Precise measurements of the _{97}^{249}Bk ground state half-life and the β^{−}-decay end-point energy

J. Chen, I. Ahmad, J. P. Greene, and F. G. Kondev

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Precise measurements of the $^{249}_{97}$Bk ground state half-life and the β^--decay end-point energy

J. Chen, I. Ahmad, J.P. Greene, and F.G. Kondev

Argonne National Laboratory, Argonne, Illinois 60439, USA

Abstract

The half-life of the ^{249}Bk ground state was determined by means of γ-ray spectroscopy, following for 728 days the growth of its α-decaying daughter nuclide ^{249}Cf. Using a chemically-purified source containing ^{249}Bk and ^{137}Cs nuclides, γ-ray singles measurements were carried out using a 25% coaxial Ge detector. The areas of the strongest 333.37- and 388.17-keV γ-ray peaks, produced in the α decay of ^{249}Cf, and the 661.66-keV peak, produced in the β^- decay of ^{137}Cs, were determined. The measured activity of the latter was used to account for geometrical and dead-time corrections to the efficiency of the spectrometer, thus minimizing the systematic uncertainties associated with long-time, γ-ray counting measurements. Using the growth with time of the ratio of the 388.17- and 661.66-keV γ ray peaks, a value of $T_{1/2}$=327.2$\pm$0.3 d (the uncertainty quoted is one standard deviation, 1σ) for the half-life of the ^{249}Bk ground state was determined. The β^-−decay end point energy of ^{249}Bk was measured with a Passivated Implanted Planar Silicon detector to be 123.6$\pm$0.4 keV .

PACS numbers: 21.10.Tg, 23.40.-s, 23.60.+e, 27.90.+b

I. INTRODUCTION

The precise knowledge of the half-life of actinide nuclides is of importance in forensic applications, where one needs to determine the quantity of a particular isotope from nuclear spectroscopic data. The need for a more precise value for the half-life of the long-lived ^{249}Bk isotope is also motivated by recent fabrications of targets of this isotope that were used in experiments aimed at searching for new superheavy elements [1, 2].

The half-life of ^{249}Bk has been measured in the past by several investigators [3, 5, 7–10], as summarized in Table I. The most recent nuclear data evaluation [11] adopts a single value of 330±4 d [8–10], while the latest edition of the Table of Isotopes recommends 320±6 d [12], based on the earlier evaluation by Schmorak [13], where only the results of Refs. [5, 7] were considered, but those of Refs. [3, 8, 9] were ignored.

The partial decay scheme of ^{249}Bk is presented in Fig. 1. This radionuclide decays mainly by β^- particle emission directly to the ground state of ^{249}Cf with no emission of γ rays [11]. The ^{249}Cf daughter disintegrates by α-particle emission, with the favored α-decay branch populating the 388-keV level of the ^{245}Cm grand-daughter, which de-excites by intense 333.37- and 388.17-keV γ rays to the first-excited level and the ground state, respectively [14]. The ^{249}Bk isotope has also a very small α-decay branch of (1.37±0.10)×10^{-3} % per ^{249}Bk decay [15], which produces the 280.36-, 308.26- and 327.45-keV γ rays in the daughter ^{245}Am nuclide with intensities of 0.10±0.01%, 0.24±0.02% and 1.06±0.06% per ^{249}Bk α decay, respectively [15]. These weak γ rays are usually not observed in γ-ray spectroscopy measurements, because of the presence of strong γ rays produced in the α decay of ^{249}Cf, and therefore, they cannot be used in direct measurements of the half-life of ^{249}Bk. However, the growth of the ^{249}Cf daughter nuclide following the β^- decay of the ^{249}Bk parent can be measured by counting the strongest 333.37- and 388.17-keV γ rays and this can in turn be used to determine the half-life of ^{249}Bk. In the present study, we have used this approach to perform a new measurement of the ^{249}Bk half-life. To reproduce the exact geometry during a series of γ-ray spectroscopy measurements, where one needs to account for identical conditions during the operation of the spectrometer over a long time period, the ^{137}Cs ($T_{1/2}$ =30.08±0.09 y [16]) nuclide was mixed with the chemically-purified ^{249}Bk sample and their activities were measured simultaneously, a technique previously used in the case of ^{252}Es [17].

The β^--decay end-point energy of ^{249}Bk was measured in the past by several investigators [3–7], as indicated in Table I. In the recent Atomic Mass Evaluation (AME2012) [18] a value of 124.6±1.4 keV is recommended. It is based on the most-precisely known experimental results of 125±2 keV, measured by Vandenbosh *et al.* [6] using a magnetic spectrometer, and 123±3 keV, reported by Glazov *et al.* [7] using a scintillation detector. We have measured the β^--particle spectrum of ^{249}Bk with a 300-μm thick, high resolution Passivated Implanted Planar Silicon (PIPS) detector and determined the end point energy with a higher precision.

II. SOURCE PREPARATION

The ^{249}Bk material was produced at the HIFR reactor of the Oak Ridge National Laboratory and a chemically-purified sample was shipped to Argonne National Laboratory. Part of the ^{249}Bk material ($\sim$1.0 μg) was mixed with 0.10 μCi of ^{137}Cs and a small drop of this solution was placed in the center of a plastic disk, and dried. The resulting mixed source was then sealed with a piece of scotch tape and attached to an aluminum plate. It was used in γ-ray spectroscopy measurements to determine the half-life of ^{249}Bk. An open, thin source was also prepared by placing $\sim$0.2 μg ^{249}Bk in the center of a 1-mm thick glass disk, where it was dried. This sample was used in electron counting in order to measure the β^--decay end point energy of ^{249}Bk.

III. EXPERIMENTAL METHODS

A. γ-ray spectroscopy measurements

Singles γ-ray spectra of the mixed ^{249}Bk-^{137}Cs source were measured with a 25% coaxial Ge detector [full width of half maximum (FWHM) of 1.8 keV at 1332.5 keV] that was shielded from room background in a cask assembled from thick lead bricks. The source was positioned in a plastic holder approximately 10 cm in front of the detector. A set of 83 mg/cm^2 Ta, 0.44 g/cm^2 Cd, 0.23 g/cm^2 Cu and 0.22 g/cm^2 Al plates was interposed between the source and the detector in order to absorb the electrons and low-energy γ rays. Such an arrangement reduced possible γ-ray coincidence summing effects to an upper limit of approximately 0.1%. The γ-ray energy and efficiency calibrations were determined using

a multi-nuclide source that contained ^{57,60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{139}Ce, ^{203}Hg and ^{241}Am (produced and calibrated by the Eckert and Ziegler Company), as well as with mono-isotopic sources of ^{152}Eu and ^{182}Ta. A sample spectrum of the ^{249}Bk-^{249}Cf source, measured 182 days after the sample preparation, is shown in Fig. 2.

B. Electron spectroscopy

Singles electron spectrum of the open ^{249}Bk source was measured in a small chamber that was kept under a vacuum of $\sim 2\times10^{-3}$ *Torr* with a 25 mm^2 area PIPS detector, whose thickness of 300 μm stopped all β^- particles emitted in the decay of ^{249}Bk (E$_\beta$=124.6±1.4 keV [18]). The measurement was carried out only a few days after the source was prepared, so that a minuscule amount of the ^{249}Cf daughter nuclide was accumulated. The typical leakage current of the detector was approximately 1 nA and the energy resolution (FWHM) was 3.0 keV. The energy calibration was performed with an open, mass-separated ^{243}Cm source that has a number of well-known discrete conversion electron lines [19, 20]. The spectra were measured at a relatively small solid angle of 0.025%, thus minimizing the random summing between the β^- particles and conversion electrons.

IV. RESULTS AND DISCUSSION

A. ^{249}Bk half-life

Gamma-ray spectra from the mixed ^{249}Bk-^{137}Cs source were measured once a week during a time period of 728 days. The duration of each individual measurement was set to be the same and was equal to 62.5 hours. Background γ-ray spectra were also measured and no γ rays were found to interfere with the main peaks identified to belong to ^{249}Cf and ^{137}Cs decays. The data analysis was carried out using the $gf3$ program of the RADWARE spectroscopy package [21]. For each spectrum the areas of the 107.19-keV (Cm K$_{\alpha2}$) and 112.12-keV (Cm K$_{\alpha1}$) x rays, and 252.84-, 333.37-, and 388.17-keV (^{249}Cf) and 661.66-keV (^{137}Cs) γ-ray peaks, were determined in exactly the same way. The spectra were also found to contain a minuscule amount of Am K x rays, produced in the α decay of ^{249}Bk. It is worth noting that while the 252.84-keV γ ray is produced in the decays of both ^{249}Cf and ^{245} Am, the 333.37- and 388.17-keV γ rays are not produced in β^- decay of ^{245}Am, as indicated

in Fig. 1. Two approaches were used in the analysis of the spectra. In the first one, a fit to a particular γ-ray peak was carried out using a peak model comprising a Gaussian, a skewed Gaussian, a smoothed step function component that accounts for the increase the background on the low-energy side of the peak, and a linear background [21]. In the second one, the peak areas were determined by means of the *sb* command of the $gf3$ program, where the counts between the left- and right-side area points for a given peak were summed, after subtraction of a linear background. We found that the areas of the 333.37-, 388.17-, and 661.66-keV γ-ray peaks that were determined by the two methods gave essentially the same value for the ^{249}Bk half-life within the statistical uncertainties.

The ratios of the 333.37- and 388.17-keV photopeak areas, S_D, to that of the 661.66-keV area, S_{Cs}, were fitted as a function of time with the well-known growth equation [22]:

$$R(t) = \frac{S_D(t)}{S_{Cs}(t)} = \frac{\epsilon_D \cdot \lambda_D \cdot N_D(t)}{\epsilon_{Cs} \cdot \lambda_{Cs} \cdot N_{Cs}(t)} = \frac{\lambda_D}{\lambda_{Cs}} \cdot \Big[A \cdot \frac{\lambda_P}{(\lambda_P - \lambda_D)} \cdot \Big(e^{-\lambda_D t} + e^{-\lambda_P t}\Big) + B\Big] \cdot e^{\lambda_{Cs} t}, \quad (1)$$

where $\lambda = \frac{ln\,2}{T_{1/2}}$ is the decay constant (subscripts P, D and Cs indicate the parent (^{249}Bk), daughter (^{249}Cf), and ^{137}Cs nuclei, respectively), $\epsilon = \epsilon_{intr} \times \epsilon_{geom} \times \epsilon_{dT}$ is the γ-ray efficiency that includes intrinsic (ϵ_{intr}), geometrical (ϵ_{geom}) and dead-time (ϵ_{dT}) contributions and A, and B are fitted constants. Values of $T_{1/2}$(D) = 351$\pm$2 y [11] and $T_{1/2}$(Cs)=30.08$\pm$0.09 y [16] were used for the ^{249}Cf and ^{137}Cs half-lives in eqn.(1), respectively. The growth curve and fit for the 388.17/661.66 pair is displayed in Fig. 3, and the analysis gave a value of $T_{1/2}$=327.2 $\pm$0.3 d for the ^{249}Bk half-life. The uncertainty given is one standard deviation (1σ). The lower part of the figure, showing the differences between the data points and the values of the fit relative to the uncertainties of the data, illustrates that the consistency between the fit and the data is within 2σ that corresponds to a deviation of about 0.2%. The fit to the 333.37/661.66 ratios gave a similar value (within the deduced uncertainties) for the half-life of ^{249}Bk.

The main advantage of the method employed here is that it allowed to account for several systematic uncertainties associated with long-counting-time γ-ray spectrometry measurements. For example, the ratios of the ^{249}Cf peaks to that of the ^{137}Cs one, automatically cancels any uncertainties due to small changes in the geometry of the measurements, as well as those associated with dead times of the data acquisition system, owing to the growth of the ^{249}Cf atoms. Changes in the intrinsic efficiency of the spectrometer were also found to be negligible. These were monitored internally by following the ratios of the 92.51- and 388.17-

keV γ rays, produced in the α decay of ^{249}Cf, which were found to be constant over the entire 728 days counting period. One should also note that there are no γ rays in coincidence with the 388.17-keV γ ray and, hence, there is no loss due to coincidence summing.

The present value for the ^{249}Bk half-life in the present work is somewhat shorter compared to that determined in the β^- activity measurements by Polyukhov *et al.* [8–10]. One should note, however, that the growth of the ^{249}Cf with time could affect the observed β^--decay counts, since a number of conversion electron lines associated with the decay of the longer-lived ^{249}Cf isotope ($T_{1/2}$ = 351±2 y [11]) would contribute to the observed activity. No such corrections were applied in the long-time measurements reported in Refs. [8–10].

B. ^{249}Bk β^--decay end-point energy

A β^--particle spectrum of ^{249}Bk collected with the PIPS detector is shown in Fig. 4. A least-squares linear fit to the Fermi-Kurie spectrum [23], shown in the insert of Fig. 4, gave an end point energy of 123.6±0.4 keV. This value is in good agreement with that of 123±3 keV measured by Glazov *et al.* [7] and 125±2 keV measured by Vandenbosch *et al.* [6], but our uncertainty is much smaller. It is worth pointing out that AME2012 [18] recommends 124.6±1.4 keV, an average of values in Refs. [6, 7]. Given the recently measured mass of ^{249}Cf, $ME(^{249}\mathrm{Cf})$=69718.1±1.3 keV [24] and the presently determined value of $Q_\beta(^{249}\mathrm{Bk})$=123.6±0.4 keV, the mass of ^{249}Bk can be obtained as $ME(\ ^{249}\mathrm{Bk})$=69841.7±1.4 keV, which is 8.9 keV lower than the recommended value in AME2012 [18]. Such a discrepancy can be attributed to inconsistencies in the available decay data, as discussed in Refs. [15, 24].

Acknowledgments

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TABLE I: Sumary of previous measurements of the ^{249}Bk half-life and the β^--decay end-point energy.

Half-life (days)	Method	Decay followed	$E_{\beta,max}$ (keV)	Method	Reference
290±20	β^- activity	∼140 days	80±20	absorption	[3]
∼365	β^- activity		100±20	absorption	[4]
314 ±8	β^- activity	∼1 year	114±15	absorption	[5]
			125±2	Fermi-Kurie	[6]
325±7	β^- activity	∼1 year	123±3	Fermi-Kurie	[7]
329±4*	β^- activity	∼600 days			[8]
330±4*	β^- activity	∼1181 days			[9, 10]

* The quoted uncertainty is two standard deviations (2σ).

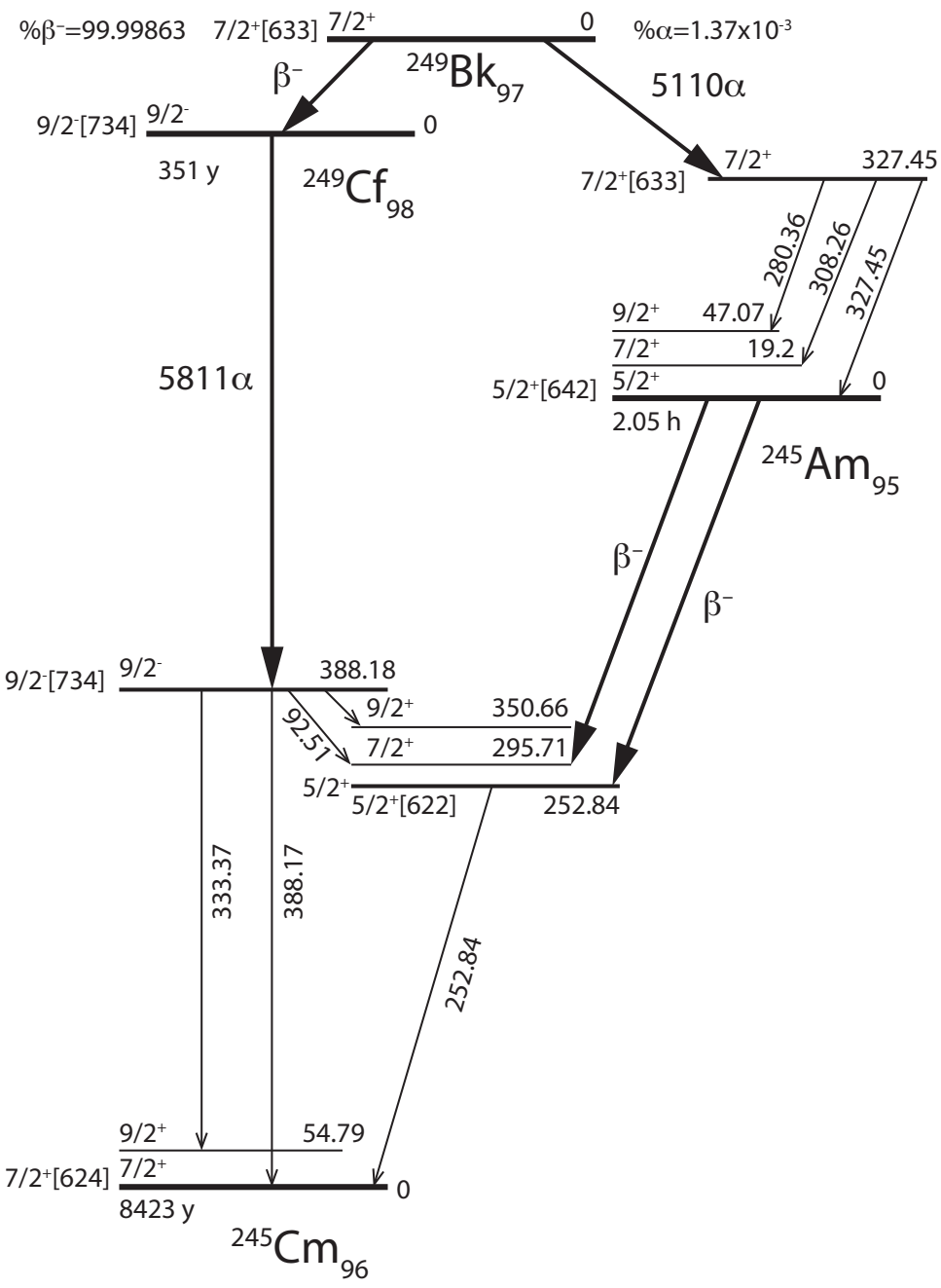

FIG. 1: Partial decay scheme of ^{249}Bk and its daughters from Refs. [11, 14, 15]. Only the main decays discussed in the present work are included.

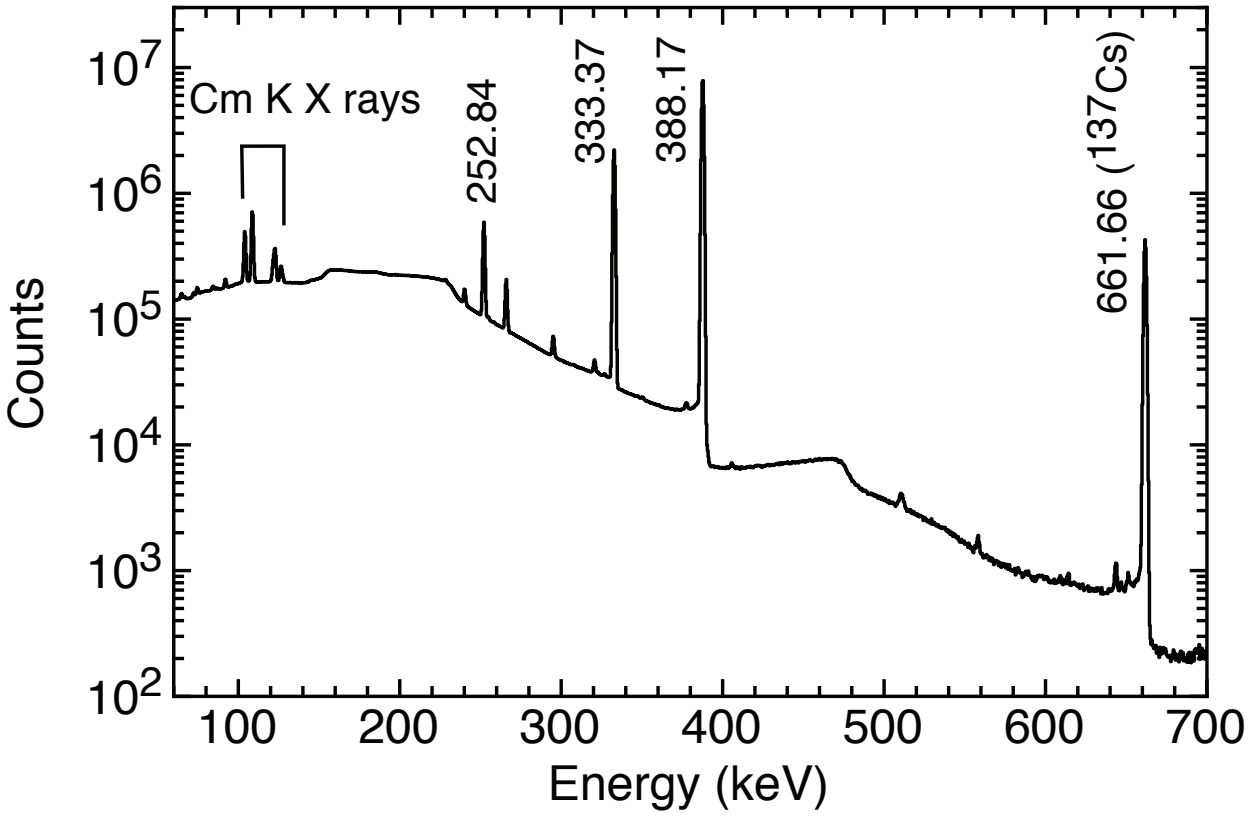

FIG. 2: Gamma-ray spectrum of the mixed ^{249}Bk-^{137}Cs source measured with a 25% coaxial Ge detector 182 days after the source was prepared. Only strong γ rays associated with the α decay of ^{249}Cf are labelled with their energies in keV, as well as the 661.66-keV γ ray produced in the β^- decay of ^{137}Cs; weak peaks are identified to belong to the ^{249}Cf α decay.

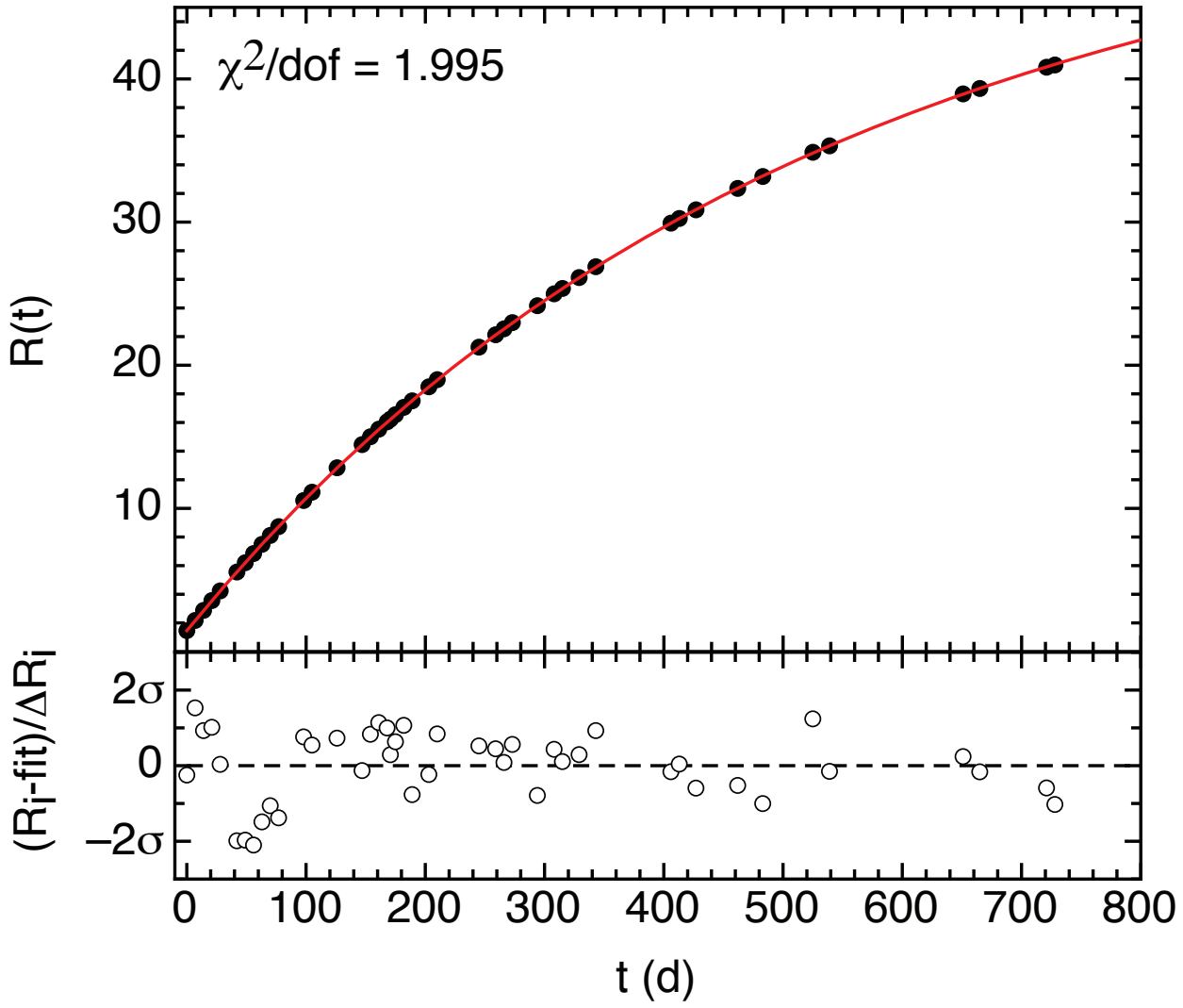

FIG. 3: Growth curve for the ratio of counts in the 388.17-keV peak of ^{249}Cf to the counts in the 661.66-keV peak of ^{137}Cs. The solid line is the best fit, from which the half-life of ^{249}Bk is derived to be $T_{1/2}$=327.2 ± 0.3 days. The lower part in the figure shows the differences between the data points and the values of the fit relative to the uncertainties of the data with $\sigma=\Delta R_i$.

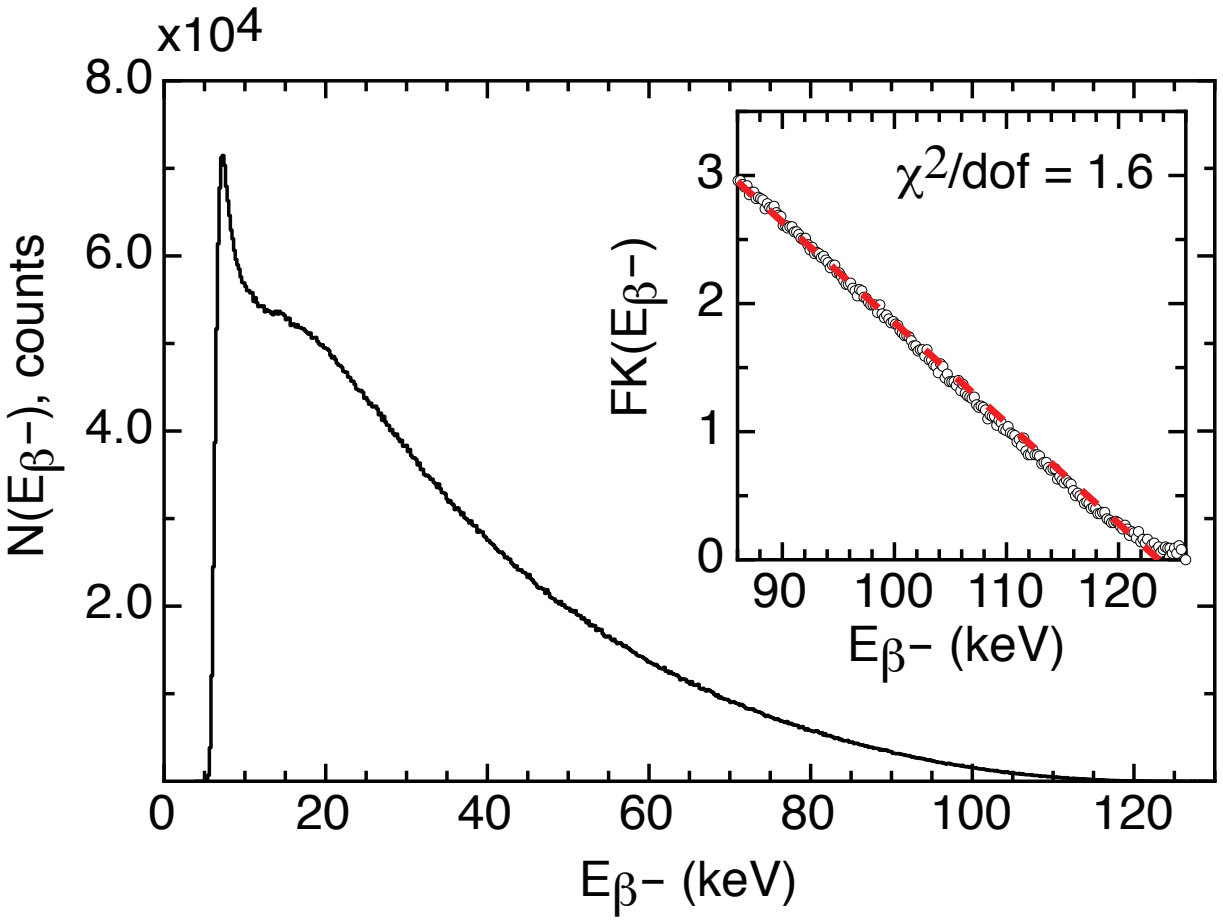

FIG. 4: A β^--particle spectrum of ^{249}Bk collected with the 300-μm thick PIPS detector. The insert shows the Fermi-Kurie (FK) plot, with the dashed line representing the result from a linear least-squares fit which gave an end point energy of 123.6 $\pm$ 0.4 keV.