

EVIDENCE FOR LONG-LIVED ISOMERIC STATES IN NEUTRON-DEFICIENT ^{236}Am AND ^{236}Bk NUCLEI

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A 5.76 MeV alpha-particle group has been observed in Am and Bk sources separated from a CERN W target. The data are interpreted due to the production of long-lived isomeric states in ^{236}Am and ^{236}Bk which decay to ^{236}Pu . The possibility of high spin states as well as of shape isomeric states is raised.

Recently [1], a consistent interpretation has been given to the secondary reaction experiments in W targets which were irradiated with 24 GeV protons. In the following we present evidence for the production, in the same target, of long-lived isomeric states in the neutron-deficient actinide nuclei, ^{236}Am and ^{236}Bk . Preliminary results of this work have been given elsewhere [2,3].

Actinide fractions from Am up to No-Lr have been separated from our (W3) [4-6] tungsten target (6 cm long, 30 g) which was irradiated in CERN with about 1×10^{18} ($\pm 20\%$) protons of 24 GeV energy. The main aim in the chemical separation [2] was to separate the heavy actinides, from Am up to Lr, from alpha emitting nuclei such as Ra, Ac, Th, U and Pu. The actinide sources were prepared [2] using the measured elution positions of the rare earth elements and known information on the actinides [7]. The decontamination factor for Th or Pu was estimated to be $\geq 10^8$; the intensity of ^{148}Gd was found to be about 4000 times larger in the Cf and Bk sources (where it should preferentially be) compared to the adjacent Es and Cm sources [2].

Alpha-particle spectra were measured, using Si surface-barrier detectors. In this paper we concen-

trated on the pronounced alpha-particle group at (5.76 ± 0.04) MeV which has been seen with the Am and Bk sources and where positive identification is possible. In fig. 1 two spectra which were measured with the Am source and two with the Bk source are given. Also shown in fig. 1 are two background spectra measured during a short period with one detector and during a long period with a second detector. Several groups (5.14, 5.3, and 5.47 MeV) are seen in addition to the very pronounced 5.76 MeV group. The 5.3 MeV could be ^{210}Po picked up from the air. Earlier, we had thought [2] that the 5.14 and the 5.47 MeV groups may be attributed to ^{239}Pu and ^{238}Pu , respectively. Further measurements showed that this is incorrect since their intensity decayed with half-lives of several years. (For the same reason most of the 5.47 MeV group could not come from ^{241}Am , though a small part of it may be contamination introduced by the manufacturer). The 5.14 MeV group could also not be due to ^{208}Po because initially its intensity grew with a half-life of about 1.5 yr, and all known decay chains to ^{208}Po have much shorter characteristic lifetimes. The 5.14 and the 5.47 MeV groups at present cannot be identified with any known activity. (The counts in fig. 1d at the low-

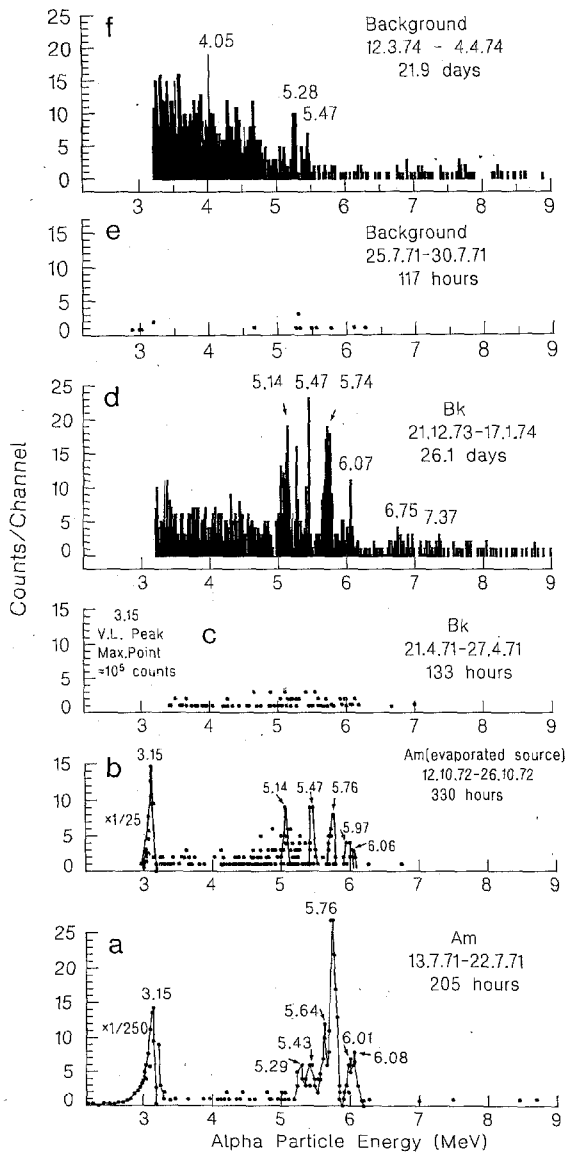


Fig. 1. Alpha-particles from Am and Bk. (a) 83 days after chemical separation, (b) after the source was evaporated, (c) 106 days after chemical separation (V.L. means very large), (d) 3 years later, (e) background spectrum measured during a short period with one detector [the same as used for (a), (b) and (c)], and (f) background spectrum measured during a long period with a second detector [the same as used for (d)].

energy region are background counts probably mainly due to electronic pick-up during the long period this measurement was taken).

The 5.76 MeV group is seen in figs. 1a, 1b and 1d but in fig. 1c there are only 3 to 4 counts in this energy

region which, however, could be background events. (To be on the safe side we therefore attributed a large error to this time normalized intensity point in fig. 2b.)

Several measurements were made over a period of about 14 years in order to identify the origin of the strong 5.76 MeV group. First, we determined the mass of the recoiling nuclei after alpha-decay in Am. The recoil velocity spectrum in coincidence with alpha-particles from 5.15 to 6.2 MeV (average energy 5.67 MeV) was measured using the time-of-flight technique [8] (fig. 6, ref. [2]), and the recoil mass obtained is $M_R = 237^{+6}_8$. This result shows that the alpha-particles seen in this region are due to the decay of actinide nuclei and not for instance from some unknown isomeric state in the rare earth region. For a consistency check in the same source served the 3.18 MeV group, which should have come from ^{148}Gd . Using the front edge of the peak in the velocity spectrum (fig. 6, ref. [2]), the mass of the recoil nuclei agrees well with 144.

In the actinide region there are four nuclei, ^{236}Pu , ^{243}Cm , ^{244}Cm and ^{249}Cf which decay with alpha-particles of about 5.76 MeV. The intensities (normalized to 30 days) of the 5.76 MeV groups in the Am and Bk sources as a function of time are shown in fig. 2. In the period between the first and the second measurement shown in fig. 2a, the Am source was evaporated in order to produce the thin source needed for the mass measurements described above. The relative intensities given in fig. 2a were obtained using the measured intensity of the 3.18 MeV alpha-particle group of ^{148}Gd ($t_{1/2} \sim 90$ yr) as a normalization, from the data given in table 1. The large difference in intensities between the first measurement as compared to the others, is due to the geometrical loss during the evaporation. The source before evaporation was in the form of chloride. Since the evaporation in vacuum took place at a temperature much higher than the melting points of the chlorides of Gd, Am and Pu (609–850°C), it is safe to assume that all the source material was evaporated and that there was no fractionation between Gd as compared to Am or Pu. Some uncertainty may arise if the sticking probability of Gd to the Ni backing was different from that of Am or Pu, or if Gd were to produce an alloy with the heated Ta crucible during the evaporation. (In order to obtain the pure decay curve in

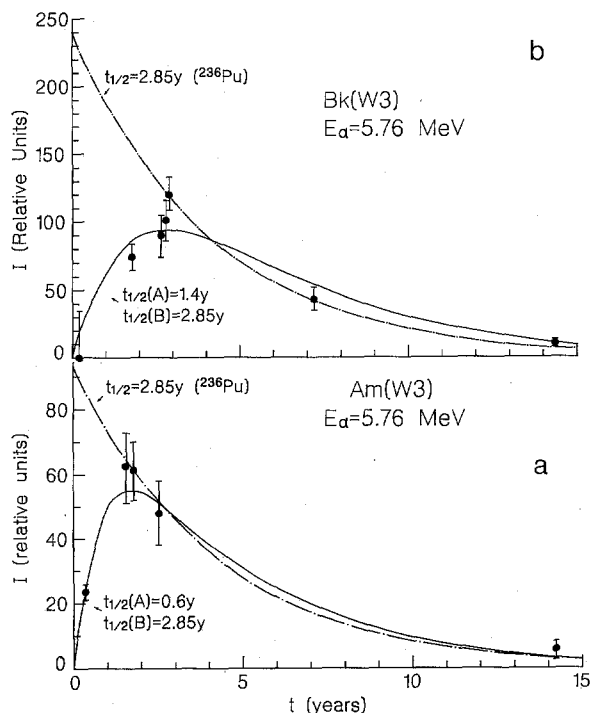


Fig. 2. (a) Decay curve of the 5.76 MeV alpha-particle group in Am. In between the first and second measurement the source was evaporated. The relative intensities were obtained by using the 3.18 MeV group of ^{148}Gd as a normalization (see text). (b) Decay curve of the 5.76 MeV alpha-particle group in Bk (see text). The first to seventh measurements were taken during 5.5, 32.9, 23.1, 14.3, 26.1, 26.8 and 60.0 days, respectively.

fig. 2a one would have to assume that while these effects did not influence the evaporation of Am or Pu, they caused an additional loss of about 70% for Gd). This hypothesis seems unlikely because Gd does

not react with Ta [9], the amount of Gd was extremely small ($\sim 10^9$ atoms) and the temperature needed was rather low (M.P. = 609°C). The Bk source was unchanged during the whole period. Most of the data shown in fig. 2b were obtained by recording event by event on paper tape (in coincidence with γ rays [2]). Because of the large intensity of the 3.18 MeV group in the Bk source, this group was not recorded on tape in order to avoid the loading of the tape, but from short term measurements taken before and after the data shown in fig. 2b it was found that the intensity of this group stayed the same. The first three measurements in fig. 2a and the first two measurements in fig. 2b were taken with one detector while the other two in fig. 2a and the other five in fig. 2b were taken with a second one. Both detectors were of the same size of 450 mm^2 and their efficiencies measured with the Am source using the 3.18 MeV group (column 3 of table 1) were the same, with a maximum systematic error of 18%. The errors given in fig. 2 are the statistical errors only.

It is seen in fig. 2 that in both cases the intensity first grew with time and then decayed by more than a factor of ten. Because of the very significant decrease in intensity ^{243}Cm , ^{244}Cm and ^{249}Cf , all with $t_{1/2} \geq 18.1\text{ yr}$, can be excluded with high certainty. But the half-life of ^{236}Pu of 2.85 yr is consistent with the data in both cases as seen from the dot-dashed lines in fig. 2. Therefore, from the measured alpha-particle energy, its half-life and the mass of the recoiling nuclei, it is concluded that the 5.76 MeV group in the Am and Bk sources is due to decay of ^{236}Pu . (The possibility of some unknown alpha decays in Am and Bk that by chance have the same energy and lifetime,

Table 1
Intensities of the 3.18 and 5.76 MeV groups as measured with the Am source.

Starting date of measurement	Length of measurement (days)	Counts/30 days	
		3.18 MeV	5.76 MeV
13 July 1971	8.5	9.3×10^4 ^{a)}	431 ^{a)}
12 October 1972	13.8	5.1×10^3	63
20 December 1972	13.3	5.1×10^3	61
20 September 1973	28.3	5.9×10^3	56
8 May 1985	60.5	5.0×10^3 ^{b)}	5.1

^{a)} The large difference between the first measurement as compared to the other measurements is due to geometrical loss during the evaporation of the source (see text).

^{b)} This value is corrected by about 10% for the decay of ^{148}Gd .

although in principle possible, seems unlikely). It, however, could not be due to contamination of ^{236}Pu in these sources. One should clearly distinguish between two possible ways of contamination: (i) contamination from the beginning, due to poor chemical separation; (ii) contamination during the long term measurements. The first possibility can be ruled out because (a) the quality of the chemical separation would allow at most 10^{-3} counts in the alpha spectra due to ^{236}Pu produced from possible U or Th contamination in the W target [10], about 5 orders of magnitude less intensity than measured. Also, according to the chemical procedure [2] if some Pu were left in the last stages of the separation it should be predominantly in the heavy actinides, for instance in the Lr-No fraction, rather than in Am. In fact no sign for 5.76 MeV group has been observed [2] in the heavy fraction; (b) a ^{236}Pu contamination from the beginning would have decayed with time, according to the dot-dashed lines in fig. 2, in strong contradiction with the experimental results. From this curve in fig. 2b we would expect 42 counts in the 5.76 MeV range in fig. 1c, where there are only 3 to 4. Contamination during the measurements may also be excluded since the source did not get in contact with any radioactive material after chemical separation, each source was kept separately air-tight under glass. Also, both of the separately stored sources would have had to be contaminated in very much the same way to reproduce the data. ^{236}Pu is not a common source and we did not have any in the laboratory. We saw no indication for ^{236}Pu in any of our other sources stored in a similar way and measured for about half a year each [11].

The best fit lines in fig. 2 assume a growing and a decaying component where the latter was fixed at the known half-life of ^{236}Pu ($t_{1/2} = 2.85$ yr). The growing times obtained are $(0.6 \pm 0.2$ yr) in Am and $(1.4 \pm 0.4$ yr) in Bk. It is impossible to interpret the data in terms of known decays, of ^{236}Np or ^{240}Cm . Both are inconsistent with our chemistry, measured half-lives and alpha spectra. Since the growth of ^{236}Pu has been observed in well separated Bk and Am sources, a consistent interpretation would be that previously unknown ^{236}Am and ^{236}Bk isotopes have been produced and decayed by electron capture (EC) or β^+ (directly or after gamma decay) to ^{236}Pu . Prior alpha decay may be ruled out since no alpha-particle

group was seen with appreciable intensity in the first or second measurements. In the case of Am one EC decay with a half-life of about 0.6 yr is consistent with the data, while in the case of Bk three decays must take place, the data are consistent with the sum of these to be around 1.4 yr. A lower limit for the ^{236}Bk half-life of about 1 month has to be assumed since otherwise we should not have seen the activity in the Bk source, as the separation between the Bk and the Cm fraction took place about 2 months after the irradiation was completed. (A 5.76 MeV group has also been observed in a Cm source. However, ^{243}Cm and ^{244}Cm also decay by alpha particles of about the same energy, but with long lifetimes. The data contain a growing component compatible with 1 yr half-life but it is difficult to extract quantitatively a growth time.)

As the lifetime of normal ^{236}Bk and ^{236}Am are expected to be several orders of magnitude shorter than our measured lifetimes, previously unobserved long-lived isomeric states must be assumed. If this is so, such isomers may exist also in other neutron-deficient heavy and superheavy nuclei. (Their assumed existence already led to a consistent interpretation of results in the superheavy elements region [1].) In principle one may consider two kinds of isomeric states, high spin states or a new kind of shape isomeric states. It should be mentioned, however, that high spin states have not been observed before in very heavy neutron-deficient actinides far from closed shells.

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