Independent Verification of Element 114 Production in the ^{48}Ca + ^{242}Pu Reaction

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Independent verification of the production of element 114 in the reaction of 244-MeV ^{48}Ca with ^{242}Pu is presented. Two chains of time- and position-correlated decays have been assigned to ^{286}114 and ^{287}114. The observed decay modes, half-lives, and decay energies agree with published results. The measured cross sections at a center-of-target energy of 244 MeV for the ^{242}Pu(^{48}Ca, 3–4n)^{287,286}114 reactions were $1.4^{+3.2}_{-1.2}$ pb each, which are lower than the reported values.

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During the last ten years the Dubna Gas Filled Recoil Separator (DGFRS) group has published numerous reports of production of superheavy elements (SHE) near $Z = 114$ in ^{48}Ca irradiations of actinide targets, as summarized in [1]. Independent verification of these observations has been of paramount importance, but due to the challenges of experiments with picobarn cross sections employing radioactive targets, confirmation results have only appeared recently. The newly implemented ^{242}Pu target facility at the Berkeley Gas-filled Separator (BGS) at the Lawrence Berkeley National Laboratory's (LBNL) 88-Inch Cyclotron has now enabled us to study the production of element 114 using the ^{242}Pu(^{48}Ca, xn)$^{290-x}$114 reaction.

A study of the ^{242}Pu(^{48}Ca, xn)$^{290-x}$114 fusion reactions was published in 2004 [2], presenting the excitation functions for the two- to four-neutron evaporation channels. Noteworthy are the relatively large cross sections reported for the three- and four-neutron evaporation channels of $\sigma_{3n} = 3.6^{+3.4}_{-1.7}$ pb and $\sigma_{4n} = 4.5^{+3.6}_{-1.9}$ pb, respectively, at a center-of-target energy of 244-MeV ^{48}Ca, which corresponds to a compound nucleus excitation energy of $E^* = 40.2$ MeV. The decay properties of $^{286-288}$114 and daughter nuclides are presented in Table I [1]. The nuclide ^{286}114 is reported to decay by either α-particle decay or spontaneous fission (SF) with a ratio of 50/50, and with a half-life of 0.13 s. The α-decay daughter ^{282}112 has been observed to disintegrate by spontaneous fission with an 82-ms half-life. Overall, 24 decay chains have been interpreted as synthesis and decay of ^{286}114. Sixteen decays of ^{287}114 have been published; the 0.48-s α-particle emission from ^{287}114 is followed by the 3.8-s α-decay of ^{283}112 and spontaneous fission of ^{279}110 ($T_{1/2} = 0.185$ s). In one of the 16 reported chains ^{279}110 underwent α-particle decay and was succeeded by another two α-particle decays and a spontaneous fission ($^{275}\mathrm{Hs} \rightarrow {}^{271}\mathrm{Sg} \rightarrow {}^{267}\mathrm{Rf}$).

The independent confirmation of important results is a fundamental part of scientific practice. To date, there have been several experimental attempts to investigate SHE production in ^{48}Ca irradiations of actinide targets. Many of these experiments [3–9] failed to produce the SHE activities presently agreed on by the DGFRS group (Table 3 in [1]). However, recently there have been two experimental confirmations of SHE production. The complete fusion of ^{48}Ca + ^{242}Pu and assumed α-particle decay of element 114 was employed by Eichler *et al.* [10] in an experiment to chemically characterize element 112 at Dubna in 2007. In addition, Hofmann [11] reported production of ^{283}112 in the ^{238}U(^{48}Ca, 3n)^{283}112 reaction. These two experiments are the only experimental confirmations of SHE since the first reports from Dubna ten years ago. This Letter reports the independent verification of the production of two isotopes of element 114 in the reaction of ^{48}Ca with ^{242}Pu.

The LBNL Advanced Electron Cyclotron Resonance source [12] produced a ^{48}Ca^{11+} beam. The ^{48}Ca^{11+} was accelerated to 263 MeV by the LBNL 88-Inch Cyclotron with typical intensities of 300–400 pnA. The beam entered the BGS through a 45 μg/cm^2 thick carbon window before passing through the targets located 1.5-cm downstream of the window. This window is the effective barrier between the beam-line vacuum and the 66-Pa helium filling gas of the separator. The targets were prepared by molecular plating, and the BGS plutonium target facility is described in more detail in [13]. A 9.5-cm diameter target wheel containing four target segments with 440, 340, 320, and 270 μg/cm^2 of ^{242}Pu (>99.9%) as PuO$_2$ on

TABLE I. Published decay properties of $^{286-288}$114 and daughter products [1].

Nuclide	Decay mode	Half-life	E_α (MeV)
^{286}114	$\alpha = 50\%$ SF $= 50\%$	$0.13^{+0.04}_{-0.02}$ s	10.19 ± 0.06
^{282}112	SF	$0.82^{+0.30}_{-0.18}$ ms	
^{287}114	α	$0.48^{+0.16}_{-0.09}$ s	10.02 ± 0.06
^{283}112	$\alpha > 10\%$ SF $< 10\%$	$3.8^{+1.2}_{-0.7}$ s	9.54 ± 0.06
^{279}Ds	$\alpha = 10\%$ SF $= 90\%$	$0.20^{+0.05}_{-0.04}$ s	9.70 ± 0.06
^{288}114	α	$0.80^{+0.27}_{-0.16}$ s	9.94 ± 0.06
^{284}112	SF	97^{+134}_{-19} ms	

2.4-μm thick Ti backing was used. Energy losses in the entrance window and targets were calculated with SRIM2008 [14]. The ^{48}Ca beam lost 2.2–3.6 MeV upon passing through the ^{242}PuO$_2$ target layer. The average center-of-target beam energy was 244 MeV, chosen because the reported cross sections for the ^{242}Pu(^{48}Ca, $3n$)^{287}114 and ^{242}Pu(^{48}Ca, $4n$)^{286}114 were largest at this energy. Using experimental masses for target and projectile [15], with the calculated mass for ^{290}114 [16], an average center-of-target compound nucleus excitation energy of 41 MeV was obtained. To disperse the heat produced in the targets, the target wheel was rotated at 12–14 Hz. During this experiment, elastically scattered beam particles were recorded by two silicon p-i-n detectors mounted ± 27 degrees from the incident beam direction, with which the product of beam dose and target thickness were monitored.

Compound nucleus evaporation residues (EVRs) recoiled out of the targets with the momentum of the beam. The transmission efficiency for an EVR to implant in the focal plane detector was estimated by a Monte Carlo simulation of EVR trajectories in the BGS, combined with experimentally measured efficiencies. For the ^{242}Pu(^{48}Ca, 3–$4n$)$^{287-286}$114 reaction the transmission efficiency is expected to be 60%. The magnetic rigidity of the EVRs was estimated to be $B\rho = 2.18$ Tm, following the discourse presented in a previous publication describing the BGS [4], and the first part of the experiment was run with the BGS magnets set for this $B\rho$. After about one-third of the running time the BGS magnet settings were changed to be sensitive to higher magnetic rigidities, placing 2.24-Tm products at the center of the detector.

After passing through the BGS, the EVRs enter the detector chamber where they pass through a multiwire proportional counter (MWPC) before being implanted in the focal plane detector. Analog signals from the MWPC, together with a time-of-flight between the MWPC and the focal plane detector, allow implantation events to be distinguished from radioactive decays of atoms previously implanted in the focal plane detector. The MWPC was filled with 500-Pa isobutane, and had a thickness equivalent to 0.5 mg/cm^2 carbon. The focal plane detector consists of 48 Si-strips (three 6×6 cm^2 cards, sixteen strips each), which provide horizontal resolution. Vertical position is determined by resistive charge division within each strip. Eight non-position-sensitive Si detector cards are located upstream and perpendicular to the focal plane detector forming a five-sided box configuration. This allows for reconstruction of α particles and fission fragments which escape the focal plane detector and stop in the upstream detectors. The efficiency for detecting full-energy α particles was approximately 75%. The geometrical efficiency for detection of a spontaneous fission decay of an implanted ion is approximately 100%. Behind the focal plane detector is a second set of silicon strip detectors, which record light, low-ionizing particles passing though the focal plane detector. These are referred to as "punch-through detectors." Data acquisition was triggered by any event registering greater than $\sim$400 keV in the focal plane detector or greater than $\sim$1.5 MeV in the upstream detectors. The detector was calibrated with α particles emitted from a source of ^{148}Gd, ^{239}Pu, ^{241}Am, and ^{244}Cm. The energy resolution was 40 keV FWHM for 5.48-MeV ^{241}Am α particles. For α particles escaping the focal plane detector and depositing their residual energy in the upstream detectors, the resolution was about 100 keV. Since all reported 114 decay chains terminated in a spontaneous fission, the efficiency for detection of a 114 decay chain was close to 100%. Immediately behind the punch-through detectors was a 2-mm thick Al vacuum window. A Ge clover γ-ray detector (with four 5-cm diameter $\times$ 8-cm long Ge crystals) was positioned directly downstream of the Al vacuum window. The efficiency for detecting SHE x rays was approximately 13% for an assumed recoil distribution centered on the focal plane detector.

To minimize the effect of random correlation of unrelated events appearing like element 114 decay chains, a fast beam shutoff was used. Upon detection of an "EVR-like event" [$5.0 < E(\mathrm{MeV}) < 20.0$, MWPC signal, no punch-through signal, no signal from upstream detectors] followed within 20 seconds by an "α-like event" with energy expected for the decay of element 114 or 112 [$9.0 < E(\mathrm{MeV}) < 10.7$, no MWPC signal, no punch-through signal] from the same detector position, the beam was switched off for 20 seconds to allow detection of subsequent decays in a nearly background-free environment. If subsequent α particles [$8.0 < E(\mathrm{MeV}) < 10.2$ MeV] were recorded during the beam off interval, the beam shutoff would be extended. During the 8-day experiment there were 34 of the 20-second beam shutoffs, none of which were extended due to detection of daughter α decays.

Two genetically correlated decay chains were observed during the experiment. Based on the observed decay properties, one decay chain was interpreted as the decay of ^{286}114, and the other one was assigned to decay of ^{287}114. Table II lists the details. The three-member decay chain observed in strip 14, listed at the top of Table II, consisted of an 11.55-MeV event coincident with the MWPC followed 0.301 seconds later by a 10.23-MeV event without a MWPC-signal. This event combination switched off the beam. A 214-MeV event (also anticoincident with the MWPC) was observed after 3.55 ms from the same position in the focal plane detector. This event chain has been interpreted as the implantation and α decay of ^{286}114 followed by the SF of the ^{282}112 daughter. Details of the four-member decay chain in the strip 7 event are listed at the bottom of Table II. A 7.73-MeV event coincident with the MWPC was observed, succeeded 815 ms later by a 3.86-MeV event anticoincident with the MWPC. After an additional 1.921 seconds, a 9.65-MeV event, without a

TABLE II. The two element 114 decay chain details.

Interpretation	E (MeV)	Δt (s)	Position (mm)	$B\rho$ (Tm)
EVR-strip 14	11.55		13.0(2)	2.28
^{286}114 α decay	10.23(4)	0.3009	12.9(3)	
^{282}112 SF	214.5	0.0036	13.1(15)	
EVR-strip 7	7.73		−2.8(4)	2.25
^{287}114 α decay	3.86(4)[a]	0.8149	−3.3(7)	
^{283}112 α decay	9.65(10)[b]	1.9208	10.1(64)	
^{279}110 SF	176	0.1854	−2.9(15)	

[a] Escape α particle depositing only partial energy in focal plane detector strip.

[b] Reconstructed from 382 keV in the focal plane strip and 9271 keV in upstream detector.

MWPC signal, followed. Finally, 185 ms later a 176-MeV event, anticoincident with the MWPC was recorded. This event chain has been interpreted as the implantation and subsequent α-particle decay of ^{287}114, α decay of ^{283}112, and SF of ^{279}Ds. The 3.86 MeV recorded for the first α particle in this event chain is lower than the 10.02 MeV reported for ^{287}114, but somewhat large for an α particle escaping the focal plane detector. This can be explained as an "escape" event, in which the α particle has a trajectory nearly parallel to the detector surface. It is likely that such a trajectory would miss the upstream detectors, landing in the 1.0–1.4 cm gap between the focal plane and upstream detectors. The measured energy of the second correlated α particle of 9.65 MeV is in agreement with the reported energy of the ^{283}112 daughter, and the following 176-MeV event can be assigned to the SF of the ^{279}Ds granddaughter. The ^{283}112 α-decay event was reconstructed from a 382-keV signal in the focal plane, coincident with 9271 keV in one of the upstream detectors. Because of the low energy recorded in the focal plane detector, there is a large uncertainty in the vertical position. The evaluated vertical position is approximately 2σ away from the vertical position of the other members of the decay chain. There may be an additional contribution to this position discrepancy due to integral nonlinearity in the lowest 5% of the analog-to-digital converter range. None of the α decays in the decay chains attributed to 114 decays were in prompt coincidence with x rays or γ rays. The SF terminating the ^{286}114 decay chain was coincident with γ rays in three of the Ge crystals, and the SF terminating the ^{287}114 decay chain was coincident with one γ ray, lending confidence to the assignment of these two events as fissions.

TABLE III. Expected numbers of random correlations for sequences: EVR-like event followed by SF, α-SF, and α-α-SF, for the two parts of the experiment, referred to by the magnetic settings of the separator. The evaluated random rates are calculated for a ±1.5-mm vertical position window and a time window of 20 seconds.

	2.18 Tm setting	2.24 Tm setting
EVR-SF	0.022	6.3×10^{-4}
EVR-α-SF	4.3×10^{-7}	3.7×10^{-8}
EVR-α-α-SF	1.0×10^{-10}	2.8×10^{-12}

The probability that the recorded events are random coincidences of unrelated events has been evaluated. For this evaluation, two simplifying assumptions have been made: (1) the EVR-like events and the α-like events are evenly distributed over the focal plane detector surface, and (2) the rates of EVR-like events and α-like events were constant at their average values. The rates of EVR-like events [$5.0 < E(\mathrm{MeV}) < 20.0$, MWPC signal, no punch-through signal, no signal from upstream detectors] in the whole detector array were 0.346 Hz at the 2.18-Tm setting and 0.0303 Hz at the 2.24-Tm setting. Alpha-decay-like events [$9.0 < E(\mathrm{MeV}) < 10.7$, no MWPC signal, no punch-though signal] were recorded at rates 5.58×10^{-3} Hz (2.18 Tm) and 5.54×10^{-3} Hz (2.24 Tm). During the course of the experiment four fission-like events [$85 < E(\mathrm{MeV}) < 300$, no MWPC signal] were recorded, 3 in the $B\rho = 2.18$-Tm portion of the experiment and 1 during the $B\rho = 2.24$-Tm portion of the experiment. Table III lists the expected numbers of random correlations that could be interpreted as element 114 decay chains: that is, the probability that a fission-like event has been preceded by an EVR, EVR-α, or EVR-α-α, in a ±1.5-mm vertical position window and time window of 20 seconds. Overall the random rates are very small, and thus it is not likely that the observed decay chains are due to random correlation of unrelated events.

The measured cross sections are $1.4^{+3.2}_{-1.2}$ pb each for ^{286}114 and ^{287}114, with 68% statistical confidence interval using Schmidt's prescription [17]. The factors contributing to systematic errors in a BGS experiment are previously described in detail and result in an additional 12% systematic uncertainty in the measured cross sections [4]. The 1.4 pb cross sections measured in this work are lower than the 4.5 pb and 3.6 pb reported by the DGFRS group for ^{286}114 and ^{287}114, respectively. Based on the 8.1-pb sum of DGFRS cross sections for ^{286}114 and ^{287}114, nearly six decay chains originating from element 114 were expected in our work. The probability that this discrepancy is due simply to statistical fluctuations can be evaluated using Eqs. 2–5 of [4]. According to this analysis, given that the DGFRS experiments observed 12 element 114 decay chains at this beam energy with a total cross section of 8.1 pb [2], the probability that our experiment results in 2 or fewer element 114 events at a total cross section of 2.8 pb is only 9%. It should be noted that both element 114 events in this Letter occurred relatively close to the high-$B\rho$ edge of the focal plane detector. The BGS focal plane image is quite large in the horizontal direction ($\sigma_h =$ 4.0 cm). Thus it is difficult to evaluate the average element 114 $B\rho$ in He gas. If the true average $B\rho$ for element 114 recoils in He gas is higher than ~2.3 Tm, much of the element 114 recoil distribution would have missed the BGS focal plane detector, resulting in the low cross section measurement.

In summary, a ^{48}Ca irradiation of ^{242}Pu targets was carried out at the LBNL 88-Inch Cyclotron using the Berkeley Gas-filled Separator. Two event chains were observed that are interpreted as the formation and decay of superheavy element 114. ^{287}114 was produced via the $^{242}\mathrm{Pu}(^{48}\mathrm{Ca}, 3n)$ reaction, and ^{286}114 was produced via the $^{242}\mathrm{Pu}(^{48}\mathrm{Ca}, 4n)$ reaction. Decay modes, lifetimes, and decay energies of the element 114 products and their daughters are consistent with those reported by the Dubna Gas Filled Recoil Separator group. Superheavy element formation cross sections measured in this work are lower than reported by the DGFRS group.

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