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Discovery of elements 113 - 118

V.K. Utyonkov [*,1] , Yu.Ts. Oganessian [1] , S.N. Dmitriev [1] , M.G. Itkis [1] ,
K.J. Moody [2] , M.A. Stoyer [2] , D.A. Shaughnessy [2] ,
J.B. Roberto [3] , K.P. Rykaczewski [4] , J.H. Hamilton [5]
for the collaboration [1,2,3,4,5,6,7]

[1] *Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna, Russian Federation*
[2] *Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, Livermore, California, USA*
[3] *Science and Technology Partnerships Directorate, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA*
[4] *Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA*
[5] *Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee, USA*
[6] *Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee, USA*
[7] *Research Institute of Atomic Reactors, Dimitrovgrad, Russian Federation*

[*] *e-mail: utyonkov@jinr.ru*

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Review of discovery and investigation of isotopes of elements 113-118 produced in the reactions of ^{48}Ca with target nuclei ^{238}U- ^{249}Cf is presented. The synthesis of the heaviest nuclei, their summary decay properties, and methods of identification are discussed. The radioactive properties of the new nuclei give evidence of the significant increase of the stability of the heavy nuclei with rise of their neutron number and approaching magic number N=184.

Keywords : superheavy elements; α-decay ; spontaneous fission.

Introduction

In 1998, experiments aimed at the synthesis of superheavy nuclei (SHN) were initiated in FLNR, JINR. In the complete-fusion reactions of ^{48}Ca projectiles with actinide target nuclei, six new superheavy elements having atomic numbers 113 through 118 were synthesized for the first time. Radioactive properties of more than 50 new heaviest nuclides were explored [1,2]. Several of these reactions were later studied in two types of experiments: in the chemical study of superheavy elements and at separators SHIP, BGS, TASCA, and GARIS.

Radioactive properties of the synthesized nuclides demonstrate a substantial increase of nuclear stability with increase of neutron number and closer approach to the predicted shell N=184. New observations establish a consistent pattern of nuclear properties in the area of the heaviest nuclides. They demonstrate the decisive role of nuclear shells and provide experimental proof of the existence of the predicted "Island of stability" of superheavy elements.

Experimental results

Decay chains of all of the nuclei synthesized hitherto in the (^{238}U-^{249}Cf) + ^{48}Ca reactions are terminated by spontaneous fission (SF) of previously unobserved isotopes. Thus, the method of the detection of consecutive decays

that lead to the known nuclei can be applied for the identification of the new ones only after independent identification of at least one isotope in the decay chain, e.g., by chemical identification of its atomic number. However, in addition to this method, approximately 20 other criteria were developed by IUPAC/IUPAP in 1991 "that must be satisfied for the discovery of a new chemical element to be recognized" [3]. Most of these criteria demonstrate, beyond a reasonable doubt, the synthesis of the nuclides with atomic numbers Z=113-118.

In our experiments, we employed the Dubna gas-filled recoil separator (DGRFS), that allows the separation of the products of complete fusion reactions from the beam of bombarding ions, elastically-scattered nuclei, and products of incomplete fusion. The detection system includes proportional chambers used to measure the time of flight (TOF) of particles and several semiconductor detectors with position-sensitive strips. The principle of operation of the separator is selection of products of the complete-fusion reaction by their charge state q in a rare gas and kinematic characteristics (mass of recoil nucleus m and its velocity v) in accordance with the separator magnetic rigidity $B\rho = mv/q$ (note, q depends linearly on v). These values are calculated for the xn-reaction channel when setting the separator's parameters. The DGFRS strongly separates forward-peaked evaporation residues (ER), products of complete-fusion reactions, within a narrow angle with a huge suppression of the products of the transfer reactions and even incomplete fusion, e.g., αxn reactions. The TOF selection in the existing separators may be complemented and reinforced by the combined measurement of recoil energy and TOF. Note, the production properties "separator", "mass separation", "angular selection", and "TOF selection" were called "assignment properties" in [3].

The first superheavy nucleus ^{289}Fl was discovered in the ^{244}Pu(^{48}Ca,3n) reaction studied at DGFRS (here and after we refer to reviews [1,2] containing references to most of earlier experimental data). The decay properties of ^{289}Fl and descendant nuclei are shown in figure 1.

With an increase of the excitation energy E^* of the compound nucleus (CN), the yields of different nuclides vary. At the lowest E^* values, the nuclides with relatively lower α-particle energies and longer half-lives T are produced (viz., ^{289}Fl). At larger projectile energies, the yield of these nuclei decreases, but the yield of other nuclides (^{288}Fl and ^{287}Fl), with higher E_α and shorter T values, increases and then decreases again with the further increase of the excitation energy. The decay properties of the nuclides, consecutively appearing with the rise of E^*, are different (compare the E_α and T values and decay modes of all of the nuclei in the chains in figure 1). In particular, the decay chains of even-Z, odd-N nuclei are longer indicating the hindrance of fission process as compared to the even-even nuclei decays. However, the difference in E_α and T values of parent nuclei $^{287-289}$Fl is rather small (by about 0.1 MeV and by factor of about 2, respectively); this observation excludes the assignment of the observed nuclides to differing types of reactions (e.g., considering that one isotope is produced in the xn channel and others – in the pxn or αxn channels).The production cross-section values as well as energies of bombarding particles [3] are comparable for all of the studied reactions, which might indicate the identity of the mechanism of all of the reactions.

For synthesis of the lighter isotopes of Fl, the reactions with lighter target nu-

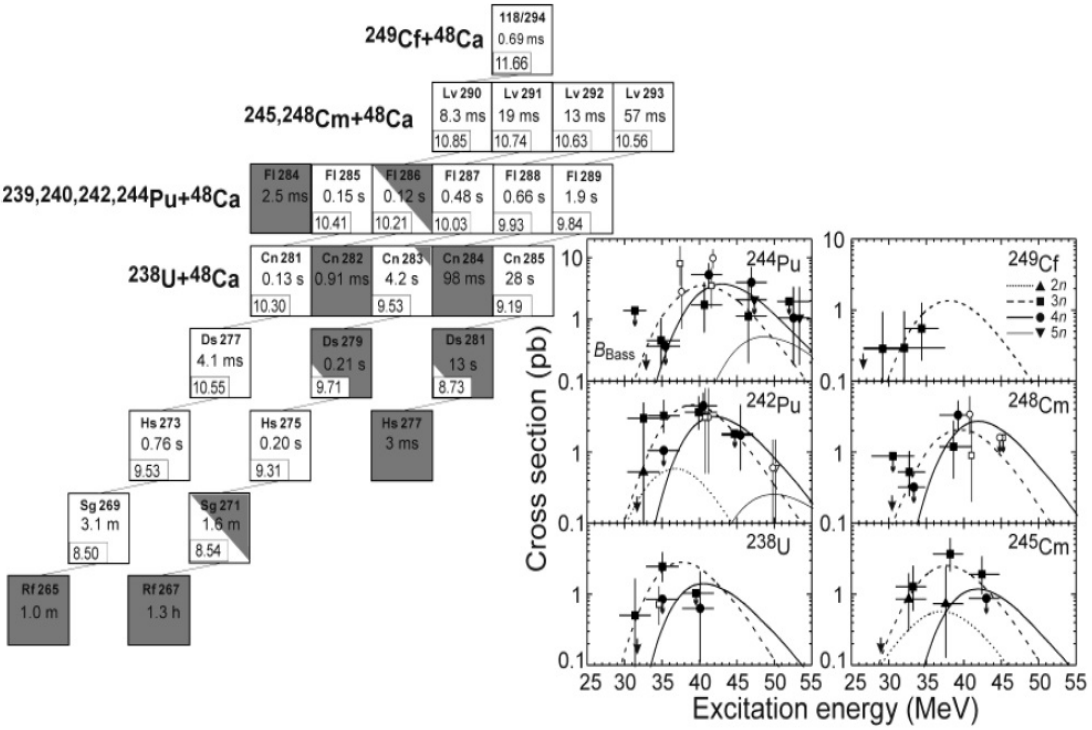

Figure 1. Left-hand side: Summary decay properties of the isotopes of even-Z elements synthesized in the given reactions. The energies of particles and half-lives are given for α-emitters (open squares). The SF nuclei are marked by grey squares. Right-hand side: Excitation functions for the 2n- to 5n-evaporation channels from the complete-fusion reactions (^{238}U - ^{249}Cf) + ^{48}Ca measured at DGFRS (solid symbols) and SHIP, BGS, and TASCA (open symbols). Lines show results of calculations [4]. Vertical error bars correspond to statistical uncertainties of the DGFRS experiments and available data from other experiments. Horizontal error bars represent the range of excitation energies. The energies at the Bass barriers (B_{Bass}) [5] are shown by arrows.

clei were studied [1,2]. In the ^{242}Pu+^{48}Ca reaction, the isotope ^{287}Fl was predominantly observed at low excitation energies while the lighter isotope (^{286}Fl) was obtained at higher projectile energies. Further increase of ^{48}Ca energy resulted in observation of the next isotope ^{285}Fl, first at BGS [6]. The same isotope was also produced at DGFRS in the reaction with a lighter Pu-isotope as the product of the ^{240}Pu(^{48}Ca,3n)^{285}Fl reaction [7]. In the same experiment carried out at higher ^{48}Ca energy as well as in the ^{239}Pu+^{48}Ca reaction, a SF nuclide was assigned to the new isotope ^{284}Fl. Thus, the excitation functions ("yield curve" in [3]) have been measured for the reactions with Pu isotopes, which, together with decay properties of the observed nuclides ("kind of decay", "branching ratio", "half-life", and "α-particle energy" in [3]), demonstrate the production of the neighboring isotopes of the same element in each reaction.

Two Fl-isotopes were observed in cross bombardments ^{244}Pu(^{48}Ca,5n), ^{242}Pu(^{48}Ca,3n)$\rightarrow^{287}$Fl and ^{242}Pu(^{48}Ca,5n), ^{240}Pu(^{48}Ca,3n)$\rightarrow^{285}$Fl, respectively. Moreover, Cn isotopes ^{282}Cn and ^{283}Cn, daughter nuclei of ^{286}Fl and ^{287}Fl, were produced in another cross-bombardment reaction ^{238}U+^{48}Ca. The identity of the decay properties observed in cross bombardments [3] definitely proves that the same nuclei were produced in different reactions. This proves that all of these nuclei were produced in the same type of reaction.

The isotopes of Lv were synthesized at DGFRS in the ^{248}Cm+^{48}Ca and ^{245}Cm+^{48}Ca reactions at different excitation energies of CNs [1,2] (figure 1). Again, the decay properties of descendants of the isotopes ^{290}Lv –^{293}Lv were

in full agreement with those found in the ^{244}Pu+^{48}Ca, ^{242}Pu+^{48}Ca and ^{238}U+^{48}Ca cross-bombardment reactions.

By now, five decay chains of element 118 were synthesized in the ^{249}Cf(^{48}Ca,3n)^{294}118 reaction studied at DGFRS[1,2,8]. Note, the descendant SF isotope ^{282}Cn was produced in the following cross reactions: ^{238}U(^{48}Ca,4n)^{282}Cn, ^{242}Pu(^{48}Ca,4n)^{286}Fl, ^{245}Cm(^{48}Ca,3n)^{290}Lv, and ^{249}Cf(^{48}Ca,3n)^{294}118.

Assignment of all of the synthesized nuclides to the products of the xn-reaction channel is justified by characteristics of DGFRS, excitation-function measurements, production of most of the nuclides in cross bombardments, as well as by decay properties of nuclei, especially their measured α-particle energies (see figure 2). The α-particle energies of even-Z isotopes investigated so far are identical and form single peaks in both cases — after production of nuclei in primary reactions and after α decays of parent nuclei (with several lower-energy peaks observed for even-odd ^{291}Lv, ^{289}Fl, and ^{283}Cn). Calculation of the Q_α values from α peaks appears to be reasonable for even-even isotopes as well as for even-Z odd-N nuclei because the energies of odd isotopes have intermediate values between those of even-even nuclides, which is in agreement with the observations for the lighter nuclei (see figures 21-26 in [9]).

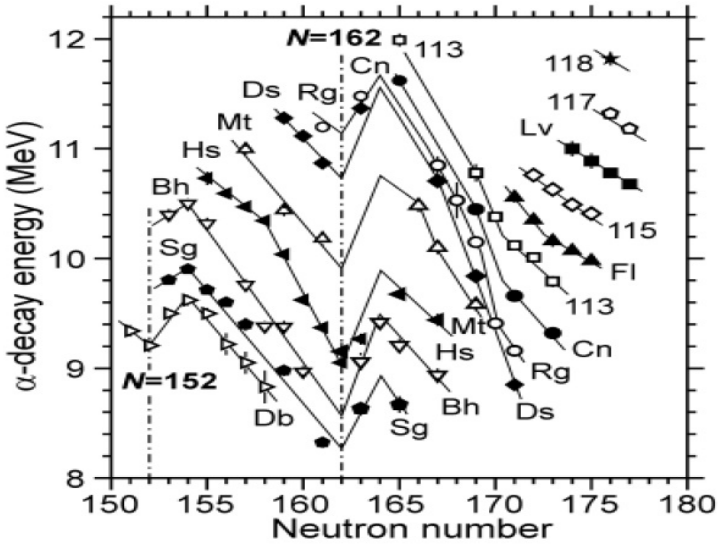

Figure 2. Measured α-decay energy vs. neutron number for the isotopes of elements 105-118 (filled and open symbols refer to even-Z and odd-Z nuclei, respectively [1,2,6-9]. The lines are drawn to guide the eye.

An important feature observed in these experiments is the fact that $Q_\alpha(N)$ systematics for isotopes of neighboring elements never intersect (see figures 25-26 in [9]). Moreover, the Q_α values for isotopes of Sg and Hs perfectly follow the trend of variation of $Q_\alpha(N)$ at the crossing of the magic neutron number N=162 (identical to what is observed for the isotopes of Ds and Rg as well as for numerous nuclides with $N \approx 152$ and 126). Therefore, the systematics of α-decay energies of even-Z elements and their descendants provides proof of identification of the Z and N of these elements. In addition, the mass numbers of nuclei can be easily established by considering their decay properties (e.g., SF is much more probable for even-N and/or Z isotopes, see figures 1 and 3) and excitation functions (e.g., products of the ^{244}Pu+^{48}Ca might be assigned to the 1n- to 3n-evaporation channel instead of 3n to 5n, but in this assumption, the product of

the ^{245}Cm(^{48}Ca,2n)^{291}Lv reaction should originate from the radiative-capture channel, which was not registered even in the cold-fusion reactions of heavy ions with ^{208}Pb and ^{209}Bi).

Furthermore, the results of experiments aimed at the chemical characterization of Cn "establish element 112 as a typical element of group 12" [2]. Thus, the atomic numbers of all of the isotopes, starting with ^{209}Lv and ending with Rf, were determined by the genetic relation [3].

In most of the reactions with even-Z target nuclei, two to three different parent nuclides were produced in the 2n- to 5n-evaporation channels. Thus, it would be reasonable to expect similar observations for the reactions with odd-Z target nuclei (see figure 3).

The heaviest odd-Z nuclei were synthesized in the ^{249}Bk(^{48}Ca,3-4n)^{293,294}117 reaction[2]. Only at high excitation energies we registered shorter decay chains of the even-N isotope ^{293}117; each consisted of three consecutive α decays terminated by SF of ^{281}Rg or 5-ms ^{277}Mt instead of α decay observed for their odd-N neighbors ^{280,282}Rg and ^{276,278}Mt.

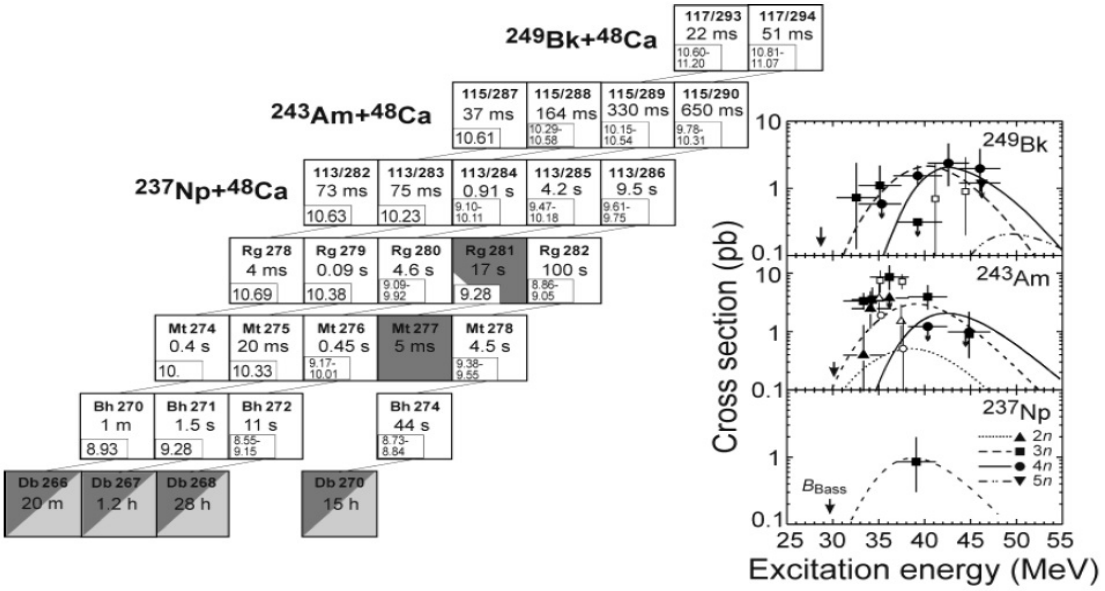

Figure 3. Same as figure 1, but for the isotopes of odd-Z elements. For Db isotopes only SF was observed however, electron capture (EC) cannot be excluded.

The observations of elements 113 and 115 were first reported in 2003-2004 [1,2]. In the reaction ^{243}Am+^{48}Ca, these two new elements were simultaneously synthesized for the first time. In new series of experiments, at the lowest ^{48}Ca energies, we detected decay chains consisting of two α decays ending in SF. Similar α-α-SF decay chains were observed for descendant nuclei in the chain starting with the isotope ^{293}117. Therefore, assignment of these chains to the product of the 2n-reaction channel, ^{48}115, seems to be the most reasonable. Note, the product of the 2n-reaction channel was clearly observed with the same yield in the ^{245}Cm(^{48}Ca,2n)^{291}Lv reaction [1,2]. At higher projectile energies, decay chains of ^{288}115, the product of evaporation of three neutrons, were registered that undergo five α-decays followed by SF. At the highest bombarding energy, we detected decay chains of ^{287}115.

The same decay chains were later produced in experiments with ^{243}Am which were performed at TASCA [10] and BGS [11]. In [12], the decay times of

nuclei in the short decay chains were re-analyzed with a newly suggested method which led to the interpretation that some of these chains might start from the isotope $^{288}115$. However, we consider that this method is incorrect. The analysis of data by method of conventional statistics demonstrate reasonable congruence of α-particle energies and decay times of nuclei assigned to $^{289}115$, $^{285}113$, and ^{281}Rg observed in the (^{249}Bk+ ^{48}Ca) and (^{243}Am+ ^{48}Ca) reactions [13].

For investigation of the region of neutron-deficient odd-Z SHN, we also studied the ^{237}Np(^{48}Ca,3n)$^{282}113$ reaction [1,2]. Similar to even-Z elements, the assignment of all of the odd-Z nuclides to the products of the xn-reaction channel is based on characteristics of the DGFRS, excitation-function measurements for the reactions with ^{243}Am and ^{249}Bk, and production of $^{289}115$ in cross bombardments, as well as decay properties of nuclei. First of all, the decay properties of all of the odd-Z nuclei clearly differ from those observed for $^{290-293}$Lv, $^{285-289}$Fl, etc. (potential products of the pxn-reaction channels or EC of parent nuclei). For odd-Z nuclei, the α spectra are more complex. However, the calculation of their α-decay energies from the highest measured α-particle energies [1,2] appears to be justified. For example, the Q_α values for odd-odd nuclei $^{288}115$-^{272}Bh, estimated or measured from the observed α-γ coincidences [10,11] are in good agreement with such calculations. The systematics (figure 2) demonstrates that the α-decay energies of all of the odd-Z nuclei are located in between and are in good agreement with the interpolations/extrapolations from neighboring even-Z nuclides with $(Z \pm 1)$ and the same neutron number. The Q_α values for Bh isotopes are located between $Q_\alpha(N$ systematics for Sg and Hs and perfectly correspond to the pattern of variation of $Q_\alpha(N$ for Bh isotopes at the crossing of N=162.

Moreover, the terminal SF nuclide ^{268}Db in the decay chain of $^{288}115$ produced in the ^{243}Am+ ^{48}Ca reaction was chemically characterized as a transactinide element [1,2]. In these experiments, the products from the irradiation of ^{243}Am by ^{48}Ca projectiles, having the same ^{248}Ca energy as in the DGFRS experiments, were collected within a larger acceptance angle of $\pm 12.5^o$ that would result in increase of collection efficiency for transfer-reaction products by a factor of more than 20. Nevertheless, after chemical extraction of transactinide elements, the SF nuclide was observed with the same decay properties and with the same cross section as at DGFRS. All of these factors allowed for the conclusion that the same isotope was observed in both the physical and chemical experiments [1,2]. Simultaneously, all of the precursors discovered in [1,2] were identified by establishing the genetic link between the ancestor and the descendant [3].

In addition one could mention that the properties of α-decaying even-even SHN are in agreement with the empirical Geiger-Nuttall relationship (see figure 26 in [1]). For SF nuclei, the TKE increases with increasing Z in agreement with the previously established dependence of TKE vs. parameter $Z^2/A^{1/3}$ (see figure 29 in [1]).

Therefore, conformity with all of the above-mentioned criteria demonstrates that the observed SHN originate from the complete-fusion reactions followed by the evaporation of the neutrons. Furthermore, the decay properties of $^{294}117$, $^{291-293}$Lv, $^{287-289}115$, $^{285-289}$Fl, and ^{283}Cn and their descendants were determined in different laboratories with use of different techniques (separators DGFRS, SHIP, TASCA, BGS, GARIS, and chemistry setups) that is in accordance

with demand of reproducibility for recognition of the discovery of new elements (see [1,2,6-8,10-12,14] and references therein).

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