

NUCLEAR SPECTROSCOPIC STUDIES OF THE 23.7 h $^{248}\text{Bk}^\dagger$

H. C. GRIFFIN

Department of Chemistry, University of Michigan, Ann Arbor, Michigan 48109

and

I. AHMAD, A. M. FRIEDMAN and L. E. GLENDENIN

Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439

Received 20 March 1978

Abstract: Main features of the decay of the 23.7 h ^{248}Bk have been determined by spectroscopy with scintillation and semiconductor detectors. The results are as follows: half-life 23.7 ± 0.2 h; branching is $(30 \pm 5)\%$ to ^{248}Cm ($\approx 23\%$ EC decay to 0^+ ground state and $\approx 7\%$ to first 2^+ state) and $(70 \pm 5)\%$ β^- decay to ^{248}Cf (5% to 592 keV $K^* = 2^-$ state and $(65 \pm 5)\%$ to the ground-state band). The β^- decay energy, Q_{β^-} , has been measured to be 860 ± 20 keV, and the electron capture decay energy, Q_{EC} , has been derived from closed cycle to be 705 ± 25 keV. The deduced $\log ft$ values of β^- and EC transitions restrict the spin of the ^{248}Bk ground state to 1, with configuration assignment $\{n[734]_{\frac{3}{2}}^-; p[633]_{\frac{1}{2}}^+\}$. It has also been deduced that the long lived isomer lies 65 ± 40 keV above the 23.7h ^{248}Bk ground state.

E

RADIOACTIVITY ^{248}Bk [from $^{248}\text{Cm}(d, 2n)$, $^{247}\text{Bk}(n, \gamma)$]; measured $T_{1/2}$, E_γ , I_γ , L X-ray-L X-ray, K_α -L X-ray, K_β -L X-ray, β^- - γ , β^- -L X-ray coin; deduced $\log ft$, Q_{β^-} , Q_{EC} , decay branching. ^{248}Cm , ^{248}Cf deduced levels, γ -multipolarity, J , π . Mass separated ^{248}Bk .

1. Introduction

Two isomers of ^{248}Bk are known. The short-lived ^{248}Bk isomer was first synthesized by Hulet ¹⁾ who deduced a half-life of 23 ± 5 h by milking the daughter ^{248}Cf from berkelium produced in a helium-ion bombardment of mixed curium isotopes. Chetham-Strode ²⁾ reported production of ^{248}Bk by the $^{247}\text{Bk}(n, \gamma)$ reaction; his characterization included half-life (16 ± 3 h), Q_{β^-} (650 ± 50 keV), and branching ratios ($\beta^-/\text{EC} = 2.4$, $< 6\%$ β^- intensity to the 42 keV 2^+ state of ^{248}Cf). A long-lived ^{248}Bk isomer was identified ³⁾ by mass spectroscopy but its decay could not be detected. A lower limit of 800 y was deduced for its α -decay. This isomer is also produced ⁴⁾ in the α -decay of ^{252}Es .

Schmorak ⁵⁾ has pointed out inconsistencies between the characteristics inferred by Chetham-Strode ²⁾ and those obtained from systematics (energies and single-

[†] Work performed under the auspices of the Division of Basic Energy Sciences of the Department of Energy.

particle state assignments) in this region. The results of the present investigation remove these inconsistencies.

2. Source preparation

Most of the measurements were made with four sources of ^{248}Bk prepared by the $^{247}\text{Bk}(n, \gamma)$ reaction. The target material consisted of a portion of the long-lived berkelium prepared by Milsted *et al.* ³⁾ containing $\approx 10^{11}$ atoms of ^{247}Bk . After irradiation the berkelium fraction was isolated and purified by extraction chromatography. Final samples had strengths of a few hundred disintegrations per sec. These yields indicate that the cross section for the $^{247}\text{Bk}(n, \gamma)$ reaction is of the order of 10^3 b.

One ^{248}Bk sample (used for obtaining Ge(Li) spectra) was produced by the irradiation of ≈ 2 mg of ^{248}Cm with 20 MeV deuterons in the Argonne 152 cm cyclotron. The beam current density was $30 \mu\text{A}/\text{cm}^2$ and the bombardment time was 16 h. The Cm target was dissolved in 10 M HNO_3 , 0.1 M KBrO_3 solution and the Bk was extracted ⁶⁾ three successive times with equal volumes of 0.14 F di-(2-ethylhexyl) orthophosphoric acid in n-heptane. The Bk was recovered from the organic phase by contact with a 10 M HNO_3 , 0.1 M H_2O_2 solution. Fission products were removed by an extraction chromatographic procedure ⁷⁾. The chemically purified Bk was run through the Argonne electromagnetic isotope separator ⁸⁾ to prepare a mass-separated ^{248}Bk sample. The sample contained $\approx 10^5$ disintegrations per minute of 23.7 h ^{248}Bk .

3. Experimental procedures and results

The decay characteristics of ^{248}Bk were determined from a variety of measurements, some of which were intended only for qualitative characterizations (such as detecting coincidences without measuring absolute intensities). However, even the qualitative measurements indicated significant features of the decay which do not agree with the conclusions of Chetham-Strode ²⁾. In particular, we found a longer half-life, a 550.7 keV transition in ^{248}Cf , and substantial intensity of L-converted transitions in both ^{248}Cm and ^{248}Cf . The half-life was determined both by β^- counting and by following the decay of the 550.7 keV γ -ray. Assignment of the γ -ray as a transition in ^{248}Cf was based on β^- - γ coincidences. Population of L-converted transitions was detected by K_β -L X-ray and β -L X-ray coincidence experiments.

The individual measurements which bear most directly on the quantitative conclusions are described in the following subsections and are summarized in sect. 4.

3.1. GAMMA-RAY SPECTROSCOPY

The γ -ray spectrum of a mass-separated ^{248}Bk sample was measured with a 25 cm^3 Ge(Li) spectrometer. Portions of the spectrum are shown in fig. 1; energies, intensities

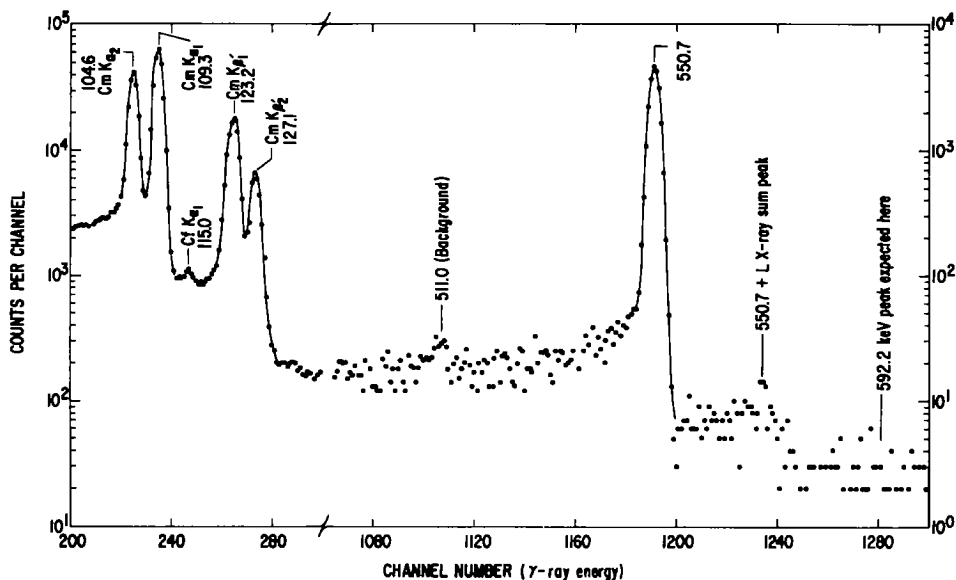


Fig. 1. Gamma-ray spectrum of a mass-separated ^{248}Bk sample measured with a 25 cm^3 Ge(Li) spectrometer. The source was placed ≈ 4 cm away from the detector end cap and the counting time was 5.0 h.

TABLE 1

K X-rays and γ -rays associated with the decay of the 23.7h ^{248}Bk

Energy (keV)	Relative intensity ^{a)}	Transition
104.6 ± 0.1	65 ± 3	Cm K_{α_2}
109.3 ± 0.1	100 (norm) ^{b)}	Cm K_{α_1}
115.0 ± 0.3	0.28 ± 0.04	Cf K_{α_1}
123.2 ± 0.2	39.0 ± 3.0	Cm K_{β_1}
127.1 ± 0.2	13.0 ± 1.6	Cm K_{β_2}
550.7 ± 0.1	51 ± 3	$592.2 \rightarrow 41.5$

^{a)} The absolute intensity of the 550.7 keV γ -ray was measured to be $5.0 \pm 0.4\%$ per 23.7 h ^{248}Bk decay.

^{b)} The total Cm K X-ray and Cf K X-ray intensities are 217 and 0.6, respectively.

and assignments are summarized in table 1. In addition to Cm K X-rays which arise from K-capture, a Cf K_{α_1} X-ray peak and a 550.7 keV γ -ray are present. The energy of the γ -ray was measured with respect to the 569.69 keV γ -ray of ^{207}Bi ; the source and the standard were counted simultaneously. We assume that since no other γ -transition in ^{248}Cf with energy greater than the K-edge has been observed, all the Cf K X-ray intensity must originate from the shake-off during β^- decay and internal conversion of the 550.7 keV transition. Using the $K_{\text{total}}/K_{\alpha_1}$ ratio ⁹⁾ of 2.15, K-fluorescence yield ¹⁰⁾ of 0.972 and a K/β^- shake-off yield ¹¹⁾ of 2×10^{-4} , we have

calculated the K-conversion coefficient of the 550.7 keV transition as 0.009 ± 0.002 . This is in good agreement with the theoretical ¹²⁾ value of 0.011 for an E1 transition. Theoretical values for a 550.7 keV M1 and E2 transition are 0.28 and 0.031, respectively.

The data in table 1 indicate that the ratio between the Cm K X-ray intensity and 550.7 keV photon intensity is 4.25 ± 0.28 . Similar measurements with scintillation detectors yielded a value of 4.0 with a statistical error of 0.1. Consideration of possible systematic error leads to an average value of 4.15 ± 0.20 . For an allowed or a first-forbidden non-unique EC transition with $Q_{EC} = 705$ keV we expect ¹³⁾ intensities of 0.74, 0.18, 0.02 and 0.06 for K, L_I, L_{II} and (M+N+...) capture, respectively. Using the above numbers we obtain the ratio of the EC decay to the 550.7 keV γ -emission as 5.79 ± 0.28 .

The β^- disintegration rate of one ²⁴⁸Bk sample was determined by two methods. First, the sample was counted in a 2π geometry gas-flow counter at various discriminator settings. The counts corresponding to energies above ≈ 40 keV were extrapolated to zero discriminator setting to give the total β^- disintegration rate. Second, the residual ²⁴⁸Cf resulting from ²⁴⁸Bk decay was assayed by α -particle spectroscopy. Although the direct β^- measurements had negligible statistical errors, the α -counts in the ²⁴⁸Cf peaks were free from any systematic errors. The γ -emission rate of the sample was determined with a NaI(Tl) spectrometer of well-known detector characteristics. The best value of the 550.7 keV γ/β^- intensity ratio, obtained from several measurements, is 0.071 ± 0.005 .

The decay of the 550.7 keV photopeak in a spectrum measured with the 25 cm³ Ge(Li) spectrometer was followed for 6 days. A weighted least-squares analysis of the peak areas gave a half-life of 23.5 ± 0.2 h. A value of 23.99 ± 0.03 h was obtained from β^- counting, but it is difficult to evaluate systematic errors in this measurement. We conclude the best value for the ²⁴⁸Bk half-life is 23.7 ± 0.2 h.

3.2. COINCIDENCE MEASUREMENTS

The relative intensities of electron capture transitions to the 0⁺ ground state and 2⁺ first excited state of ²⁴⁸Cm were determined from coincidences among Cm X-rays. The spectra displayed in fig. 2 were measured with two 7.6 cm \times 7.6 cm NaI(Tl) detectors. Coincidence correlations were obtained with a two-parameter analyzer in which digital gates were set on one of the parameters. In principle the desired branching ratio can be obtained from either the K _{β} -L X-ray coincidence intensity or the L X-ray versus L X-ray coincidence intensity. Although both coincidence spectra were analyzed, the latter is less sensitive to uncertainties in peak shapes.

The L X-ray versus L X-ray coincidences arise from two types of events — L-capture to a level which deexcites predominantly by L-converted transitions and K-capture (only K _{α} emission is accompanied by L-shell vacancies) to a similar level. In order to avoid dependence on absolute geometries and the efficiency of the coincidence

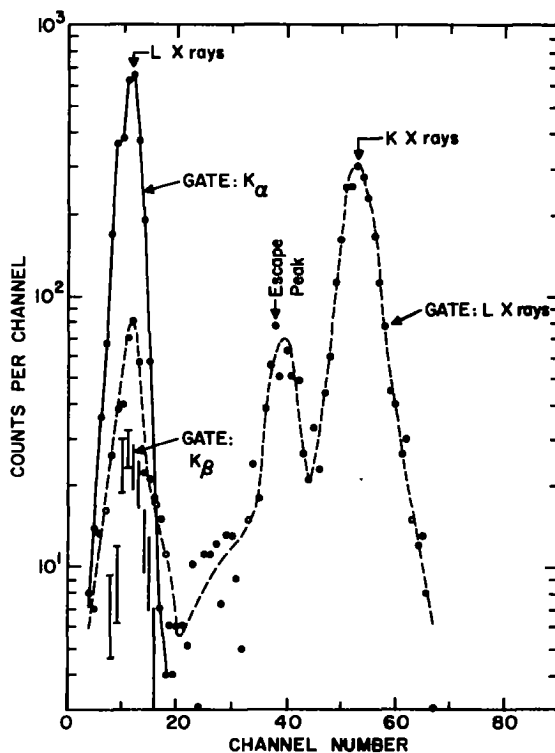


Fig. 2. The ^{248}Bk γ -ray spectra measured with a 7.6 cm \times 7.6 cm NaI(Tl) detector in coincidence with Cm K_α (denoted by \bullet), Cm K_β (error bars) and (Cm+Cf) L X-rays (\circ). The first two spectra have 0 to 1 count per channel beyond L X-ray region.

circuit, the L X-ray-L X-ray coincidence spectrum was compared with the L X-ray spectrum gated by the K_α peak. The latter arises primarily from atomic processes following K-capture. In the analysis we have used a value of 0.54 ± 0.01 for the average L-fluorescence yield and assumed that it is relatively insensitive to the distribution of primary vacancies among L-subshells ¹⁴). Experimental values of fluorescence yields in Cm are 0.28, 0.55 and 0.63 for the L_I , L_{II} and L_{III} subshells, respectively. Theoretical values of conversion coefficients and capture intensities from various shells were also used in these analyses. The analysis of the L X-ray-L X-ray coincidence data indicates that $(18 \pm 2)\%$ of EC decays populate the 2^+ level in ^{248}Cm . Analysis of the K_β -L X-ray coincidence spectrum gives a value of $(35 \pm 10)\%$ for this intensity. From the weighted average we obtain a value of $(22 \pm 5)\%$ for the EC intensity at 2^+ state.

The data bearing on the β^- branching to the 0^+ and 2^+ levels in ^{248}Cm are rather qualitative. A comparison of the ^{248}Bk L X-ray singles spectrum (fig. 3) with Cm L X-ray spectrum in coincidence with K X-ray and Cf L X-ray spectrum gated by β^- particles indicates that the contributions from the two elements are almost equal. The end point of the β^- spectrum (fig. 4) measured in coincidence with L X-rays

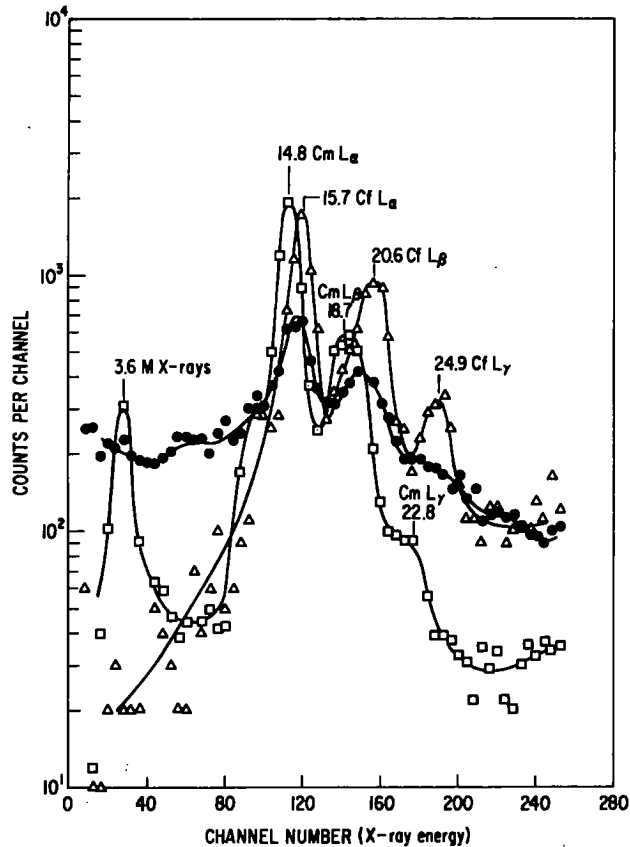


Fig. 3. The ^{248}Bk L X-ray spectra measured with a proportional counter. Spectrum denoted by \bullet represents singles spectrum and the spectra denoted by \square and \triangle were obtained in coincidence with Cm K X-rays and β^- particles, respectively.

suggests that the L-converted transitions in ^{248}Cf are fed predominantly by high energy β^- transitions, from which we infer that the Cf L X-rays originate from the 2^+ level in ^{248}Cf . From a quantitative analysis of coincident β^- spectra we find that the total intensity of the β^- group feeding the 2^+ level is at least three times that attributed to the 550.7 keV γ -ray intensity but less than one-half the total β^- intensity.

3.3. THE β^- SPECTROSCOPY

The β^- spectra, more quantitative than those given in fig. 4, were obtained to determine Q_{β^-} for ^{248}Bk decay and to confirm the assignment of the 550.7 keV γ -ray. The relevant spectra are displayed in fig. 5. The Fermi-Kurie plot of the ^{248}Bk singles spectrum measured with a Si(Li) detector (curve A) shows significant curvature which we assume to arise from edge effects in and scattering from the detector. The energy

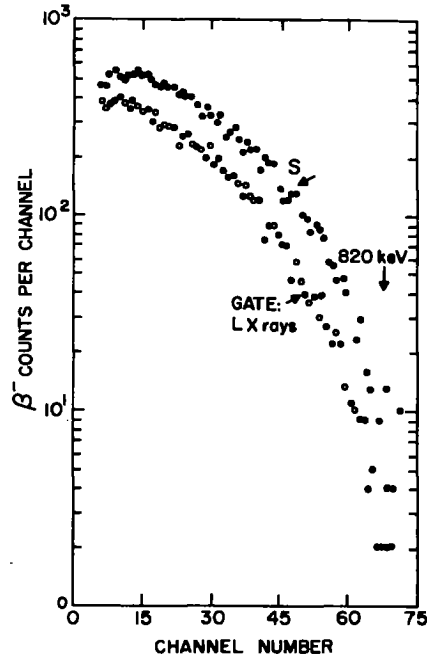


Fig. 4. The ^{248}Bk β^- spectrum measured with a Pilot B scintillator in coincidence with Cm and Cf L X-rays. The β^- singles spectrum (S) is shown for comparison.

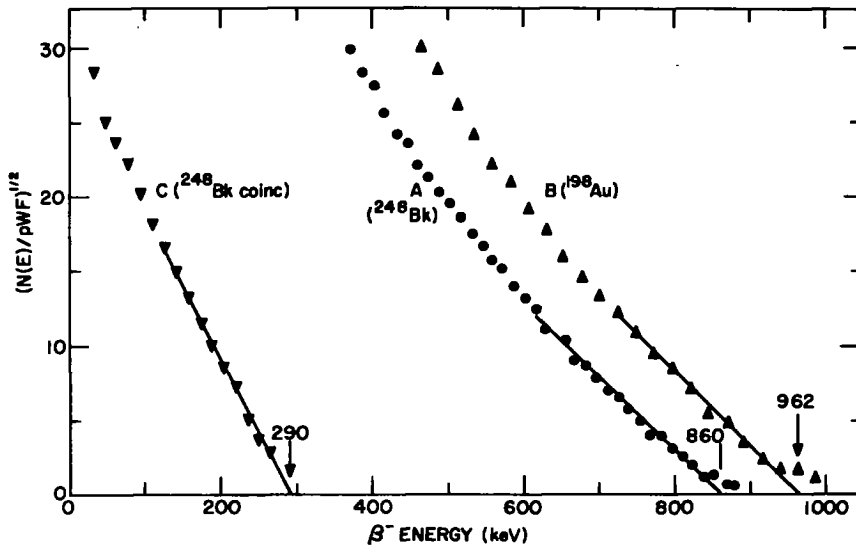


Fig. 5. Fermi-Kurie plot of a ^{248}Bk β^- singles spectrum measured with a Si(Li) detector. A ^{198}Au spectrum measured with the same detector is shown for comparison. The β^- spectrum C was obtained with a liquid scintillator in coincidence with the 550.7 keV γ -ray.

calibration was obtained with ^{207}Bi conversion electron lines. A ^{198}Au spectrum was used to establish the linear range of the Fermi-Kurie plot. Since the energy range used for the extrapolation is large compared with the 41.5 keV energy difference between the 0^+ and 2^+ states of ^{248}Cf , the observed end point can be interpreted as a weighted average of the true end points of the transitions to these two states. These data yield a value of 870 ± 20 keV for Q_{β^-} .

For the measurement of β^- transitions leading to the 550.7 keV γ -ray, the ^{248}Bk sample was dissolved in a liquid scintillator and γ -rays were detected with a NaI(Tl) detector. The energy calibration and the resolution response function of the liquid scintillator were measured with several conversion-electron sources. The Fermi-Kurie plot of the resolution-corrected spectrum is shown as curve C of fig. 5. The observed end-point energy of 290 keV is interpreted as a sum of the true β^- end-point energy and the energies of the internal conversion electrons and X-rays from the $2^+ \rightarrow 0^+$ transition in ^{248}Cf . The analysis of curve C gives a true end-point energy of 257 keV and the ground-state β^- decay energy of 849 keV ($257 + 592$). The above two measurements of Q_{β^-} agree within the uncertainties assigned to them and yield the average value of 860 ± 20 keV. The measured end-point energy of the β^- spectrum in coincidence with Cf L X-rays and the fact that the 550.7 keV γ -ray is in coincidence with Cf L X-rays strongly suggest that the 550.7 keV γ -transition terminates at the 41.5 keV 2^+ state of ^{248}Cf .

4. Discussion

The decay scheme of the 23.7 h ^{248}Bk deduced from the present investigation is shown in fig. 6. The characteristics of the ground-state bands of ^{248}Cm and ^{248}Cf are known from α -decay studies^{5, 15}) of ^{252}Cf and ^{252}Fm . A comparison of EC and β^- intensities to the 550.7 keV γ -intensity leads to absolute intensities of $(5.0 \pm 0.4)\%$, $(70 \pm 5)\%$ and $(30 \pm 5)\%$ for the 550.7 keV γ , total β^- and total EC transition, respectively. Measurements of the branching in EC decay give $(7 \pm 2)\%$ intensity to the 2^+ state and $(23 \pm 4)\%$ to the ^{248}Cm ground state. A similar analysis gave β^- intensity of $(45 \pm 10)\%$ and $(20 \pm 10)\%$ to the 0^+ and 2^+ members of ground-state band; the total intensity to the ground-state band is $(65 \pm 5)\%$.

The β^- spectral measurements and γ versus L X-ray coincidences indicate that the 550.7 keV E1 transition deexcites a 592.2 keV level to the 41.5 keV level. The absence of parallel transitions ($< \frac{1}{300}$ of the 550.7 keV γ -intensity) implies that the 592.2 keV state has $I^\pi = 2^-$. The comparative half-lives¹⁶) for EC and β^- transitions constrain the ground state of ^{248}Bk to $I = 1$. The configuration for ^{248}Bk ground state can be deduced from the known single-particle orbitals in the neighboring odd-mass nuclei. The ground state of ^{247}Cm is known¹⁷) to be the $\frac{5}{2}^- [734]$ neutron orbital and that of ^{247}Bk is the $\frac{3}{2}^- [521]$ proton state with the $\frac{7}{2}^+ [633]$ orbital lying only 41 keV away¹⁸). From the coupling of the above neutron and proton orbitals only one $I = 1$ state is formed. We, therefore, assign the ^{248}Bk ground state to the $\{n[734]\frac{5}{2}^- ; p[633]\frac{7}{2}^+\}$ configuration.

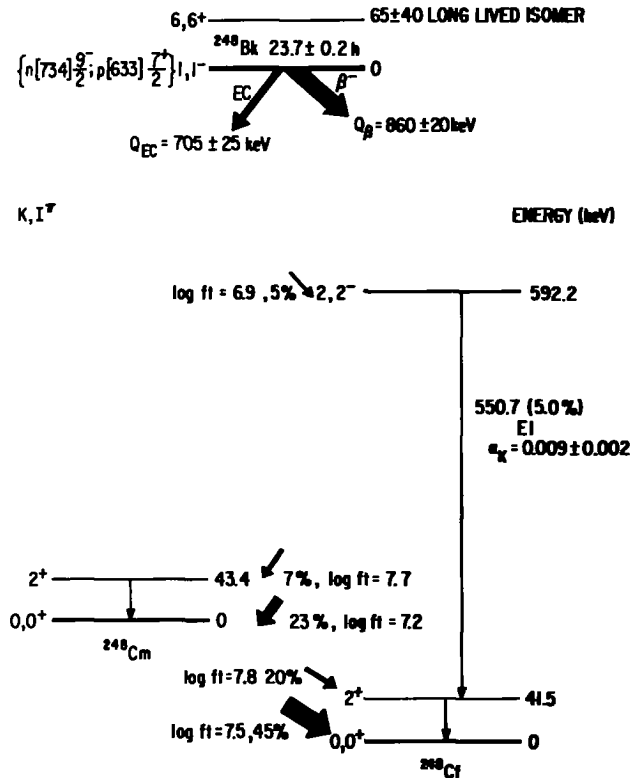


Fig. 6. Decay scheme of the 23.7h ^{248}Bk deduced from the present work.

Because of its low energy we interpret the 592.2 keV state as a $K^\pi = 2^-$ octupole vibrational state. It should be pointed out that as such it would be the lowest 2^- octupole vibration known in the actinide region. This state was also observed in the $^{249}\text{Cf}(d, t)$ reaction ¹⁹⁾ and it was given the same assignment. It was shown that this $K^\pi = 2^-$ band contains a large contribution from the $\{n[734]_{\frac{9}{2}}^-; n[622]_{\frac{5}{2}}^+\}$ configuration. With this assignment the β^- transition to the 592.2 keV state is equivalent to a transition between $p[633]_{\frac{7}{2}}^+$ and $n[622]_{\frac{5}{2}}^+$ orbitals in the neighboring odd-mass nuclei. The $\log ft$ value of EC transition involving these two states in ^{249}Es decay ⁹⁾ is determined to be 7.3, which is close to the value of 6.8 observed in ^{248}Bk β^- decay. The small difference between the two values could be accounted for in terms of pair occupation probabilities of orbitals involved in the transitions.

The β^- decay energy of 860 ± 20 keV combined with the ^{248}Cm - ^{248}Cf mass difference of 154 ± 10 keV yields a value of Q_{EC} as 705 ± 25 keV for ^{248}Bk EC decay. The Cm-Cf mass difference was derived from $E_{\alpha_0} (^{248}\text{Cf}) = 6262 \pm 5$ keV [ref. ¹⁵⁾], $Q_{\alpha} (^{248}\text{Cm}) = 5161 \pm 5$ keV [ref. ⁵⁾] and the ^{244}Pu - ^{244}Cm mass difference of 1358 ± 7 keV [ref. ⁵⁾]. A similar analysis based on $Q_{\alpha} (^{252}\text{Es} \text{ to long-lived } ^{248}\text{Bk}) = 6738 \pm 3$

keV [ref. 4)], $Q_{EC}(^{252}\text{ES}) = 1290 \pm 30$ keV [ref. 4)] and $Q_{\alpha}(^{252}\text{Cf}) = 6217 \pm 1$ keV [ref. 5)] indicates that Q_{EC} for the long-lived ^{248}Bk is 770 ± 30 keV. Therefore the long-lived ^{248}Bk isomer lies 65 ± 40 keV above the 23.7 h ^{248}Bk ground state. This energy difference is comparable to the error but it is much smaller than the difference implied by the value of Q_{β^-} reported by Chetham-Strode²⁾. Other evidence indicates that the excitation energy of the long-lived isomer is unlikely to be as much as 100 keV. Milsted *et al.*³⁾ obtained a lower limit of 10^{11} s for the partial half-life of the long-lived ^{248}Bk isomer for the production of ^{245}Cf . Fields *et al.*⁴⁾ obtained $I, K^{\pi} = 6, 6^{+}$ for the long-lived isomer. Therefore ^{248}Cf production could arise either by direct β^{-} decay of long-lived isomer or by a E5 isomeric transition to the 23.7 h ^{248}Bk followed by β^{-} decay to ^{248}Cf . The half-life of a 100 keV E5 transition is found to be substantially less than 10^{11} s on the basis of single-particle estimate¹³⁾. This half-life argument suggests that the energy difference between the ground states of the two isomers is less than 100 keV.

The authors thank J. Lerner for the Isotope Separator preparation of the ^{248}Bk sample and H. Diamond, K. F. Flynn and R. K. Sjoblom for their help in chemical purifications and activity determination.

References

- 1) E. K. Hulet, *Phys. Rev.* **102** (1956) 182
- 2) A. Chetham-Strode, Jr., Lawrence Radiation Lab. Report UCRL-3322, Ph.D. thesis (1956)
- 3) J. Milsted, A. M. Friedman and C. M. Stevens, *Nucl. Phys.* **71** (1965) 299
- 4) P. R. Fields, I. Ahmad, R. F. Barnes, R. K. Sjoblom and Wm. C. McHarris, *Nucl. Phys.* **A208** (1973) 269
- 5) M. R. Schmorak, *Nucl. Data Sheets* **17**, no. 3 (1976) 391
- 6) D. F. Peppard, S. W. Moline and G. W. Mason, *J. Inorg. Nucl. Chem.* **4** (1957) 344
- 7) I. Ahmad, R. F. Barnes, R. K. Sjoblom and P. R. Fields, *J. Inorg. Nucl. Chem.* **34** (1972) 3335
- 8) J. Lerner, *Nucl. Instr.* **102** (1972) 373
- 9) I. Ahmad, R. K. Sjoblom and P. R. Fields, *Phys. Rev.* **C14** (1976) 218
- 10) I. Ahmad and R. K. Sjoblom, *Proc. ERDA Symp. on X- and gamma-ray sources and applications*, CONF-760539, Ann Arbor, Michigan, 19-21 May 1976, p. 249
- 11) T. A. Carlson, C. W. Nestor, Jr., T. C. Tucker and F. B. Malik, *Phys. Rev.* **169** (1968) 27
- 12) R. S. Hager and E. C. Seltzer, *Nucl. Data* **A4** (1968) 1
- 13) C. M. Lederer, J. M. Hollander and I. Perlman, *Table of isotopes*, 6th ed. (Wiley, NY, 1967)
- 14) W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price and P. Venugopala Rao, *Rev. Mod. Phys.* **44** (1972) 716
- 15) I. Ahmad, unpublished results
- 16) N. B. Gove and M. J. Martin, *Nucl. Data Tables* **A10** (1971) 206
- 17) P. R. Fields, I. Ahmad, A. M. Friedman, J. Lerner and D. N. Metta, *Nucl. Phys.* **A160** (1971) 460
- 18) I. Ahmad, A. M. Friedman, R. R. Chasman and S. W. Yates, *Phys. Rev. Lett.* **39** (1977) 12
- 19) S. W. Yates, R. R. Chasman, A. M. Friedman, I. Ahmad and K. Katori, *Phys. Rev.* **C12** (1975) 442