now under construction,[42] has been designed to provide a reliable capability for checking components for defects in production mode. Most of the components are small (less than one pound), hand-held parts containing metal-encased high explosives, although larger parts (up to twenty pounds) will be handled as needed. In the past, a 3 MeV Van de Graaff machine has been used at Pantex as a neutron source, but the upgrade is needed due to frequent maintenance problems. The Pantex Neutron Facility is designed around 150 mg of ^{252}Cf. ^{252}Cf was selected at Pantex as a reliable neutron system with reduced cost of operation and maintenance, as compared with a Van de Graaff- or reactor-based system.[43]

2.3.2.2 U.S. Army Research, Development, and Engineering Center

Neutron radiography is also used by the U.S. Army at the Research, Development, and Engineering Center at Picatinny Arsenal, New Jersey.[44] The facility at Picatinny Arsenal uses ^{252}Cf sources to perform NR examinations of castings, turbine blades, and metal parts with explosive interiors. ^{252}Cf sources were selected as simple, very dependable neutron sources of well-known strength that yield immediate information on sensitive materials. Many of the materials examined are classified or restricted materials, so offsite transfer for examination at a reactor-based facility not only increases cost and schedule delays, but also adds undesirable security risk. The research center at Picatinny Arsenal also uses ^{252}Cf in neutron scattering experiments to examine resins in

Table 2.5 Application of NR to aircraft maintenance inspection

Problem to detect	Degree of applicability
General metal structure:	
Cracks	Confirmed application
Welds	Confirmed application
Bolt holes	Confirmed application
Entrapped moisture	Potential application
Honeycomb damage	Confirmed application
Engine components	Confirmed application
Corrosion:	
Initial onset of corrosion	Individual cases
Surface corrosion	Confirmed application
Galvanic lap corrosion	Confirmed application
Intergranular corrosion	Potential application
Stress corrosion	Potential application
Formation of carbon fiber composite structures:	
Entrapped moisture	Confirmed application

Source: Mast, H.-U., "The Joint German Neutron Radiography Assessment Programme—Results, Conclusions and Suggestions," in *Neutron Radiography* (J. Barton, ed.), Gordon and Breach Science Pubs., Langhorne, PA 1994.

explosives cartridge casings and in NAA studies to examine propellants and explosives for the presence of contaminants.

2.4 MEDICAL APPLICATIONS OF ^{252}Cf SOURCES

2.4.1 Treatment of Cancer with ^{252}Cf—Brachytherapy

Among the earliest studies of the use of ^{252}Cf were those leading to treatments for cancer. Preliminary work on the dosimetry of fast neutrons and other radiations emanating from very small sources of californium implanted as needles or seeds was followed up by work on the biological effectiveness of this new form of radiation. It was quickly found that neutrons could be more effective than gamma rays in treating tumors under certain

circumstances; consequently, after protocols were developed and facilities constructed, clinical trials were begun. Early results proved to be disappointing, until investigators at the University of Kentucky Medical Center discovered that advanced, hypoxic tumors, which are the most deadly form of cervical cancer, shrank much more rapidly when first exposed to neutrons and then to x-rays or gamma rays. This enhanced shrinkage, which has now been repeatedly demonstrated, results in greatly improved recovery statistics.

By 1985, over 450 patients had been treated in clinical trials. Early results from 1975–1985 showed for the new procedure a 5-year survival rate of 54% versus a 12% rate for conventional radiotherapy. This translates to 189 lives saved at this institution alone by 1985. More recently, Maruyama et al. reported a 45% 5-year survival with only 4% complications in advanced carcinoma of the cervix, which is believed to be the leading malignancy in females in some countries.[45, 46] To date, more than a thousand patients have been treated in the U.S.,[47] saving 400–500 lives. Worldwide, the number of lives saved using ^{252}Cf is much greater.

Treatment of certain cancers through interstitial and intracavitary implants, known as brachytherapy, continues to be used with ^{252}Cf clinically to treat:[48, 49]

•glioblastoma multiforme (malignant brain tumors),
•gynecological and genital tumors,
—cervical cancers,
—uterine cancers,
—breast cancers,
—vaginal cancer,
—cancer of the penis,
—urethral cancer,
—prostate cancer,
—cancer of the pelvic wall,
•anal/rectal cancer,
•cancer of the bladder,
•skin cancers,
•lymph node metastasis,
•head and neck cancers,
—tongue cancer,
—lip cancer,
—floor of mouth,
—oral cavity mucosa cancer,
—advanced tonsillar oropharyngeal carcinoma,
—parotid gland tumors,
—paranasal sinus tumors,
—salivary gland tumors,
•esophageal tumors,
•bronchial cancers,
•soft tissue sarcomas, and
•cancer of the pancreas

Results from initial therapy trials have been promising [47, 50] and have stimulated others to consider this treatment method. There is no substitute for ^{252}Cf in the brachytherapy (also called Seaborg therapy) of certain cancers, which is now being efficiently used in developed nations around the world.[51, 52, 46, 47] Moreover, ^{252}Cf will be more widely useful in interstitial implant therapy as tiny, high activity needle sources become available.

Some institutions involved in brachytherapy treatment and research are listed below.

- University of Kentucky. First institution to use ^{252}Cf for cancer treatment—1975. Pioneered highly successful work on hypoxic tumors. A ^{252}Cf treatment facility was constructed in 1985. The Markey Center continues to perform medical treatment and research programs using ^{252}Cf.

- Other United States cancer centers using ^{252}Cf

—Allegheny Hospital, New York
—Anderson Hospital, Houston, Texas
—Baylor College of Medicine, Houston, Texas
—Harper Hospital, Wayne State University, Detroit, Michigan

—J. G. Brown Center, Louisville, Kentucky
—Rose Center, Lexington, Kentucky
—Sloan-Kettering Center, Dayton, Ohio

- Foreign countries using ^{252}Cf

—Australia
Prince Henry's Hospital, Melbourne
—Japan
Cancer Institute Hospital, Tokyo
Keio Center, Keio
Teikyo University, Tokyo
—Lithuania
Scientific Research Institute of Oncology, Vilnius
—Netherlands
TNO Institute
—Russia
Research Institute of Medical Radiology, Academy of Medical Sciences, Obninsk
—Thailand
Chiang Mai University
—The Czech Republic
Research Institute of Clinical and Experimental Oncology, Brno
—United Kingdom
Gray Laboratory
Hammersmith Hospital

- United Nations, Vienna

—Scientific Committee on the Effects of Atomic Radiation

2.4.2 Boron Neutron Capture Therapy (BNCT)

Some researchers have[53, 54, 55] suggested that the combination of ^{252}Cf brachytherapy with ^{10}B loading of the tumor could increase the tumor radiation dose because of a boost from the $^{10}B(n,\alpha)\ ^{7}Li$ reaction. For clinical trials of fast-fission neutron therapy, nuclear reactors are the best source of neutrons. However, the expense of nuclear reactors and the impracticality of locating them in a hospital or in an urban area make ^{252}Cf an attractive choice for routine therapy. ^{252}Cf has been used with encouraging results in the intracavitary and interstitial treatment of different cancers. With the higher-activity ^{252}Cf sources that are available, external neutron beam therapy or a combination of photon and neutron therapy can be used to treat superficial as well as deep seated carcinomas.

BNCT is gaining support from cancer radiation therapists throughout the world for the treatment of some of the most fatal cancers (gliomas, melanomas, and hepatomas). Currently existing BNCT techniques, combined with ^{252}Cf brachytherapy, might be used to extend the lives of cerebral glioma patients dying throughout the world. However, further studies of boron compounds and irradiation sources are needed for this therapeutic method to become more effective with a wider range of cancers.

2.4.3 Study of Cancer in Mice with ^{252}Cf

Researchers are using ^{252}Cf to gain a better understanding of cancer. At the University of Kentucky, Feola and coworkers study genetic effects and leukemogenesis with ^{252}Cf radiation.[56,57] In Japan, Watanabe and coworkers[58,59] study the effects of the dose rate and the energy of fission neutrons on tumorigenesis in mice exposed to ^{252}Cf neutron irradiation. Ito[60] has used ^{252}Cf to study the relative biological effectiveness of fission neutrons for inducing liver tumorigenesis in mice in comparison to other radiation, such as γ-rays from ^{60}Co. Ito also studied the occurrence of tumors in the offspring of irradiated parents, gaining information about cancer-prone genetic trait activation through ^{252}Cf irradiation of mice. Hence, ^{252}Cf is not only used in treating cancer, but also to gain knowledge of how it is formed, how it is inherited, and how it may be affected.

2.5 ^{252}Cf-PLASMA DESORPTION MASS SPECTROMETRY (PDMS)

Studies begun over twenty years ago of the chemistry that takes place in a nascent fission track have led to a breakthrough in the mass spectrometry of biopolymers. The resultant mass spectrometer, which uses ^{252}Cf as the ion source, has been commercialized. There are close to 200 ^{252}Cf-PDMS systems now in operation throughout the world, primarily in biological research labs. Over the years, the contributions from several labs has led to a solid fundamental understanding of the physics of ion desorption from fission tracks. ^{252}Cf-PDMS developments have recently been summarized along with contemporary research in the U.S., France, Germany, Austria, and Sweden.[61] Some results of recent ^{252}Cf-PDMS activities are highlighted below.

- ^{252}Cf-PDMS has been applied to measure the kinetics of the conversion of coals to soluble products under model liquefaction conditions.[62] The improved mass sensitive detector system requires only small samples (about 500 mg) and showed some advantages over infrared and ultraviolet detection.
- ^{252}Cf-PDMS has been favorably compared with field ionization mass spectrometry and gel permeation chromatography.[63]
- ^{252}Cf-PDMS has been successfully applied to various cyclodextrins,[64] providing information on the molecular structure of these thermally labile compounds.
- Advances in the application of ^{252}Cf-PDMS have led to the use of the technique in clinical quantitation studies, development of a microscopic method for measuring chemical homogeneity, in situ applications where serial modifications can be made and studied in a sequential fashion and the utilization of the primary fragmentation patterns to correlate with the primary structure.[65]

Future research, development and applications will likely focus on the chemistry of the ^{252}Cf-PDMS process and the understanding and control of gas-phase reactions that occur in the ejection plume.[65] The in situ modification technique will likely be applied to new approaches to pharmacokinetic studies and to cell surface interactions where ^{252}Cf expands the capabilities of in vitro studies of biological processes.

^{252}Cf-PDMS devices are commercially available from a company in Sweden and from SUMI, a company in the Ukraine, Russia. The cost is comparable to other types of mass spectrometers.

2.6 FISSIONABLE MATERIALS ASSAY

2.6.1 ^{252}Cf Shufflers

Several new applications of ^{252}Cf have appeared recently, including monitoring low-density wastes for fissionable materials, such as uranium and plutonium. Neutron-chamber detectors, also known as passive-active neutron (PAN) shufflers, or ^{252}Cf shufflers, have so far been used to monitor vehicles, to assay large crates of waste, and to assay 55 gallon drums of nuclear facility waste for uranium and transuranium elements. A shuffler is a nondestructive assay instrument used to determine the fissile content of materials. It places an isotopic source of neutrons near the material to induce fissions, withdraws the source, and counts the delayed neutrons. The source is shuffled in this way until a sufficient number of delayed neutrons have been counted.

The PAN shuffler technique is generally applied to difficult assay cases. The amount of fissile material present may be very small (a few milligrams), the rate of spontaneously emitted neutrons may be low, or the highest assay precision may be required in spite of material inhomogeneity. In all these cases, gamma-ray backgrounds, self-shielding, or matrix effects can make gamma-ray assays impractical, favoring the use of shufflers. Materials ranging from highly radioactive spent-fuel assemblies to low-level waste drums have been assayed with ^{252}Cf shufflers, as have leached hulls, various process materials, scrap, and waste.

Detector applications in the area of nuclear safeguards and waste management involve measuring neutrons from fission and (α,n) reactions with well-moderated neutron proportional counters (often embedded in a slab of polyethylene). Other less-moderated geometries are useful for detecting both bare and moderated fission-source neutrons with good efficiency. The neutron chamber is an under-moderated detector design comprising a large, hollow, polyethylene-walled chamber containing one or more proportional counters. Neutron-chamber detectors are relatively inexpensive, can have large apertures (usually through a thin chamber wall), and offer very good detection efficiency per dollar.

Design calculations for PAN shuffler type devices have been made for some years[66,67,68] and some facilities have recently begun to appear.[68] The theoretical background for shufflers and techniques for practical applications have been reported elsewhere,[69] including procedures for assaying mixtures of fissile isotopes, inhomogeneous materials, and flowing liquids. ^{252}Cf shufflers have been used to assay fissile uranium and plutonium using active neutron interrogation and then counting the induced delayed neutrons. The ^{252}Cf shuffler at Los Alamos National Laboratory (LANL) is used to assay fissile uranium and plutonium by using active neutron interrogation followed by counting of the induced delayed neutrons.

In recent years, both passive- and active-neutron nondestructive assay systems have been used to measure the uranium and plutonium content in drums.[70] Because of the heterogeneity of the wastes, representative sampling is not possible and nondestructive analysis methods are preferred over destructive analysis. Active-neutron assay systems are used to measure the fissile isotopes such as ^{235}U, ^{239}Pu, and ^{241}Pu. There are some active systems that utilize 14-MeV neutron generators with delayed-neutron counting or with the differential die-away technique. There are also passive assay systems that use gamma-ray scanning, while others (for example, passive drum counters) use passive-neutron signals. The add-a-source technique is also used in some cases to improve the accuracy of passive measurements.[71] The add-a-source technique combines passive-neutron counting plus coincidence counting (from fissile material such as plutonium) with multiplicity counting (from an added ^{252}Cf source) to improve accuracy and low-level detectability limits.

2.6.2 Existing ^{252}Cf Shuffler Applications

2.6.2.1 Automated Box/Drum Waste Assay at Savannah River

At the Westinghouse Savannah River Company (WSRC) 321M facility, a ^{252}Cf shuffler waste-assay system has been made a part of material access and accountability boundary.[72] A ^{252}Cf pass-through type shuffler integrated with a conveyor handling system processes waste across the material access and accountability boundary. The WSRC shuffler screens waste boxes and drums for ^{235}U. An automated conveyor loads waste containers into the shuffler, and upon verification, transfers the containers across the boundary. Verification consists of a weight measurement followed by active neutron interrogation. Containers that pass low-level waste criteria are conveyed to an accumulator section outside the boundary. Any container that fails to meet the waste criteria is rejected and sent back to the load station for manual inspection and repackaging. The system, which is automated, significantly reduced personnel operating costs by eliminating the need for security forces at the boundary during waste transfer. The system also eliminates the chance for mix-up between measured and nonmeasured waste.

2.6.2.2 Uranium Billet Assay at Savannah River

WSRC also has a Scrap ^{252}Cf Shuffler used to assay U-Al cores of billets and a newer ^{252}Cf shuffler specifically designed for assaying billets with improved precision. Assay results are part of the materials accountability and quality control information on the billets, which are destined for extrusion into reactor fuel tubes. The shufflers alternately irradiate a billet with neutrons from ^{252}Cf, then count delayed neutrons emitted from the billet. Neutrons from ^{252}Cf are sufficiently energetic to penetrate a large, dense billet and uniformly assay the ^{235}U throughout the billet's volume. An assay is completed in just 8 minutes. Measurement precision, uncertainties, and the procurement and operation of these instruments have been reported elsewhere.[68]

2.6.3 Neutron Multiplicity Coincidence Counting

^{252}Cf is also used as a neutron source for calibration of neutron multiplicity counters, as well as neutron coincidence counters. Such counters are now in use and are still developing, with ^{252}Cf playing an important role. Multiplicity counters are being used for a number of purposes, such as in determining the fissile mass of samples of plutonium-bearing materials from pyrochemical processes[73,74] and for assay of homogeneous and geometrically simple material containing ^{235}U.[71] Pyrochemical process materials are difficult to measure using conventional neutron counting methods because of significant self-multiplication and variable (α,n) reaction rates. Multiplicity counters measure important characteristics of the neutron multiplicity distribution that make it possible to determine sample mass even when multiplication and (α,n) rate are unknown.

Neutron coincidence counters provide for passive assay of small plutonium samples by neutron coincidence counting. Some designs can be used with a glove box sample-well (in-line application) and/or in freestanding mode. LANL performs design calculations, testing and precalibration for coincidence counters. One such counter is in use at the Plutonium Fuel Production Facility in Japan.[75]

2.6.4 Subcriticality Measurements with ^{252}Cf

^{252}Cf-source-driven noise analysis is a method for determining the subcriticality of fissile systems. The measurement provides a parameter (k) that is directly related to the criticality state of fissile systems such as nuclear reactor fuel arrays. The ^{252}Cf-source-driven noise analysis method has been proven useful in measurements of subcritical configurations of fissile systems for a variety of applications, such as:

- initial fuel loading of reactors,
- quality assurance of reactor fuel elements,
- fuel preparation facilities,
- fuel processing facilities,
- fuel storage facilities,
- zero-power testing of reactors, and
- verification of calculational methods for assemblies (with neutron $k < 1$).

^{252}Cf-source-driven noise analysis can verify the criticality safety margins of spent LWR fuel configurations, and be used to justify burnup credit for spent reactor fuel transportation/storage casks. A high sensitivity of noise-measured parameters to small changes in fissile systems has been observed. Monte Carlo calculations of noise-measured parameters have been used to evaluate subcriticality measurements with ^{252}Cf sources against commercially available detectors, with favorable results.

Source chambers containing ^{252}Cf at the required source intensity for LWR fuel applications have been constructed and have operated successfully for ten years and can be fabricated to fit into control rod guide tubes of PWR fuel elements. Fission counters especially developed for spent-fuel measurements are also available, and a portable subcriticality measurement system employing the ^{252}Cf-source-driven noise analysis method with calculational validation capability has been recently assembled.[76]

2.7 EDUCATIONAL APPLICATIONS OF ^{252}Cf

Californium has been used since 1969 when Georgia Tech received a 119 microgram source prepared at the Savannah River Plant.[77] This loan was approved by the AEC to promote the use of ^{252}Cf for educational purposes. The loan also allowed for the subsequent loan of the source to other educational institutions. The earliest uses of ^{252}Cf were in courses and labs, as well as in special projects. Typical course subjects included nuclear reactor engineering, nuclear radiation detection, nuclear engineering lab, radiation attenuation, environmental radioactivity, applied radiological physics, and the chemistry of nuclear technology. Special projects were undertaken by faculty members investigating applications for river water analysis, well-logging, on-line ore sorting, and neutron spectrum studies. Since that time, educational uses of ^{252}Cf have grown tremendously. Examples of some educational applications are highlighted below:

Francis Marion University. ^{252}Cf is used as a neutron generator in the health physics program at Francis Marion University, which has one of the larger undergraduate health physics programs in the nation. Students in courses on nuclear physics, nuclear radiation physics, and health physics use a ^{252}Cf loaded device to learn NAA, study thermal neutron/fast neutron flux ratios, and to make neutron flux determinations and mappings.[78]

Oak Ridge Associated Universities. ^{252}Cf is used to provide training and education on the properties of neutrons and neutron activation. ^{252}Cf is considered an essential component that makes the facility unique.[79]

In all, there are 78 sources totaling over 7 mg of ^{252}Cf on loan (from the REDC at ORNL) to 35 educational institutions in the U.S. Some of these use their sources for calibration, dosimetry, or research purposes. Yet, while many universities are equipped with reactor facilities that can be used for education as well as research, still others must rely upon ^{252}Cf sources as the foundation for their nuclear science and/or health physics programs.

2.8 MISCELLANEOUS STUDIES

^{252}Cf is used to study spent-fuel detector performance.[80] Instruments to measure radiation from spent-fuel assemblies have been developed in the United States and in France with different objectives (safeguards and criticality safety) that have led to different designs. Small capsules of ^{252}Cf and ^{137}Cs placed in a fuel pin are moved through an array of fuel pins. By raising the source pin, axial profiles are measured to determine the responses of instruments to complete assemblies. The measurements show the relative contributions to the detectors' responses of neutrons and gamma rays from different pin locations. Sums of these measurements simulate total responses and how the instruments are correlated. Neutron absorbing pins containing gadolinium can also be inserted into the assembly to measure the dampening of the neutron count rates. These studies are useful in understanding the responses of spent fuel and fresh mixed-oxide fuel stored underwater with poison rods.

^{252}Cf is also used as a neutron source in destructive testing of instruments for space, response to weapons effects, and other applications where sub-atomic particle damage is a factor. An example of the latter is the ^{252}Cf shuffler where ^{3}He proportional counter tubes are degraded by neutron irradiation.[81]

2.9 OTHER APPLICATIONS

It should be noted that there are other applications being made of ^{252}Cf than those detailed in this report. In addition to the important applications that were described, there are some applications, such as calibration and dosimetry, which account for many of the ^{252}Cf sources that are used. ^{252}Cf is extensively used in fission fragment and half-life studies. Also, there are application areas that may use few sources, but are nonetheless important, such as research on a ^{252}Cf power source for space use. No doubt, there are uses of ^{252}Cf of which the authors are unaware.

3. CONCLUSIONS

As a very compact source with a highly concentrated and reliable spectrum of neutrons, ^{252}Cf has found a wide variety of important uses. This report has summarized many, though not all, of these uses. For many of these applications, such as brachytherapy, fuel rod scanning, and online elemental analysis, ^{252}Cf is unique and irreplaceable. For others, such as reactor startup, it is simply the best available alternative. Some applications have fostered new multimillion-dollar-a-year business ventures, such as online analyzers, PDMS detectors, and shufflers. Some applications, such as brachytherapy and explosive or contraband detectors, even save lives. Some uses of ^{252}Cf underpin curricula and research at educational institutions. All of these far-reaching beneficial uses were made possible through the combined efforts of researchers, developers, and entrepreneurs, worldwide, and with the continued support of the U.S. DOE.

It is clear that ^{252}Cf is an important material that is having a worldwide impact. Yet, it is produced in only two places in the world: the United States and Russia. Continued production of this material is strongly needed and supported by many persons, nuclear facilities, academic institutions, and businesses for whom and for which its availability is essential or important.

4. ACKNOWLEDGMENTS

The authors gratefully acknowledge the aid and support received from numerous persons and organizations during the development of this report. The production of transplutonium elements, including ^{252}Cf, in the High Flux Isotope Reactor and the subsequent chemical separation and purification of the product elements in the Radiochemical Engineering Development Center, is supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research, and the U. S. Department of Energy (DOE). The distribution of ^{252}Cf by means of loans is made possible by the base support of the Materials Management Team, Office of Nuclear Weapons Management, Office of Defense Programs, and DOE. The distribution of ^{252}Cf by means of sales to the commercial market is under the aegis of the Isotope Production and Distribution Program, Office of Nuclear Energy and DOE. These DOE programs are administered through the Oak Ridge National Laboratory and the Chemical Technology Division.

Appreciation is expressed for personal direction and guidance from J. E. Bigelow and R. G. Wymer; for administrative support from V. B. Brown; and for consultation and perspective from J. Barton, R. Fox, D. Froom, E. F. Janzow, T. L. Janzow, R. D. MacFarlane, N. L. McElroy, R. Reda, L. S. Robinson, T. Tillinghast, R. C. Woodward, and others; for permissions from the American Nuclear Society; and to K. J. Notz, whose earlier, unpublished manuscript provided the inspiration for this new look at a most versatile radioisotope.

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