Synthesis of the New Element with Z=117

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Abstract. The synthesis of the new chemical element with atomic number Z=117 is presented. The isotopes ^{293}117 and ^{294}117were produced in fusion reactions between ^{48}Ca and ^{249}Bk. The ^{249}Bk was produced in the High Flux Isotope Reactor and chemically separated at Oak Ridge. Decay chains involving eleven new nuclei were identified by means of the Dubna Gas Filled Recoil Separator. The measured decay properties show a strong rise of stability for super-heavy nuclei toward N=184.

1. Introduction

The stability of superheavy elements (SHE) is strongly influenced by the shell structure of neutrons and protons, see e.g., [1,2]. In the absence of corrections for shell structure, the liquid drop model predicts nuclei with Z≥100 should not exist. However, isotopes of elements having atomic numbers up to Z=118 have been observed [1]. Indeed, beyond the especially stable, spherical double-magic ^{208}Pb (double closed shells Z=82 and N=126), theoretical predictions are that N=184 should be spherical magic number that would give special stability to nuclei. The proton magic number is predicted in different approaches to be 114, 120, and 126 to form an Island of Stability. The synthesis of new elements with neutron number (N) approaching 184 provide important tests of the nuclear structure models used to predict closed spherical shells in the heaviest elements.

Reactions between doubly-magic ^{208}Pb and singly-magic ^{209}Bi target nuclei and stable neutron-rich projectiles such as ^{64}Ni or ^{70}Zn were used for the synthesis of new heavy elements. In these cold fusion reactions isotopes with Z •113 and N•165 [3, 4], stabilized by the Z=108 and N=162 shell gaps for deformed shapes were observed (see Fig. 1) . The dramatic drop of the production cross section with increasing Z practically excludes the continuation of such experiments for heavier elements.

A new method of synthesizing superheavy elements, with Z≥112 and neutron numbers closer to the predicted spherical shell closure at N=184, was pioneered at the Flerov Laboratory of Nuclear Reactions (FLNR) of Joint Institute for Nuclear Research (JINR) [1, 5]. Four new isotopes of

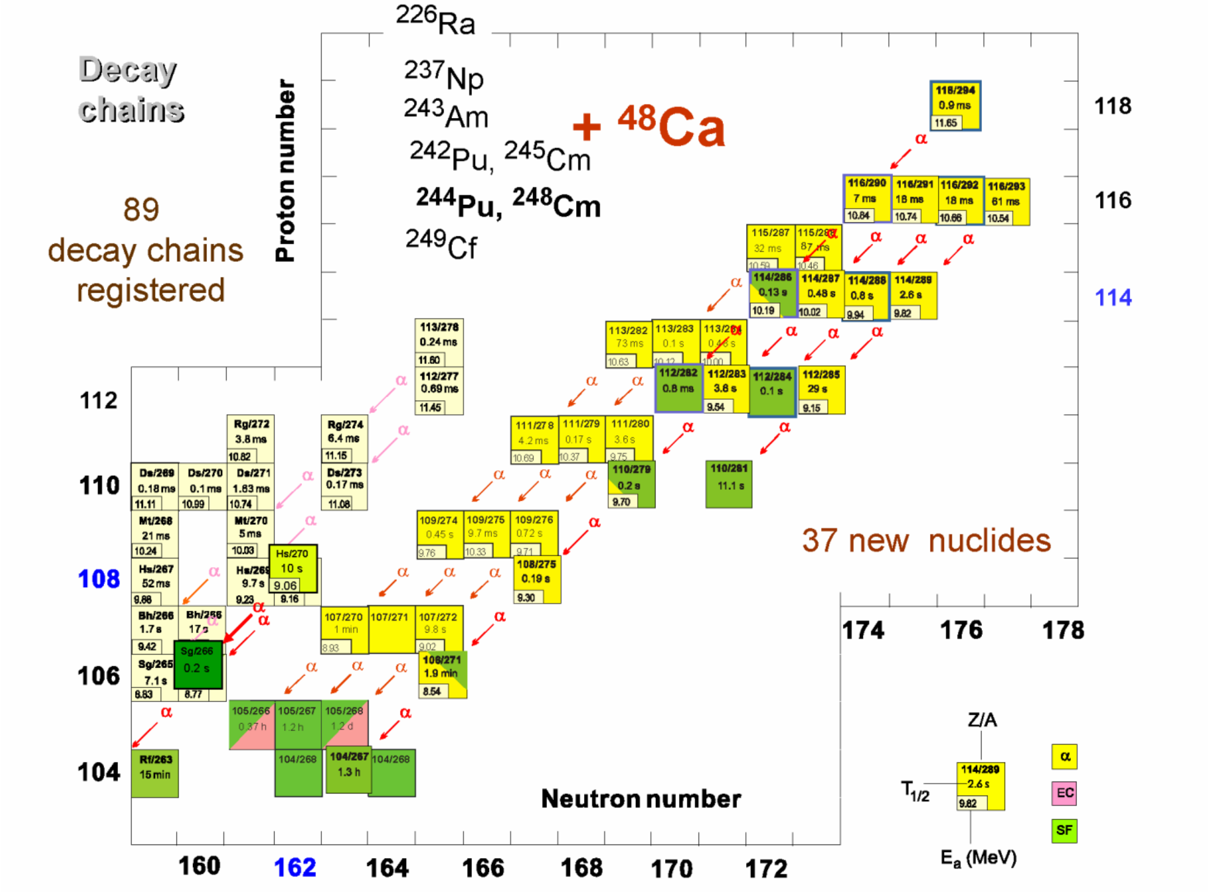

Fig. 1 Superheavy elements produced in cold fusion (pale yellow) [3, 4] and hot fusion (bright yellow on right) [1] reactions. The number of decay chains and nuclides were prior to this work.

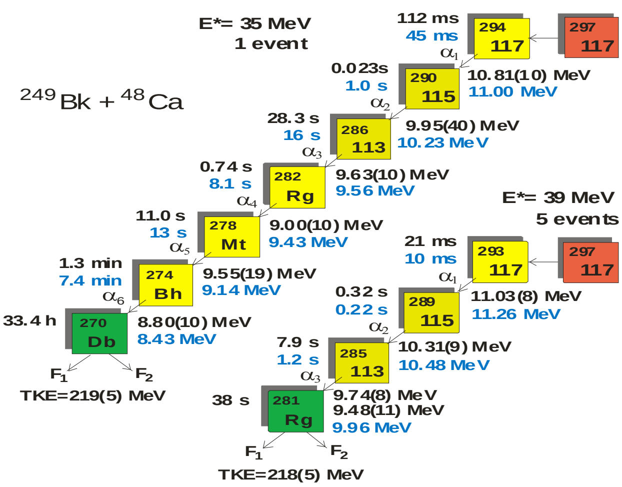

Fig. 2 Observed decay chains interpreted as originating from the isotopes A=294 and A=293 (average of five events) of the new element Z=117 [10]. The deduced and predicted [11] lifetimes (= $T_{1/2}$/ln 2) and α-particle energies are shown in black and blue, respectively. Taken from [10].

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element Z=112 and fourteen isotopes of new elements with Z=113-116 and 118 were identified [1] by using heavy-ion fusion reactions of doubly-magic ^{48}Ca projectiles and actinide targets of U-Cm and Cf, respectively (see Fig. 1). The element with Z=117 was missing because of difficulty in obtaining the short lived ^{249}Bk for target material. The probabilities of formation and the decay properties of these 18 new nuclei provide evidence of a considerable increase in nuclear stability with increasing neutron number in the nucleus. The identification and decay properties of the Z=112, 114 isotopes obtained at Dubna (1, 5) have been recently confirmed in several independent experiments [6-9]. Here we describe the synthesis of ^{293,294}117 (N=176,177) isotopes in the ^{48}Ca + ^{249}Bk 4n and 3n reactions (see Fig. 2). If (p,xn) reactions had occurred, then known isotopes of 116 would be seen and they were not, so 117 is correct. The observed α-decay chains show increasing stability (longer half-lives) for eleven new isotopes that end in the spontaneous fission (SF) of ^{281}Rg (T_{SF} = 26s) and ^{270}Db (T_{SF} •1d) [10]. The decay properties of the observed eleven neutron-rich isotopes demonstrate the decisive role of the shell effects in the stability of the heaviest nuclei approaching N = 184.

2. Experimental Procedures and Results

The ^{249}Bk material ($T_{1/2}$ of 320 d) was produced at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). Targets of Cm and Am were irradiated with a flux of 2.5 x 10^{15} n/cm^{2}s as part of an ongoing ^{252}Cf campaign. Following a three-month cooling period, extensive chemical separations were carried out over the next three months. The Bk chemical fraction, separated and purified at the Radiochemical Engineering Development Center (REDC) at ORNL, shown in Fig. 3, contained 22.2 mg of ^{249}Bk, only 1.7 ng of ^{252}Cf, and no other detectable impurities.

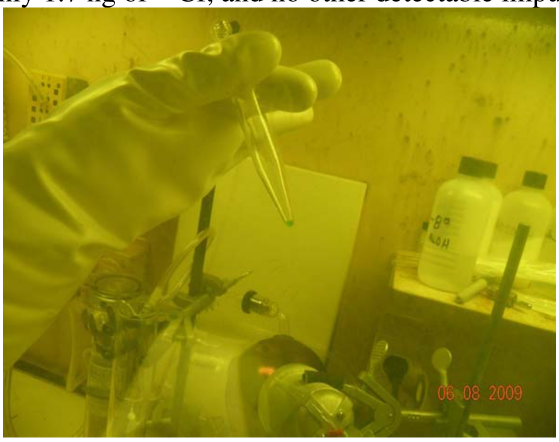

Fig. 3 The 22 mg of ^{249}Bk at the end of the separation at REDC (Oak Ridge).

Six arc-shaped targets, each with an area of 6.0 cm^2, were made at the Research Institute of Atomic Reactors (Dimitrovgrad, RF) by depositing Bk nitrate onto 0.74-mg/cm^2 Ti foils to a thickness of 0.31 mg/cm^2 of ^{249}Bk. The targets were mounted on the perimeter of a disk that was rotated at 1700 rpm perpendicular to the beam direction while the beam was wiggled vertically up and down over the target. The experiments were performed employing the Dubna Gas-Filled Recoil Separator [1, 12] and the heavy-ion cyclotron U-400 at JINR.

More details of the experiment than in our initial publication [10] will be given in a forthcoming paper [13]. The basic features are presented here. Evaporation residues (ER) passing through the separator with an overall transmission about 35% were registered by a time-of-flight system with a detection efficiency of 99.9%, and were implanted in a 4 cm x 12 cm Si-detector array with 12 vertical position-sensitive strips surrounded by eight 4 cm x 4 cm side detectors. The position-averaged detection efficiency was 87% of 4π for α-particles emitted from implanted nuclei. If an α-particle was detected only by a side detector (its position was lost), the total energy was estimated as a sum of energy measured by the side detector and half of the threshold energy (•0.5 MeV), with its total energy uncertainty increased to ± 0.4 MeV. The position resolution (FWHM) of the strip detector was •1.2 mm when registering correlated decay chains of the ER-$\alpha_1-\alpha_2-\alpha_3$-SF type. The background rate in the detector was reduced by switching off the beam for at least 3.5 minutes after a recoil signal was detected with an implantation energy expected for Z=117 ERs, followed by an α-like signal with an energy between 10.7 MeV and 11.4 MeV, in the same strip, within a 2.2-mm-wide position window. For 252-MeV ^{48}Ca projectiles, the excitation energy of the compound nucleus ^{297}117 is estimated to be E* = 39 MeV, near the expected maximum for the total ER cross section (sum of 3n and 4n evaporation channels [1]). A 70 day irradiation at this beam energy was performed from July 27 to October 29 to give a total beam dose of 2.4 x 10^{19}.

Five position-correlated decay chains were observed in the 252-MeV ^{48}Ca irradiation; in each case, two or three α-decays were observed between the time of arrival of the ER and the detection of SF. The averaged decay properties of the five events assigned to the ^{293}117 isotope are shown in Fig. 2. The average α energy of the first five α-particles emitted following the recoil implantations is $E_{\alpha 1}$=11.03 ± 0.08 MeV and $T_{\alpha 1}$=14(+11,-4) ms. The average α energy emitted by the daughter nuclei and detected in three out of five chains is E_{α_2}=10.31 ± 0.09 MeV with T_{α_2}=0.22(+0.26,-0.08) s. The third α-transition was observed to have E_{α_3}=9.74 ±0.08 MeV and E_{α_3}=9.48±0.11 MeV, and T_{α_3}=5.5(+5.0,-1.8) s. All five decay chains ended in spontaneous fission with T_{SF}=26(+25,-8)s.

The maximum cross section is expected for the 4n evaporation channel for the E*=39 MeV excitation energy. The observed decay chains are assigned to originate from the isotope ^{293}117. This assignment is thus supported by the systematics of the cross sections σ_{xn}(E*) measured previously for the production of superheavy isotopes with Z=108, 112-116, and 118 in ^{48}Ca-induced reactions [1], by calculations made for the evaporation residues of the reaction ^{249}Bk + ^{48}Ca [14-16], and by the result of our ^{249}Bk + ^{48}Ca experiment performed at lower beam energy (discussed next). In the E_α energy range between 8.8 MeV and 11.3 MeV, where we expect α-particles of the first five transitions 117$\rightarrow$115$\rightarrow$113 $\rightarrow$111$\rightarrow$109$\rightarrow$107, the counting rate was 0.17/s (with beam on) and 10^{-3}/s (beam off) for the whole area of the front detector. The position window defined by the resolution of the detector is about 0.005 of the entire detector area, so background rates in a given detector "pixel" are small. The total numbers for random sequences [17] imitating each of the observed five decay chains were calculated to be 6 x 10^{-6}, 10^{-3}, 10^{-5}, 3 x 10^{-11} and 3 x 10^{-11}.

Next, the experiment was run at a ^{48}Ca energy of 247 MeV for 70 days with a total beam dose of 2 x 10^{19}. The excitation energy of the compound nucleus ^{297}117 was approximately 35 MeV, which favors the 3n reaction channel. A new decay chain with six consecutive α-decays and ending in SF was detected, see Fig 2. In this chain, the Z=111 great-granddaughter nucleus emitted an α-particle with $E_{\alpha 4}$=9.00 MeV instead of undergoing SF. Then at least two more α-transitions followed, and after about 33 hours, a fission event was recorded. The latter observation is significantly different from the known decay properties of ^{294}(118) nucleus [1] and it was made when about 75% of the ^{249}Bk still remained in the target. Therefore, we assign the chain to the decay of the neighboring odd-odd nucleus ^{294}117. The chance probability for this chain was 6 x 10^{-4}.

The decay properties of the neighboring isotopes ^{293}117 and ^{294}117, their daughters ^{289}115 and ^{290}115, and granddaughters ^{285}113 and ^{286}113 are essentially the same but change significantly for the great-granddaughter nuclei. In spite of a strong hindrance resulting in a relatively long half-life, SF is a principal decay mode of the odd-even nucleus ^{281}111 (see fig. 2). However, the heavier isotope ^{282}111 undergoes α-decay. The SF decay of ^{281}111 can be understood by comparing the present results with the properties of the neighboring even-Z nuclei. In the T_{SF} (N) systematics, the decrease in the half-life with increasing neutron number for nuclei with N>162 changes to a strong increase in stability as N approaches the spherical shell at N=184 [18]. Minimum values of T_{SF} are characteristic in the transition region N=168-170. These T_{SF} have minimum values because the effect of nuclear shells is at a minimum. For example, the Z=110 darmstadium isotopes with N=169 and 171 and the

Z=112, N=170, 172 copernicium isotopes, undergo SF rather than α-decay [1]. The odd-Z isotopes of elements 113 and 115 with N=169-173 have a preference for α-decay [19, 20] because of their high hindrance of SF for nuclei with odd number of protons and the relatively low T_α. Only in isotopes of elements 105 is SF observed where the α-decay half-live exceeds 10^5 s for ^{268}Db. The reaction ^{249}Bk+^{48}Ca yields daughter nuclei that originate from the evaporation residues ^{293}117 and ^{294}117 with one or two extra neutrons compared with those produced in the lower-Z reactions. Approaching closer the N=184 shell should yield a decrease in their decay energy Q_α and an increase in T_α with respect to the neighboring lighter isotopes at the same Z. This behavior is clearly observed experimentally for all the isotopes with Z≥111, for the ^{293}117 chain and in Q_α for ^{294}117. The decay times for Z≥112 in the ^{294}117 chain are far longer than those in the ^{293}117 chain, see Fig.4. By analogy with the neighboring even-Z isotopes, all the nuclei in the ^{293}117 and ^{294}117 decay chains with Z>111 and N≥172 should undergo α-decay. With its odd proton, the nucleus ^{281}111 (N=170) is in a "critical" region, and may avoid SF only because of the hindrance produced by its odd proton. However, in spite of a hindrance of 3 x 10^4 with respect to its even-even neighbor ^{282}112, the isotope ^{281}111 has a probability b_{SF} ≥ 83% for SF. So, even the high hindrance caused by its odd proton does not "save" this nucleus from SF because of the weakening of the stabilizing effect of the N=162 and N=184 neutron shells. The presence of an extra, unpaired neutron in the neighboring isotope ^{282}111 further hinders SF relative to the α-decay of this nucleus. The experimental Q_α and half-lives T_α are presented in Figs. 4a,b for isotopes with Z= 111, 113, 115 and 117. As N increases, Q_α decreases with a considerable increase in T_α. The isotopes of elements 111 and 113 exhibit an especially strong growth of T_α (N). Except for ^{281}111, all the nuclides shown in Fig. 4, are α-emitters; with T_α smaller than T_{SF} to indicate the high stability of

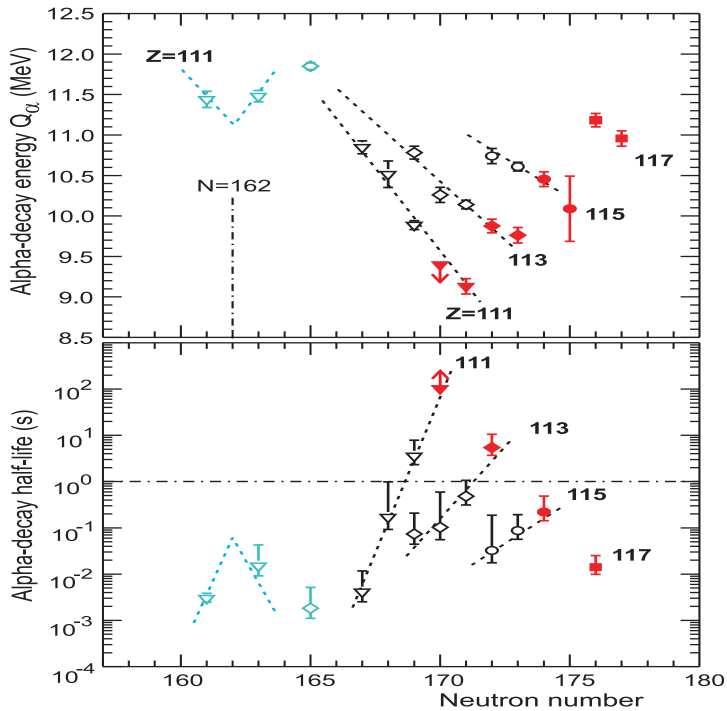

Fig. 4 Alpha-decay energies and b) half lives vs. neutron number for the isotopes of elements with Z=111-117. All the nuclides with N>165 were produced in ^{48}Ca induced reactions. In red are shown the results obtained in the present work. In the plot (b) the values of T_α(exp) are given only for the nuclei produced in the decay of the isotope of ^{293}117 (5 events). Figures are taken from [10].

the superheavy nuclei with respect to SF. By comparing the experimental and theoretical α-particle energies in Fig. 2, one sees that the macroscopic-microscopic calculations of the masses of the superheavy nuclei [11] are in a good agreement with our experiment for all the nuclei in the decay chains of the isotopes of element 117. The production cross sections for the nuclei of element 117 in the reaction ^{249}Bk+^{48}Ca are σ =0.5(+1.1,-0.4) pb and σ=1.3(+1.5,-0.6) pb at E*=35 MeV and E*=39 MeV, respectively. These results are similar to previously measured cross sections for the reactions of ^{233,238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm and ^{249}Cf targets with ^{48}Ca projectiles [1].

In summary, we have synthesized a new chemical element with atomic number 117 in the fusion of ^{249}Bk and ^{48}Ca. Two isotopes of element 117, with atomic masses 293 and 294 were observed to undergo α-decay with E_α=11.03(8) MeV and 10.81(10) MeV and half lives 14(+11,-4) ms and 78(+370,-36) ms, respectively. Their sequential α-decay chains ended in spontaneous fission of ^{281}Rg (T_{SF} ~ 26 s) and ^{270}Db (T_{SF} ~ *1d*s), respectively. Our knowledge of the properties of odd-Z nuclei in the region of the most neutron-rich isotopes of elements 105 to 117 is significantly expanded by our eleven newly identified isotopes which have increased stability with larger neutron number N. Investigations of the chemistry of superheavy elements and their place in the Periodic Table are opened up by their longer half-lives. The new isotopes, with superheavy nuclides previously synthesized in ^{48}Ca reactions, demonstrate the critical role of nuclear shells and provide experimental verification for the existence of the predicted "Island of Stability" for superheavy elements.

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