Time-Dependent Multiplicity for Cf-252 Neutron Source

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Abstract

A Cf-252 neutron source is widely used for testing and calibrating neutron multiplicity counters. General equations for calculating the time-dependent strength and multiplicity of Cf-252 sources are deduced based upon the decay model of Cf-252, Cf-250, and their daughter products Cm-248 and Cm-246. With the nuclear data of these four nuclides, an instance of a long-aged (> 40 a) Cf-252 source is presented to illustrate the variation of the strength and multiplicity with time; the calculation results indicate that the 1st, 2nd, and 3rd moment factorials of the neutron multiplicity are significant decreased compared to the ones of nuclide Cf-252. For verification, a neutron multiplicity counting experiment for a Cf-252 source (I#) and another Cf-252 source (II#) with 17.1-year service life was carried out respectively using a thermal neutron multiplicity counter. The measured results are consistent with the calculation results from equations. The results from this study help to understand the changes in attributes with respect to time for any Cf-252 source while making appropriate corrections for obtaining accurate calibration results.

Key words: Cf-252 neutron source, Cf-250, Cm-248, decay model, neutron strength correction, neutron multiplicity correction, neutron multiplicity counting

1. Introduction

A Cf-252 neutron source is widely used in the field of nuclear physics, nuclear engineering and nuclear technology [1,2,3]. In most conventional application scenarios, users only need the absolute neutron strength of the Cf-252 source as the basic input parameter; however, for non-destructive neutron multiplicity counting, aside from the calibration of the detector efficiency with the Cf-252 source strength, the neutron multiplicity data is also needed to calibrate the coincidence fraction, dead time, and other parameters necessary for multiplicity counting [4].

The neutron strength of the Cf-252 source in the range of 10^5 to 10^{10} n/s can be calibrated with an expanded uncertainty from 2-3.5% (k=2) by the manganese bath method. Then the neutron strength with a short service life (<15 a) can be calculated with enough accuracy from the single Cf-252 decay mode with the activity information and the service life at the time of delivery.

Since the half-life of the Cf-252 nuclide is 2.645 a and the half-life of its isotope Cf-250 is 13.08 a, when the service life of the Cf-252 source is greater than 15 a, the neutron strength is not only determined by the single Cf-252 nuclide decay correction, but also contributed from Cf-250 and their decay daughters Cm-248 and Cm-246. Frankly, the time-dependent strength of the Cf-252 source was well investigated and documented in reference [5,6,7].

The neutron multiplicity distribution of a Cf-252 source with a service life is also determined by the neutron multiplicity from the spontaneous fission (SF) of the Cf-252, Cf-250, and their decay daughter Cm-248 and Cm-246. However, their neutron multiplicities are different from each other, and the real neutron multiplicity of a Cf-252 source should not simply represented with the Cf-252 nuclide SF multiplicity parameters if the service life is getting more than 15 a. As the time-dependent multiplicity for a Cf-252 source is not well investigated and documented, the motivation of this paper is to fulfil this research blank.

For the sake of completeness, general equations for calculating the time-dependent strength were repeated here and the neutron multiplicity of Cf-252 source are deduced based on the decay model of Cf-252, Cf-250, and their daughter Cm-248 and Cm-246. With the nuclear data of these four nuclides, an instance of a long-aged (> 40 a) Cf-252 source is presented to illustrate the variation of the strength and multiplicity with time, and the calculation results indicate that the 1st, 2nd, and 3rd moment factorials of the neutron multiplicity are significant decreased compared to the ones of nuclide Cf-252. For verification, the neutron multiplicity counting experiment for this Cf-252 source (I#) and another Cf-252 source (II#) with a 17.1-year service life was carried out respectively with a thermal neutron multiplicity counter. The measured results were consistent with the results calculated from equations. This helps us to understand the changes in time-dependent characteristics for any Cf-252 source, and to make proper corrections on neutron multiplicity when a long-aged Cf-252 source is used for multiplicity counter calibration.

2. Simulation of a Long-Aged Source

2.1 Nuclear Data

The main isotopes of Cf-252 source are Cf-250 and Cf-251, of which Cf-251 does not exhibit SF. Cf-252 and Cf-250 have α decay pathway, respectively producing decay daughters Cm-248 and Cm-246, which also exhibit SF. The basic nuclear data of these four nuclides are shown in Table 1, which are cited from ENDF/B-VIII.0[8]. In Table 1, λ refers to the decay constant and η refers to probability of SF.

Table 1 Basic nuclear data for Cf-252, Cf-250 Cm-248 and Cm-246

Nuclide	Half-life/a	λ /s^{-1}	η /%	Nubar	Prompt Nubar
Cf-252	2.645	8.30E-09	3.092	3.7676	3.757
Cf-250	13.08	1.68E-09	0.077	3.52	3.51
Cm-248	3.48E5	6.31E-14	8.39	3.161	3.13
Cm-246	4760	4.64E-12	0.0263	2.948	2.93

Defining $P(\nu)$ as the neutron multiplicity of SF, the 1st, 2nd, and 3rd moment factorials (ν_1, ν_2 and ν_3) of neutron multiplicity are defined as followings:

$$\nu_1 = \sum_{\nu=1}^{max} \nu P(\nu). \tag{1}$$

$$\nu_2 = \sum_{\nu=2}^{max} \nu(\nu-1)P(\nu). \tag{2}$$

$$\nu_3 = \sum_{\nu=3}^{max} \nu(\nu-1)(\nu-2)P(\nu). \tag{3}$$

The neutron multiplicity of these four nuclides are shown in Table 2, which are cited from reference [9]. The ν_1, ν_2, and ν_3 of nuclides Cf-250, Cm-248 and Cm-246 are significantly smaller compared with those of Cf-252.

Table 2 Prompt neutron multiplicity of SF for Cf-252, Cf-250 Cm-248 and Cm-246

P(ν)	Cf-252	Cf-250	Cm-248	Cm-246 *
0	0.002177	0.003819	0.0065	0.015211
1	0.025987	0.036543	0.0600	0.076693
2	0.125119	0.167337	0.2228	0.262585
3	0.274046	0.29453	0.3493	0.344768
4	0.305081	0.298273	0.2521	0.217966
5	0.185474	0.14514	0.0898	0.075556
6	0.0659	0.047222	0.0175	0.00722
7	0.014292	0.004017	0.0019	0
8	0.001822	0.003119	0.000067	0
9	0.000102	0	0	0
10	5E-07	0	0	0
total	1.0000	1.0000	1.0000	1.0000
ν_1	3.757	3.510	3.130	2.930
ν_2	11.952	10.344	7.971	6.940
ν_3	31.668	25.192	16.056	12.705

* The sum of the probability was 1.0005 in original reference and corrected to unity.

2.2 General Equations for Time-dependent Strength and Multiplicity

The Cf-252 source is produced via irradiation in a reactor, followed by separation and purification steps prior to encapsulation and final delivery to the end user. The reference initial time-zero for a source is defined when the Cf-252 source was purified and encapsulated. Treating the Cf-252 neutron source as separate Cf-252, Cf-250 and Cm-248 and Cm-246 sources, so the SF rate *F* of a Cf-252 source as a function of time is governed by following equation:

$$F(t) = F_{252}(t) + F_{250}(t) + F_{248}(t) + F_{246}(t), \tag{4}$$

in which,

$$F_{252}(t) = \lambda_{252} N_{252}(0) e^{-\lambda_{252} t} \eta_{252},$$

$$F_{250}(t) = \lambda_{250} N_{250}(0) e^{-\lambda_{250} t} \eta_{250},$$

$$F_{248}(t) = \lambda_{248} N_{252}(0)\left(1 - e^{-\lambda_{252} t}\right)(1 - \eta_{252})\eta_{248}, \text{ and}$$

$$F_{246}(t) = \lambda_{246} N_{250}(0)\left(1 - e^{-\lambda_{250} t}\right)(1 - \eta_{250})\eta_{246}.$$

where *F* refers to SF rate, and *N* refers to number of atoms of nuclides, and λ refers to the decay constant, and η refers to probability of SF, and *x* in subscript refers to isotope x.

The average moment factorials $\overline{\nu_1}$, $\overline{\nu_2}$, and $\overline{\nu_3}$ for neutron multiplicity as a function of time for a Cf-252 source is governed by the following equations:

$$\overline{\nu_1(t)} = \frac{F_{252}(t)\nu_{252,1} + F_{250}(t)\nu_{250,1} + F_{248}(t)\nu_{248,1} + F_{246}(t)\nu_{246,1}}{F(t)}, \quad (5)$$

$$\overline{\nu_2(t)} = \frac{F_{252}(t)\nu_{252,2} + F_{250}(t)\nu_{250,2} + F_{248}(t)\nu_{248,2} + F_{246}(t)\nu_{246,2}}{F(t)}, \text{ and} \quad (6)$$

$$\overline{\nu_3(t)} = \frac{F_{252}(t)\nu_{252,3} + F_{250}(t)\nu_{250,3} + F_{248}(t)\nu_{248,3} + F_{246}(t)\nu_{246,3}}{F(t)}. \quad (7)$$

From equations (2), (3), and (4), the relative contribution of SF neutrons from Cf-250, Cm-248 and Cm-246 gradually increases as the service time of the Cf-252 source increases, resulting in the average first, second, and third moment factorials of neutron multiplicity for the Cf-252 source gradually decreasing.

2.3 An instance of Long-Aged Cf-252 Source

A Cf-252 source (I#) was delivered on 16 August 1981, with a 4.31E7 s-1 neutron emission rate, and has been used with a service life more than 41 years until the end of 2022. The number of atoms of the Cf-252 and Cf-250 was measured [10] and was 4.452E16, 7.746E15 respectively at the date of delivery.

With Equation (4), the time-dependent neutron strength for this Cf-252 source is shown in Figure 1 and the neutron contribution of the four nuclides are listed in Table 3.

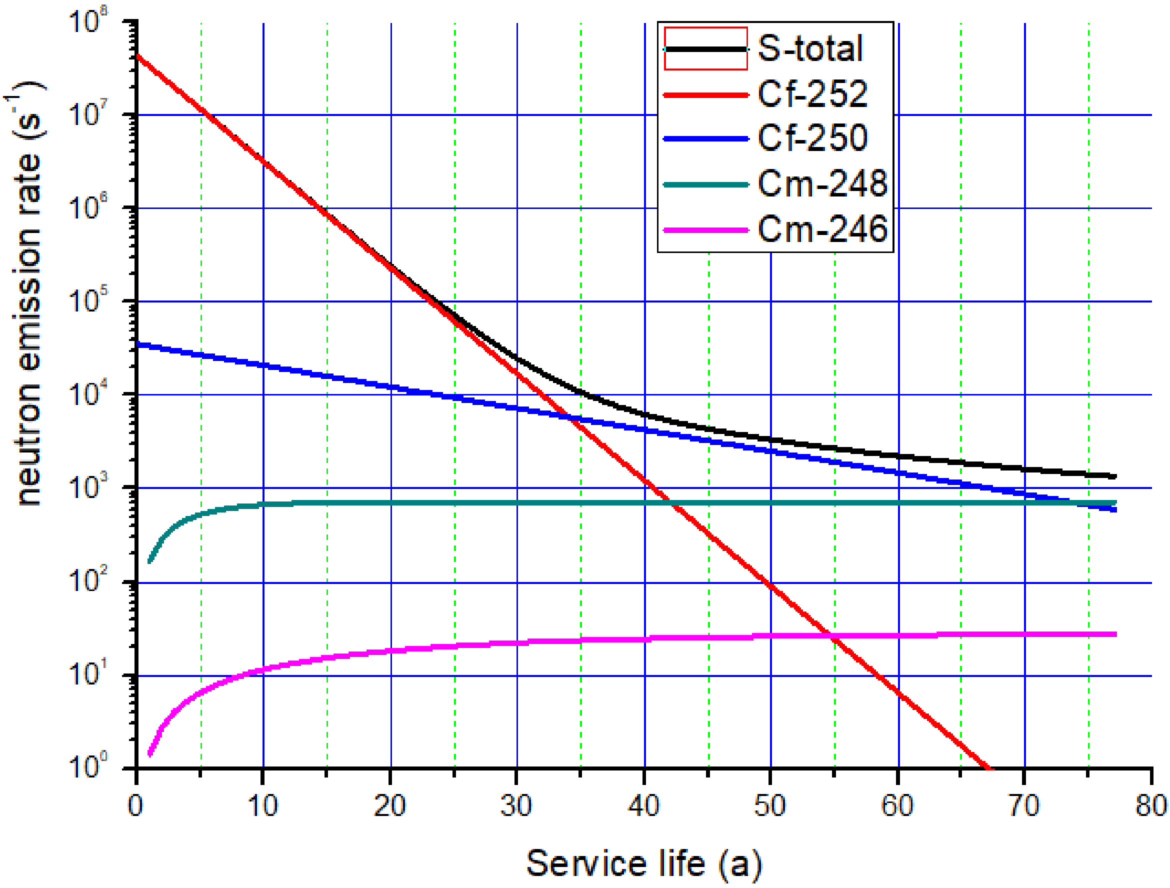

Fig. 1 Time-dependent neutron strength for the Cf-252 source

Table 3 Time-dependent neutron contribution for the Cf-252 source

Service life /a	Total SF rate /s^{-1}	Total neutron emission rate /s^{-1}	Neutron contribution from/%			
			Cf-252	Cf-250	Cm-248	Cm-246
0.0	1.14E7	4.30E7	99.92	0.08	0.00	0.00
10.0	8.38E5	3.15E6	99.32	0.66	0.02	0.00
20.0	6.42E4	2.40E5	94.62	5.07	0.30	0.01
30.0	6.68E3	2.44E4	67.63	29.33	2.94	0.09
40.0	1.76E3	6.16E3	19.51	68.42	11.67	0.40
50.0	9.68E2	3.31E3	2.64	74.88	21.70	0.78

As showing in Figure 1 and Table 3, for this long-aged Cf-252 source with service life of 40 a, the contribution of SF neutron from nuclide Cf-252 was less than 20 %, and the most SF neutron contribution was from nuclide Cf-250 with about 70%, even the neutron from Cm-248 accounted for about 12%. This changes of neutron contribution apparently made the neutron multiplicity of this Cf-252 source changed accordingly.

With Equations (5), (6) and (7), the time-dependent average first, second, and third moment factorials of neutron multiplicity for the Cf-252 source is shown in Figure 2 and their relative difference to nuclide Cf-252 are listed in Table 4.

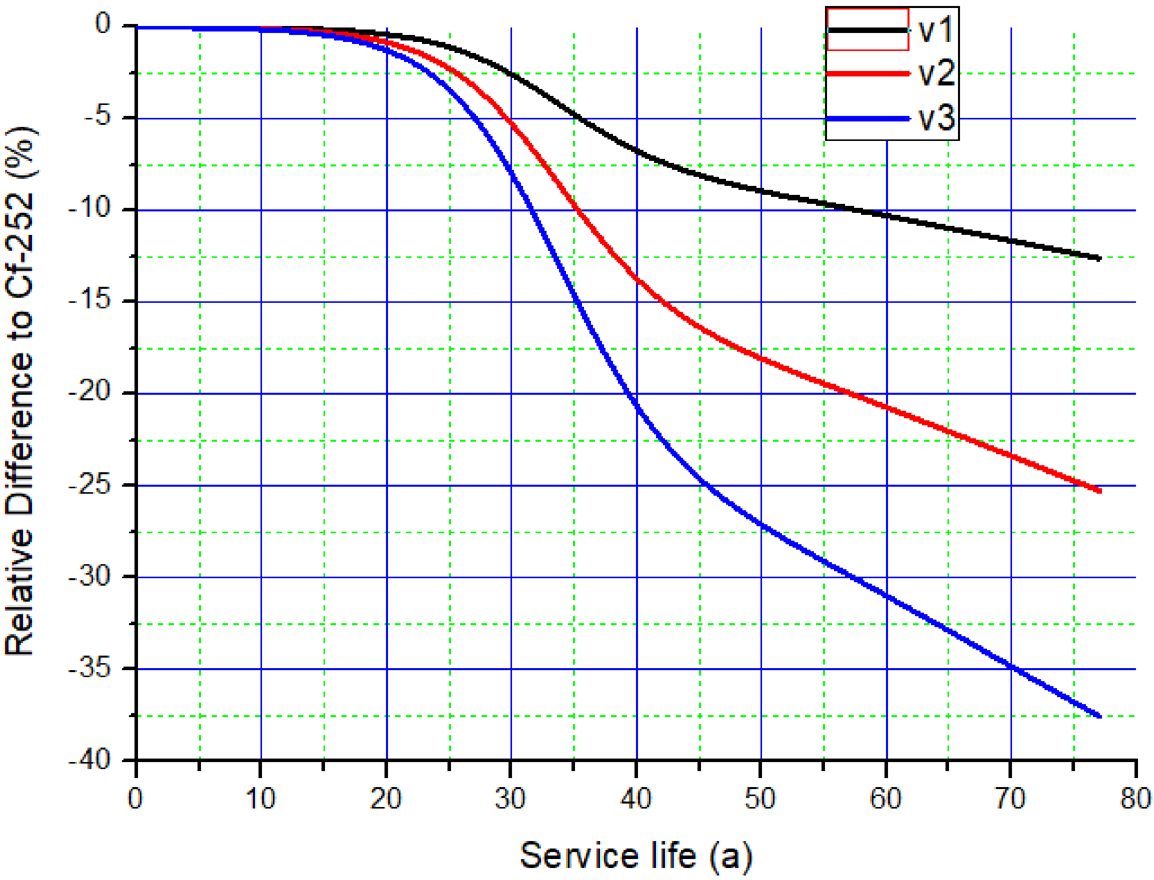

Fig. 2 Time-dependent neutron multiplicity for the Cf-252 source

Table 4 Prompt neutron multiplicity vs. time for the Cf-252 source

Service life /a	$\bar{\nu}_1$	$\bar{\nu}_2$	$\bar{\nu}_3$	Relative bias to nuclide Cf-252 /%		
				$\frac{\Delta\bar{\nu}_1}{\nu_1}$	$\frac{\Delta\bar{\nu}_2}{\nu_2}$	$\frac{\Delta\bar{\nu}_3}{\nu_3}$
0.0	3.757	11.950	31.662	-0.01	-0.01	-0.02
10.0	3.755	11.939	31.618	-0.05	-0.10	-0.16
20.0	3.741	11.850	31.261	-0.42	-0.85	-1.29
30.0	3.659	11.318	29.132	-2.61	-5.30	-8.01

40.0	3.503	10.312	25.126	-6.76	-13.72	-20.66
50.0	3.421	9.792	23.079	-8.94	-18.07	-27.12

As showing in Figure 2 and Table 4, it indicates that the 1st, 2nd, and 3rd moment factorials of the neutron multiplicity for this long-aged (more than 40 a service life) Cf-252 source are significant decreased compared to the ones of nuclide Cf-252.

The accuracy of the strength and multiplicity for the Cf-252 source with single Cf-252 decay mode correction within 20 a service life was less than 6% and 1.5%, respectively. If the Cf-252 was still used for calibration beyond 20 a service life, the strength and multiplicity for this Cf-252 source should be calculated to the experiment date with Equations (4) through (7) to obtain more accurate calibration results, especially in the scenario of multiplicity counter calibration.

3. Experimental Verification

3.1 Description of the Experiments

The neutron multiplicity counting experiment for the Cf-252 source (I#) mentioned above and another Cf-252 source (II#) with a 17.1-year service life was carried out respectively on 5 September 2022. The II# Cf-252 source was relatively newer and used as a reference for comparison and verification.

The parameters of these two Cf-252 sources are listed in Table 5.

Table 5 Original parameters of these two Cf-252 sources on date of delivery

	Date of delivery	Number of atoms of Cf-252	Number of atoms of Cf-250	Neutron emission rate s^{-1}
a)I #	16 August 1981	4.452E16	7.746E15	4.31E07
b)II #	27 June 2005	1.24E15	2.30E14	1.20E06

a) These data cited from reference [10], originally the uncertainty was not provided, but can be deduced from the reference. The uncertainties for all the data are 2%.
b) The data was corrected with the I# source and the uncertainties for all the data are propagated to 2%.

Their calculated parameters upon the experiment date are shown in Table 6.

Table 6 Calculated parameters of these two Cf-252 sources on the experimental date*

	Service life a	SF rate s^{-1}	Neutron emission rate s^{-1}	$\overline{\nu_1}$	$\overline{\nu_2}$	$\overline{\nu_3}$
I #	41.1	1.62E03	5.664E03	3.490	10.232	24.809
II #	17.2	3.65E03	1.37E04	3.748	11.892	31.428

* The uncertainties for all the data are 2%, excluding the data of service life.

A well-type, single ring He-3 tube thermal neutron multiplicity counter with 8.79% (1±2%) detection efficiency was used and the signal was recorded and analyzed with a multiplicity analyzer. The source was placed on the center of the counter respectively for multiplicity measurements. The sketch map of the experiments is shown in Figure 3. The measuring time was long enough to ensure the statistical uncertainty of the measured Single, Double, and Triple counts (refers to S, D, and T, respectively) were less than 1%.

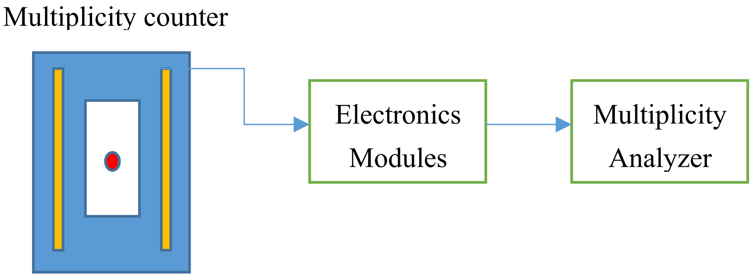

Fig. 3 Sketch map of the verification experiments

3.2 Results and Discussions

The measured S, D and T results for the I# source was 497 (1±0.024%) cps, 32.5 (1±0.23%) cps, and 1.15 (1±1.32%) cps, and 1208 (1±0.01%) cps, 86.6 (1±0.09%) cps, and 3.28 (1±0.62%) cps for the II# source. Based on the mathematic algorithm of the neutron multiplicity counting, the measured S, D and T are governed by the following equations:

$$S = F\varepsilon\nu_1 \qquad (8)$$

$$D = \frac{1}{2} f_d F \varepsilon^2 \nu_2 \qquad (9)$$

$$T = \frac{1}{6} f_t F \varepsilon^3 \nu_3 \qquad (10)$$

in which ε refers to detection efficiency, and F refers to the SF rate, and f_d, f_t refer to the coincidence fraction respectively.

With Equation (8)-(10) and the multiplicity counter parameters, the $\overline{\nu_1}$, $\overline{\nu_2}$, and $\overline{\nu_3}$ for these two sources were deduced and listed in Table 7.

Table 7 Measured results and relative difference

	$\overline{\nu_1}$	$\overline{\nu_2}$	$\overline{\nu_3}$	Relative difference to calculation results /%		
				$\overline{\nu_1}$	$\overline{\nu_2}$	$\overline{\nu_3}$
I#	3.489 (2.0%*)	10.138 (4.0%)	24.675 (6.2%)	-0.05	-0.92	-0.54
II#	3.769 (2.0%)	11.989 (4.0%)	31.167 (6.0%)	0.57	0.82	-0.83

* 2.0% in parentheses is the propagated uncertainty for the measured values.

The measured results are excellently consistent with the calculation results with less than 1% relative difference.

4. Conclusion

The general equations for simulating the time-dependent strength and multiplicity of a Cf-252 source were deduced based on the decay model of nuclides Cf-252, Cf-250, and their daughters Cm-248 and Cm-246. This simulation equations were used to a Cf-252 source with more than a 40-year service life to illustrate the variation of the strength and multiplicity with respect to time and verified with a neutron multiplicity experiment. The measurement results of $\overline{\nu_1}$, $\overline{\nu_2}$, and $\overline{\nu_3}$ for both this long-aged Cf-252 (I#) source and another newer Cf-252 (II#) source were consistent with the simulation results within a 1% relative difference.

This study established a set of integrated simulation equations for both neutron strength and its multiplicity calculation/correction to a long-aged (more than 15 a) Cf-252 neutron source. It helps us understand the changes in attributes with respect to time for any Cf-252 source and make proper corrections. Specifically, these parameters are essential for neutron multiplicity counter calibration in the field of nuclear material assay and accountability. It must be addressed that the number of atoms for Cf-252 and Cf-250 must be known at the date of the last separation of californium from curium and manufactured for delivery.

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6. References

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