Discovery of the New Element Z=117 and Confirmation of 115

Yu. Ts. Oganessian¹, J. H. Hamilton² and V. K. Utyonkov¹ for the collaboration

¹Joint Institute for Nuclear Research, RU-141980, Dubna, Russian Federation,
²Department of Physics and Astronomy, Vanderbilt University, USA and Collaboration with Oak Ridge National Laboratory, Lawrence Livermore National Laboratory and Research Institute of Atomic Reactors
j.h.hamilton@vanderbilt.edu

Abstract. The discovery of the new chemical element with atomic number Z=117 is presented. The isotopes 288\(^{117}\) and 294\(^{117}\)were 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 superheavy nuclei toward N=184. The reaction \(^{48}\)Am+\(^{28}\)Ca was studied at three energies. Twenty one decay chains of \(^{288}\)115 and one decay chain of \(^{288}\)115 were observed to confirm the earlier discoveries of Z=115 and 117.

1 Introduction

The shell structure of neutrons and protons strongly influences the stability of superheavy elements (SHE) see e.g., [1,2]. Without shell structure corrections, the liquid drop model predicts nuclei with Z≥100 should not exist. Theoretical predictions indicate that N=184 should be the next spherical magic number that would give special stability to nuclei with different calculations predicting proton magic numbers of 114, 120 and 126 to form the 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.

Cold fusion reactions (one neutron evaporation) between doubly-magic \(^{208}\)Pb and singly-magic \(^{209}\)Bi target nuclei and stable neutron-rich projectiles such as \(^{66}\)Ni and \(^{70}\)Zn were used to synthesize new heavy elements with Z= 108 - 113 and N≤165 [3, 4], stabilized by the Z=108 and N=162 shell gaps for deformed shapes. The production cross sections dropped dramatically with increasing Z, such that practically synthesis was not possible for heavier elements.

The Flerov Laboratory of Nuclear Reactions (JINR) [1, 5] pioneered a new method of synthesizing superheavy elements, with Z≥112 and neutron numbers closer to the predicted spherical shell closure at N=184. Four new isotopes of element Z=112 and fourteen isotopes of new elements with Z=113 to 116 and 118 were identified [1] by using heavy-ion fusion reactions of doubly-magic \(^{48}\)Ca projectiles and long lived (years) actinide targets of U to Cm and Cf, respectively. The element with Z=117 was missing because there is no long lived berkelium for a target. 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 JINR [1,5] identification and decay properties of the Z=112, 114 isotopes have been recently confirmed in several independent experiments [6-9]. The synthesis of \(^{293,294}\)117 (N=176,177) isotopes in the \(^{48}\)Ca + \(^{209}\)Bk 4n and 3n reactions are presented here. No (p,xn) reactions to known isotopes of 116 were seen so our 117 assignment is correct. The observed longer half-lives for the \(\alpha\) decay chains show increasing stability for our eleven new isotopes that end in the spontaneous fission (SF) of \(^{281}\)Rg (T\(_{SF}\) = 26 s) and \(^{270}\)Db (T\(_{SF}\)≈1 d) \[10,11\]. The decisive role of the shell effects in the stability of the heaviest nuclei approaching N = 184 is strongly supported by our new data.

Studies of the \(^{241}\)Am and \(^{29}\)Ca reactions at three excitation energies yield 3, 6 and 12 decay chains of \(^{288}\)115 to confirm the discovery of this new element.

2 Experimental procedures
The $^{249}$Bk material ($T_{1/2} = 320$ d) was produced at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). Irradiation of Cm and Am with a flux of $2.5 \times 10^{15}$ n/cm$^2$/s were made as part of an ongoing 8 month $^{252}$Cf production campaign. After a three-month cooling period, extensive chemical separations at the Radiochemical Engineering Development Center at ORNL yielded 22.2 mg of $^{249}$Bk with only 1.7 ng of $^{252}$Cf, and no other detectable impurities.

Six arc-shaped $^{249}$Bk targets, each with a thickness of 0.31 mg/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. The targets mounted on the perimeter of a disk were rotated at 1700 rpm perpendicular to the beam direction. In addition, the beam was wiggled vertically up and down over the target. The experiments were performed employing the heavy-ion cyclotron U-400 at JINR and the Dubna Gas-Filled Recoil Separator [1,12].

The basic features of our work are presented here with details in refs. 10,11. After passing through the separator with an overall transmission about 35% evaporation residues (ER) were registered by a time-of-flight system with a detection efficiency of 99.9%. Then they 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 as seen in Fig. 1. The position-averaged detection efficiency was 87% of $4\pi$ for $\alpha$-particles emitted from implanted nuclei.

The total energy of an $\alpha$-particle detected only by a side detector (its position was lost) was estimated as a sum of the energy measured by the side detector and half of the threshold energy (≈0.5 MeV), with a total energy uncertainty of $\pm 0.4$ MeV. The position resolution (FWHM) of the strip detector was $\leq 1.2$ 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 minutes. After a recoil signal was detected with an implantation energy expected for $Z=117$ ERs, followed by an $\alpha$-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 the $^{48}$Ca beam was switched off to reduce the background. A 70 day irradiation with 252-MeV $^{48}$Ca projectiles was performed to give a total beam dose of $2.4 \times 10^{19}$. At this energy 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]).

### 3 Results

Five position-correlated decay chains were observed in the 252-MeV $^{48}$Ca irradiation. In each case, two or three $\alpha$-decays were observed between the time of arrival of the ER and the detection of SF. Fig. 2 gives the averaged decay properties of the five events assigned to the $^{293}$117 isotope. All five decay chains ended in spontaneous fission with $T_{SF} = 26(+25,-8)$ s.

For the $E^* = 39$ MeV the maximum cross section is expected for the 4n evaporation channel. Thus the five observed decay chains are assigned to originate from the isotope $^{293}$117. This assignment is supported by the systematics of the cross sections $\sigma_{4n}(E^*)$ measured previously for the production of superheavy isotopes with $Z=108, 112-116,$ and 118 in $^{48}$Ca-induced reactions [1].
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 lifetimes (τ ≈ T_{1/2}/ln 2) and α-particle energies are shown in black and blue, respectively. Taken from [10].

by calculations made for the evaporation residues of the reaction ^249\text{Bk} + ^{48}\text{Ca} [14-16], and by the result of our ^249\text{Bk} + ^{48}\text{Ca} experiment performed at lower beam energy (discussed next). The total numbers for random sequences [17] imitating each of the observed five decay chains were calculated to be 6 × 10^{-6}, 10^{-3}, 10^{-5}, 3 × 10^{-11} and 3 × 10^{-11}.

Then, the experiment was run at a ^{48}\text{Ca} energy of 247 MeV for 70 days with a total beam dose of 2 × 10^{19}. The excitation energy of approximately 35 MeV of the compound nucleus ^{297}117 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_α=9.00 MeV instead of undergoing SF, followed by two more α-transitions, and after about 33 hours, a fission event. The latter observation is significantly different from the known decay properties of ^{281}111 and ^{290-293}117 nuclei [1]. We assign the chain to the decay of the neighboring odd-odd nucleus ^{294}117. The chance probability for this chain was 6 × 10^{-11}.

The decay properties of the neighboring ^{293}117 and ^{294}117 isotopes, their daughters ^{293}115 and ^{290}115, and granddaughters ^{287}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 ^{284}111 (see fig. 2). However, the heavier isotope ^{285}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. The T_{SF} have minimum values because the effect of nuclear shells is at a minimum. For example, the Z=110 darmstadtium isotopes with N=167 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 [19-21] 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-life exceeds 10^7 s for ^{268}\text{Db}. The ^{249}\text{Bk}+^{48}\text{Ca} reaction yields daughter nuclei that have 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 and the ^{294}117 chain are far longer than those in the ^{293}117 chain, see Figs. 2 and 3. 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. The odd proton 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 × 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 "keep" 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 additional unpaired neutron in its 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. 3a,b. As N increases, Q\(_{α}\) decreases and T\(_{α}\) increases. 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. 3, are α-emitters; with T\(_{α}\) smaller than T\(_{SF}\) to indicate the high stability of the superheavy nuclei with respect to SF. The macroscopic-microscopic calculations of the masses of the superheavy nuclei [13] are in a good agreement with our experiment for all the isotopes in the decay chains of element 117 (see Fig. 2). The production cross sections for the nuclei of element 117 in the reaction \(^{286}\)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 order to firmly establish the discovery of the new element Z=115 [19,20], the reaction \(^{243}\)Am+\(^{48}\)Ca\(→^{288}\)115 has been studied at excitation energies E* of 40, 36 and 33 MeV in the same way as the 117 experiment discussed above. The decay chains shown in Fig. 4 were observed at E*=40 MeV with α energies and life-times the same as seen earlier [20]. At 36 MeV six decay chains were observed and at 33 MeV 12 decay chains. The beam dosages were 3.7×10\(^{18}\), 3.3×10\(^{18}\) and 1.7×10\(^{19}\). All of the 21 chains have same α energies and half-lives within the error limits. The cross-sections for these energies are shown in Fig. 5 along with theoretical calculations [22]. Also shown is the cross section of the 2n channel to \(^{289}\)115 which was observed at E*=33 MeV. This is the same isotope populated in the α decay of \(^{291}\)117. These excitation data provide clear evidence to confirm the discovery of the elements Z=115 and 117.

In summary, we have synthesized a new chemical element with atomic number 117 in the fusion of \(^{248}\)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), 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, together with superheavy nuclides previously synthesized in reactions with \(^{48}\)Ca, demonstrate the critical role of nuclear shells and provide experimental verification for the existence of the predicted "Island of Stability" for superheavy elements. In a recent experiment, we confirmed the discovery of Z=115 by bombarding \(^{244}\)Am with \(^{48}\)Ca at excitation energies of 40, 36 and 33 MeV. At these respective energies we have seen three, six, and twelve decay chains of \(^{288}\)115, and determined their cross sections.

We are grateful to the JINR Directorate and the U-400 cyclotron and ion source crews for their continuous support of the experiment. We acknowledged the support of the Russian Federation Agency of Atomic Energy, grants RFBR Nos. 07-02-00029, 09-02-12060, 09-03-12214, the U.S. Department of Energy through Contracts DE-AC05-00OR2272 (ORNL) and DE-AC52-07NA27344 (LLNL), and grants DE-FG-05-88ER40470 (Vanderbilt University) and DE-FG07-01AL67358 (UNLV). These studies were performed in the framework of the Russian Federation/U.S. Joint Coordinating Committee for Research on Fundamental Properties of Matter.
FUSION11

Fig. 4. Two of the three decay chains of $^{288}\text{I}^{115}$ observed in the reaction of $^{243}\text{Am}$ and $^{48}\text{Ca}$ at an excitation energy 40 MeV.

Fig. 5. Experimental and theoretical [22] (dotted curves) cross sections in pb for 2n, 3n, 4n and 5n reactions of $^{48}\text{Ca}$ with $^{245}\text{Cm}$, $^{243}\text{Am}$ and $^{242}\text{Pu}$ targets. The $^{243}\text{Am}$ results include the earlier [19,20] results and our new results.

References