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Neutron Sources for Standard-Based Testing

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Neutron Sources for Standard-Based Testing

The DHS TC Standards and the consensus ANSI Standards use ^{252}Cf as the neutron source for performance testing because its energy spectrum is similar to the ^{235}U and ^{239}Pu fission sources used in nuclear weapons. An emission rate of 20,000 ± 20% neutrons per second is used for testing of the radiological requirements both in the ANSI standards and the TCS. Determination of the accurate neutron emission rate of the test source is important for maintaining consistency and agreement between testing results obtained at different testing facilities. Several characteristics in the manufacture and the decay of the source need to be understood and accounted for in order to make an accurate measurement of the performance of the neutron detection instrument. Additionally, neutron response characteristics of the particular instrument need to be known and taken into account as well as neutron scattering in the testing environment.

General properties

^{252}Cf is an intense neutron emitter that is routinely doubly encapsulated in compact, cylindrical source capsules. ^{252}Cf decays by alpha emission (96.91% probability) and spontaneous fission (3.09% probability) which results in an overall half-life of 2.645 years. The average neutron energy is 2.13 MeV and the most probable energy is 0.70 MeV. Californium source activity may be seen quoted in three different ways: in μCi (or Bq), in micrograms (μg) or in neutron flux units (n/s in 4π). When Californium source activity is quoted in μCi (or Bq) it includes all disintegrations that produce alpha particles or neutrons. The relationship between ^{252}Cf activity units is as follows: ^{252}Cf emits 2.314x10^{6} neutrons per second per microgram, its specific activity is 536 μCi/μg and 1 μCi of ^{252}Cf yields 4,316 n/s. The H*(10) neutron dose rate from 1 μg of ^{252}Cf at one meter without any shielding is 2.55 mrem/h. The gamma rays contribute another 0.140 mrem/h. The dose rates (and emission rates) may slightly vary among different ^{252}Cf sources depending on their age, isotopic composition and encapsulation (these are discussed later). The basic nuclear data for Californium isotopes is provided in Table 1.

Table 1. Basic Nuclear Data for Cf Isotopes

Nuclide	Half-Life ($T_{1/2}$)	a-Decay Branching Fraction	Spontaneous Fission (SF) Branching Fraction	Average Neutron Yield per Fission (SF)	Total Neutron Emission Rate [n/(g.s)]
^{249}Cf	351 y	≈ 1.0	5.2x10^{-9}	3.4	2.676x10^{3}
^{250}Cf	13.20 y	0.99921	0.00079	3.53	1.117x10^{10}
^{251}Cf	898 y	≈ 1.0	9.0x10^{-6}	3.7	1.954x10^{6}
^{252}Cf	2.645 y	0.96904	0.03096	3.768	2.314x10^{12}
^{253}Cf	17.81 d	0.0031	Unknown	Unknown	8.406x10^{4}
^{254}Cf	60.5 d	0.00299	0.99701	3.93	1.232x10^{15}

The energy spectrum of ^{252}Cf can be described by the Watt equation:

$$N(E) = e^{-E/a}\sinh(\sqrt{bE}),$$

where E is the neutron energy in MeV and for ^{252}Cf, a=1.18 MeV and b = 1.03419 MeV^{-1}. The average neutron energy is 2.13 MeV and the most probable energy is 0.70 MeV.

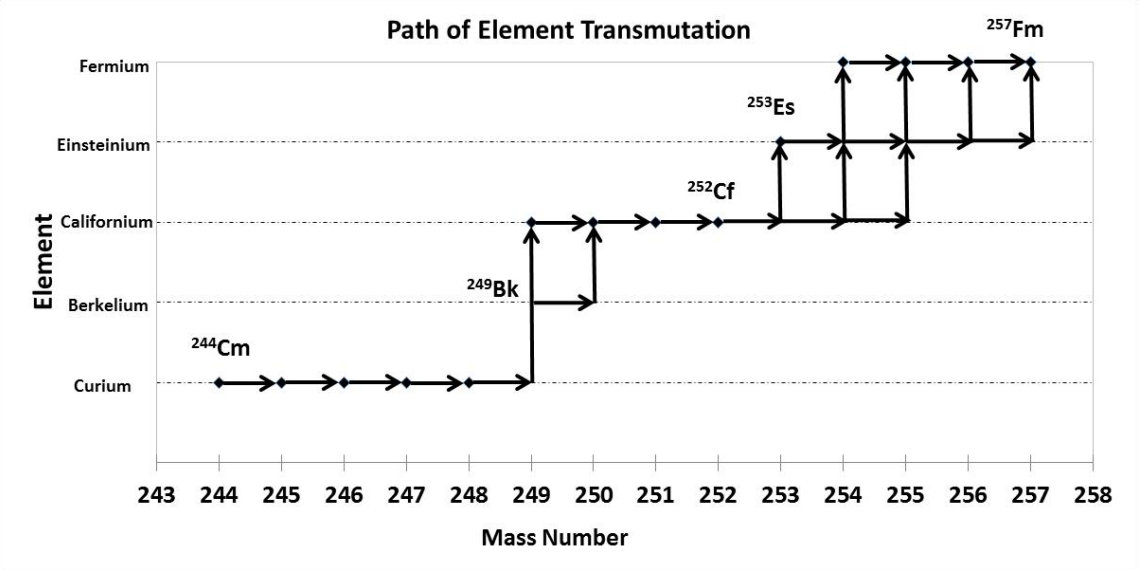
Production

Californium properties, production, supply and applications are reviewed in several reports and presentations [1-5]. Californium is produced in two facilities world-wide: at the High Flux Isotope Reactor (HFIR) located at the Oak Ridge National Laboratory (ORNL) in Tennessee, USA and at the Research Institute for Atomic Reactors (RIAR) in Dimitrovgrad, Russia (Figure 1).

Figure 1. Oak Ridge National Laboratory facility in USA and Research Institute for Atomic Reactors in Dimitrovgrad, Russia

Californium produced at ORNL supplies approximately 70% of the demand for the material. ^{252}Cf is produced in HFIR and then processed in the adjacent Radiochemical Engineering Development Center (REDC) located also at ORNL. The HFIR produces Californium by transmutation from curium oxide targets. The transmutation involves two processes: a). neutron absorption, which increases the nuclear mass by 1 and b). beta decay, which increases the atomic number by 1. For example, ^{244}Cm absorbs a neutron to become ^{245}Cm, and then ^{245}Cm absorbs a neutron to become ^{246}Cm and so on up to ^{249}Cm. At this point the beta decay replaces the neutron absorption by ^{249}Cm because the half-life of ^{249}Cm (64.2 min) is too short to allow further capture of a neutron before ejecting a beta particle and becoming ^{249}Bk. Then the ^{249}Bk either captures a neutron and becomes ^{250}Bk which decays to ^{250}Cf or ^{249}Bk absorbs a neutron to become ^{250}Bk which beta decays into ^{250}Cf. From this point consecutive neutron absorptions produce californium isotopes up to ^{255}Cf (see Figure 2).

Figure 2. Path of element transmutation in the High Flux Isotope Reactor (ORNL)



The transplutonium elements produced at HFIR are chemically separated and purified in a highly sophisticated, remotely operated process. As a result of the Californium production technology each irradiated batch has a mixture of californium isotopes. The isotopic composition of the californium sources is usually given in mass units (e.g. μg). A typical composition of a new ^{252}Cf source from ORNL in atom % is given in Table 2.

Table 2. Isotopic composition of a new source

Nuclide	Isotopic Composition (atom %)
^{249}Cf	4.32
^{250}Cf	10.82
^{251}Cf	3.31
^{252}Cf	81.50
^{253}Cf	0.04
^{254}Cf	0.01

The basic nuclear data for Californium isotopes is provided in Table 1.
With time, Californium isotopes in the source alpha decay to Curium isotopes. The ^{245}Cm, ^{246}Cm, ^{247}Cm and ^{248}Cm isotopes in Table 3 come from the alpha decay of the ^{249}Cf, ^{250}Cf, ^{251}Cf and ^{252}Cf isotopes. The isotopic composition of some older (>15 years) ^{252}Cf sources provided by ORNL is shown in Table 3.

Table 3. Isotopic composition of several old ^{252}Cf sources from ORNL assayed after 15 or more years after production

Source ID	Batch ID	Cf-249 µg	Cf-250 µg	Cf-251 µg	Cf-252 µg	Cm-245 µg	Cm-246 µg	Cm-247 µg	Cm-248, µg
SR-CF-3050OR	CXCF598	1713.53	2042.972	1346.968	728.84	49.52	2345.77	15.09	31262.97
SR-CF-3048OR	CXCF579	2991.615	1819.724	1364.31	445.657	93.12	2322.96	16.45	25621.12
SR-CF-3046OR	CXCF561	2358.67	1379.51	1050.985	338.088	79.42	1975.77	13.7	27092.87
SR-CF-3040OR	CXCF537	2320.446	1956.731	1540.457	331.765	79.78	2890.96	20.49	29154.7
SR-CF-3039OR	CXCF537	1593.869	1344.041	1058.11	227.883	57.7	2146.49	14.81	25345.68
SR-CF-3047OR	CXCF561	1554.705	909.297	692.752	222.849	52.35	1302.32	9.03	17858.13
SR-CF-3037OR	CXCF537	1462.013	1232.852	970.576	209.031	53.24	1986.61	13.67	23897.37
SR-CF-3036OR	CXCF537	1432.616	1208.063	951.06	204.828	52.17	1946.67	13.39	23416.86
SR-CF-3038OR	CXCF537	1419.588	1197.076	942.411	202.965	51.39	1911.78	13.19	22574.26
SR-CF-3041OR	CXCF537	1195.11	1007.784	793.389	170.871	40.95	1481.35	10.52	14788.53
SR-CF-3042OR	CXCF537	1105.295	932.047	733.764	158.029	38.02	1378.05	9.77	13917.49
SR-CF-3034OR	C68EST	2776.956	1442.62	1419.684	126.897	116.85	2917.81	23.06	30037.88
SR-CF-3035OR	CXCF537	769.917	649.238	511.119	110.079	27.55	1018.86	7.07	11604.34

After the chemical separation from Curium the californium mixture is plated onto a palladium alloy wire. An appropriate amount of this wire, dependent on the neutron activity desired, is cut and then doubly encapsulated into small zirconium alloy or stainless steel containers (see Figure 3).

Figure 3. ^{252}Cf capsules

Availability and commercial suppliers

The current picture of ^{252}Cf source supply was shaped in May 2008 when the DOE announced that it planned to halt the production of ^{252}Cf in HFIR in ORNL beyond 2008 [3]. The DOE announcement had arisen because the US government's need for ^{252}Cf had diminished and the DOE decided to stop funding the ^{252}Cf program in order to cut costs. However, a reversal in the DOE position on ^{252}Cf was achieved because of a prompt response by industry users to this potential disruption in supply. In May 2009 the DOE, the industry users and the source providers all came to an agreement to make up the resulting funding gap to continue the ^{252}Cf production program at ORNL. In order to recuperate the cost of funding the gap in the ^{252}Cf production cost at ORNL, the commercial source suppliers have significantly raised the prices for ^{252}Cf sources. New sources are available through the following US commercial vendors:

Frontier Technology Corporation
1641 Burnett Drive
Xenia, OH 45385
Phone: 937-376-5691
Fax: 937-376-5692
Email: info@frontier-cf252.com
www.frontier-cf252.com http://www.frontier-cf252.com/californium.html

QSA Global, Inc.
40 North Avenue
Burlington, MA 01803
Phone: 888-272-2000
Fax: 781-359-9179
http://www.qsa-global.com/sources/home.aspx

Eckert & Ziegler
Isotope Products, Inc.
24937 Avenue Tibbitts
Valencia, CA 91355 (USA)
Phone: +1 661 309 1010
Fax: +1 661 257 8303
E-Mail: isotope@ezag.com
http://www.ezag.com/home/products/isotope-products/industrial-sources.html

General Electric Company
Vallecitos Nuclear Center
6705 Vallecitos Road
Sunol, CA 94586
Phone: 925-862-4292
Fax: 910-341-2892
http://www.ge.com/nuclear/>www.ge.com/nuclear/

Sealed ^{252}Cf sources were available for loan to agencies and subcontractors of the U.S. government and to universities for educational, research, and medical applications. The ^{252}Cf Lease/Loan program was established in the 1970s to provide low-cost access to ^{252}Cf sources. The ^{252}Cf Lease/Loan program has had uncertain funding for the past few years and had been functioning in a reduced capacity, with no new source leases, and source returns performed with full cost recovery to the loanee/leasee. Recently, funding for the ^{252}Cf source lease program was discontinued by DOE and no ^{252}Cf sources are available for leasing.

Decay

The basic nuclear data used by the Radiological Engineering Development Center at ORNL for Cf isotopes are provided in Table 1. The emission rates given in Table 1 can be calculated by the following expression:

$$\text{Emission rate} = \nu*(\text{SF Branching Fraction})*[(\ln 2)/T_{1/2}]*[6.02214\text{x}10^{23}/(\text{At. Wt.})] \quad (1)$$

To get the neutron emission rate in [n/(g.s)], the half-life must be in seconds. ν is the average neutron yield per fission, "At.Wt." is the atomic weight of the isotope.

Since the amount of Californium isotopes is usually given in mass units (e.g. μg, grams,) the masses of the isotopes present in the Californium source mixture need to be decay corrected to get the current amounts. To obtain the activity of each isotope the following formula can be used

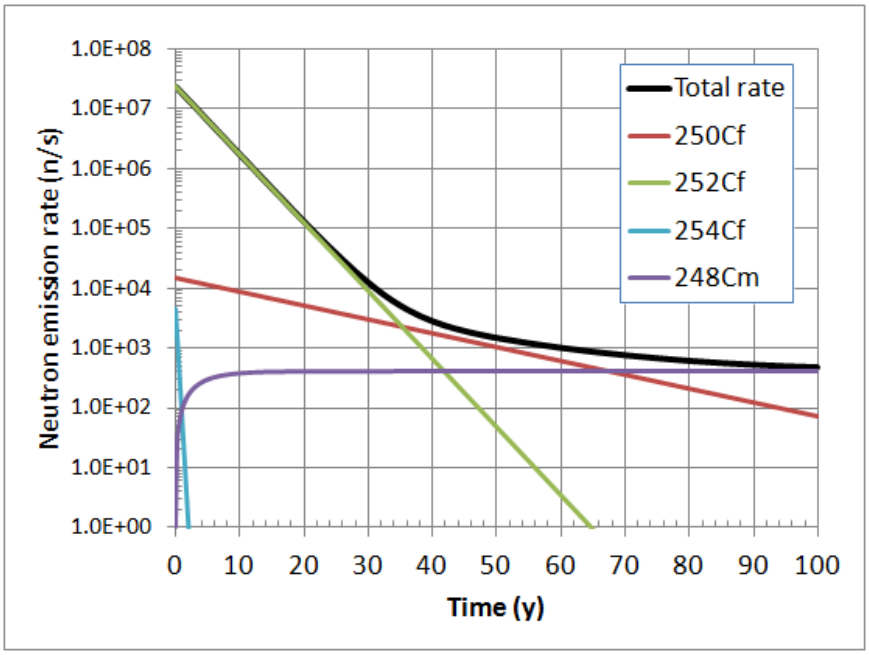
$$A(\text{Bq}) = [\ln(2)/(t_{1/2}(\text{s}))] * \text{mass(g)} * 6.02\text{E}23 / \text{atomic number}, \quad (2)$$

where the half-life is in seconds, mass is in grams, and the resulting activity A (the number of disintegrations per unit time) is in Becquerels. The atomic number is an approximation to the atomic mass term used in Eq. 1. If needed the activity may be easily converted from Bq into Ci by dividing by 3.7E10. If the Californium source has known mixtures of Curium isotopes expressed in mass units (e.g. μg), their activity can also be calculated using equation (2). The overall activity (disintegrations/unit time) of the californium source is the sum of the activities of all isotopes present in the mixture. According to ISO 8529 the relative standard uncertainty in the ^{252}Cf half-life is 0.5% to 0.7%. After about two half-lives (~ 5 y), the uncertainty in the half-life will result in a relative standard uncertainty in the source strength of about 1%. The uncertainties in the half-lives of other Cf isotopes may be even larger. Given the limited supplies of pure isotopes of Californium as well as other experimental limitations it may not be possible to carry out the desired measurements to the degree of perfection that may be desired. It is therefore recommended that ^{252}Cf sources be recalibrated every five years. ISO 8529 requires that the source strength of a ^{252}Cf source shall be corrected for radioactive decay on a day-to-day basis.

In practice, only ^{250}Cf needs to be considered as a competing neutron source as the other Californium isotopes are either too short or long-lived in comparison to ^{252}Cf. However, the contribution of ^{250}Cf to the overall neutron emission rate is negligible for Cf sources less than about 15 years old. The alpha decay daughter of ^{252}Cf, ^{248}Cm has a long half-life ($3.48\text{x}10^5$ y) but a relatively high spontaneous fission probability (8.4%) and so contributes to the overall neutron emission rate. But even after 25 y this contribution is only about 1% of the total neutron yield. Note that the above time scales are comparable to the typical recommended working lifetime of 15 y for Cf sources. But if old (>15y) Cf sources are to be used in standards-based performance testing, an initial neutron emission calibration will be required (due to the propagation of the uncertainty in the ^{252}Cf half-life and the generally unknown contribution from ^{250}Cf). Thereafter, increasingly frequent recalibration will be required as the relative ^{248}Cm contribution increases and the overall neutron yield no longer follows a predictable exponential decrease.

Figure 4 shows the neutron yield with time of a Cf source with the initial composition shown in Table 2. The absolute emission rate shown in this Figure is based on an initial ^{252}Cf content of 10 μg.

Figure 4. Neutron emission rate of Cf source with time



Due to the ±20% tolerance on the neutron emission rate, a new ^{252}Cf source will have a maximum usable life of about 1.5 y in regard to neutron standard-based testing. However, a source's useful lifetime can be extended by ganging weaker sources together to reach the required neutron emission rate. A much older Cf source where ^{250}Cf and ^{248}Cm are important contributors will have a longer useful working life due to the increase in effective half-life but corrections to the neutron yield need to be carefully made.

Nuclear decay data

There is a spread among the nuclear decay data measured by different researchers and there are multiple evaluations of the "best" values. In 2002 DOE standardized all nuclear data for inventory reporting and safeguards applications to one source – the sixth edition of Nuclear Wallet Cards (2000) in PDF version maintained by the Nuclear Data Center of Brookhaven National Laboratory (http://www.nndc.bnl.gov/wallet/wcdoe.html). This version will be "frozen" in time and will be the official, DOE, reference source for nuclear decay information until replaced by a new DOE Standard. The data in the Nuclear Wallet Cards (2000) of interest to ^{252}Cf neutron sources includes the half-lives and branching rations of Californium and Curium isotopes.

Source anisotropy

Since most Cf sources are encapsulated in cylindrical capsules, their emission perpendicular to the cylinder axis and along the radial axis will differ. Additionally, source material in the capsule may not be uniformly distributed. Source guide tubes if present may also contribute to this anisotropy. Source anisotropy may be estimated experimentally by measurement of the neutron emission rate in different directions but is better suited to a Monte Carlo calculation. Typically, Cf source anisotropy ranges from a few percent to 10% even up to 14%.

Neutron scattering

Room scatter by the floor and walls of the counting facility may be determined by radiation transport calculations or by measurements and it is specific for each individual facility (room). This contribution is difficult to standardize among different testing facilities. The air attenuation (air outscatter) and air inscatter of neutrons increases approximately linearly with the source-detector distance. Annex D of ISO 8529 gives the net effect (inscatter minus outscatter) for ^{252}Cf and several ISO- recommended radionuclide neutron sources. In all cases, their effect can be reduced by minimizing source-detector distance. Support structures should be light with little or no hydrogenous materials. For all scatter contributions, the spectral and angular distribution is different from that of the original source spectrum. Therefore, the relative contribution of the scattered neutrons to the reading of the device is dependent upon the energy and angular dependence of the response of the particular device.

Consideration of neutron scatter is especially of concern when the Cf source is unmoderated (as specified in current versions of the ANSI and TC Standards). When used in this fashion, the source of thermal neutrons upon which most instruments rely for detection, is not the Cf source per se but the neutrons which have undergone subsequent energy-loss collisions within the test facility (e.g. concrete floors and walls). If the instrument under test is not surrounded by a hydrogenous moderator (e.g. Polyethylene) or supported on a PMMA phantom, it's these thermalized room-return neutrons which are detected by the instrument. A detector that is surrounded by a moderator or supported on a phantom is less influenced by room-return neutrons but nevertheless in order to ensure that the Cf source is the primary source term, the distance to any scattering surface – particularly hydrogenous materials – should be maximized. Be aware also, that even though a concrete wall might be several meters away, it may be a significant source term if its surface area is large i.e. the angle subtended by the wall is large.

In summary,

- Maximize distance from any scattering materials (especially those containing hydrogen) – such as concrete walls.
- The distance above concrete floors should also be maximized.
- Avoid use of any organic materials (polyethylene, Lucite, etc...) to support the Cf source or instrument under test.
- Be aware, that neutron scattering can be minimized but is not likely to be eliminated entirely. As a result, direct comparison of instrument performance results at different facilities may not be straightforward.

Determination of neutron emission rate

Neutron emission rate is best established using the manganese bath technique at a neutron metrology facility such as NIST. For a new ^{252}Cf source, a simple exponential decay correction (λ=0.2621 y^{-1}) will give an accurate neutron emission rate for many years. An older source (>15 y), will require frequent recalibration as the neutron emission rate increasingly deviates from a single exponential decay.

NIST usually calibrates neutron sources in the range of 10^5 to 10^{10} n/s with an expanded uncertainty from 2-3.5% (k=2) by the manganese bath method. Weaker sources (10^4-10^5) n/s may be calibrated but the uncertainties will be larger. Commercial suppliers calibrate the emission rate of ^{252}Cf neutron sources to 5-8% using cross calibration or a transfer-standard calibration source.

Photon contributions

Photons are produced within the source during spontaneous fission and following alpha decay. The low energy fluence is attenuated by the stainless encapsulation but there is an appreciable fluence of higher

energy photons and overall the photon fluence is about 50% of the neutron fluence. However, from a dose perspective, the photon dose is about 5% of the neutron component. Secondary photons will also be produced through neutron-induced inelastic collisions and capture gamma reactions with material comprising and within the test facility.

Alternative neutron sources

Although ^{252}Cf is currently used for standard based testing because of its spectrum similarity with ^{235}U and ^{239}Pu it has some disadvantages: a relatively short half-life which necessitates frequent source replacement (~1.5 years for standard-based testing). Additional considerations are cost and long term availability. A comprehensive review of alternative neutron sources for standard-based testing is provided in [7]. Alternative neutron sources include other spontaneous neutron sources, sources based on (α,n) reactions and neutron generators based on the (d,d) reaction. Candidate spontaneous sources are listed in Table 4 taken from [7]. From that list Curium isotopes look like the most promising candidates. ^{242}Cm and ^{244}Cm were produced in large quantities in 1990s and ^{248}Cm sources are manufactured in Russia, however limited quantities are produced annually. The IEC recommended ^{244}Cm but no suppliers were identified.

Table 4. Possible candidate spontaneous fission sources for ^{252}Cf replacement

Isotope	Half-life (y)	SF probability	Average Energy (MeV)	Average neutrons per fission	n/s per gram	mg required for 2E04 n/s source
^{252}Cf	2.645	0.0392	2.13	3.7655	2.31E12	8.66E-06
^{254}Cf	0.166	0.9969	~2	3.93	1.23E15	1.63E-08
^{250}Cm	7400	0.86	1.83	3.31	2.03E10	9.83E-04
^{250}Cf	13.08	7.7E-04	~2	3.53	1.10E10	1.82E-03
^{248}Cf	0.914	2.90E-05	2.32	3.34	5.65E09	3.54E-03
^{248}Cm	3.48E05	0.0839	1.95	3.11	4.00E07	0.500
^{242}Cm	0.446	6.37E-08	2.10	2.52	1.97E07	1.02
^{244}Cm	18.1	1.37E-06	2.11	2.75	1.13E07	1.77
^{246}Cm	4780	2.63E-04	2.07	3.18	9.40E06	2.13
^{249}Bk	0.880	4.76E-10	2.09	3.6	1.0E05	190
^{236}Pu	2.85	1.37E-09	2.24	2.13	5.73E04	349
^{238}Pu	87.74	1.85E-09	2.02	2.22	2.60E03	7.69E03
^{249}Cf	350.6	5.20E-09	~	3.4	2.7E03	8.5E03

Neutron sources based on the (α,n) reaction include ^{241}Am-Be, ^{241}Am-B, ^{241}Am-Li, Pu-F_4. Such sources are readily available from source suppliers; however their neutron energy spectra are different from the fission spectra of ^{235}U and ^{239}Pu. The neutron spectrum of (α,n) sources can be modified to mimic the moderated fission spectrum to a certain extent. ANSI and Technical Capabilities standards are moving in the direction of using a moderated neutron source (^{252}Cf) using 4 cm of high density polyethylene. From

the neutron sources based on (α,n) reaction, ^{241}Am looks like the best choice as the alpha emitter and B and Be as best options as target nuclei. Compact neutron generators based on the (d,d) reaction (E_n=2.45 MeV) are also a potential future option as an alternative source. A decision on a ^{252}Cf replacement for standard-based testing should consider the spectral similarity to fission spectrum, half-life and cost, specific activity, availability and supply in quantities required and interference of other isotopes and gammas.

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