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Production and decay of the heaviest nuclei ^{293,294}117 and ^{294}118

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Two years after the discovery of element 117 we undertook a second campaign using the ^{249}Bk+^{48}Ca reaction for further investigations of the production and decay properties of the isotopes of element 117 on a larger number of events. The experiments were started in the end of April 2012 and are still under way. This paper presents the results obtained in 1200 hours of experimental run with the beam dose of ^{48}Ca of about 1.5×10^{19} particles. The ^{249}Bk target was irradiated at two energies of ^{48}Ca that correspond to the maximum probability of the reaction channels with evaporation of three and four neutrons from the excited ^{297}117. In this experiment two decay chains of ^{294}117 ($3n$) and five decay chains of ^{293}117 ($4n$) were detected. In the course of the long-term work, ^{249}Cf – the product of decay of ^{249}Bk (330 d) – is being accumulated in the target. Consequently, in the present experiment we also detected a single decay of the known isotope ^{294}118 that was produced during 2002–05 in the reaction ^{249}Cf(^{48}Ca,$3n$)^{294}118. The obtained results are compared with the data from previous experiments. The experiments are carried out in the Flerov Laboratory of Nuclear Reactions, JINR, using the heavy-ion cyclotron U400.

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The production and spectroscopic studies of the heaviest elements is one of the most rewarding and challenging investigations in nuclear physics. The discoveries of new superheavy atoms expand simultaneously the Periodic Table of the Elements and the Segre Chart of the Nuclei. New data on decay properties are helping to verify the predicted existence of the so called "Island of Stability" of superheavy elements. The stabilizing effect of the expected N=184 neutron shell closure found previously for even-Z nuclei [1] is supported by the experimental observation of a decrease of the decay constant for α emission with increasing neutron number. This observation was found to apply to the decays of odd-Z isotopes created during the discovery of elements Z=115 and 117 [2–5]. Longer lifetimes of nuclei located closer to N=184 can facilitate the measurements of chemical properties when the heaviest atoms are produced in sufficient amounts.

However, since the cross sections to create superheavy nuclei are at the picobarn and sub-picobarn level, experiments utilizing the current technology last several months and usually result in only few identified nuclei. Therefore, verification of low-statistics data and investigation of new isotopes are of utmost importance. The continuation of experiments results in optimization of the production methods through the determination of excitation functions, as demonstrated in recent studies of the ^{243}Am+^{48}Ca reaction [4, 5].

These goals motivated our new search for Z=117 isotopes among the ^{249}Bk+^{48}Ca reaction products. The discovery of element 117 in the complete-fusion reaction ^{249}Bk+^{48}Ca was reported in 2010 [3]. In experiments performed between July 27, 2009, and March 1, 2010, at two projectile energies of 252 MeV and 247 MeV in the middle of the target, corresponding to the excitation energies (E^*) of the compound nucleus ^{297}117 of 39 MeV and 35 MeV, respectively, two isotopes of element 117 were synthesized for the first time.

The excitation energy E^*=39 MeV of the ^{297}117 complete-fusion nucleus is close to the maximum yield of the $4n$-evaporation channel as expected from the comparison with other fusion-evaporation reactions with ^{48}Ca [1]. Indeed, we synthesized five decay chains of the odd-even isotope ^{293}117 at an excitation energy of 39 MeV. An α decay of ^{293}117 was followed by two consecutive α decays of previously unknown ^{289}115, ^{285}113, and was terminated by the spontaneous fission (SF) of ^{281}Rg; the decay sequence spanned an interval of about one minute.

From the well-established behavior of the excitation functions measured for numerous reactions, it followed that a reduction of the projectile energy should result in a decrease of the cross section for the $4n$ channel but permit observation of a heavier isotope with lower α-particle energy and longer lifetime, the product of the $3n$ channel. In an odd-odd nucleus, fission is suppressed compared to α emission because of the unpaired nucleons, resulting in a longer α decay chain. Indeed, at the 35 MeV excitation energy, we produced one longer decay chain of the odd-odd isotope ^{294}117. In this chain, the great-granddaughter nucleus with Z=111 did not undergo SF, but instead emitted an α particle. It was followed by two more α transitions and then, after about 33 h, a spontaneous fission event was recorded.

For more precise measurement of the radioactive properties of ^{293,294}117 and their descendant nuclei we started a new series of experiments. Here we present results of the two runs performed at ^{48}Ca beam energies of 247 MeV and 252 MeV.

As in the previous experiments from 2009–2010 [3], the ^{249}Bk was produced at Oak Ridge National Laboratory (ORNL) through the intense neutron irradiation of Cm and Am seed material. In 2010–2011, two irradiations were performed at the High Flux Isotope Reactor. The first irradiation of four targets lasted for approximately 250 days. The second irradiation of five targets lasted for only one month (August 2011), but created about half of the total ^{249}Bk material. The Bk fraction was chemically separated from all irradiated targets and was purified at the Radiochemical Engineering Development Center at ORNL, see ref. [3] for a description of irradiations and chemical procedures. Half of the total ^{249}Bk material, 12.7 mg, was shipped in early March 2012 to JINR Dubna. It contained 0.51 mg of ^{249}Cf (β^- decay product of ^{249}Bk) and only 0.45 ng of ^{252}Cf, and no other detectable impurities. Six arc-shaped targets, each with an area of 5.4 cm^2, were made at the Research Institute of Atomic Reactors (Dimitrovgrad, RF) by depositing BkO_2 onto 0.72 mg/cm^2 Ti foils to a thickness of 0.33 mg/cm^2 of ^{249}Bk. The targets were mounted on the perimeter of a disk 12 cm in diameter that was rotated at 1700 rpm perpendicular to the beam direction. The experiments were performed employing the Dubna gas-filled recoil separator (DGFRS) and the heavy-ion cyclotron U-400 at JINR.

Based on results of previous experiments [3] in which the ion charge of element 117 recoils was measured to be $6.1^{+0.3}_{-0.2}$, we used a magnetic rigidity of the separator of 2.395 T·m during the whole run. This is close to the magnetic rigidity of about 2.36 T·m used in the experiments on the synthesis of element 118 [6].

The detection system was modified to increase the position granularity of the detectors, which reduces the probability of observing sequences of random events that mimic decay chains of synthesized nuclei. The new focal-plane detectors consisted of two 6×6 cm^2 silicon detectors each having 16 strips with position sensitivity in vertical direction. These detectors were surrounded by six similar 6×6 cm^2 side detectors without position sensitivity. Behind the focal-plane detectors, which had a thickness of 0.3 mm, a pair of veto detectors similar to the side detectors was mounted for the detection and rejection of signals from high-energy long-range charged particles (α's, protons, etc produced in direct reactions of projectiles with the DGFRS media) which can pass through the separator without being detected by the time-of-flight system. The full-width-at-half-maximum (FWHM) energy resolution of implantation detector was 34 to 73 keV, while the summed signals recorded by the side and implantation detectors had an energy resolution of about 83 to 117 keV. The FWHM position resolution of the implantation detector were 1.1-1.8 mm for ER-α and 0.5-1.2 mm for ER-SF signals, compare [3]. Other experimental conditions, including the method of calibration of the detectors, were the same as in [3]. In order to reduce the background rate in the detector, the beam was switched off for at least 3 min after a recoil signal was detected with $E_{\rm ER}$=7–18 MeV followed by an α-like signal in the focal-plane detector within energy intervals of 10.7–11.3 MeV and 10.0–10.7 MeV and time intervals of 0.4 s or 2.0 s, respectively, in the same strip, within a 3.2 mm wide position window.

The experiment was performed from April 23 to July 13, 2012, at two ^{48}Ca projectile energies of 252 MeV and 247 MeV (mid-target) with total beam doses of 1.2×10^{19} particles and 3.4×10^{18} particles, correspondingly. With the energy spread of the incident cyclotron beam, the small variation of the beam energy during irradiation, and the energy losses in the target (3.0 MeV), we expected the resulting ^{297}117 compound nuclei to have excitation energies of 37.0–41.9 MeV and 32.8–37.5 MeV, respectively. Excitation energies of the compound nuclei are calculated using the masses in [7, 8]. The beam energy losses in the separator's entrance window (0.71 mg/cm^2 Ti foil), target backing, and target layer were calculated using the nuclear data tables in [9, 10].

The decay properties of four nuclei originating from the $4n$-evaporation reaction product, ^{293}117, measured in the five similar decay chains observed in the first run at E^*=39 MeV and seven nuclei in two decay chains originating from heavier isotope ^{294}117 observed at E^*=35 MeV are shown in Fig. 1(a) and Fig. 1(b), respectively. In two cases (chains 1 and 3 in Fig. 1(a)) the α particles of the parent nucleus ^{293}117 were detected by both the focal-plane and side detectors with $E_{\rm f-p}$=3.457 MeV, $E_{\rm s}$=7.443 MeV ($E_{\rm f-p}$+$E_{\rm s}$=10.900 MeV) and $E_{\rm f-p}$=1.376 MeV, $E_{\rm s}$=9.738 MeV ($E_{\rm f-p}$+$E_{\rm s}$=11.114 MeV), respectively. In the first chain, a side-only event with E_α=9.99 MeV was found between α particles with energies 10.90 MeV and 9.86 MeV; in the third chain this α particle was not detected. Thus, in both of these cases, the beam was not switched off. In the three other cases, the daughter products of ^{293}117 (^{289}115, ^{285}113, and ^{281}Rg) were observed when the beam was off and thus associated with a very low counting rate of background events.

Despite the detection of two decay chains completely during beam-on intervals, the expected total number of random events ER–$\alpha_{1,\rm on}$–$\alpha_{3,\rm on}$–SF$_{\rm on}$ is lower than 10^{-5}. The expected numbers of random sequences of the types

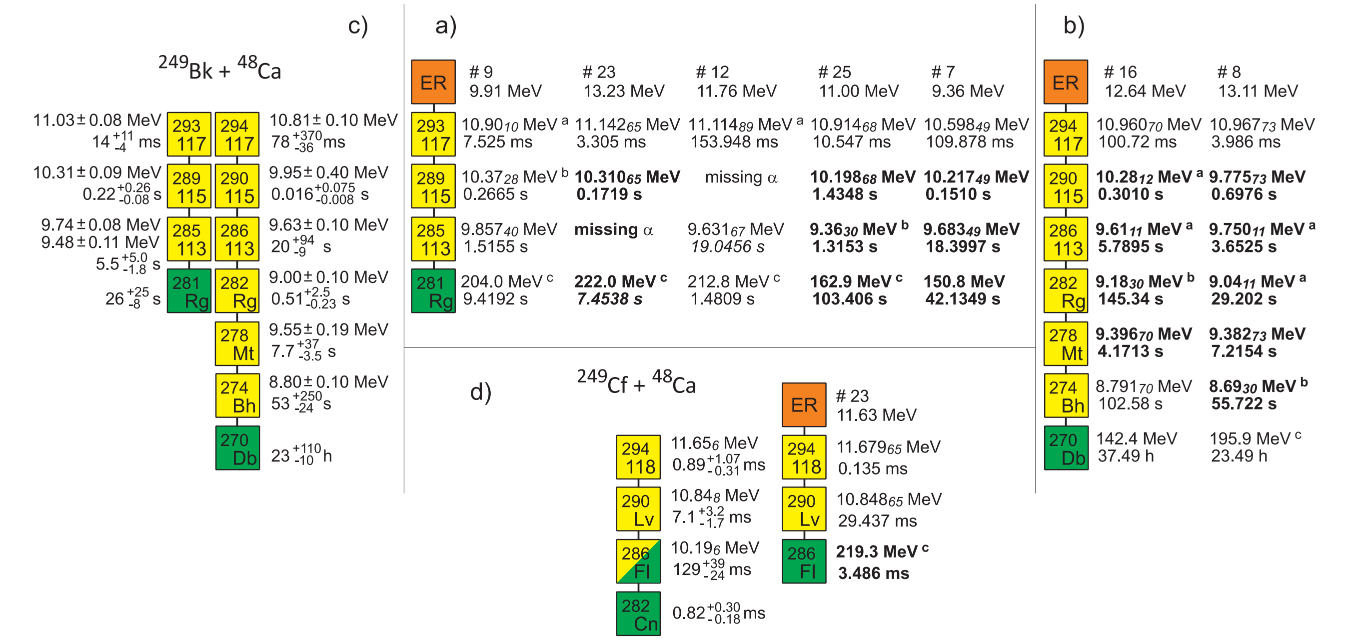

FIG. 1: (color). (a) Decay properties of the isotope $^{293}117$ and its decay products observed in this work. The first rows show ER energies and detector strip numbers. Subsequent rows provide α-particle and SF fragment energies and the time intervals between events. Bold events were registered during a beam-off period. The α-particle energy errors are shown by smaller italic numbers. Time intervals for events following a "missing α" were measured from preceding registered events and are shown in italics ([a] α particle registered by both focal-plane and side detectors, [b] escaped α particle registered by side detector only, [c] fission event registered by both focal-plane and side detectors). (b) The same for isotope $^{294}117$. (c) Average decay properties ($T_{1/2}$ and E_α) of $^{293}117$–^{281}Rg and $^{294}117$–^{270}Db observed in [3]. (d) Average decay properties ($T_{1/2}$ and E_α) of $^{294}118$–^{282}Cn determined in the reaction ^{249}Cf+^{48}Ca [1, 6] (left-hand panel) and properties of $^{294}118$–^{286}Fl measured in this work (right-hand panel).

ER–$\alpha_{1,\mathrm{on}}$–$\alpha_{2,\mathrm{off}}$–SF$_{\mathrm{off}}$ (2nd and 4th decay chains) and ER–$\alpha_{1,\mathrm{on}}$–$\alpha_{2,\mathrm{off}}$–$\alpha_{3,\mathrm{off}}$–SF$_{\mathrm{off}}$ (5th chain) are lower than 5×10^{-10} and 10^{-14}, respectively [11]. Note, only eight α particles with E_α=9.4–10.6 MeV and five SF events were registered by the whole focal-plane detector during the total beam-off time interval (t=2$\times$10^5 s), among them, five α particles and three SF events belong to the decay chains of $^{293}117$.

The loss of two α particles (marked "missing α" in Fig. 1(a)) in five decay chains consisting of three α decays each, is in agreement with the 87% efficiency of the detector array for observing full-energy α particles. The probability of the random appearance of a beam-on signal in one of the six side detectors with E=9.6–10.3 MeV and within 1-s time interval of a triggering event in strip 9 was about 8%. We tentatively assigned the side-only signal in chain 1 to $^{289}115$; its total energy was estimated to be the sum of the energy measured by the side detector and half of the threshold energy of the focal-plane detector (0.77 MeV for strip 9) with the uncertainty in determining the total energy increased to $\approx$0.3 MeV (68% confidence limit). In the fourth decay chain, the third α decay, $^{285}113$, was also registered by the side detector only but during very low background conditions. The probability of its random origin is about 1%.

The other nine α particles were detected solely by the focal-plane detector. The position deviations between ER signals and two α particles detected by both the focal-plane and side detectors, nine α decays registered entirely by the focal-plane detector as well as five SF events, were in full agreement with the position resolutions of detectors for consecutive ER-α and ER-SF signals.

In two of the decay chains of $^{294}117$, the 3n-evaporation product, all twelve α decays were registered (see Fig. 1(b)). Six α particles were absorbed by only the focal-plane detector, four α particles were detected by both the focal-plane and side detectors, and two α particles were detected by the side detector only. The last α decay (^{274}Bh) in the first decay chain was detected when the beam was back on ($\Delta t_{\alpha1-\alpha6}$ >3 min). But the probability of this event as a random particle plus two beam-off α particles detected by the side detectors only (without signal in the focal-plane detector) is 0.01–0.03. Within a limited event count of $^{294}117$, for which only a single event was observed in [3], the properties of nuclei in the new decay chains point to the same activities arising from $^{294}117$ detected in two experiments using ^{294}Bk target. Note, as it was discussed in [3], in the both new decay chains we observed longer lifetimes for $^{290}115$ and ^{282}Rg compared with the values detected in the first experiment. The summary of radioactive properties of nuclei observed in the reaction ^{249}Bk(^{48}Ca,3n)^{294}117 [3] are shown in Fig.1(c).

The radioactive properties of nuclei observed in the reaction ^{249}Bk(^{48}Ca,4n)^{293}117 in 2010 [3] and this work, as well as in the reaction ^{243}Am(^{48}Ca,2n)^{289}115 [4, 5], are shown in Table I. The radioactive decay properties of ^{293}117 and all descendant nuclei discovered in 2010 [3] were completely confirmed by registration of five new decay chains in this new series of experiments. One can see in Fig. 1 that the results of the five events in the first experiment are in good agreement with the data of this work. Moreover, the α-particle energies and decay times of the isotopes ^{289}115, ^{285}113, and ^{281}Rg registered after the α decay of the parent nucleus ^{293}117 in the reaction ^{249}Bk+^{48}Ca and synthesized directly in the reaction ^{243}Am+^{48}Ca [4, 5] are comparable. Therefore, the isotope ^{289}115 was produced in two reactions with target nuclei ^{243}Am and ^{249}Bk as 2n-evaporation product in the first case and as a daughter nucleus after the α-decay of heavier parent nuclide ^{293}117 in the second case. The observed spread of α energies, often clearly exceeding the energy resolution of detectors is most likely related to fine structure in α decays. However, the experiments with better energy resolution, better statistics and the observation of α-γ correlations are needed to corroborate this interpretation.

The cross sections for the 3n- and 4n-evaporation channels at E^*=35 and E^*=39 MeV were measured to be $\sigma_{3n} = 3.6^{+6.1}_{-2.5}$ pb and $\sigma_{4n} = 2.0^{+2.2}_{-1.0}$ pb in this work that are larger but within experimental uncertainties when compared with the previous results of $\sigma_{3n} = 0.5^{+1.1}_{-0.4}$ pb and $\sigma_{4n} = 1.3^{+1.5}_{-0.6}$ pb [3]. These cross section values are consistent with the results of previous experiments where cross sections for the reactions of ^{238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm, and ^{249}Cf targets with ^{48}Ca beams have been measured [1–6].

One also should note that target isotope ^{249}Bk (Z=97) decays into ^{249}Cf (Z=98) with a half-life of T_β=330 d. During a long experiment, the percentage of ^{249}Bk in the target decreases and the quantity of ^{249}Cf becomes larger. This creates an opportunity to produce Z=118 isotopes during the same experiment in the ^{249}Cf+^{48}Ca reaction [6] after sufficient ^{249}Cf material accumulates in the target layer.

The isotope ^{294}118 of the new element 118 was produced for the first time in the reaction ^{249}Cf+^{48}Ca [6]. With 245 MeV ^{48}Ca projectiles, one decay chain of ^{294}118 was observed. An increase of the ^{48}Ca energy to 251 MeV resulted in an increase of the cross section of the 3n-evaporation channel and two additional ^{294}118 atoms were synthesized. In two cases ^{294}118 underwent two consecutive α decays terminated by the spontaneous fission of ^{286}Fl. In the third decay chain three α decays of ^{294}118, ^{290}Lv, and ^{286}Fl were detected that were followed by the SF of ^{282}Cn. Such a decay pattern of ^{294}118 is consistent with the radioactive properties of ^{286}Fl (a 50-% fission branch was determined for its decay in [12, 13]) and ^{282}Cn [12–15].

In addition to two nuclei ^{294}117 at 247-MeV ^{48}Ca, we observed one more decay chain in strip 23. This decay chain consists of an evaporation residue ($E_{\rm ER}$=11.63 MeV), two α decays registered during beam-on interval by the focal-plane detector and a spontaneous fission within beam-off pause caused by the ER-α_2 pair (see Fig. 1(d)). The decay properties of the nuclei in this chain agree well with those determined for ^{294}118 and its descendant nuclei ^{290}Lv and ^{286}Fl (fission branch of 50%) [1, 6, 14, 15].

The ingrowth of ^{249}Cf in the ^{249}Bk target material at the end of this run was 28%. According to the previously obtained cross-section data on the production of the nuclide ^{294}118 [6], as well as that of the other superheavy nuclides produced in the ^{48}Ca-induced reactions, ^{294}118 can be observed in the reaction ^{249}Cf(^{48}Ca,3n) over the whole energy range from E^*=26.6 MeV [6] to about 40 MeV [1–6, 12–15]. Taking into account the buildup of ^{249}Cf in the preceding [3] and present experiments, where ^{294}118 was not registered, and the total beam dose of ^{48}Ca of 5.9×10^{19} (effective beam dose is about 1.6×10^{19} for 0.3-mg/cm^2 target), the detected decay chain of ^{294}118 corresponds to $\sigma_{3n} = 0.3^{+0.7}_{-0.26}$ pb for the total excitation-energy interval of 26.6–37.5 MeV for ^{297}118. This value is in good agreement with cross sections measured for this reaction at 245 MeV (E^*=26.6–31.7 MeV, $\sigma_{3n} = 0.3^{+1.0}_{-0.27}$ pb) and 251 MeV (E^*=32.1–36.6 MeV, $\sigma_{3n} = 0.5^{+1.6}_{-0.3}$ pb) [6].

In conclusion, the discovery of a new chemical element with atomic number 117 that was reported for the first time in 2010 [3] has now been corroborated through the observation of additional five decay chains originating from the 4n-evaporation product, ^{293}117, and two decay chains of the 3n channel, ^{294}117, of the reaction ^{249}Bk+^{48}Ca. The radioactive decay properties of the eleven isotopes ^{293,294}117, ^{289,290}115, ^{285,286}113, ^{281,282}Rg, ^{278}Mt, ^{274}Bh, and ^{270}Db measured in this work are in full agreement with the results of the first experiment [3] as well as with the decay data determined for ^{289}115, ^{285}113, and ^{281}Rg measured in the cross-bombardment reaction ^{243}Am(^{48}Ca,2n)^{289}115 [4, 5]. The average cross sections for the production of ^{293}117 and ^{294}117 nuclei in the ^{249}Bk+^{48}Ca reaction at E^*=39 MeV and 35 MeV determined from the observation of 10 and 3 events amounts to $1.5^{+1.1}_{-0.5}$ pb and $1.1^{+1.2}_{-0.6}$ pb, respectively. The earlier reported discovery of the heaviest known element 118 [6] was confirmed by the observation of one more decay chain of ^{294}118 and its daughter nuclei ^{290}Lv and ^{286}Fl. The decay properties of the heaviest nuclei were determined more accurately and demonstrate once more the decisive role of nuclear shell effects in the stability of superheavy nuclei.

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TABLE I: Decay properties of nuclei originating from ^{294}118, ^{294}117, and ^{293}117.

Z	N	A	No. observed[a]	Decay mode, branch (%)[b]	Half-life[c]	E_α (MeV)[d]
118	176	294	4(4/4)	α	$0.69^{+0.64}_{-0.22}$ ms	11.66±0.06
117	177	294	3(3/3)	α	50^{+60}_{-18} ms	10.81–10.97
	176	293	10(10/10)	α	27^{+12}_{-6} ms	10.60–11.14
116	174	290	11(11/11)	α	$8.3^{+3.5}_{-1.9}$ ms	10.85±0.07
115	175	290	3(3/3)	α	$0.24^{+0.28}_{-0.09}$ s	9.78–10.28
	174	289	11(11/11)	α	$0.38^{+0.18}_{-0.10}$ s	10.20–10.54
114	172	286[e]	25(20/12)	α/SF:50/50	$0.12^{+0.04}_{-0.02}$ s	10.19±0.06
113	173	286	3(3/3)	α	$8.7^{+10.4}_{-3.1}$ s	9.61–9.75
	172	285	14(12/12)	α	$5.6^{+2.2}_{-1.2}$ s	9.48–10.18
112	170	282[e]	12(12/-)	SF	$0.82^{+0.30}_{-0.18}$ ms	
111	171	282	3(3/3)	α	40^{+49}_{-14} s	9.00–9.18
	170	281	14(12/-)	SF	22^{+8}_{-5} s	
109	169	278	3(3/3)	α	$5.2^{+6.2}_{-1.8}$ s	9.38–9.55
107	167	274	3(3/3)	α	54^{+65}_{-19} s	8.69–8.80
105	165	270	3(3/-)	SF	22^{+26}_{-8} h	

[a] Number of observed decays and number of events used for calculations of half-lives and α-particle energies, respectively.

[b] Branching ratio is not shown if only one decay mode was observed.

[c] Error bars correspond to 68%-confidence level.

[d] For odd-Z nuclei the energy range of α particles detected by the focal-plane or both the focal-plane and side detectors is shown.

[e] Decay properties of these nuclei are in agreement with those measured in two decay chains in [14, 15].

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