Investigation of ^{252}Cf sources with a Ge gamma-ray detector

Frank B.E. Becker*, Ron Dagan and Christian M. Marquardt

Karlsruhe Institute of Technology (KIT), Institute for Nuclear Waste Disposal, Hermann-von-Helmholtz-Platz 176344 Eggenstein-Leopoldshafen, Germany

*Corresponding author: frank.becker@kit.edu

Abstract

This study investigated the feasibility of combining high-resolution gamma-ray spectroscopy with the simulation capabilities of the Nucleonica Nuclear Science Portal with the aim to determine the properties of Cf sources. In this contribution, we present the results for a 20-month-old and a 49-year-old Cf source. In particular, the question arises whether the neutron emission rate can be determined using gamma-ray spectroscopy.

Introduction

Determining accurately the neutron dose is one of the crucial issues of radiation protection. ^{252}Cf, partly decaying by spontaneous fission, is a powerful neutron source. It is widely used e.g. as a calibration standard and even for mobile, portable purposes. To determine the neutron emission rate or dose, several devices such as neutron moderation spheres and dosemeters using thermoluminescence detectors, or optically stimulated luminescence detectors, as well as etched-track and nuclear emulsion detectors are commonly employed for passive neutron area dose monitoring. Active devices such as gas-filled, scintillation and semiconductor detectors perform direct measurements of neutron dose rate. Moderators are used to thermalize fast neutrons in order to profit from the higher interaction cross sections of thermal neutrons. For example, a polyethylene moderator sphere with a He-3 recoil-proton counter tube in its center is such a device. Unfortunately, the measurement results of these devices are subject to greater uncertainties of up to 20–30%. In this study, we investigated the possibilities of high-resolution gamma-ray spectroscopy to determine the properties of Cf sources including the neutron emission rate. Due to the huge amount of spontaneous fission products produced by the decay of ^{252}Cf, highly dense gamma-ray lines are expected in the corresponding spectra. For this reason, and given the overlapping gamma rays, high-resolution experimental devices and advanced simulation tools are required to account for the contribution of each decaying isotope to the gamma-ray spectra. A high-purity germanium (HPGe) detector was used to record gamma-ray spectra. The experiments were carried out for ^{252}Cf sources, including a 20-month-old and a 49-year-old source, and an available HPGe detector. The Nucleonica Nuclear Science Portal[1] offers tools such as e.g. 'Decay Engine' and 'Gamma Spectrum Generator', which allow for calculating the decay products of ^{252}Cf, their age-dependent activities and gamma-ray spectra with a detector response model. The inclusion of simulated gamma-ray spectra offers a new advantageous approach. ^{252}Cf sources also contain other Cf isotopes like ^{249}Cf ($t_{1/2}$ = 350.6 a), ^{250}Cf ($t_{1/2}$ = 13.08 a), ^{251}Cf ($t_{1/2}$ = 898 a) and ^{252}Cf ($t_{1/2}$ = 2.645 a). In this regard, the basic decay schemes of these isotopes are shown in Figure 1.

Materials and methods

Gamma-ray spectra of a 49-year-old Cf source were measured in the controlled area of the Institute for Nuclear Waste Disposal at the Karlsruhe Institute of Technology (KIT-INE) (experimental setup see Figure 2). In another measurement campaign of KIT-INE[3], a 20-month-old Cf source was investigated at the calibration laboratory of the Safety and Environment Department at KIT.

A portable well-type HPGe detector system manufactured by Mirion Technologies (Canberra) was used. The source-detector distance was 20 cm ensuring a low dead time for the detector. The measurement time

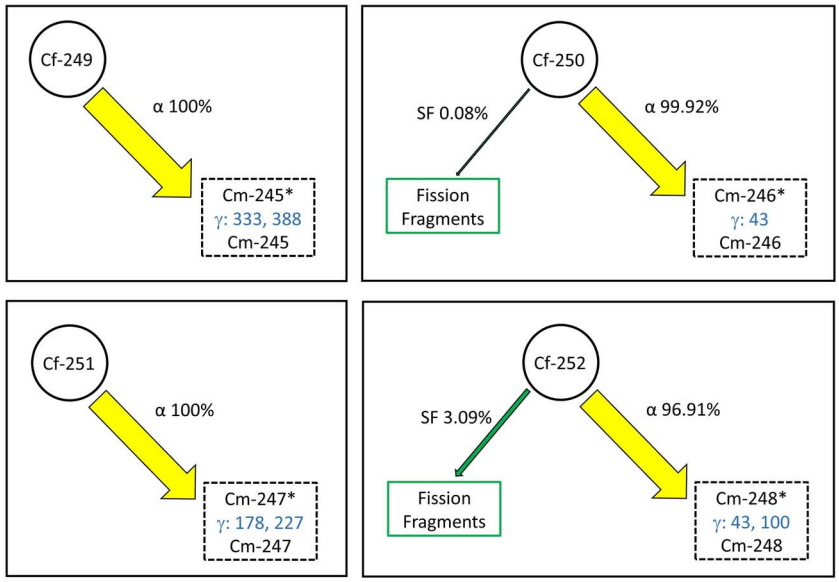

Figure 1. Basic decay schemes of ^{249}Cf, ^{250}Cf, ^{251}Cf and ^{252}Cf. Excited nuclear states after alpha decay are marked with an asterisk. SF stands for spontaneous fission. Subsequent gamma-decay energies are given in keV. The data are taken from reference[2].

Figure 2. Experimental setup for the measurement with the old source and a HPGe detector.

was 22 h. A ^{152}Eu source was employed for energy calibration and for the determination of the detector efficiency. The details of the measurement of the 20-month-old source are given in Wu *et al.*[3].

The source information provided by the available certificates is as follows: for the 20-month-old source, the activity amounts to 198 MBq for the reference of 1 October 2018. The cylindrical double-encapsulated stainless steel source has a diameter of 7.8 mm and a height of 10 mm. The active diameter amounts to 3.2 mm. For the 49-year-old source, only the activity of 560 MBq dated to 19 January 1973 is provided.

The Nucleonica Nuclear Science Portal[1] was used to support the analysis of the gamma-ray spectra. The WEB application Nucleonica allows the simulation of experimental setups from a basic point of view as shown in Figure 3. Gamma-ray spectra are modeled with a point-like source and a basic geometric arrangement of the source, filters and detector. The model geometry of the coaxial detector was obtained with the 'Gamma Spectrum Generator' (GSG)[4].

The GSG approach is based on the idea that the following aspects are of second order and therefore can be neglected. Those aspects include the gamma-ray self-attenuation, pile-up effects due to both true and random coincidence summing, losses of counts due to the dead time of the spectrometer and non-linearity of multichannel analyzers. Based on those assumptions, a specially developed and validated program was used by the programmers of Nucleonica to create different databases for gamma-spectrum simulation. It includes large sets of peak-to-total and continuum-to-total efficiency ratios and parameterized continuum profiles calculated on grids with detector crystal dimensions, gamma-ray energies and source-to-detector distances. More details concerning this methodology are described by Berlizov *et al.*[5]

A typical detector configuration setting for GSG looks as follows[4] (see also Figure 3): source to detector distance—>200 mm, Crystal type—> HPGe, Crystal length—>58 mm, Contact length—>36 mm, Contact diameter—>36.0 mm, Crystal diameter—>56.5 mm, Absorbing filter layers—>0 mm, Detector Input window—> Aluminum 0.5 mm, Crystal packaging—> Vacuum 3.0 mm, Inactive layer/reflector—> Germanium 0.8 mm, Number of spectrum channels—>8192, Channel to energy conversion factor—>0.3 keV per channel, Energy resolution at 122–>1.3 keV, Energy resolution at 1332–>2.3 keV.

In Nucleonica, the 'Decay Engine'[6] provides simulated ^{252}Cf decay data, and gamma-ray spectra can be generated using the Detector Response Model in

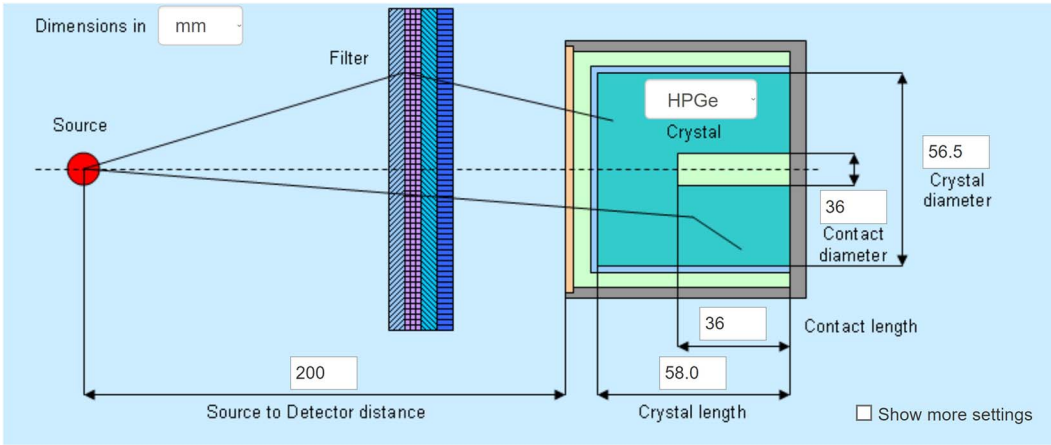

Figure 3. Example for a Ge detector simulation setup in Nucleonica GSG.

the GSG app. The nuclear data for the ^{252}Cf source simulation were taken from the JEFF3.1 library[7]. This data library is today the commercially certificated database for the spectra dealt with in the current work.

Nuclide mixtures containing the decay and fission products of ^{252}Cf after a certain age and their activities were registered. From these data, the corresponding gamma-ray spectra were simulated with GSG. Subsequently, the obtained data were compared with the measurement results.

It should be emphasized that the advantage of Nucleonica is that it does not employ Monte Carlo methods. This allows relatively fast and reasonable calculation of results as long as the measurement setup is in accordance with the validated configuration of the GSG model. Strictly speaking, the use of Monte Carlo methods, which is beyond the scope of this work, will allow more complexity, but will increase considerably the uncertainty of several parameters that need to be carefully checked depending on the problem, as shown for example in the report of the AAPM Research Committee Task Group 268[8].

Results

In Figure 4, the measured gamma-ray spectra of the 20-month-old and 49-year-old sources are depicted. The spectra show a high line density. However, in this investigation, we focus on gamma-ray transitions, which are recommended for the determination of the source composition[9]. To determine the activity of ^{252}Cf, it is recommended by Gehrke *et al.*[9] to use the gamma-ray line of the fission product ^{140}La ($t_{1/2}$ = 1.68 d) at 1596 keV. See also Table 2 in Gehrke *et al.*[9] for an overview of the gamma rays of the fission products of ^{252}Cf. In the case of the 49-year-old source, however, the intensity of this gamma radiation corresponds to the sum of the ^{140}La amounts from the spontaneous fission of ^{252}Cf and ^{250}Cf. In contrast, the contribution from ^{250}Cf is negligible for the 20-month-old source and the 1596 keV gamma ray is attributed to ^{252}Cf decay alone. The gamma-ray transitions as indicated in the spectra of Figure 4 can be assigned to the following Cf isotopes (see also Figure 1): for ^{251}Cf—178 keV (after the alpha decay into ^{247}Cm) and for ^{249}Cf—333, 388 keV (after the alpha decay into ^{245}Cm).

Determining the amount of ^{250}Cf by gamma spectroscopy is still a challenge, but important for older sources. For the 20-month-old source, only ^{252}Cf (1596 keV peak of ^{140}La) and ^{249}Cf could be identified after its alpha decay via the 333 and 388 keV transition of ^{245}Cm* to ^{245}Cm in the spectra. ^{251}Cf could not yet be identified at this age of the source, but only becomes measurable via the 178 keV gamma-ray line after 3–5 years of age[9]. For the 49-year-old source, gamma-ray transitions of the corresponding Cm isotopes attributed to ^{249}Cf, ^{251}Cf and ^{252}Cf, as well as ^{250}Cf alpha decay are clearly visible in the spectra.

Source composition

From the line intensities of the spectra, we obtained the following results for the 20-month-old source[3]: the ratio of ^{252}Cf to ^{249}Cf amounts to 0.16 with a ^{252}Cf activity of 0.2 GBq. For the 49-year-old source, such an assignment was not possible as the thickness and active area of the encapsulation are unknown. For the older source, however, we were able to investigate the intensity ratios for ^{137}Cs and ^{132}I, as their gamma-ray lines at 662 and 668 keV are energetically very close to each other, so an efficiency and attenuation correction is not necessary.

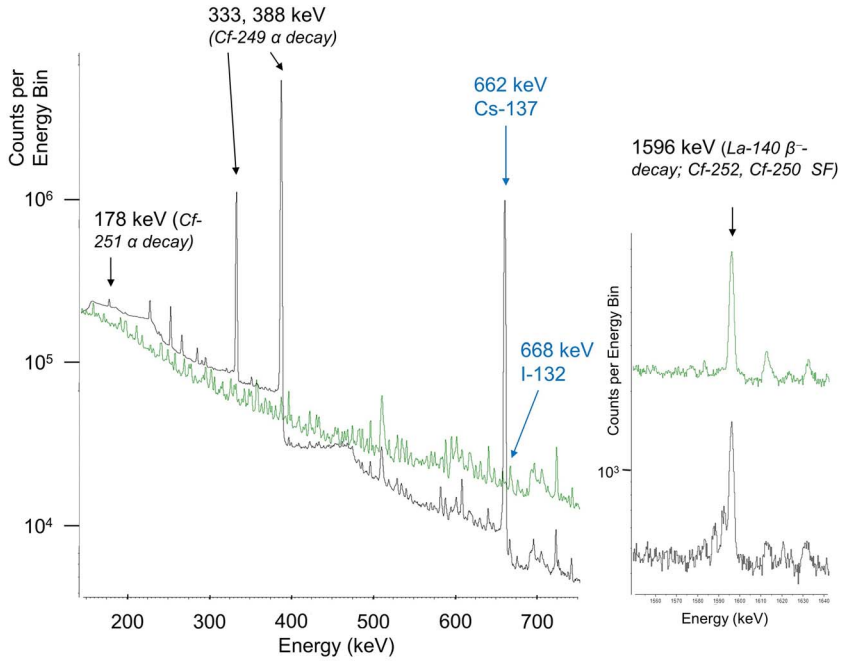

Figure 4. Measured gamma-ray spectra of the 49-year-old (black line) and 20-month-old source (green line, higher background at high energies) in the energy range of 100–800 keV and around 1596 keV (for details, see text).

Age determination

For the age determination, the ratio of the intensities in the gamma-ray spectra of the ^{137}Cs peak at 662 keV and of the ^{132}I peak at 668 keV are proposed by Gehrke *et al.*[9] For our 20-month-old source, the 662 keV peak was not clearly visible in the spectra. Therefore, the gamma-ray transitions and the evaluation of the 133.5 keV peak (mainly due to the beta decay ^{144}Ce/^{144}Pr) in relation to peaks at 497.1 (mainly due to the beta decay ^{103}Ru/^{103}Rh) or 724.3 keV (multiplet, dominated by the beta decay ^{105}Ru/^{105}Rh and ^{145}Ce/^{145}Pr), in combination with Nucleonica simulations, were used to determine the age as 20 months[3]. For the old source with an age of 49 years, the age determination should be applicable by evaluating the ratios of the ^{137}Cs peak at 662 keV and the ^{132}I peak at 668 keV as a function of age. However, from our previous investigations[3] with Nucleonica, different age-dependent activity ratios are predicted. This is illustrated in Figure 5 with the plot of these ratios against the source age. The red curve shows the results from the growth and decay equations are given by Gehrke *et al.*[9], while the black curve is generated by Nucleonica simulations. For ages larger than 30 years, both curves differ significantly. To deduce a reliable age, it must be determined which of the two curves is valid and better describes reality. For that, we measured the activity ratio 662 keV/668 keV of the 49-year-old source, as indicated with the green circle in Figure 5. This data point does not fit with the curve according to Gehrke's method, but follows the trend of the curve from our Nucleonica simulations. The difference in the curve progression over 30 years is explained by taking into account different numbers of contributing nuclides and decay chains. Obviously, the consideration of only one or a few decay chains to the isotope production in older sources is no longer sufficient, and the influence of as many decay pathways as possible should be included. Therefore, we have set the 'Accuracy Factor' in the 'Decay Engine' of Nucleonica to the maximum possible.

Neutron emission and dose rate

For the 20-month-old source, the ^{252}Cf activity of 0.2 GBq converts into a neutron emission rate of 2.5×10^7 neutrons per second. A corresponding neutron dose rate at 50 cm amounts to 1 mSv per h. Note that in this case only the ^{252}Cf activity was taken into account, as the amount of ^{250}Cf is negligible for the 20-month-old source. However, for our old source, the amount of ^{250}Cf becomes important[10].

After the publication of Roberts and Jones[10], the atom number ratio of ^{250}Cf to ^{252}Cf of a newly manufactured source varies from 0.174 to 0.242. With Nucleonica, we calculated the age-dependent ratio of the neutron emission rate (nn) to the intensity of

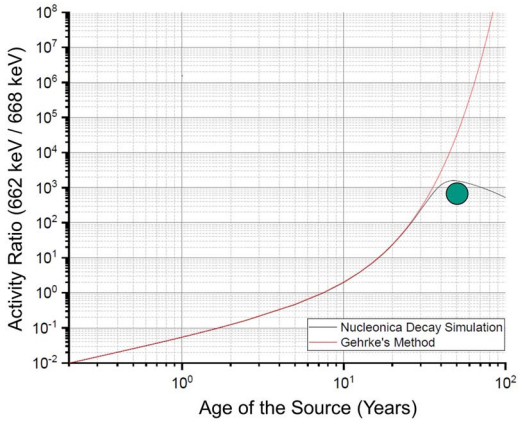

Figure 5. Activity ratio curves for the age determination of ^{252}Cf sources (for details, see text).

1596 keV ^{140}La gamma-ray line. In Figure 6, results for the atom number ratios 0.174 (red circles) and 0.242 (black triangles) are plotted up to a source age of 100 years. As a result, knowing the age of the ^{252}Cf sources and measuring the ^{140}La gamma-ray transition intensity, the neutron emission rate can be determined quite easily and accurately. In our theoretical approach, the ratio is normalized to one for the date of source production. For measurements, the scaling of the ratio must be determined according to the individual experimental setup, i.e. the absolute efficiency of the detector and the shielding of the source capsule must be taken into account. The difference between the two theoretical curves in Figure 6 is at most <1%. In addition, even if the age of the source is not known, but is below 30 years (as an assumed extended maximum for use), nn is determined with an uncertainty of about 3%. To this assignment, one has to add the uncertainty of the measurement. This depends first on the number of counts in the 1596 keV gamma-ray peak in the spectra, which can be kept below 1% if a peak content of at least 10^4 counts is reached. Furthermore, the absolute efficiency of the detector must be considered. This can be determined with calibrated gamma-ray sources or is often provided by the detector manufacturer. The absolute peak efficiency is supposed to be determined with an accuracy of 3%[11, 12]. Another source of uncertainty is the thickness of the source capsule. However, the high gamma-ray energy of 1596 keV causes comparatively low absorption in materials, so an uncertainty of 1 mm in a steel capsule would have an effect of about 5% on the gamma-ray transmission in the material.

In total, an overall accuracy of 5–10% for the neutron emission rate seems to be achievable with this method.

Conclusions

Our Cf source gamma-ray spectroscopy studies are still ongoing and further experiments for deeper insight are envisaged. The studies described and the theoretical predictions for determining the neutron-emission rate via the 1596 keV gamma-ray line of ^{140}La are supposed to be verified or continued with measurements of other well-described ^{252}Cf sources. The simultaneous measurement of the gamma spectrum and the neutron spectrum (e.g. with Bonner spheres) would be a rather complex but more profound method to consider. At present, we conclude the following: to handle the high line density in the spectra of Cf sources, we successfully employed the Nucleonica simulation toolkit. Matching the experimental results with the simulated ones considerably helps to take a large number of nuclides into account.

Information about the Cf isotopes that make up the source can be obtained from the line intensities extracted from the spectra. However, we face limitations for the 20-month-old source with regard to the ^{251}Cf low-energy gamma rays, which are not visible in the spectra. Hence, only the amount of ^{252}Cf and ^{249}Cf could be determined. Concerning the 49-year-old source, detailed information on the source capsule is missing. Although gamma-ray lines of ^{252}Cf, ^{251}Cf and ^{249}Cf could be identified, no quantitative conclusions could be drawn in this regard. Finally, the identification of ^{250}Cf is lacking for both sources. Its amount has a strong influence on the properties of old Cf sources concerning the neutron-emission rate[10]. The identification of this isotope using gamma-ray spectroscopy is a task yet to be solved and will be addressed in future studies. Concerning the age determination of Cf sources, we found discrepancies with the literature in our previous investigation[3]. The measurement result of this study confirms our new approach for the age determination curve. Unfortunately, the age determination for old sources is fraught with high uncertainties. To improve the accuracy, we aim to develop more suitable activity ratio curves by Nucleonica simulations and the analysis of measured gamma-ray spectra. This means that we search for other pairs of gamma rays, as successfully realized in our previous study for the 20-month-old source.

Nevertheless, we could show that the neutron-emission rate, which is a basis for dose rate estimations, can be assessed via the intensity of 1596 keV ^{140}La gamma-ray line in the measured spectra. An

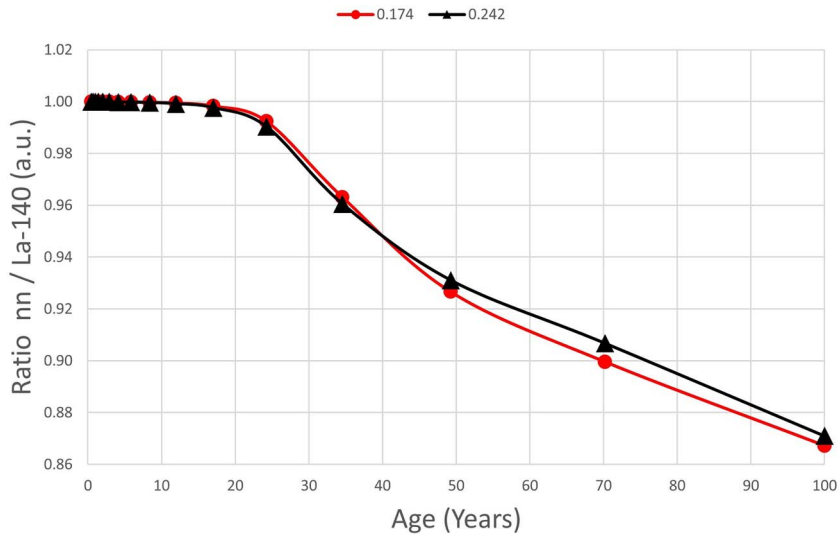

Figure 6. Determination of the neutron emission rate (nn) via the intensity of 1596 keV ^{140}La gamma-ray line (for details, see text).

uncertainty estimation showed that the accuracy of this measurement could be better than that of the usual devices for determining the ambient dose equivalent rate of neutrons.

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