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Experiment on the Synthesis of Element 113 in the Reaction ^{209}Bi(^{70}Zn,n)^{278}113

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The convincing candidate event of the isotope of the 113th element, ^{278}113, and its daughter nuclei, ^{274}111 and ^{270}Mt, were observed, for the first time, in the ^{209}Bi + ^{70}Zn reaction at a beam energy of 349.0 MeV with a total dose of 1.7×10^{19}. Alpha decay energies and decay times of the candidates, ^{278}113, ^{274}111, and ^{270}Mt, were (11.68 ± 0.04 MeV, 0.344 ms), (11.15 ± 0.07 MeV, 9.26 ms), and (10.03 ± 0.07 MeV, 7.16 ms), respectively. The production cross section of the isotope was deduced to be 55^{+150}_{-45} fb (10^{-39} cm^2).

KEYWORDS: new element ^{278}113, new isotopes ^{274}111 and ^{270}Mt, gas-filled recoil separator
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Finding the new isotopes of very heavy elements, including new elements, and studying their decay properties are interesting subjects in both nuclear physics and nuclear chemistry. Since 2002, we have investigated the production and decay of ^{271}Ds ($Z = 110$) and ^{272}111 using ^{208}Pb(^{64}Ni,n) and ^{209}Bi(^{64}Ni,n) reactions, respectively,[1,2] at the RIKEN Linear Accelerator Facility (RILAC). Subsequently, we studied the isotope of the 112th element using the ^{208}Pb(^{70}Zn,n)^{277}112 reaction.[3] Our results clearly confirmed the production of the isotopes reported by Hofmann and coworkers,[4–9] and provided new spectroscopic information on the isotopes and their daughter nuclei.

As an extension of our previous work, we performed experiments aimed at synthesizing an isotope of larger atomic number, $Z = 113$, using the ^{209}Bi + ^{70}Zn reaction. During irradiation, we observed one α-decay chain that can be assigned to subsequent decays from ^{278}113, using the genetic correlation of α-decays connected to the known nuclides, ^{266}Bh and ^{262}Db.

The production of element 113 was first reported by Oganessian *et al.*[10] in 2004 using the ^{243}Am(^{48}Ca,xn) ($x = 3, 4$) reaction at the Flerov Laboratory of Nuclear Reaction (FLNR) of Joint Institute of Nuclear Research (JINR), Russia. They reported that the observed decay chains were consistent with the subsequent decays from ^{288}115 produced by the 3n evaporation channel and from ^{287}115 produced by the 4n evaporation channel. In these chains, three atoms of ^{284}113 and one atom of ^{283}113 were consequently assigned to be α-decay daughters of ^{288}115 and ^{287}115, respectively. The chains ended by spontaneous fission of previously unknown dubnium isotopes, ^{268}Db and ^{267}Db. To confirm both the atomic number and mass number of these isotopes, several types of further experiments such as measurement of the excitation function, chemical separation of long-lived dubnium isotopes, and direct mass measurement of the products using an isotope separator on-line system, are scheduled at FLNR.[11]

The results of the present work strongly indicate the synthesis of the 113th element, even though the number of chains observed was only one, because the chain ended with known nuclides. More chains are expected to be observed in further experiments.

The present experiment, which started on September 5, 2003, was interrupted on December 29, 2003, and then restarted on July 8, 2004 and continued until August 2, 2004. The net irradiation time was 79 days.

A ^{70}Zn ion beam of 352.6 MeV was extracted from RILAC. The beam energy was determined by measuring magnetic rigidity in a 90° bending magnet and by a time-of-flight method. The absolute accuracy was ±0.6 MeV. The drift in the beam energy during the whole beam time was ±0.3 MeV. The beam intensity was monitored by measuring projectiles elastically scattered by the targets with a PIN photodiode mounted at 45° with respect to the incident beam direction at a distance of 1.28 m from the target position. The typical beam intensity on the target was 2.4×10^{12} s^{-1}.

Targets were prepared by vacuum evaporation of metallic bismuth onto carbon backing foils of 30 μg/cm^2 thickness. The thickness of the bismuth layer was about 450 μg/cm^2. The targets were covered by 10-μg/cm^2-thick carbon to protect them from sputtering. The energy loss of the beam in the target was estimated to be 5.4 MeV using range and stopping power tables.[12] Beam energy at the half-depth of the targets was estimated to be 349.0 MeV. Sixteen targets were mounted on a rotating wheel of 30 cm diameter. The wheel was rotated during irradiation at 2000 rpm.

The reaction products were separated in-flight from the beam using a gas-filled recoil ion separator, GARIS,[1] and

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were guided into a detector box placed at the focal plane of GARIS. The separator was filled with helium gas at a pressure of 86 Pa. The value of the magnetic rigidity ($B\rho$) of GARIS for evaporation residue measurement was set at 2.09 Tm.

The focal plane detection system consists of two sets of a timing detector and a silicon semiconductor detector box (SSD box). The timing detector is an assembly of microchannel plates (MCP) that detect secondary electrons emitted from a thin foil by impact of ions passing through the foil. The details of the timing detector were described elsewhere.[2)] The SSD box placed downstream of the timing detectors consists of five silicon detector plates. The dimensions of each detector are 60 mm × 60 mm. One of the silicon detectors, which faces the direction of incoming particles, is placed at the bottom of the SSD box and consists of 16 strip detectors (PSD). The dimensions of each strip detector are 3.75 mm × 60 mm. The strip detectors are position sensitive along the longer dimension. Four other detectors (SSDs) are set to detect decaying particles from the reaction products implanted in the PSD.

Evaporation residues are implanted in the PSD after passing through the two timing counters. The timing signal was used for two purposes. One is to measure the time of flight (TOF) of incoming particles for rough estimation of their mass number together with energy signals from the PSD. The second purpose is to identify decay events originating from implanted nuclei in the PSD. The events without signals from either timing detector are regarded as the decay events.

The detection system was periodically checked by measuring the several α-decay lines of the transfer-reaction products, such as ^{211}Po, ^{212}At, and ^{213}Rn, setting the $B\rho$ of GARIS at 1.67 Tm, and keeping all the other conditions the same as those for the actual measurement. Energy calibration of the detectors for decay α-particles was performed simultaneously.

We observed one event of implantation of an evaporation residue (ER) in the PSD followed by four consecutive α-decays terminated by a spontaneous fission decay. The measured positions of all six sequential events were within the spatial resolution of the PSD.[1)] The observed energies, time differences between events, and positions of each decay are summarized in Table I, together with those of the implantation event. The energies have been calibrated for α-particles. Therefore, the listed energies for the ER and spontaneous fission are not accurate because possible effects of pulse height defect are not taken into account. The events were registered in strip #12 of the 16 strips in the PSD.

For the α_1 and α_4, the α particles were detected only by the PSD. The energy resolution measured using only the PSD was 39 keV in full width at half maximum (FWHM). For the α_2 and α_3, the decaying α particles were ejected from the PSD and implanted to the SSD. Therefore, the energies were measured partly by the PSD and the residual ones by the SSD. The resolution in these cases was 66 keV FWHM. For the fifth decay (hereafter, we use SF to represent the fifth decay), the energy was measured using only the PSD. For all the five decay events, no TOF signal registered from the timing detectors was associated.

The probabilities of accidental coincidence between the implantation of ER and individual decays were estimated as follows. The counting rate of decay events, which yielded no TOF signal for a decay energy greater than 8 MeV in the run (Run #196) and the strip (strip #12) where the decay chain was observed, was 8.5×10^{-4} s^{-1}. That for a decay energy greater than 100 MeV was 9.6×10^{-6} s^{-1}. With the position window of 0.6 mm/60 mm (= 0.01) and time differences of 0.344 ms for α_1, 9.6 ms for α_2, 16.8 ms for α_3, 2.5 s for α_4, and 43.4 s for SF, the probabilities of accidental coincidence were evaluated to be 2.9×10^{-9}, 8.2×10^{-8}, 1.4×10^{-7}, 2.1×10^{-5}, and 9.5×10^{-6}, for α_1, α_2, α_3, α_4, and SF, respectively. The singles counting rate of the PSD was 2 s^{-1} at the typical beam intensity.

For the ER event, TOF between the timing counters was 44.6 ns. The time resolution of the system was measured to be 0.5 ns FWHM for 5.5 MeV α-particles. The resolution is expected to be better for ER, where the energy measured by the PSD was 36.75 MeV. Figure 1 shows a scatter plot of energy and TOF for the run (Run #196) in which the decay chain was observed. In the figure, only events detected by strip #12 of the PSD are plotted. The point corresponding to the implantation event is indicated by a circle with an arrow. Loci corresponding to projectile-like particles ($A \approx 70$) and

Table I. Observed events. ΔE_{sum}: Energy resolution in FWHM. See text. ΔT: Time differences between events. Position: measured from the bottom of the detector.

	E_{PSD} (MeV)	E_{SSD} (MeV)	E_{sum} (MeV)	ΔE_{sum} (MeV)	ΔT	TOF (ns)	Position (mm)
ER	36.75	—	36.75	—	—	44.6	30.33
α_1	11.68	—	11.68	0.04	0.344 ms	—	30.49
α_2	6.15	5.00	11.15	0.07	9.26 ms	—	30.40
α_3	1.14	8.89	10.03	0.07	7.16 ms	—	29.79
α_4	9.08	—	9.08	0.04	2.47 s	—	30.91
SF	204.1	—	204.1	—	40.9 s	—	30.25

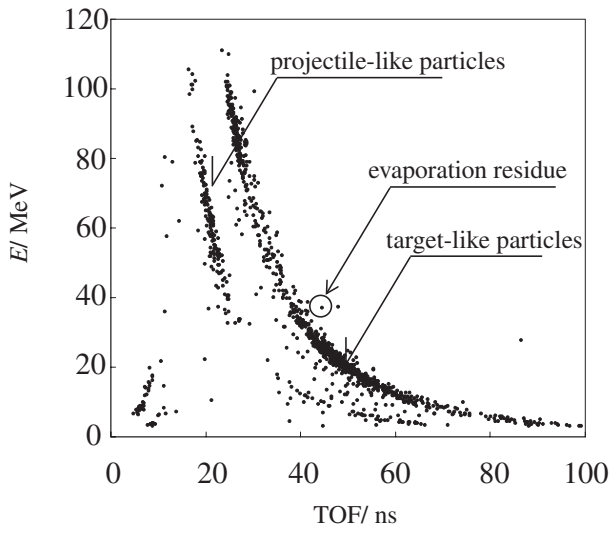

Fig. 1. Two-dimensional plot of energy measured by PSD vs TOF measured by timing counters for the run (run #196, time period = 12.7 h), in which the decay chain was observed. Events detected only by strip #12 of PSD are shown. A point corresponding to the implantation event is shown by a circle with an arrow. Loci corresponding to projectile-like particles ($A \approx 70$) and target-like particles ($A \approx 209$) are also shown.

target-like particles ($A \approx 209$) (target recoil and transfer reaction products) are seen in the figure. The implantation event is well separated from the locus of the target-like particles that may contribute to the background of the measurement. The figure indicates that the mass of the implanted particle is higher than that of the target-like particles. The mass number of ER was roughly estimated to be 280 ± 15 using a method similar to that described in ref. 2.

The total dose of the ^{70}Zn projectile was 1.7×10^{19}. The production cross section of this specific event was deduced to be 55^{+150}_{-45} fb (10^{-39} cm^2) using the transmission efficiency of GARIS of 0.8.[2)] Indicated uncertainty in the cross section is only statistical one with 1σ (68% confidence level).

The experiment was designed to produce the isotope, ^{278}113, by the one-neutron evaporation channel in the ^{209}Bi + ^{70}Zn complete fusion reaction. On the basis of a systematic study of the most probable reaction energies for the one-neutron evaporation channel in the ^{208}Pb(^{64}Ni,n)^{271}Ds,[1)] ^{209}Bi(^{64}Ni,n)^{272}111,[2)] and ^{208}Pb(^{70}Zn,n)^{277}112[3)] reactions, a reaction energy of 349.0 MeV at the half-depth of the targets was adopted to maximize the relevant cross section for producing ^{278}113. The corresponding energy of the center of mass frame is 261.4 ± 2.0 MeV, where ± 2.0 MeV indicates the range of reaction energy due to the energy loss of the beam in the bismuth targets. The excitation energy (E^*_{CN}) of the compound nucleus, ^{279}113, is calculated to be 14.1 ± 2.0 MeV using the predicted mass for a compound nucleus[13)] and the experimental masses of the beam and target.

If we assume that the implanted nucleus followed by the decay chain observed in the present study is the product of the ^{209}Bi(^{70}Zn,n)^{278}113 reaction, the members of the decay chain can be assigned as ^{278}113, ^{274}111, ^{270}Mt, ^{266}Bh, and ^{262}Db. The probability that the decaying α particle escapes in the backward direction and gives no signal below the threshold of PSD is 15%.[1,2)] Therefore, the members can also be possibly assigned as further α-decay products, such as ^{258}Lr. However, this possibility is excluded in the present case on the basis of the decay property of ^{258}Lr.

The isotope ^{262}Db is known to decay by α and spontaneous fission and branching ratios of $\approx$64% and $\approx$33%, respectively, with a half-life ($T_{1/2}$) with a 1σ error of 34 ± 4 s.[14)] The measured decay time of 40.9 s for SF, corresponding to $T_{1/2} = 28^{+140}_{-13}$ s, is well consistent with the reported value.

The α-decay of ^{266}Bh produced by the ^{249}Bk + ^{22}Ne reaction was reported by Wilk *et al.*[15)] This decay was followed by the α-decays of ^{262}Db and ^{258}Lr. The decay energy (E_α) and decay time for ^{266}Bh were 9.29 ($\pm \sim 0.10$) MeV and 0.87 s, respectively. The decay time, 0.87 s, corresponds to $T_{1/2} = 0.6^{+2.9}_{-0.3}$ s. The measured decay time of α_4 in the present work, 2.47 s, corresponding to $T_{1/2} = 1.7^{+8.2}_{-0.8}$ s, agrees within the 1σ error with the value reported by Wilk *et al.* Considering the rather wide distribution of α-decay energies from odd-odd nuclei, as shown in the case of ^{272}111,[2)] our present α_4 ($E_\alpha = 9.08 \pm 0.04$ MeV) does not contradict the value reported in ref. 15. It should be noted that only one α-decay event was reported in this reference.

The possibility of a radiative capture process (zero-neutron evaporation channel) to form the observed decay chain could not be excluded purely on the basis of the property of the measured SF. The isotope ^{263}Db, which is a decay product of ^{279}113 after four sequential α-decays, is also known to decay by spontaneous fission ($\approx$57%) and α-decay ($\approx$43%) with a half-life of 27^{+10}_{-7} s,[14)] consistent with the measured time for SF. However, this channel is excluded by considering the excitation energy of the compound nucleus. The calculated value, $E^*_{CN} = 14.1 \pm 2.0$ MeV, is too high to populate the ground state of the compound nucleus by only γ-ray emission, because the calculated one-neutron separation energy of the compound nucleus is only 7.5 MeV, using the same mass prediction.[13)]

The two-neutron separation energy of the compound nucleus is calculated to be 13.8 MeV using also the same theoretical prediction.[13)] The free energy for two-neutron emission is calculated to be only 0.3 MeV in this case. Therefore, the two-neutron evaporation channel is also excluded in this excitation energy because the phase volume of this channel is much smaller than that of the one-neutron evaporation channel.

The possibility of a one-proton evaporation channel leading to the product of ^{278}112 is excluded by comparing the observed decay time of SF with that of ^{262}Rf which decays by 100% spontaneous fission with a half-life of 47 ± 5 ms,[16)] greatly shorter than the observed value, $T_{1/2} = 1.7^{+8.2}_{-0.8}$ s.

In conclusion, the reaction product, followed by the decay chain observed in our experiment, was considerd to be most probably due to the ^{209}Bi(^{70}Zn,n)^{278}113 reaction. As a result, the members of the decay chain were consequently assigned as ^{278}113, ^{274}111, ^{270}Mt, ^{266}Bh, and ^{262}Db. The decay chain is shown in Fig. 2, together with decay energies (E_α and/or E_{SF}) and decay times.

In summary, a convincing candidate event of the isotope of the 113th element, ^{278}113, and its daughter nuclei, ^{274}111 and ^{270}Mt, were produced, for the first time, by the ^{209}Bi + ^{70}Zn reaction at a beam energy of 349.0 MeV with a total

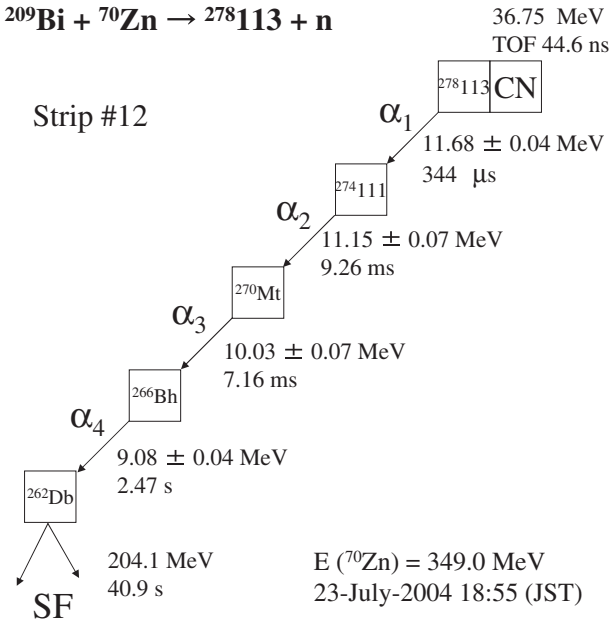

Fig. 2. Decay chain observed in irradiation of ^{209}Bi targets by ^{70}Zn projectiles. Measured energies and decay times are indicated in the figure.

dose of 1.7×10^{19}. The result led to the identification of an isotope of the 113th element for the first time. The production cross section of the isotope by the studied reaction was deduced to be 55^{+150}_{-45} fb (10^{-39} cm^2).

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