Synthesis of Superheavy Nuclei in the ^{48}Ca + ^{244}Pu Reaction

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In bombardment of ^{244}Pu with ^{48}Ca, we observed a decay sequence consisting of an implanted heavy atom, three subsequent α decays, and a spontaneous fission (SF), all correlated in time and position. The measured α energies and corresponding time intervals were $E_\alpha = 9.71$ MeV ($\Delta t = 30.4$ s), 8.67 MeV ($\Delta t = 15.4$ min), and 8.83 MeV ($\Delta t = 1.6$ min); for the SF ($\Delta t = 16.5$ min), the total deposited energy was approximately 190 MeV. The α-particle energies together with the decay times and SF terminating the chain offer evidence of the decay of nuclei with high atomic numbers. This decay chain is a good candidate for originating from the α decay of the parent nucleus ^{289}114, produced with a cross section of about 1 pb.

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A fundamental outcome of macro-microscopic nuclear theory is the prediction of an "island of stability" of superheavy elements. Calculations performed over more than 30 years with different versions of the nuclear shell model predict substantial enhancement of the stability of heavy nuclei when approaching the closed spherical shells $Z = 114$ and $N = 184$. Isotopes of element 114 with neutron numbers from 174 to 176, close to the predicted spherical nuclei and, consequently, being relatively stable, can be produced in the fusion reaction of ^{244}Pu with ^{48}Ca ions [1].

With the doubly magic ^{48}Ca projectile, the resulting compound nucleus ^{292}114 is weakly excited; its excitation energy at the Coulomb barrier is about 30 MeV. Consequently, nuclear shell effects are still expected to persist in the excited nucleus, increasing the survival probability of the evaporation residues (EVRs), as compared to "hot fusion" reactions ($E^* \approx 45$ MeV), which were used for the synthesis of heavy isotopes of elements with atomic numbers $Z = 106$, 108, and 110 [2]. Additionally, the high mass asymmetry in the entrance channel should decrease the dynamical limitations on nuclear fusion arising in more symmetrical reactions.

In spite of these advantages, past attempts to synthesize new elements in ^{48}Ca-induced reactions with actinide targets gave only upper limits for their production [3]. The first positive result was obtained in 1998 when two spontaneous fission (SF) events were observed in the ^{48}Ca + ^{238}U reaction with an integrated beam dose of 3.5×10^{18} ions. These were assigned to the decay of a new isotope of element 112 produced in the reaction ^{238}U(^{48}Ca,3n)^{283}112 ($N = 171$) with a cross section of 5 ± 2 pb [4].

In the ^{48}Ca + ^{244}Pu reaction the ^{292}114 compound nuclei are expected to deexcite by emission of two to four neutrons. From calculations by Pustylnik, based on the experimental cross sections of hot fusion reactions, and diffusion model calculations by Wada [5] the maximum cross section for producing evaporation residues in the ^{48}Ca + ^{244}Pu reaction is expected to be the $3n$-evaporation channel at an excitation energy of 35 MeV. The absolute cross sections of the xn channels are estimated with larger uncertainty; the calculated cross section at the maximum of the $3n$ channel varies from 1 to 10 pb.

Smolańczuk *et al.* [6,7], who successfully reproduce the decay properties of the most neutron-rich known heavy nuclei, have calculated that the even-even isotopes ^{288}114 and ^{290}114 will have partial α-decay half-lives $T_\alpha = 0.14$ s and 0.7 s, respectively. Their predicted SF half-lives are considerably longer: $T_{\mathrm{SF}} = 2 \times 10^3$ s and 4×10^5 s, respectively. For their daughter nuclei—isotopes of element 112—the values of T_α and T_{SF} are more comparable, but α decay should still prevail. The α-decay granddaughters—the isotopes of element 110—are expected to decay primarily by spontaneous fission. For the odd isotopes, in particular for the ^{289}114 nucleus, the predictions are less definite; the odd neutron can lead to hindrance of α decay and, especially, spontaneous fission. Here one expects competition between the two decay modes in the daughter products with $Z \leq 112$ and somewhat longer chains of sequential α-decays than in the case of the neighboring even-even isotopes.

We note that T_α calculations by Möller *et al.* [8] for $^{288-290}$114 give values exceeding those of [6,7] by orders of magnitude (e.g., T_α of 7×10^4 s for ^{289}114). This, however, does not change the expected decay pattern for these isotopes of element 114 and their daughters. We can expect a sequence of two or more α decays terminated by a spontaneous fission as it recedes from the stability region around $N = 184$.

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We present here initial results of an experiment to synthesize nuclei with $Z = 114$ in the vicinity of predicted spherical nuclear shells in the complete fusion reaction ^{48}Ca + ^{244}Pu.

A ^{48}Ca^{5+} beam was extracted from the ECR ion source and injected into the Dubna U400 heavy ion cyclotron. The average intensity of the ion beam on the target was 4×10^{12} pps at the material consumption rate of about 0.3 mg h^{-1}. The beam energy was determined with a precision of $\sim$1 MeV, by measuring the energies of scattered ions, and by a time-of-flight (TOF) technique.

The targets consisted of the enriched isotope ^{244}Pu (98.6%) in the form of PuO_2, deposited onto 1.5-μm Ti foils to a thickness of $\sim$0.37 mg cm^{-2}. Each target had an area of 3.5 cm^2 in the shape of an arc segment with an angular extension of 40° and an average radius of 60 mm. Nine targets were mounted on a disk that was rotated at 2000 rpm across the beam direction.

We used a ^{48}Ca bombarding energy of 236 MeV, corresponding to the calculated maximum of the $3n$-evaporation channel to form the isotope ^{289}114. EVRs recoiling from the target were separated in flight from beam particles and various transfer-reaction products by the Dubna Gas-filled Recoil Separator [9], passed through a TOF system, and were implanted in the focal-plane detectors. At a beam intensity of 4×10^{12} pps, the overall counting rate of the detector system was 15 s^{-1}. The principal sources of events with a TOF signal are the scattered target nuclei and target-like transfer reaction products. Background events without a TOF signal, which can imitate α particles from decay of implanted nuclei, can be due to light low-ionizing particles (n, p, α, etc.) produced in direct nuclear reactions and the small fraction of recoils not detected by the TOF chambers. The collection efficiency of the separator was estimated from the results of test experiments in the bombardment of $^{\rm nat}$Yb and enriched $^{204,206-208}$Pb targets with ^{48}Ca ions. We deduce that 40% of the recoiling $Z = 114$ nuclei formed in the ^{244}Pu target would be implanted in the focal-plane detector.

The TOF detector was used to measure the time of flight of recoiling nuclei (detection efficiency of $\sim$99.7%) and to distinguish the focal-plane detector signals of particles passing through the separator from those of the radioactive decay of previously implanted nuclei. The focal-plane detector consisted of three 40×40 mm^2 silicon *Canberra Semiconductor* detectors, each with four 40-mm-high $\times$ 10-mm-wide strips having position sensitivity in the vertical direction. To increase the detection efficiency for α particles escaping the focal-plane detector, we arranged eight detectors of the same type without position sensitivity in a box surrounding the focal-plane detector. Employing these side detectors increased the α-particle detection efficiency to $\sim$87% of 4π. A set of three similar "veto" detectors was situated behind the front detectors in order to eliminate signals from low-ionizing light particles, which sometimes pass through the focal-plane detector without being detected in the TOF system.

Alpha-energy calibrations were periodically performed using the α peaks from nuclides produced in the test reactions mentioned above. The fission-energy calibration was obtained by detecting fission fragments from the SF of ^{252}No [10]. The energy resolution for detection of α particles in the focal-plane detector was $\approx$45 keV; for detection by the side detectors of α particles escaping from the focal plane detector, the energy resolution was $\approx$180 keV. We determined the position resolution of the signals of correlated decays of nuclei implanted in the detectors: For sequential α-α decays the FWHM position resolution was 1.0 mm; for correlated EVR–α signals, 1.4 mm; and for correlated EVR–SF signals, 1.2 mm.

The experiment was performed during November and December 1998. Over a period of 34 days a total of 5.2×10^{18} projectiles was delivered to the target.

In the analysis of the experimental data, we assumed that the island of stability of superheavy nuclides has a border at which nuclei are unstable against spontaneous fission. As long as any α-decay chain leads to the edge of the island of stability, it should be terminated by spontaneous fission. In test experiments, we observed that 95% of SF events from the ^{252}No implants produced in the ^{206}Pb + ^{48}Ca reaction are characterized by total deposited energy exceeding 130 MeV (without corrections for the pulse-height defect). Four such events were observed in the ^{244}Pu + ^{48}Ca bombardment.

Two events, with measured energies $E = 149$ MeV and $E = 153$ MeV, were detected 1.13 and 1.07 ms, respectively, after the implantation of corresponding position-correlated recoil nuclei. For one of these SF events both fission fragments were registered by the focal-plane and side detectors. We assign these events to the spontaneous fission of the 0.9-ms ^{244mf}Am isomer, a product of transfer reactions with the ^{244}Pu target. Another signal was registered at the bottom edge of the sensitive layer of the outermost detector strip. Moreover, alpha particles escaping from this point of the focal plane detector could be registered by side detectors with an efficiency of only about 7%. In this case, the analysis of the preceding correlated events is strongly complicated, as a significant part of the information is lost.

The last SF event was observed as two coincident signals (two fission fragments) with energy deposited in the focal-plane detector $E_{\rm F1} = 120$ MeV and in the side detector $E_{\rm F2} = 52$ MeV; $E_{\rm tot} = 172$ MeV. Correcting for pulse-height defect using calibration data mentioned above, this would mean a total deposited energy of 190 MeV. We searched the data backwards in time from this event for preceding α-particles in the same position with $E_\alpha >$ 8 MeV [6–8]. The entire position-correlated decay chain is shown in Fig. 1. An α particle was detected in the focal-plane detector 30.4 s after the implantation of a recoil nucleus in the middle of strip 8. The measured recoil energy

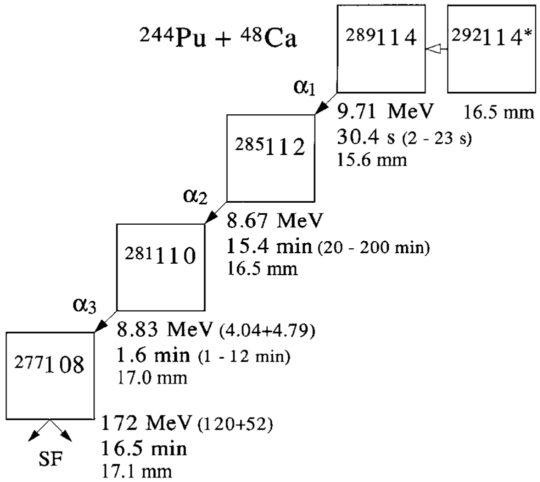

FIG. 1. Time sequence in the observed decay chain. The expected half-lives corresponding to the measured E_α values for the given isotopes are shown in parentheses following the measured lifetimes. Hindrance factors of 1 and 10 were assumed for α decay of nuclei with an odd neutron number. Positions of the observed decay events are given with respect to the top of the strip.

(6.1 MeV), TOF signal (26 ns), and roughly estimated resulting mass value ($\sim$320 amu) are consistent with those expected for a complete-fusion EVR, as determined in the calibration reactions. The energy of the first α particle was $E_\alpha = 9.71$ MeV. A second α particle, having an energy $E_\alpha = 8.67$ MeV, was observed at the same location after 15.4 min. A third α particle, escaping the front detector leaving an energy $E_{\alpha 1} = 4.04$ MeV and absorbed in the side detector with $E_{\alpha 2} = 4.79$ MeV ($E_{\mathrm{tot}} = 8.83$ MeV), was measured 1.6 min later. Finally, 16.5 min later, the SF event was observed.

All 5 signals (EVR, $\alpha_1, \alpha_2, \alpha_3$, SF) appeared within a position interval of 1.6 mm, which strongly indicates that there is a correlation among the observed decays. The chi-squared per degree of freedom (with three degrees of freedom) for the consecutive position differences of the candidate decay chain is such that only 21% of the chi-squared distribution is greater than this value.

Assuming that the decay sequence for a valid event will terminate with SF, we developed a Monte Carlo technique to estimate the probability of the candidate event being due to random correlations. Artificially, 10^5 fissions were inserted into the data, distributed at random positions and times over the entire detector array and entire experiment duration. We searched the 34 min preceding each random fission for three α-particle-like signals with energies 8.5–10.0 MeV and one EVR-like event preceding the α events. All four of these events had to be within 2.0 mm of the artificial fission and meet the position criteria at greater than 95% confidence level to be considered a possible random correlation. The probability per fission of finding such a correlated event was determined to be $P_{\mathrm{err}} = 0.006$. With the given energy window and no time restriction within the 34-min interval, many random sequences were found that could not be proposed as the decay of $Z = 114$ or nearby elements. By applying the Geiger-Nuttall relationship, we imposed a lifetime window for each α event. Requiring that the hindrance factor must be between 1 and 10 for each α energy reduced P_{err} to 6×10^{-4}.

Another P_{err} calculation was performed for strip 8 at the position in which the candidate event occurred, following the procedure described in Ref. [11]. For a position-correlation window of 1.6 mm the signals from EVR-like events were observed with a frequency of 1.3 h^{-1}. The signals of α-like events with $E_\alpha = 8.1 - 10.5$ MeV occurred with a frequency of 1 h^{-1}. Thus, calculated from event rates alone, with event-to-event time distances restricted to actual measured values, the probability of this decay sequence being caused by the chance correlation of unrelated events in strip 8 is 1×10^{-4}.

All events of the decay chain are correlated in time and position and match the decay of a superheavy nucleus that is predicted by theory. For the whole decay chain the basic rule for α decay, defining the relation between Q_α and T_α, is fulfilled. This can be seen in Fig. 1 where the expected half-lives, corresponding to the measured α-particle energies for the isotopes with the specified atomic numbers of the genetically related radionuclides, are shown. For the calculation of half-lives, the formula of Viola and Seaborg with parameters from Refs. [6,7] has been used, with hindrance factors of 1 and 10. The detected sequential decays have larger $T_{1/2}$ vs E_α values than the known radioactive nuclides. The best candidate for the parent nucleus is the even-odd isotope ^{289}114, produced in the 3n-evaporation channel. This one event corresponds to a cross section of about 1 pb. The decay properties of the observed nuclei are also in agreement with calculations [6,7], assuming reasonable hindrances for the decay of nuclei with odd neutron numbers.

In our experiment we observed a four-member decay sequence. If we assume that it actually consisted of five decays (the spontaneous fission was due to ^{273}106), the probability of missing any one of the four α events is about 34%, but the probability of missing any *particular* α event in the chain and observing the other three is only about 8.5%.

The lifetimes of the new isotopes, in particular ^{285}112 and ^{281}110, appear to be approximately 10^6 times longer than those of the known nuclei ^{277}112 and ^{273}110 [2,12], which have eight fewer neutrons. Therefore, the observed decay properties of the synthesized nuclides, together with the data obtained earlier for ^{283}112 [4], can be considered the first experimental proof of the existence of enhanced stability in the region of superheavy elements.

During the performance of the experiments we lost two of our most active colleagues, Dr. V. B. Kutner and Dr. B. I. Pustylnik. We address our deepest appreciation of the great contribution our friends gave to the completion of the present work. We are indebted to our late colleague Dr. Yu. A. Lazarev who greatly contributed to this research at the initial stages. We are grateful to the JINR Directorate, in particular to Professor Ts. Vylov and Professor V. G. Kadyshevsky for the help and support we got during all stages of performing the experiment. We express our thanks to Dr. V. Ya. Lebedev and Dr. S. N. Dmitriev for developing methods for preparation of the metal Ca samples for the ECR-ion source, and also to V. I. Krashonkin, V. I. Tomin, A. M. Zubareva, and A. N. Shamanin, for their help in preparing and carrying out the experiment. We would like to express our gratitude to the personnel of the U400 cyclotron and the associates of the ion-source group for obtaining an intense ^{48}Ca beam. The Livermore authors thank their Russian hosts for their hospitality during the experiment. The help of Dr. E. K. Hulet in the early phases of our collaboration is gratefully acknowledged. This work has been performed with the support of the Russian Foundation for Basic Research under Grant No. 96-02-17377 and of INTAS under Grant No. 96-662. The ^{244}Pu target material was provided by the U.S. DOE through ORNL. Much of the support for the LLNL authors was provided through the U.S. DOE under Contract No. W-7405-ENG-48. These studies were performed in the framework of the Russian Federation/U.S. Joint Coordinating Committee for Research on Fundamental Properties of Matter.

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