Quantitative determination of ^{252}Cf

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Sealed ^{252}Cf sources in the microCurie to milliCurie range are routinely used in industry and research. At present, no reliable analytical method is available for precise determination of the activity. Very strong sources can be analyzed by neutron counting but the error is large. We propose to use gamma-ray spectroscopy for such analysis. In particular, high-energy gamma rays (above 1 MeV) of fission fragments in equilibrium with the source are very convenient because they have little absorption in materials surrounding the source. We have measured the gamma/alpha ratio for ^{252}Cf with an uncertainty of better than 5 %. The experiment involved the prepration of several thin ^{252}Cf sources, alpha pulse height analysis, determination of alpha decay rate, and the measurement of gamma singles spectrum with a well shielded germanium spectrometer. The measured ratio and the gamma ray spectroscopy of unknown samples can provide the activity of ^{252}Cf in these samples.

1. Introduction

For nuclear structure studies of the fission fragments with large germanium detector arrays, about 100 μCi ^{252}Cf ($t_{1/2}$ = 2.64 y) sources are needed. The fission fragments have energies of 70 - 120 MeV and because of the large velocity of the fragments gamma rays are Doppler broadened. In order to reduce the Doppler broadening, the fragments should stop in a material in a very short time. Thus the fission source must be in intimate contact with some material. We have produced such sources and used them successfully in experiments with large germanium dectector arrays [1]. The ^{252}Cf source was electroplated on an iron foil at Oak Ridge National Laboratory. At Argonne National Laboratory, about 100 μg/cm^2 indium was evaporated on a similar iron foil. This foil was placed on the foil with the electroplated Cf, and the sandwich was passed through a rolling machine. The high pressure brought the Cf into intimate contact with indium [2] and iron. Gamma-ray spectra of this source showed no Doppler broadening. While electroplating the sample it was important to know the activity in the source. In principle, the activity of such a source can be determined by measuring its gamma singles spectrum with a calibrated Ge spectrometer, and using the counts in one of the clean γ-ray peaks. In this procedure, one can use the mass yield [3,4] of one of the abundant isotopes, the γ-ray branching [5–9] of a known strong γ ray in its decay, and the fission decay branch of ^{252}Cf. However, the γ-ray spectrum of the fission source is quite complex and usually either the peaks are not well resolved and/or they contain contributions from several isotopes. Also, if the source is not sealed, the fission fragments may escape beacuse of their large energies and thus the source will not have fission products in secular equilibrium. The ^{252}Cf γ-ray spectrum may contain additional lines due to the excitation of surrounding matarials by fission neutrons. For this reason, the above procedure is difficult to use. We have measured the gamma/alpha ratio for ^{252}Cf decay which provides a simple technique for the quantitative determination of ^{252}Cf in a sample.

2. Experimental methods and results

There were two goals of the present measurements. The first goal was to identify strong clean gamma lines in the γ-ray spectrum of ^{252}Cf

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and measure their relative intensities for different source configurations. The second goal was to determine the absolute intensities of the γ rays identified as good candidates. For the first part, three sealed sources were prepared. The first two sealed sources, labelled in table 1 as Sealed I and Sealed II, were prepared by electroplating the ^{252}Cf activity on 20 mg/cm^2 Au foils. Each of these foils was then covered by a similar gold foil on which 100 μg/cm^2 In was evaporated and then the sandwich was pressed under a rolling machine. In these two sources, the fission fragments stopped in Au in 1-2 ps thus eliminating Doppler broadening of γ-ray peaks. The third sample was prepared by placing ^{252}Cf solution in a 10-ml conical bottle, drying the solution, and closing the bottle. The ^{252}Cf material was produced in the High Flux Isotope Reactor at Oak Ridge National Laborartory and it contained small quantities of ^{249}Cf ($t_{1/2}$ = 351 y), ^{250}Cf ($t_{1/2}$ = 13.1 y) and ^{251}Cf ($t_{1/2}$ = 900 y). The isotopic composition of the source material was determined at Oak Ridge by mass spectrometry.

For the second part, thin sources were prepared on Pt disks by either molecular plating or by spreading the activity with tetraethylene glycol. The α-particle spectra of these samples were measured with a 25 mm^2 Passivated Implanted Planar Silicon (PIPS) detector which had intrinsic resolution (FWHM) of 10 keV. The high-resolution spectrum was used to determine the isotopic composition of the source material. We determined that 97.4±0.3 % of α particles belong to the ^{252}Cf decay which is in excellent agreement with the value of 97.3±0.2 % determined by mass spectrometry. The alpha spectra were also measured with a 100 mm^2 PIPS detector at low geometry (source to detector distance = 8.5 cm) which was determined with a calibrated source. The isotopic composition of the source and its strength were then used to determine the absolute intensities of fission γ rays.

Gamma-ray spectra of all ^{252}Cf sources were measured with a 25 % Ge detector placed in a low-background shield. The energy and efficiency calibrations were made with a calibrated mixed source containing ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y. A 2.6 g/cm^2 beryllium plate was placed between the source and the detector in order to absorb β^- particles. Beryllium was used as the absorber because low-Z materials produce less Bremsstrahlung. For the open sources, the source-to-detector distance was 5.2 cm and for the sealed sources it was between 8 and 10 cm. The gamma-ray spectrum of a dry ^{252}Cf source in a closed bottle measured with the 25 % Ge detector is displayed in Fig. 1.

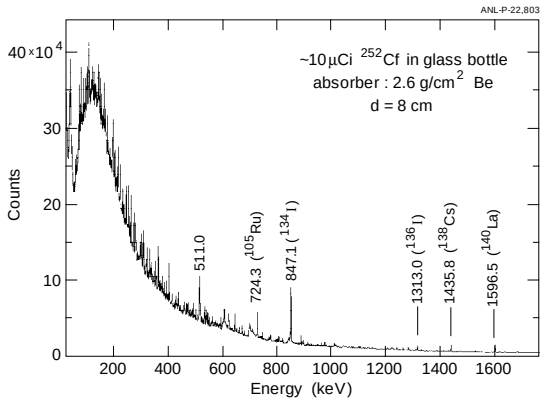

Figure 1. Gamma-ray spectrum of a 10-μCi ^{252}Cf source in a glass bottle measured with a 25% Ge detector.

An expanded region of this spectrum containing the strongest and cleanest peaks is shown in Fig. 2 (a). In Fig. 2 (b), we show a spectrum of an open source for comparison. This open source was covered with a 1-mm thick aluminum plate 6 days before the counting. It is clear that in Fig 2 (a) the ^{140}Ba-La is in equilibrium with ^{252}Cf decay but not in Fig. 2(b). Gamma-ray spectra of the two sealed sources, one source in closed bottle, and an open source were measured. The spectra of the open source were measured at different intervals after the source was covered with the Al plate. The relative intensities of a few strong isolated γ rays determined from these measurement are given in table 1.

The data in table I show that the relative intensities of 1435.8 and 1596.5 keV γ rays are the

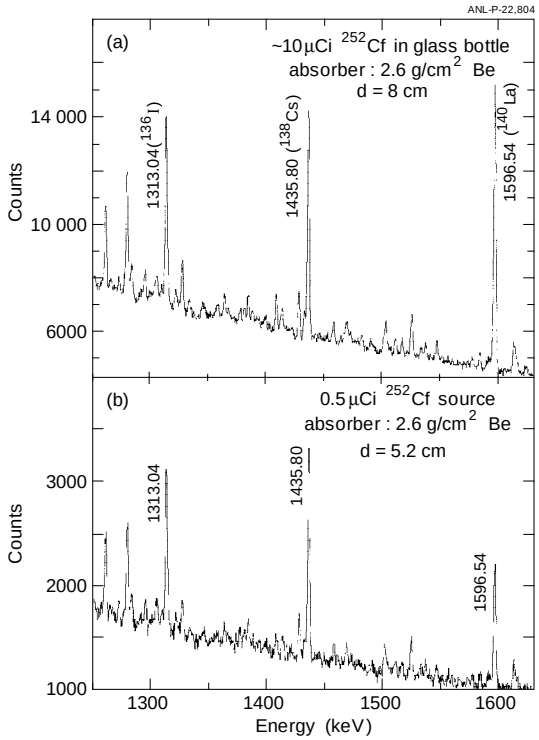

Figure 2. a) An expanded region of the γ-ray spectrum displayed in Fig. 1 showing strongest peaks in the high-energy clean region. b) γ-ray spectrum a 0.5 μCi ^{252}Cf open source measured with a 25% Ge detector 6 days after the source was covered with Al.

same in the three sealed sources where none of the fission fragments escape the source containment. For the open source, the spectrum measured soon after it was covered with the Al plate shows that the 1596.5 keV intensity is only about one half of the intensity measured for sealed sources. The reason for this difference is that in the case of ^{138}Cs ($t_{1/2}$ = 32.2 min), 6 days are long enough to establish secular equilibrium but not for ^{140}Ba-La, which has a half-life of 12.746 d. We have measured the intensity of the 1596.5 keV γ ray at regular intervals and have compared them with values expected from a growth curve for ^{140}Ba-La. In the case of an open source one half of the

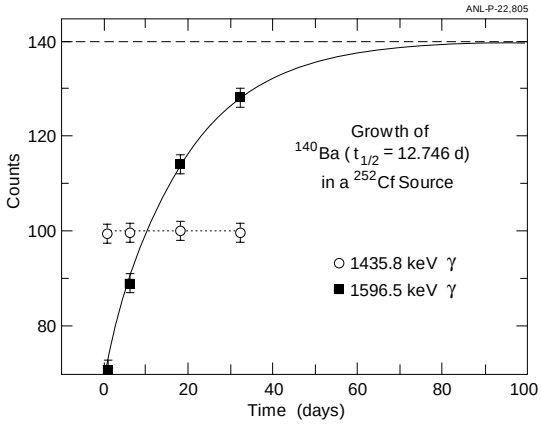

Figure 3. Growth of the ^{140}Ba-La 1569.5-keV peak calculated using a half-life of 12.746 d is shown by the solid curve and measured values by the filled squares. The intensity of the 1435.8-keV γ ray is denoted by open circles.

fission products enters the source and stops there and other half escapes.

Using this information and the half-life of ^{140}Ba (12.746 d) we have calculated the growth of the 1596.5-keV gamma intensity which is displayed in Fig. 3. The measured quantities are shown as filled squares. The good agreement indicates that our assumption is correct and also that there is no loss of any fission fragment. The intensity of the 1435.8 keV γ ray is represented by open circles. The constancy of this intensity indicates that ^{138}Cs reached secular equilibrium within a day as expected from its half-life.

As shown in the table, the relative intensity of the 1313.0 keV γ ray is smaller for the open source and the source in the glass bottle than the intensity for the Sealed I and Sealed II sources. This decrease is most likely due to the Doppler broadening [10]. Independent yields of isobars have been tabulated by Wahl [4]. From this table we can determine direct population of levels in ^{136}Xe, ^{138}Ba and ^{140}Ce in the prompt fission of ^{252}Cf. We see that there is no direct population of ^{138}Ba and ^{140}Ce but there is appreciable direct

Table 1
Relative Intensities of γ rays in ^{252}Cf Fission.

Isotope	Half-life	Gamma energy	Relative Intensity				
		keV	glass	Sealed I	Sealed II	Open Early[a]	Open Late[b]
^{136}I	45 s	1313.04	76±2	82±2	83±2	77±3	74±3
^{138}Cs	32.2 min	1435.80	100 (norm)	100 (norm)	100 (norm)	100 (norm)	99±3
^{140}Ba-La	12.746 d	1596.54	142±3	138±3	142±3	86±3	130±3

[a] The spectrum was measured 6 days after the source was covered.
[b] The spectrum was measured 32 days after the source covered.

population of levels in ^{136}Xe. In the direct population of these levels, γ rays are emitted from moving fragments producing Doppler broadening of γ-ray peaks. Because of the broadening, this contribution is lost from the narrow gamma line.

From the present analysis we have determine the gamma/alpha ratio for the 1435.8 keV γ ray as (1.38±0.0.06)x10^{-3}, which corresponds to an absolute intensity of 0.134±0.006 % per ^{252}Cf decay. This value can also be calculated from the fission branch (3.09 %), cumulative yield of ^{138}Cs (5.82 %) and gamma-ray probability of the 1435.8-keV γ ray (76.3 %). We calculate a value of 0.137±0.010 % which is in excellent agreement with the measured intensity. For a sealed source, we obtain an intensity of 0.19±0.01 % for the 1596.5-keV γ ray using the relative intensities in table 1, which agrees well with the value of 0.18±0.01 % obtained from the fission yield.

3. Summary

We have measured the relative intensities of some clean gamma lines in the fission of ^{252}Cf and find reproducible results for the 1435.8 and 1596.5 keV γ rays. For an open source the intensity of the 1596.5 keV γ ray depends on the length of the time since the source was covered to stop the fission fragments. Also, the intensity of the 1313.0 keV γ ray was found to have different values for an open and properly sealed source. The absolute intensity of the 1435.8 keV γ ray was found to be constant as a function of time for an open source measured a few days after it was covered. We recommend that the absolute intensity of this γ ray reported above be used for quantitative analysis of ^{252}Cf sources. These γ rays, however, do not have as clean Gaussian shape as γ rays from α or β decay sources. The results will therefore vary depending on how the peak areas are determined.

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REFERENCES

1. D. Patel, A. G. Smith, G. S. Simpson, R. M. Wall, J. F. Smith, O. J. Onakanmi, I. Ahmad, J. P. Greene, M. P. Carpenter, T. Lauritsen, C. J. Lister, R. F. Janssens, F. G. Kondev, D. Seweryniak, B. J. P. Gall, O. Dorveaux and B. Roux, J. Phys. G. **28** (2002) 649.
2. C. Teich, A. Jungclaus and K. P. Lieb, Nucl. Instr. Meth. **A418** (1998) 365.
3. K. F. Flynn, J. E. Gindler and L. E. Glendenin, J. Inorg. Nucl. Chem **37** (1975) 881.
4. A. C. Wahl, At. Nucl. Data Tables **39** (1988) 1.
5. A. C. Crouch, At. Data Nucl. Data Tables **19** (1977) 419.
6. J. Blachot and C. Fiche, At. Data Nucl. Data

Tables **20** (1977) 241.
7. W. W. Bowman and K. W. MacMurdo, At. Data Nucl. Data Tables **13** (1974) 90.
8. L. E. Glendenin, J. E. Gindler, I. Ahmad, D. Henderson, J. Meadows, Phys. Rev. **C22** (1980) 152.
9. Table of Radioactive Isotopes, E. Browne and R. B. Firestone editor, John Wiley & Sons, New York (1986).
10. A. G. Smith, W. R. Phillips, J. L. Durell, W. Urban, B. J. Varley, C. J. Pearson, J. A. Shannon, I. Ahmad, C. J. Lister, L. R. Morss, K. L. Nash, C. W. Williams, M. Bentaleb, E. Lubkiewicz, and N. Schulz, Phys. Rev. Lett. **73** (1994) 2540.