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RESULTS OF THE EXPERIMENT ON CHEMICAL IDENTIFICATION OF Db AS A DECAY PRODUCT OF ELEMENT 115

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Результаты эксперимента по химической идентификации элемента Db как продукта распада элемента 115

Впервые проведена химическая идентификация Db — конечного продукта распада изотопа 115-го элемента в реакции $^{243}\mathrm{Am}(^{48}\mathrm{Ca}, 3n)\,^{288}115$. Эксперимент выполнен на ускорителе У-400 ЛЯР, ОИЯИ. Мишень из ^{243}Am (1,2 мг/см^2) облучалась пучком $3{,}4 \cdot 10^{18}$ ионов с энергией 247 МэВ в середине слоя мишени. Продукты ядерных реакций собирались в медном сборнике, поверхностный слой которого после снятия на токарном станке растворяли в концентрированной HNO_3. Элементы 5-й группы сорбировались на катионообменной смоле Dowex 50×8 с последующей их десорбцией 1 М раствором HF, образующей анионные фторкомплексы с элементами 5-й группы. Элюент упаривали на полиэтиленовой пленке (0,4 мкм), которую помещали между парой полупроводниковых детекторов, окруженных ^{3}He-счетчиками для регистрации α-частиц, осколков деления и нейтронов. В эксперименте наблюдалось 15 событий спонтанного деления с $T_{1/2} = 32^{+11}_{-7}$ ч, которые мы относим к ^{268}Db. Сечение образования в реакции $^{243}\mathrm{Am} + {}^{48}\mathrm{Ca}$ равно $4,2^{+1,6}_{-1,2}$ пб. Полученные результаты согласуются с результатами эксперимента по синтезу 115-го элемента на газонаполненном сепараторе ядер отдачи, в которых впервые наблюдался изотоп ^{268}Db после пяти последовательных α-распадов материнского ядра. Таким образом, данные настоящего эксперимента являются независимым доказательством синтеза 115-го, равно как и 113-го, элемента в реакции $^{243}\mathrm{Am} + {}^{48}\mathrm{Ca}$.

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Dmitriev S. N. et al. E12-2004-157
Results of the Experiment on Chemical Identification of Db as a Decay Product of Element 115

For the first time the chemical identification of Db as the terminal isotope of the decay element 115 produced via the $^{243}\mathrm{Am}(^{48}\mathrm{Ca}, 3n)\,^{288}115$ reaction was realized. The experiment was performed on the U400 cyclotron of FLNR, JINR. The ^{243}Am target was bombarded with a beam dose of $3.4 \cdot 10^{18}$ ^{48}Ca projectiles at an energy of 247 MeV in the center of the target. The reaction products were collected in the surface of a copper catcher block, which was removed with a lathe and then dissolved in concentrated HNO_3. The group 5 elements were separated by sorption onto Dowex 50×8 cation-exchange resin with subsequent desorption using 1M HF, which forms anionic fluoride complexes of group 5 elements. The eluant was evaporated onto 0.4 μm thick polyethylene foils which were placed between a pair of semiconductor detectors surrounded by ^{3}He neutron counters for measurement of α particles, fission fragments and neutrons. Over the course of the experiment, we observed 15 spontaneous fission events with $T_{1/2} = 32^{+11}_{-7}$ h which we attribute to ^{268}Db. The production cross section for the $^{243}\mathrm{Am} + {}^{48}\mathrm{Ca}$ reaction was $4.2^{+1.6}_{-1.2}$ pb. These results agree with the original element 115 synthesis experiment where ^{268}Db was first observed as the terminal isotope following the five consecutive α decays from the $^{288}115$ parent nucleus at the Dubna gas-filled separator. The data from the present experiment give independent evidence fot the synthesis of element 115 as well as element 113 via the $^{243}\mathrm{Am} + {}^{48}\mathrm{Ca}$ reaction.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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Relatively long half-lives of isotopes of elements 105–116 obtained [1–6] in reactions between ^{48}Ca and ^{248,245}Cm, ^{244,242}Pu, ^{243}Am and ^{238}U and chemical properties of SHE predicted theoretically provide possibilities for new experiments devoted to the chemical identification of SHE, study of their chemical properties, combination of chemical and physical methods for SHE synthesis.

All of the new nuclides were synthesized employing physical techniques. Thin target layers (~ 0.3 mg/cm^2) of the isotopically enriched actinide isotopes were irradiated by ^{48}Ca beam of an accurately preset energy. Recoil nuclei knocked out of the target were separated from ^{48}Ca ions and various reaction products by means of kinematic gas-filled separator that was tuned to transmit the complete fusion products with an efficiency of about 40%. The decay of nuclei was registered by position-sensitive silicon detectors mounted in the separator's focal plane. Correlated decays of single atoms, i.e., chains of sequential α decays and spontaneous fission ($\alpha - \alpha - \alpha - \ldots$ SF) registered by the detector array refer to the unknown nuclides. Their identification is based on their radioactive decay properties and the mechanism of the reaction leading to their production, in particular, on the characteristic dependence of the yield of the neutron-evaporation products on the excitation energy of the compound nucleus. Investigating these dependences needs time-consuming measurements of the production cross sections of the nuclei of interest at various energies of the ^{48}Ca ion beam.

At the same time, chemical identification of any isotope in the observed decay chains could give us the identification of atomic numbers of all the nuclei in the chain and provide an independent evidence of the discovery of the new element(s). Such an opportunity is open for element 115.

An isotope of element 115 with mass number 288 was synthesized [6] in the reaction $^{48}\text{Ca} + {}^{243}\text{Am} \rightarrow {}^{288}115 + 3n$. It undergoes five sequential α decays, that end in spontaneous fission of ^{268}Db (Fig. 1).

The sum time of the five α-transitions is about 20 s. The half-life of the terminal spontaneously fissioning nuclide ^{268}Db estimated from the three observed events is $T_{1/2} = 16^{+19}_{-6}$ h. With a few observed events, as was discussed in the paper [6], one cannot exclude that ^{268}Db could undergo α decay, leading to spontaneously fissioning ^{264}Lr. On the other hand, direct production of ^{268}Db via transfer of 25 nucleons (10 protons and 15 neutrons) to ^{243}Am target from ^{48}Ca at the energy close to the Coulomb barrier is practically impossible [7].

In the DGFRS experiment the thickness of ^{243}Am target was 0.3 mg/cm^2, the transmission efficiency was about 40%. In case of chemical experiment the target thickness can be increased up to ≈ 1.0 mg/cm^2 and efficiency of isolation can be expected about 75%. Thus 1–2 events per day could be expected (Table 1).

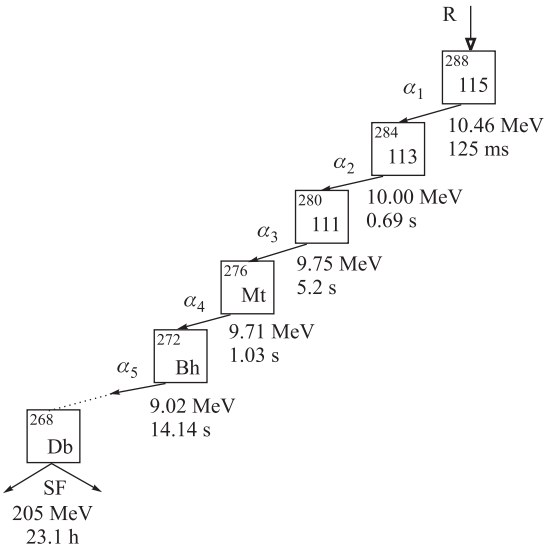

Fig. 1. Decay of $^{288}115$ observed with Dubna Gas-Filled Recoil Separator (DGFRS) [6]

Therefore, a relatively long lifetime of ^{268}Db and its quite characteristic decay mode (SF) allow us to propose [8] an experiment on chemical identification of Db as descendant product of decay of element 115 which is produced in the reaction $^{48}\mathrm{Ca} + {}^{243}\mathrm{Am}$ with the cross section of only about 3 pb ($3 \cdot 10^{-36}$ cm^2).

According to its atomic configuration ([Rn]5d^{14}6p^{3}7s^2), Db belongs to the 5th group of the Periodic Table, thus being heavier homologue of Nb and Ta. By studying the 34-s ^{262}Db [9], it was established that Db, like Nb and Ta, is

Table 1

	DGFRS	Chemistry
Target thickness	0.36 mg $\cdot$ cm^{-2}	$\sim$ 1.0 mg $\cdot$ cm^{-2}
Energy range	3.3 MeV	10 MeV
Transmission	$\sim$ 35%	$\approx$ 75% (expected)
Beam dose	$4.3 \cdot 10^{18}$	—
Events number	3	—
Decay mode	SF	SF
Half-life	16^{+19}_{-6} h	—
Cross section	$2.7^{+4.8}_{-1.6}$ pb	—

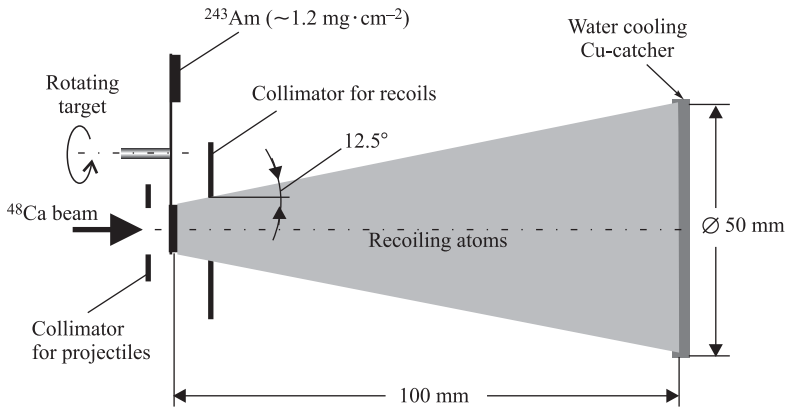

Fig. 2. The scheme of an irradiation of ^{243}Am target

well absorbed on glass from concentrated HNO_3. In the processes of extraction by Aliquat 336 [10] from the chloride solutions, its behavior is most close to that of Nb and differs from Ta and Pa (pseudo-homologue), while in the extraction from the fluoride solutions it is analogous to Nb and Ta and differs from Pa. In general, one observes the theoretically predicted [11, 12] inversion of the properties within the group of homologues with the transition from 5d to 6d elements, i.e., in its chemical properties Db is most close not to Ta but to Nb.

For the chemical identification, the element should be separated according to its group properties. For this purpose, we developed and used in experiments the method of sorption extraction of elements of group 5, as anionic fluoride complexes. Bearing in mind that the $Z = 105$ isotope of interest undergoes SF, we paid special attention to separating group 5 elements from actinides and, first of all, from spontaneously fissioning isotopes of californium, ^{252}Cf ($T_{1/2} = 2.65$ y, $b_{\mathrm{SF}} = 3.1\%$) and ^{254}Cf ($T_{1/2} = 60.5$ d, $b_{\mathrm{SF}} = 99.7\%$).

The experiment has been performed employing the beam of the FLNR (JINR) U400 cyclotron in June, 2004. The principal scheme of the set-up for target irradiation is shown in Fig. 2.

The 32-cm^2 rotating target consisted of the enriched isotope of ^{243}Am (99.9%) in the oxide form. The target material was deposited onto 1.5-μm Ti foils to a thickness of 1.2 mg/cm^2 of ^{243}Am. The target was bombarded by ^{48}Ca ions with an energy corresponding to 247 MeV in the middle of target layer and average intensity of $5 \cdot 10^{12}$ ions/s. A collimator, 10 mm in diameter, limited the irradiated area. The recoiling nuclei of the reaction products passed through

the second 12-mm collimator, 10 mm from the target, and stopped in a copper catcher. The latter, 50 mm in diameter, was positioned on the beam axis, 100 mm downstream the target. The efficiency of collecting the reaction products in such a geometry (capture angle $\pm 12.5^{\circ}$) was close to 100%. The range of recoils in copper catcher did not exceed 3–4 μm. All in all, eight identical experimental runs with duration of 20 to 45 h were performed.

In each run, after the end of irradiation, the catcher was brought to the radiochemical laboratory. The catcher surface was accurately cleaned of the aerosol particles carrying ^{243}Am and afterwards a 7- to 10-μm upper layer (corresponding to 120–180 mg of Cu) was cut from its surface using a microlathe. Then the copper chips were dissolved in HNO_3. The obtained nitric solution was a complex system containing high content of copper (catcher material), products of the reactions of ^{48}Ca with Cu, implanted fission fragments, as well as products of multinucleon transfer reactions, including the spontaneously fissioning ^{252}Cf and ^{254}Cf. For spectrometric control of the behavior of elements of group 5 and actinides, we added into the solution the aliquots of the nitrates of the radionuclides, ^{92m}Nb ($T_{1/2} = 10.15$ d), ^{177}Ta ($T_{1/2} = 56.6$ h), ^{169}Yb ($T_{1/2} = 32$ d), and ^{167}Tm ($T_{1/2} = 9.25$ d).

Isolation of group 5 elements from the nitric solution included the following basic stages:

- Separation of the reaction products from the macro component (copper) via their co-sedimentation with lanthanum hydroxide at $pH = 7$ in ammonium media; copper was kept in solution as ammonium complex. For the preparation of thin spectrometric sources with minimum content of ballast material at the final stage, the procedure was repeated. The obtained sediment was then dissolved in 2 M solution of HNO_3.

- Separation of group 5 elements from lanthanum and actinides by their sorption from nitric solution on cation-exchange resin Dowex 50×8 with further desorption of group 5 elements as fluoride-anionic complexes by 2 ml of 1 M solution HF. The obtained solution was then evaporated down to the volume of 0.1 ml.

- Preparation of the thin sources (working samples) for further measurements by depositing the solution from a capillary onto a polyethylene foil (0.4 μm thick, 15 mm in diameter), with subsequent drying in a hot helium stream.

In control experiments with non-irradiated copper catchers and added radionuclides ^{92m}Nb, ^{177}Ta, ^{167}Tm and ^{169}Yb the present technique has shown to extract group 5 elements with chemical yield of 90% and suppression of lanthanides by a factor of $\sim 10^5$. From spectrometric measurements with thin sources, ^{92m}Nb

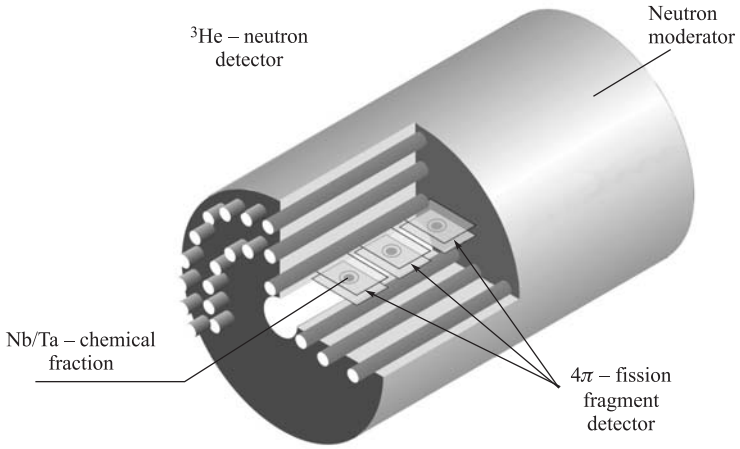

Fig. 3. Scheme of the neutron detector

and ^{177}Ta were isolated with efficiency of $(85 \pm 5)\%$ and $(75 \pm 5)\%$, respectively with suppression of actinides by a factor of $\geqslant 8 \cdot 10^3$ (the value estimated from the detection limit of ^{169}Yb of working samples).

All in all, the chemical procedure took 2 to 3 h, starting from the end of irradiation till the beginning of measurements by detectors.

For the registration of α particles and spontaneous fission fragments we used a detection module including 4 identical chambers, each with two semiconductor detectors. The detectors with an area of 6 cm^2 were mounted with 4-mm spacing in front of each other. The sample under study was put into the spacing between detectors. All the chambers were positioned inside a neutron detector, in order to register neutrons from spontaneous fission. The neutron detector (Fig. 3) had 72 ^{3}He counters that were positioned in three layers apart from cylinder axis. The detector array was calibrated with sources of ^{248}Cm and ^{252}Cf. The efficiency of detecting fission fragments by semiconductor detectors was about 90%, neutrons were detected with $\sim 40\%$ efficiency. In the course of the 330-h test run before the experiment no background events were detected.

At eight runs with irradiation of ^{243}Am target by ^{48}Ca ions (with a total dose of $3.4 \cdot 10^{18}$) we detected 15 events of spontaneous fission. The measurements were carried out for 910 h. All the 15 events were appeared in 174-h interval after the beginning of the measuring.

No SF events were detected in subsequent 736 h. The time of irradiations, beam dose and the number of SF events observed for each sample are given in Table 2. The half-life of 32^{+11}_{-7} h determined from the time distribution of SF

Table 2. **The results of the experiment**

N	Irradiation time, h	Beam dose of ^{48}Ca ions, ions	Energy of fission fragments E_1/E_2, MeV	Number of neutrons detected for each SF event	Time of detection, h
1	20	$2.5 \cdot 10^{17}$	120/126	2	20
2	22	$3.7 \cdot 10^{17}$	–/86	1	74
3	22	$3.4 \cdot 10^{17}$	131/124 116/122	1 2	15 72
4	22	$2.9 \cdot 10^{17}$	104/120 97/125 100/128	1 1 1	22 29 51
5	38	$6.7 \cdot 10^{17}$	117/118 108/107 110/104 –/76	2 3 0 2	6 9 15 68
6	23	$3.9 \cdot 10^{17}$	120/114	2	39
7	22	$3.6 \cdot 10^{17}$	—	—	—
8	45	$7.4 \cdot 10^{17}$	119/110 118/105 65/58	2 2 3	5 93 174

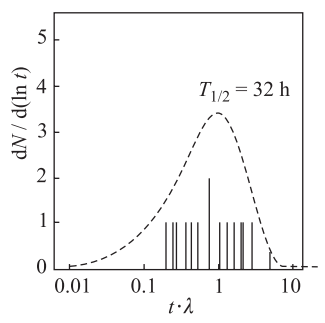

Fig. 4. Time distribution of SF events in the ^{243}Am + ^{48}Ca reaction

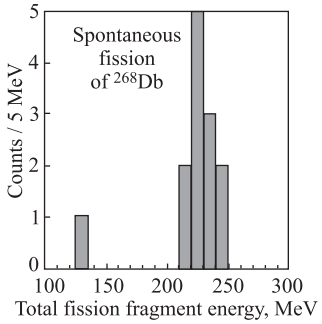

Fig. 5. The total kinetic energy of fission fragments

events (Fig. 4) agrees with the half-life obtained in the physical experiment [6] within statistical errors, see Table 3.

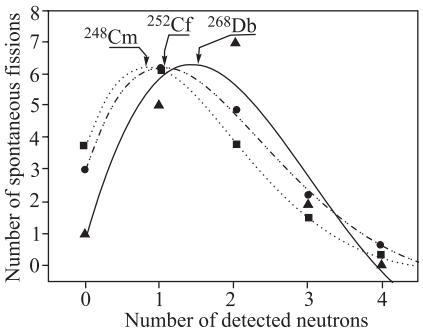

Fig. 6. Number of neutrons registered by ^{3}He detectors per SF decay (without taking into account the detector efficiency): ▲ — samples 1–8 (Table 2); • — experimentally measured distribution of ^{252}Cf thin sources; ■ — experimentally measured distribution of ^{248}Cm thin sources. The lines are shown for visual clearness. The arrows show distribution maxima. The data for ^{248}Cm and ^{252}Cf are in the 1:10000 scale

The total kinetic energy of fission fragments ($\overline{\text{TKE}}$) determined as the sum of amplitudes of the time-coincident signals from both detectors, corrected for energy loss in source and backing layers was about 235 MeV (Fig. 5). This result also agrees with the data of physical experiment ($\overline{\text{TKE}} \sim 225$ MeV). The average neutron multiplicity per fission was $\nu \sim 4.2$ (Fig. 6). The both parameters, i.e., the high $\overline{\text{TKE}}$ value and the high neutron multiplicity, give evidence for the fission of a rather heavy transactinide nuclide. Note, that for spontaneous fission of ^{248}Cm $\overline{\text{TKE}} = 181$ MeV, $\nu = 3.14$; for ^{252}Cf $\overline{\text{TKE}} = 185$ MeV, $\nu = 3.75$.

The ninth experiment was carried out under the same conditions (^{48}Ca-ion beam dose $\approx 2.9 \cdot 10^{17}$), with the same beam energy as in the previous eight runs, but *without chemical separation* of the ^{243}Am$+^{48}$Ca reaction products. The experiment was aimed at determining the background of spontaneously fissioning nuclei (mainly Cf isotopes) implanted into the catcher. After the end of irradiation the catcher surface was cleaned of ^{243}Am-carrying aerosol particles and put in contact with solid-state track detector. In the course of long-term exposition, the detectors were changed and subjected to physical-chemical treatment, in order to develop latent tracks of SF fragments.

The counting rate was 2 events per day, in 30 days after the end of the irradiation 63 SF events were detected. With our separation factor of group 5 elements from actinides more than $8 \cdot 10^3$, the probability of detecting a single SF event from actinide isotopes (including Lr) in 174 h is less than 0.1%.

In total, the obtained data allow one to state undoubtedly that all the 15 events of spontaneous fission detected in the present experiment are due to the isotope of Db, the final descendant product of decay of the new element 115.

From the yield of spontaneously fissioning nuclei the cross section of producing the mother nucleus of element 115 — the product of neutron evaporation in the ^{243}Am$+^{48}$Ca reaction, can be determined to be about 4 pb ($4 \cdot 10^{-36}$ cm^2). This agrees with the value measured in the experiments with the gas-filled separator ($\sigma_{3n} \sim 3$ pb).

Table 3. **The results of the «physical» and «chemical» experiments of the study ^{48}Ca + ^{243}Am reaction**

	«Physical» experiment [6]	«Chemical» experiment
Separation method	Kinematic separator	Radiochemical separation
Separation efficiency	$\approx 40\%$	$\approx 80\%$
Registration	Decay chains of nuclei with $Z = 115$	SF nuclei with $Z = 105$
Energy of ^{48}Ca-ion beam in the middle of target layer	246 MeV	247 MeV
Total ion beam dose (ions)	$4.5 \cdot 10^{18}$	$3.4 \cdot 10^{18}$
Thickness of ^{243}Am target	0.36 mg/cm^2	1.2 mg/cm^2
The number of events of the decay observed in the experiment	3	15
Cross section of producing the mother nucleus of element 115	$\sim 2.7^{+4.8}_{-1.6}$ pb	$4.2^{+1.6}_{-1.2}$ pb
Half-life of 105 element	16^{+19}_{-6} h	32^{+11}_{-7} h
Total kinetic energy of fission fragments ($\overline{\text{TKE}}$)	~ 225 MeV	~ 235 MeV
The average neutron multiplicity per fission	—	4.2
Identification method of the SF-decays nuclei in ^{48}Ca + ^{243}Am reaction	The characteristic dependence of the yield of the neutron-evaporation products on the excitation energy of the compound nucleus ($Z = 115$)	Isolation of the group 5 elements ($Z = 105$)

Table 3 presents the data of the two experiments aimed at the determination of properties of the spontaneously fissioning isotope of element 105 that ends up the chain of the sequential α decays of element 115 produced in the reaction ^{243}Am + ^{48}Ca. The properties of the nuclide ^{268}Db obtained in the decay chains of element 115 that were observed in the experiments with the gas-filled recoil separator, agree, in all the measured parameters, with the data of the present chemical experiment, which determines its atomic number. It should be noted

that, due to the high efficiency of the chemical separation of the reaction products and the possibility of employing relatively thick target layers, the yield of the isotopes of superheavy elements provided with the present experimental technique is about a factor of 5 higher than with kinematic separators.

Thus, the data of the present experiment give an independent evidence for the synthesis of element 115 as well as 113 element in the reaction ^{243}Am+^{48}Ca.

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