Armenian Journal of Physics, 2015, vol. 8, issue 3, pp. 102-111

SEARCHING FOR LONG-LIVING RARE PRODUCTS OF THE ^{252}Cf SPONTANEOUS FISSION

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Received 19 May 2015

Abstract – The long-living products of the ^{252}Cf spontaneous fission are searched for with the help of a Ge(Li) detector irradiated by a 25.5 year old source in the low-background laboratory in the Avan salt mine (Yerevan). No significant photopeaks from the decays of ^{125}Sb, ^{134}Cs, ^{146}Pm, ^{150}Eu, ^{152}Eu and ^{158}Tb are observed in the detector response function analyzed in the energy range of 400÷1400 keV. The upper limits of their cumulative yields are estimated. Strong photopeaks are observed for two long-living isotopes, ^{137}Cs and ^{154}Eu . The content of the ^{137}Cs photopeak corresponds to its cumulative yield of $(5.07\pm0.35)\cdot10^{-2}$, in agreement with the available data, while the contents of the ^{154}Eu photopeaks correspond, on an average, to the yield of $(1.84\pm0.17)\cdot10^{-2}$, which is much larger than expected from the model predictions. The upper limits for the rates of the sodium (^{22}Na) and cobalt (^{60}Co) radioactivity of the ^{252}Cf nucleus relative to the rate of the α-decay are estimated to be $1.6\cdot10^{-5}$ and $3.5\cdot10^{-7}$, respectively.

Keywords: spontaneous fission, rate of radioactive decay

The spontaneous fission of heavy nuclei is an extremely complex many-body, many-channel process. The main observables that reflect the underlying mechanisms of this process are the mass and charge distributions of the fission fragments. The number of potentially possible fragments is enormously large, reaching about one thousand. To date, only a small fraction of them is observed experimentally, because of very low yields (both independent and cumulative) for the overwhelming part of fission fragments (see for review [1] and [2] and references therein). For instance, in the case of the ^{252}Cf nucleus the cumulative yields are measured only for less than one hundred fission fragments [1]. Any new experimental information on the latter will be rather useful for testing the model-based predictions for the mass and charge distributions of the fission fragments, in particular, in the regions far from the maximum of these distributions, where the available experimental data are rather scarce.

The aim of this paper is to obtain first experimental restrictions on the cumulative yields of a number of long-living fission products (with half-life $T_{1/2}$ exceeding a few years) accumulated as a result of the spontaneous fission processes in a ^{252}Cf source during long time (25.5 years). The search for long-living products was carried out by detection of γ-quanta accompanying their β-decays by a Ge(Li) detector installed in the low background laboratory in the Avan salt mine (in Yerevan), 242 meters underground (about 655 m.w.e.). The description of the experimental setup can be found in [3], [4] and [5]. The procedure of the analysis of the response function of the Ge(Li) detector is described in [6].

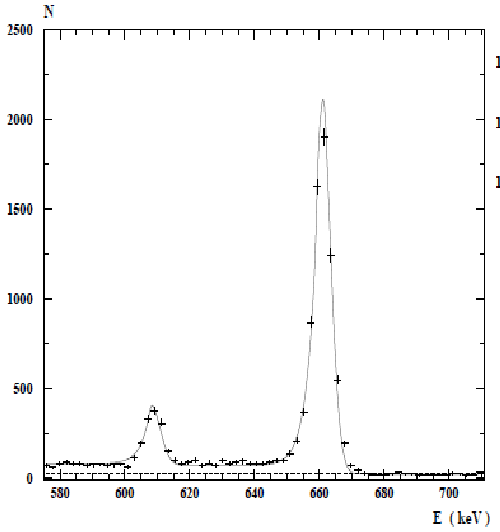
The ^{252}Cf fission source used in this work is prepared 25.5 years ago with an initial activity of $2\cdot10^5$ Bq. The detection of its γ-radiation was performed in two runs of t_m = 256 and 160 hours counting time. The detector response function, built during these runs, is contributed by gammas from the prompt de-excitation of fission fragments, gammas from the decay of short-living fission products, as well as gammas from the decay of a fraction of long-living products which (or their precursors in the corresponding decay chain) had not escaped from the source surface but have been accumulated in the source volume during 25.5 years. In addition to these runs, complementary measurements were performed, detecting for t_m = 161 hours the γ-radiation from the source empty container, the radiation activity of which is almost exceptionally related to the long-living fission products escaped from the surface of the source and accumulated on the inner surface of the container during a period (about 24 years) when the source was being kept in the container. As it will be discussed below, the detector response function can be also influenced by the environmental background radiation.

The long-living fission products involved in present study are listed in Table 1, along with their comparatively intense γ-lines to be looked for in the detector response function. Besides nuclides-candidates to the conventional binary fission, Table 1 also includes long-living light nuclei ^{22}Na and ^{60}Co which presumably can be produced in very asymmetric binary fission (so called cluster radioactivity) or in the ternary fission of ^{252}Cf. Note, that we did not include in Table 1 several γ-lines which overlap with other expected intense lines, both associated with the other fission fragments or the environmental background radiation caused, in particular, by the presence of a variable content of the radon gas in the salt mine, as well as by inelastic scattering of fission neutrons in the constructive materials of the setup (some examples will be discussed below).

Table 1. Half-lives and emitted γ-quanta energies of nuclides considered. The data are taken from [7-9].

Nuclide	Half-life (in years)	Energy of gammas (in keV)
^{22}Na	2.6	1274.5
^{60}Co	5.27	1173.5; 1332.5
^{102m}Rh	3.74	475.1; 631.3; 697.5; 766.8; 1046.6; 1103.2; 1112.8
^{108m}Ag	438	433.9; 614.3
^{125}Sb	2.76	427.9; 463.4; 600.6; 632.0
^{134}Cs	2.07	604.7; 795.9
^{137}Cs	30.08	661.7
^{146}Pm	5.53	453.9; 735.9; 747.2
^{150}Eu	36.9	439.4; 584.3
^{152}Eu	13.54	778.9; 964.1
^{154}Eu	8.6	873.2; 996.3; 1004.8; 1274.4
^{158}Tb	180	994.1

Unlike all long-living products of the ^{252}Cf self-fission, the ^{137}Cs is known to belong to the most favorable ones, with the cumulative yield Y_c , measured in several experiments, around $5{\cdot}10^{-2}$ [1], in agreement with the predicted value of $Y_c = 5.05{\cdot}10^{-2}$ [2]. Our data plotted in Figures 1 and 2 (for the registered energy range of 575÷710 keV) are consistent with this feature, exhibiting the dominance of the ^{137}Cs $E_\gamma = 661.7$ keV line in the detector response function inferred both in measurements with the container (without the source) and with the source (without the container). Besides this main photopeak, our data exhibit another, lower-energy peak, corresponding to the 609.3 keV line of ^{214}Bi, a progeny of the ^{226}Rn decay chain. The data presented in Figure 1 can be fitted including into the fit function the response functions of the mentioned two γ-lines (being parameterized as described in [5] and [6]) and a practically energy-independent (in the considered energy range) low-level environmental background. The fit result is depicted by the solid curve in Figure 1.



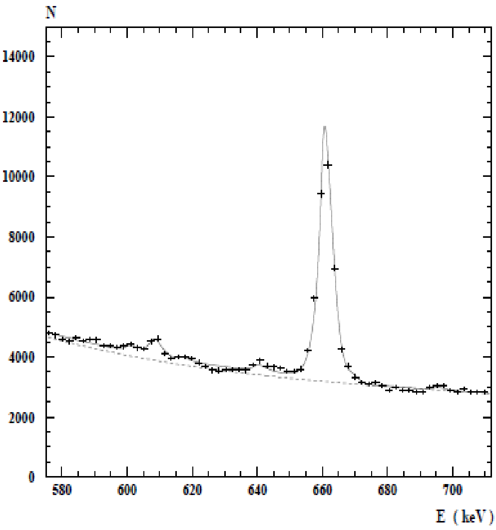


Figure 1 (left). The detector response function in the registered energy range of 575÷710 keV in the measurements with the source container. The solid curve is the fit result (see the text). The dashed line is the fitted background.

Figure 2 (right). The same as Figure 1, but for the measurements with the source.

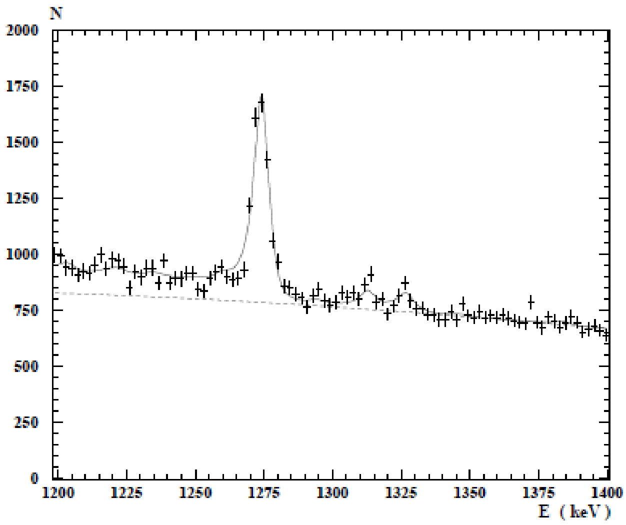
The description of the data plotted in Figure 2 is more complicated due to the appearance of several noticeable photopeaks corresponding to short-living fission products, as well as due to the large contribution from continuum background. The latter originates mainly from a superposition of a large number of long-term tails of the response functions of higher-energy γ-lines corresponding to the prompt de-excitation of fission fragments, as well as the decays of short-living fission products. The continuum background can be parameterized by an exponential function. In order to reproduce the peculiarities in the observed spectrum, we inserted into the fit function fixed contributions from the most favorable short-living fission fragments, which were calculated using the corresponding cumulative yields Y_c , the intensity I_γ and the detection efficiency ε_γ of decay photons, as well as the expected number of fission acts during the measurement time t_m . The values of Y_c were taken from predictions [2] which are, for favorable fission fragments, proved to be quit consistent with experimental data (if any) [1]. The most prominent considered contributions from short-living isotopes (with $T_{1/2}$ not exceeding a few hours) are visible in the detector response function, including: ^{140}Cs (602.2 keV), ^{112}Ag (617.4 keV), ^{142}La (641.3 keV), and a group of three nuclides, ^{101}Mo (696.8 keV), ^{132}Sb (696.8) and ^{132m}Sb(696.8 keV). The included contributions from ^{97}Nb (657.9 keV) and ^{132}I (667.7 keV) are obscured by the wings of the ^{137}Cs photopeak. The contributions to be found from the fitting procedure concern the lines 609.3 keV belonging to ^{214}Bi and 661.7 keV belonging to ^{137}Cs, as well as 669.6 keV due the inelastic scattering of fission neutrons in the shielding copper layers, ^{63}Cu(n,n′γ)^{63}Cu. The contribution from the latter is barely visible in Figure 2. The fit result is depicted by the solid curve in Figure 2.

The number n_γ of counts corresponding to the ^{137}Cs photopeak, inferred from the fitting procedure, is related to the cumulative yield Y_c of ^{137}Cs via the following simplified expression, valid for the case when the measurement time t_m is much smaller than the inverse decay constants of ^{252}Cf and ^{137}Cs, $1/\lambda_S$ and $1/\lambda_F$, respectively:

$$n_\gamma = Y_c (1-\eta_{esc}) I_\gamma \varepsilon_\gamma f N_0 t_m \lambda_S \lambda_F [\exp(-\lambda_S t_a) - \exp(-\lambda_F t_a)] / (\lambda_S - \lambda_F), \qquad (1)$$

where t_a is the source age, N_0 is the initial number of ^{252}Cf nuclei, f is the branching ratio of the ^{252}Cf spontaneous fission, η_{esc} is the probability that the fission fragment ^{137}Cs (or, more exactly, its precursors ^{137}I and ^{137}Xe the individual yields of which significantly exceed that for ^{137}Cs) escapes from the source surface. Note, that the detection efficiency ε_γ for the 661.7 keV line in our case is equal to $\varepsilon_\gamma = (3.3 \pm 0.2) \cdot 10^{-3}$ [5]. The expression (1) was also applied to the measurements with the container, replacing the coefficient $(1-\eta_{esc})$ by η_{esc} and introducing a small correction due to its cooling time (a short period during which the container was empty). The fit of the data collected with the source during 256 and 160 hours results in the following estimations for the product of Y_c and $(1-\eta_{esc})$: $Y_c(1-\eta_{esc})=(3.53 \pm 0.12) \cdot 10^{-2}$ and $(3.43 \pm 0.12) \cdot 10^{-2}$, respectively, where the quoted errors reflect both statistical ones and uncertainties in the energy calibration of the detector. On the other hand, the fit of the data collected with the container leads to $Y_c\,\eta_{esc}=(1.61\pm0.13)\cdot10^{-2}$. Combining these values, one infers $\eta_{esc}=0.32\pm0.02$ for the escaping probability averaged over more prominent products (with the atomic weight A=137) of the ^{252}Cf spontaneous fission, while for the cumulative yield one obtains $Y_c = (5.07\pm0.35)\cdot10^{-2}$, where the quoted error also includes the uncertainty in the efficiency ε_γ (±6%). The quoted value is in agreement with the available estimations [1], [2].

In order to search for signals and infer restrictions on the cumulative yields of other long-living isotopes listed in Table 1, we analyzed the detector response function in the following, relatively narrow energy regions: (410÷490) keV, (575÷710) keV, (730÷830) keV, (830÷950) keV, (950÷990) keV, (990÷1092) keV, (1092÷1150) keV, and (1200÷1400) keV. Besides ^{137}Cs, strong photopeaks were observed (in measurements with the source) only for ^{154}Eu at the all energies listed in Table 1: E_γ = 873.2, 996.3, 1004.8 and 1274.4 keV, the latter being the most intense one (I_γ = 0.348). In Figure 3 the response function in the energy range (1200÷1400) keV is plotted, exhibiting a strong photopeak of the 1274.4 keV line. It is slightly contaminated by a group of three low-intensity lines belonging to short-living isotopes ^{144}La(1276.3 keV), ^{95}Sr(1277.4 keV) and ^{138}I(1277.5 keV). The contributions of the latter, as well as other relatively intense lines belonging to the most favorable short-living fission products, are fixed in the fit function (as described above for the case of the energy range of 575÷710 keV). The contributions to be found from the fitting procedure concern the E_γ = 1274.4 keV line of ^{154}Eu, as well as the 1327.0 keV line originating from the background reaction $^{63}Cu(n,n'\gamma)^{63}Cu$. Similar fitting procedures were also performed for two other energy ranges, (830÷950) keV and (990÷1092) keV, where the lines E_γ = 873.2 keV, 996.3 keV and 1004.8 keV of ^{154}Eu are enclosed. The fit result for the range of (990÷1092) keV is shown in Figure 4.



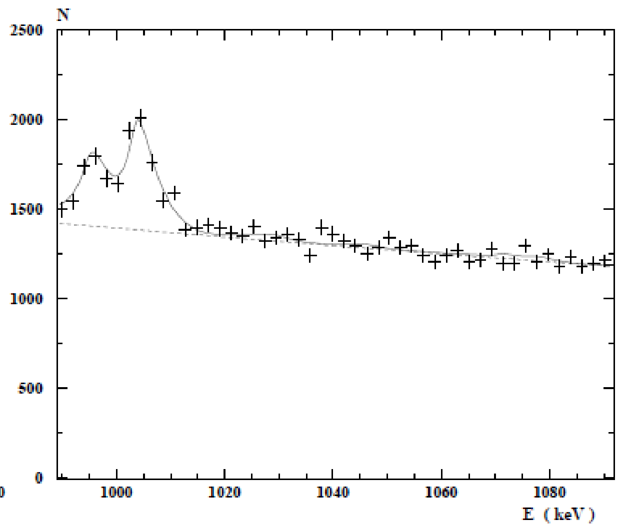


Figure 3 (left). The detector response function in the registered energy range of 1200÷1400 keV in the measurements with the source. The solid curve is the fit result (see the text). The dashed line is the fitted background.

Figure 4 (right). The same as Figure 3, but for the energy range of 990÷1092 keV.

The extracted values for the cumulative yield of ^{154}Eu (which for the case of this isotope practically coincides with its independent yield) multiplied by the factor $(1-\eta_{esc})$ turned out to be quite consistent for the all three energy ranges and for both measurement runs (for 256 and 160 hours). The value of $Y_c(1-\eta_{esc})$ averaged over these six independent estimations is equal to $Y_c(1-\eta_{esc})=(1.84\pm0.13)\cdot10^{-2}$. What concerns the measurements with the container, for none of aforementioned lines a significant photopeak was observed in the response function; for the averaged value of $Y_c\,\eta_{esc}$ the following restriction was inferred: $Y_c\,\eta_{esc}=(0.5\pm1.0)\cdot10^{-4}$, the upper limit within two standard deviations being $2.5\cdot10^{-4}$. The corresponding restriction for the escaping probability for the fission fragment ^{154}Eu is $\eta_{esc} < 0.013$, much smaller than the value of $\eta_{esc} = 0.29\pm0.03$ for A=137 isobars. The stopping range of a ^{154}Eu ion from the ^{252}Cf spontaneous fission is expected to be by about 1.5 times smaller than that for ^{137}I and ^{137}Xe, the main precursors of ^{137}Cs (see [10] for review). The large difference between escaping probabilities can be realized under assumption that the mean potential (geometrical) path of the fission fragments in the source volume exceeds sufficiently the range of ^{154}Eu, being, however, simultaneously compatible with those for ^{137}I and ^{137}Xe, allowing a fraction of them (about 30%) to escape from the source surface. Unfortunately, the luck of information about the constructive features of the source used in this study doesn't allow us to make quantitative estimations. It should be also pointed out, that the inferred yield $Y_c = (1.84\pm0.17)\cdot10^{-2}$ of ^{154}Eu (where the quoted error also includes the uncertainty in the efficiency ε_γ) exceeds by about four orders of magnitude the predicted value of $2.58\cdot10^{-6}$ [2]. Further studies are needed to realize the reason of this discrepancy. Unlike ^{137}Cs and ^{154}Eu, no significant signals corresponding to the γ-lines of other long-living isotopes listed in Table 1 were visible in the detector response function. Nevertheless, we tried to describe the latter including into the fit procedure an additional fit parameter corresponding to the contribution from the each of long-living isotopes under consideration. The fitted contributions were averaged over all runs and relevant γ-lines and, finally, the upper limit of Y_c was estimated within two standard deviations from its estimated mean value. These upper limits are presented in Table 2. We have to point out that, unlike other cases, the value quoted for ^{22}Na is based only on the measurements with the container, because in the measurements with the source its 1274.5 keV line could not be identified due to its obscuring by the 1274.4 line of ^{154}Eu (see Table 1); in this case the upper limit of Y_c was inferred under assumption that the escaping probability η_{esc} is close to 1 for the lightest fission fragments. Our data on ^{22}Na and ^{60}Co complete to a some extent the rather scarce list for the upper limits of the rates of the ^{252}Cf cluster radioactivity including the following light nuclei (in the range of $20 \leq A \leq 60$): ^{28}Mg, ^{43}K [11] and ^{46}Ar, ^{48}Ca [12]. It should be also noted, that the γ-lines from almost all heavy nuclides listed in Tables 1 and 2 (except ^{146}Pm and ^{158}Tb) were observed in the recent study [13], where a 37.5 year old ^{252}Cf source was used. However, no data concerning their yields are quoted in [13].

In conclusion, first experimental restrictions are presented for a number of long-living heavy products of the ^{252}Cf spontaneous fission. While the inferred cumulative yield $(5.07\pm0.35)\cdot10^{-2}$ for ^{137}Cs agrees with available experimental data and the model prediction, that for ^{154}Eu, estimated to be $(1.84\pm0.17)\cdot10^{-2}$, exceeds by about four orders of magnitude the predicted value. New measurements are needed, especially with a very fresh ^{252}Cf sample, to ascertain whether it, due of the technology of its preparation, is initially contaminated by an amount (if any) of ^{154}Eu not related to the ^{252}Cf spontaneous fission. First experimental restrictions on the rates of the sodium (^{22}Na) and cobalt (^{60}Co) radioactivity of ^{252}Cf are obtained; the corresponding decay constants normalized to that for α-radioactivity are limited as $\lambda(^{22}\text{Na})/\lambda(\alpha) < 1.6\cdot10^{-5}$ and $\lambda(^{60}\text{Co})/\lambda(\alpha) < 3.5\cdot10^{-7}$.

Table 2. Experimental restrictions on the yields of long-living isotopes, compared with predicted yields (if any) [2].

Nuclide	Y_c (this work)	Y_c (predicted)
^{22}Na	$< 5.0\cdot10^{-4}$	–
^{60}Co	$< 1.1\cdot10^{-5}$	–
^{102m}Rh	$< 1.6\cdot10^{-4}$	–
^{108m}Ag	$< 2.5\cdot10^{-3}$	–
^{125}Sb	$< 6.3\cdot10^{-3}$	$1.76\cdot10^{-4}$
^{134}Cs	$< 2.2\cdot10^{-3}$	–
^{137}Cs	$(5.07\pm0.35)\cdot10^{-2}$	$5.05\cdot10^{-2}$
^{146}Pm	$< 2.2\cdot10^{-3}$	$1.59\cdot10^{-8}$
^{150}Eu	$< 1.6\cdot10^{-4}$	–
^{152}Eu	$< 1.0\cdot10^{-3}$	–
^{154}Eu	$(1.84\pm0.17)\cdot10^{-2}$	$2.58\cdot10^{-6}$
^{158}Tb	$< 4.6\cdot10^{-3}$	$1.33\cdot10^{-5}$

Acknowledgment. The work was supported by State Committee Science MES RA, in frame of the research project № 13-1C245. One of the authors (H.G.) is grateful to Amur Margaryan for useful discussions.

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