The Use of ^{252}Cf for Calibrating Safeguards Monitors

Jeff Chapman[1], Stephen Croft[2]
[1]Oak Ridge Associated Universities
Oak Ridge, TN USA
[2]Canberra Industries, Inc.
Meriden, CT USA
E-mail: chapmanj@orau.gov, scroft@canberra.com

Abstract:

Spontaneous fission ^{252}Cf neutron sources are commonly used to calibrate and test the operating performance of safeguards instruments and portal monitors. ^{252}Cf is preferred over (α,n) sources for many operational and safety reasons, but most importantly can be designed to emit an energy-dependent spectrum that closely matches that of ^{240}Pu. ^{240}Pu is the pre-dominant neutron emitter of plutonium metal and oxides. Because portal monitors and other border monitoring devices are designed to detect low neutron fluence rates at distances of 1-3 metres from the detector, several characteristics in the manufacture and decay of the source need to be understood and accounted for in order to make an accurate measurement of the performance: isotopic composition and ^{250}Cf to ^{252}Cf evolution, age, traceability, angular dependence of the neutron flux, and design and manufacture of the moderating cover are examples. This paper describes these issues and presents the range of possible error terms in the utilization of these physically small, relatively weak, (10^4 n.s^{-1}) neutron sources for calibration of portal monitors.

Keywords: ^{252}Cf; calibration; californium; plutonium

1. Introduction

For calibration of neutron coincidence counters and multiplicity counters, ^{252}Cf is used as a surrogate for ^{240}Pu, the principle isotope of interest in either low or high burnup plutonium fuels. In 1993 Croft [1] presented the results of Harwell Laboratory's work with ^{252}Cf as a calibration source for passive neutron counters. His work presents a comprehensive review of the issues and effects observed, in time, with the neutron multiplicity distribution, mean neutron energy, the Reals to Totals ratio, and the overall effect of source isotopic composition, particularly as it influences the neutron output over time. In passive neutron counters it is imperative to understand and account for these effects so that an accurate representation of the counter's performance relative to a mixture of spontaneously fissioning isotopes found in safeguarded and waste materials of plutonium (^{238}Pu/^{240}Pu/^{242}Pu).

In the last few years, a significant effort is underway for a large-scale development and deployment of homeland defense and border monitoring equipment. Two types of neutron detection systems have been deployed: large, fixed-geometry portal monitors and hand-held radioisotope identifiers (RIDs). Most of these systems use moderated ^{3}He proportional counters to detect fission neutrons. Various algorithms are used to analyze the signal neutron pulse train and alarm when the rate is determined to be greater than the background rate. ^{252}Cf sources are used to calibrate the systems for efficiency and to test overall system performance. Performance-based testing and calibration with ^{252}Cf provides assurances to the instrument users that IAEA Significant Quantities (SQs)[1] can be effectively detected and interdicted, as necessary.

[1] As defined under the terms of the NPT (Non-Proliferation Treaty, 1968), an IAEA significant quantity is "the approximate quantity of nuclear material in respect of which, taking into account any conversion process involved, the possibility of manufacturing a nuclear explosive device cannot be excluded."

This paper discusses the calibration issues of ^{252}Cf relative to the performance testing of these devices for international safeguards and interdiction, and presents an additional framework to the original efforts of McGarry and Boswell. [2]

2. ^{252}Cf Source Characteristics

In the testing of instruments, the nominal neutron count rate specified is between 10^4 and $2x10^4$ neutrons sec^{-1} $(4\pi\ Sr)^{-1}$ [see Section 3].

2.1. Isotope Atom Fractions

The isotopic composition of a ^{252}Cf source changes from target batch to target batch. While ^{252}Cf is produced in two facilities world-wide, we focus on the production facility at Oak Ridge National Laboratory, where over 95% of the targets are irradiated. [3], [4]. The isotopic composition of recent campaigns varies only slightly as shown in Table 1.

	Batch Code						
Cf Isotope	CXCF-598	CXCF-669	CXCF-579	CXCF-561	CXCF-537	CXCF-531	CAMP68
^{249}Cf	4.32	1.87	6.7	6.72	4.72	5.84	5.76
^{250}Cf	10.82	9.1	9.63	9.36	10.19	10.01	9.22
^{251}Cf	3.31	2.78	2.97	2.91	3.04	3.01	2.85
^{252}Cf	81.5	86.13	80.63	80.98	82.04	81.12	81.99
^{253}Cf	0.04	0.0005	0.03	0.02	0.01	0.01	0.165
^{254}Cf	0.01	0.008	0.04	0.01	0.01	0.01	0.018

Table 1. Atom Fractions (%) from ORNL., (1998-2002, Martin R.)

The atom fractions of the principle spontaneous fissioning isotopes of ^{252}Cf and ^{250}Cf are fairly constant between recent campaigns, as shown in Table 1. However, when targets of a different origin were used in the 1980s and early 1990s, the atom fractions were substantially different (particularly for the Russian-produced material). In fact, it was the case during testing of our portal monitors at an off-site facility, that a source from the 1980s was used and inaccurately 'calibrated' to the test date of interest. As the source ages it is very important to account for the neutron output of the ^{250}Cf isotope as well, as shown later in this paper.

2.2. Decay Properties of the Source

Literature values for isotopic properties are found in Table 2. The value for ^{251}Cf is estimated from systematics.

Cf Isotope	Half-Life (y)	Spontaneous Fission Branching Fraction
^{249}Cf	350.6	5.2×10^{-9}
^{250}Cf	13.08	7.7×10^{-4}
^{251}Cf	898	9×10^{-6}
^{252}Cf	2.645	3.1×10^{-2}
^{253}Cf	0.0515	-
^{254}Cf	0.165	9.97×10^{-1}

Table 2. Decay Properties of Constituent Isotopes.

3. Neutron Testing Requirements

A number of new test plans and requirements have been published in the last year, either by the IAEA, the IEC, or ANSI. We focus on ANSI here, American National Standards Institute, standards N42.34 and N42.35. [5], [6].

For RIDs (ANSI N42.34), the specification (§8.3.3) calls for the use of an un-moderated ^{252}Cf source of 0.01 µg. The source when placed 250 mm from the instrument must activate an alarm within 2 seconds (no repeatability tests required). At this distance from the instrument, the source produces a neutron dose rate of "approximately 0.3 mrem h^{-1}," the standard indicates (where 1 mrem is approximately 0.1µSv). No specific neutron output is provided nor is a requirement on the uncertainty of the source output. The assumption is that the source used is always "fresh" and free of other Cf isotopes and that it is "NIST traceable. This is often not the case and therein lies the problem.

For portals (ANSI N42.35), the specification calls for a ^{252}Cf source with an intensity of $2x10^4$ neutrons sec^{-1} $(4\pi\ Sr)^{-1}$ ± 20% (implied at 2 standard deviations from the mean). The source is used un-moderated, and is moved through the portal at a specified rate and distance. The alarm probability must be greater than or equal to 0.90 at a 95% confidence.

With a "fresh" source we see that each of the reference test cases specified uses a different source, given a nominal neutron yield 2.34×10^6 neutrons sec^{-1} $(4\pi\ Sr)^{-1}$ per µg ^{252}Cf. The RIDs use 2.34×10^4 n/s while the portals, 2×10^4 n/s, a 17% difference that creates an operational inconvenience to a test laboratory or during calibration – one would hope the same source would satisfy each of the requirements. In addition, the explicit details of source certification and expression of uncertainty should be given for these test sources, especially at such low yields, where the source cross-calibration techniques described by McGarry and Boswell [2] are pushed by today's practices. Simply, it is very difficult to produce certified reference standards at these rates with desired levels of precision. The ANSI test procedures do no allow for variations in excess of ± 20% (2σ), nor do they allow for decay of the source with time. The standards are essentially written such that a test, with Cf, can be conducted over a short period of time (e.g. months).

4. Source Certification

We present this information here not to question the quality of the manufacturer's work, but rather to give the end user of the source some thoughts to consider when actually using the source and applying it to "certification" test results. As we understand it, californium source suppliers normally produce a "source certificate" in one of two ways, though discussions and improvements are underway as we speak.

The default is to send a certificate showing that the source neutron yield is nominal. When a 20,000 $n.s^{-1}$ source is ordered, for example, the source certificate states that the nominal yield is 20,000 $n.s^{-1}$ at a 10% nominal uncertainty. The source is actually yielding between 18,000 $n.s^{-1}$ and 22,000 $n.s^{-1}$, with a Poisson variation of roughly 140 $n.s^{-1}$ (1σ). This is a very large uncertainty to deal with, given the conditions of the test. In portal monitors, for example, where the probability of an alarm must satisfy the 0.90 probability at a 95% confidence, a source yielding 18,000 $n.s^{-1}$ will either fail, or the system is over-designed (and therefore possibly too costly).
The second way is for the manufacturer to certify the source against a secondary reference material. The customer, for example, receives the same source as in the "default" case, but the certificate reports the actual neutron yield measured. That is to say, instead of 20,000 $n.s^{-1}$, the certificate may read 19,137 $n.s^{-1}$. The real capability to claim better than 10% uncertainty on the (inter-calibration) measurement is probably pushing the limit of the measurement device used by typical source vendors---again this is being evaluated.

In either case, we have had the traditional luxury of performing very accurate self-calibration measurements in our neutron multiplicity counters (using for example the Large Efficient Multiplicity Counter or LEMC) and we will present those results within the next year. As a result, we've been able to predict quite accurately what the actual neutron yield is. Others have not had this luxury.

5. Decay Corrections

The first and most significant judgement error that has been made is using sources that have been "laying around" in the back vault for years. Normally, the end user will decay correct the source activity assuming only ^{252}Cf is present. For sources older than 7-8 years, this calculation will underestimate the neutron yield because it inadequately accounts for the ^{250}Cf contribution, which decays much more slowly (13.08 y half-life).

The proper formalism for treating the neutron yield, with time is to account for the ^{250}Cf, in conjunction with the more rapidly decaying isotope, ^{252}Cf:

$$S(t_2) = S(t_1)e^{-\lambda_{252}(\delta t)}\left[\frac{1 + R_1 e^{\lambda_{eff}(\delta t)}}{1 + R_1}\right]$$

$S(t_n) = \textit{neutron rate at time, } t_n$

$\delta t = t_2 - t_1$

$\lambda_{252} = \textit{decay const } {}^{252}Cf$

$\lambda_{250} = \textit{decay const } {}^{250}Cf$

$\lambda_{eff} = \lambda_{252} - \lambda_{250}$

$R_1 = S_{250}(t_1) / S_{252}(t_1) \qquad R_1 = 0.004742$

6. Summary

In the science of calibrating passive neutron counters, we have been able to make very accurate measurements and predictions of the source yield as a function of decay time because we measure directly the single, doubles, and triples rate using multiplicity counting methods, and, have had access to well characterised reference materials. As a result, we've not been that insistent on the manufacturer's to tighten up their uncertainty estimates of neutron yield; furthermore, the neutron rates have been high enough to exceed statistical issues that are encountered with sources of 10^4 n.s^{-1}. However, in the case of calibrating and testing the newly developed homeland defense devices, the low-level count rate and the fact that we tend to use old sources for testing, creates new problems when certifying the instruments. Greater attention is needed on the issue of source certification, expressions of uncertainty, and instrument performance. In the case where old sources are used, source decay should be performed correctly, as presented here.

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