Production of cold target-like fragments in the reaction of $^{48}Ca + ^{248}Cm$

H. Gäggeler,* W. Brüchle, M. Brügger, M. Schädel, K. Sümmerer, and G. Wirth
Gesellschaft für Schwerionenforschung, D-6100 Darmstadt, Federal Republic of Germany

J. V. Kratz, M. Lerch, Th. Blaich, G. Herrmann,† N. Hildebrand, and N. Trautmann
Institut für Kernchemie, Universität Mainz, D-6500 Mainz, Federal Republic of Germany

D. Lee, K. J. Moody,‡ K. E. Gregorich, R. B. Welch,** and G. T. Seaborg
Nuclear Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

D. C. Hoffman,§ W. R. Daniels, and M. M. Fowler
Isotope and Nuclear Chemistry Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

H. R. von Gunten
Radiochemisches Laboratorium, Universität Bern, CH-3000 Bern 9, Switzerland
and Eidgenössisches Institut für Reaktorforschung, CH-5303 Würenlingen, Switzerland
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Yields for isotopes of Rn through Pu have been measured in the reaction $^{48}Ca + ^{248}Cm$ at an energy of 248–263 MeV (1.04–1.10 times the Coulomb barrier). Despite the low bombarding energy, high and essentially constant integral yields of about 1 to 2 mb for the elements Rn through U were observed. There is evidence that these nuclides are produced with little excitation energy.

I. INTRODUCTION

Recent attempts to produce superheavy elements by cold fusion between ^{48}Ca and ^{248}Cm have failed to yield evidence for fusion evaporation residues at a cross section limit of 10^{-34}–10^{-35} cm^2 within a half-life window between microseconds and years.[1] This result does not necessarily indicate a lower stability than that assumed so far for the compound nucleus $^{296}116$ which may be produced in this reaction. The evolution from entrance channel to surviving evaporation residues from a spherical compound nucleus is not known well enough. It may be that this very heavy system no longer fuses at energies near the Coulomb barrier due to dynamical hindrance (extra-push concept) (Refs. 2 and 3) or that the compound nucleus produced at some 20–40 MeV of excitation energy is entirely lost by prompt fission. It is therefore of importance to investigate the $^{48}Ca + ^{248}Cm$ reaction in some detail at energies near the interaction barrier to learn more about the interplay between different reaction channels. In a previous publication[4] we reported on transcurium isotope yields from the reactions $^{40}Ca + ^{248}Cm$ and $^{48}Ca + ^{248}Cm$ at energies between 0.9 and 1.35 times the Coulomb barrier. In this paper we present results of below-target cross sections for isotopes of Rn through Pu from the reaction of ^{48}Ca with ^{248}Cm at energies near the Coulomb barrier.

II. EXPERIMENTAL

Two series of experiments were performed at Lawrence Berkeley Laboratory (LBL) and at Gesellschaft für Schwerionenforschung (GSI) using radiochemical methods and on-line gas-jet transport of short-lived reaction products combined with electronic detection systems. The target array described in Ref. 4 was used for the radiochemical experiments and that described in Ref. 7 was used for the on-line experiments. Two targets containing 1.5 and 1.7 mg/cm^2 of ^{248}Cm in the form of curium oxide (96.5% ^{248}Cm, 3.5% ^{246}Cm) on 2.4 mg/cm^2 thick Be foils were used. The energy of the ^{48}Ca ions in the targets covered a range of 248–263 MeV with the exception of the ROMA experiments (see below), where the energy within the targets was 233–249 MeV. These energy ranges give the energy spread within the target and were measured with surface barrier detectors. The width of the ^{48}Ca beam energy distribution after leaving the ^{248}Cm target was measured to be about 13 MeV (FWHM).

For the off-line radiochemical experiments, products recoiling from the target were collected on 6–10 mg/cm^2 thick copper foils subtending laboratory angles from 0° to 60°. Near the barrier this angular acceptance should catch all targetlike products.

The chemical procedures that were used are described in Ref. 6. Six to eight hours were needed for the chemical separation and purification of the elements of interest. Chemical yields were assumed to be $(35 \pm 9)\%$ on the basis of yield determinations of the procedures. Final samples were counted alternately for γ and α decay using Ge(Li) and Si-surface barrier detectors. The γ-ray data were analyzed with standard computer programs to obtain absolute decay rates. Final cross sections were calculated with a set of programs which take into account chemical yields; the beam history to correct for growth and decay during bombardment, chemical separation times from possible precursors, target thickness, and beam integral.

The α peaks were integrated and decay rates were converted into cross sections applying the same programs.

In the case of the on-line experiments at GSI, the recoil atoms were thermalized in a gas-jet recoil chamber designed to collect both fusion and transfer products in the gas phase. The recoil atoms were swept out of the chamber and transported by Ar carrier gas within a few seconds through a teflon capillary (1 mm inner diameter and 40 to 60 m long) to two different detection systems. A cryogenic system[5] was used to determine yields of several Rn isotopes. The gas-jet transport system was coupled to a quartz tube filled with quartz powder and heated to 1000 °C to trap nonvolatile reaction products and then to a tube filled with tantalum powder kept at 800 °C to remove traces of water. The gaseous products were then condensed on a cooled surface kept at 50 K. This temperature allows quantitative adsorption of Rn without retaining the Ar carrier gas. Alpha events were registered in an annular detector placed in front of the cooled surface. Using absolutely calibrated Rn sources, the overall efficiency for transport, condensation, and α counting of this cryogenic system was determined to be 4%. During irradiation, α spectra were recorded on line. After the end of irradiation, successive spectra were taken to follow the decay of condensed reaction products.

Detection of short-lived Ra, Ac, and Th nuclides was performed with the second system, the rotating multidetector apparatus (ROMA).[7] It is designed to collect continuously recoil atoms attached to KCl clusters in the Ar carrier gas on thin (70 μg/cm^2) polypropylene foils mounted on a rotating wheel. This wheel was equipped with eight equally spaced foils around the circumference and was stepped every five seconds to position the foils between pairs of surface barrier detectors. The large number of short-lived daughter nuclides in equilibrium and the time lapse between neighboring detector pairs were used to extract initial decay rates of short-lived isotopes assuming a transport time between the gas-jet collecting chamber and the ROMA apparatus of about 10 sec. After the end of irradiation, the wheel continued rotation and α-particle spectra were accumulated periodically for 25 h. These spectra were used to determine activities of longer-lived nuclides. Approximately 300 d after the end of bombardment, a single foil from the wheel was counted for 150 h with a surface barrier detector. Activities due to ^{228}Th, ^{248}Cf, ^{252}Cf, and ^{252}Es were observed in addition to sputtered ^{248}Cm and some long-lived Po isotopes. With this information it was possible to normalize the relative cross sections from the ROMA experiment to the absolute actinide cross sections from the off-line radiochemical experiments.[4] In the normalization it was assumed that the cross sections measured relative to each other do not vary significantly between the energy ranges of 233–249 MeV and 248–263 MeV used in the ROMA and radiochemical experiments, respectively.

III. RESULTS AND DISCUSSION

Table I summarizes the measured cross sections for isotopes of Rn, Ra, Ac, Th, U, and Pu produced in the reaction of ^{48}Ca+^{248}Cm at a bombarding energy of 248–263 MeV. The errors listed in Table I include statistical uncertainties, and uncertainties in the counting efficiencies and chemical yields. In the case of Pu, data were obtained from two different experiments performed at LBL and GSI. The cross sections of ^{243}Pu, ^{245}Pu, and ^{246}Pu agree within the quoted errors although there seems to be a small systematic difference between the sets of data. In Fig. 1 the cross sections of Table I are depicted together with the data for transcurium yields[4] which were measured in the same experiments. For Pu, the weighted averages from Table I are shown. Despite the low bombarding energy ($1.04 \leq E/B \leq 1.10$) remarkably high cross sections were obtained for nuclides far below the targets. Peak cross sections of some 200 μb were measured for ^{219}Rn, ^{222}Ra, ^{225}Ac, and ^{227}Th. We conclude that their production by transfer reactions with possible target contaminants such as Pb or Bi is highly unlikely, because these isotopes are more neutron-rich compared to normal transfer products observed in heavy ion induced reactions with, e.g., Bi targets.[8] The measured isotope yields were fitted with Gaussian distributions (solid lines in Fig. 1). The transcurium isotope distributions are described reasonably well by Gaussians with a full-width at half maximum (FWHM) of 2.5 mass units. The distributions for the below-curium elements can be fitted by Gaussians with FWHM's of 5 to 6 mass units. No data exist for Fr,

TABLE I. Cross sections for below-target nuclides from ^{48}Ca+^{248}Cm at E_{lab}=248–263 MeV.

Nuclide	Cross section (μb)	Nuclide	Cross section (μb)
^{210}Rn	19±9	^{225}Th[a]	65±33
^{211}Rn	52±12	^{226}Th[a]	132±14
^{212}Rn	188±19	^{227}Th[a]	172±44
^{219}Rn	209±100	^{228}Th[a]	125±34[b]
^{220}Rn	126±40	^{230}U	0.6±0.1
^{221}Rn	45±17	^{237}U	69±22
^{222}Rn	10±3	^{236}Pu	3±1
^{221}Ra[a]	172±30	^{238}Pu	120±70[c]
^{222}Ra[a]	188±31	^{243}Pu	572±143[d]
^{223}Ra[a]	157±46		980±260[e]
^{223}Ac[a]	109±14		776±282[f]
^{224}Ac[a]	167±43	^{245}Pu	290±74[d]
			410±110[e]
^{225}Ac[a]	185±23		350±130[f]
^{226}Ac[a]	164±20	^{246}Pu	128±34[d]
			160±50[e]
			144±59[f]

[a]Energy within target 233–249 MeV. *Relative* yields from this measurement normalized to *absolute* yields from Ref. 4 for a 248–263 MeV bombardment (see the text).
[b]Corrected for contribution from ^{228}Ac precurser.
[c]Corrected for contribution from ^{238}Np precurser.
[d]Measurement from LBL.
[e]Measurement from GSI.
[f]Average value from LBL and GSI data.

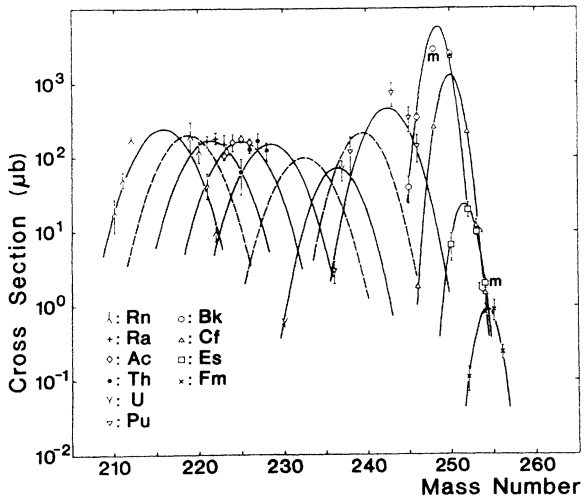

FIG. 1. Independent yields for below-target nuclides (this work) and above-target nuclides (from Ref. 4) for $^{48}Ca+^{248}Cm$ at a bombarding energy of $E_{lab}=248-263$ MeV. The solid lines show best-fit Gaussian curves through the measured isotope yields at fixed Z. They have significantly different widths with 2.5 u (FWHM) for Bk through Fm (above Cm elements), 5.0 for U and Pu, and 5.5 for Rn, Ra, Ac, and Th (below Cm elements), respectively. The dashed lines reveal interpolated yield curves for Fr, Pa, and Np.

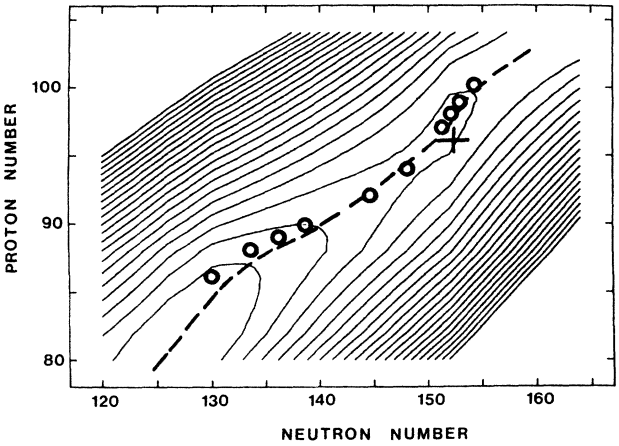

FIG. 2. Calculated potential energy surface for $^{48}Ca+^{248}Cm$ including shell effects and deformations of the separating fragments. Angular momentum effects were neglected ($l=0$) and R_{min} was taken as 11 fm (for details see Ref. 12). Shown are equipotential lines with a spacing of 10 MeV. The cross represents the injection point (^{248}Cm). The dashed line shows the bottom of the potential energy surface. The circles depict the experimentally determined maxima (most probable product masses A_p) of the Gaussian curves from Fig. 1.

Np, and Pa and the dotted curves present estimated yield curves. The observed significant change in the widths of the Gaussian curves between trans- and below-curium elements may indicate that they are produced by different reaction mechanisms.

The reaction of ^{48}Ca with ^{238}U has been studied using detector techniques.[9,10] At a bombarding energy of 5.4 MeV/u (1.13 E/B), which is comparable to our energy, a rather flat mass distribution between about $A=50$ and 220 was observed. This mass distribution is attributed to quasifission.[9,10] No information was gained on the N/Z ratio of products from this reaction channel because the applied detector technique allowed determination of mass yields only. The occurrence of quasifission is known to be correlated with a significant dynamical suppression of compound nucleus formation.[10,11] Our observation of rather constant yields from Pu to Rn suggests a mass distribution similar to that observed[9,10] in the $^{48}Ca+^{238}U$ system. This might be evidence for a hindrance to fusion in the $^{48}Ca+^{248}Cm$ reaction and could explain why no superheavy elements were observed in previous experiments.[1]

The maxima of the Gaussian distributions, A_p, from Fig. 1 agree well with predictions made on the basis of potential energy surface calculations. Such a model was shown[12] to describe A_p values for deep-inelastic transfer reactions. In Fig. 2 the results of a potential energy surface calculation for $^{48}Ca+^{248}Cm$ are shown taking into account Coulomb, centrifugal, and nuclear potentials of the entrance and exit channels for touching spherical nuclei and Q_{gg} values which were corrected for pairing effects.[12] The cross in Fig. 2 shows the injection point of the system before any interaction ($^{248}_{96}Cm$). It is expected that the evolving system proceeds from this point toward the valley of the potential energy surface. This corresponds to the highest excitation energies, i.e., highest level densities for the fragments in the exit channel. Indeed, the experimentally deduced A_p values (circles) closely follow the bottom of the potential energy valley. The slight displacement of the A_p values for far below curium elements toward lower neutron numbers compared to the minimum potential energy predictions is due to neutron evaporation from the primary fragments. The differences, $A_p'-A_p=\Delta A$, where A_p' corresponds to the bottom of the potential energy surface (dashed line), give an estimate of the number of neutrons evaporated by the primary fragments and, hence, an estimate of the excitation energies of surviving heavy products. For the elements U through Fm, the ΔA values are not significantly different from zero which indicates that these nuclides are produced with low excitation energy, i.e., essentially "cold." For the elements Rn through Th the number of evaporated neutrons increases to about 2, thus indicating excitation energies of about 15–20 MeV in the heavy fragments. This is in line with the dip in cross section around uranium (see Fig. 1). This local minimum most likely results from losses of primary fragments by prompt fission. Indeed, if one assumes those products to be produced with some 15–20 MeV excitation energy, uranium isotopes around $A=236$ are expected[13] to fission away with about 50% probability, whereas lower Z elements should have survival probabilities larger than 90%. The cold formation of the heavy fragments is not expected to be due to a secondary effect, i.e., due to sequential fission of more highly excited primary species, because very little sequential fission was reported for similar collisions of ^{48}Ca,

TABLE II. Calculated excitation energies for heavy fragments from $^{48}Ca+^{248}Cm$ at $E_{lab}=248-263$ MeV.

Element	Primary mass A_p'[a]	Ground-state Q_{gg} value (MeV)	Fragment kinetic energy[b] (MeV)	Excitation energy E^* for heavy fragment[c] (MeV)
Rn	218	+75.1	260.0	17–27
Ra	223.5	+57.7	249.5	12–22
Ac	227	+46.9	244.4	8–18
Th	230.5	+40.9	238.6	8–18
U	236.5	+25.4	226.6	5–16
Pu	242.25	+12.9	214.4	5–16
Bk	249	−9.9	192.0	5–16
Cf	250.25	−16.9	184.0	6–17
Es	252	−31.9	176.1	0–11
Fm	254.75	−39.6	168.2	0–12

[a]From Fig. 2 (dashed line).

[b]Calculated with: $E_{out}=1.438Z_3Z_4/1.16(A_3^{1/3}+A_4^{1/3}+2)$, where Z_3,Z_4 and A_3,A_4 are the charge and mass numbers of the separating fragments.

[c]Calculated for the bombarding energies 248 and 263 MeV with

$$E^*(A_4)=[A_4/(A_3+A_4)](E_{c.m.}+Q_{gg}-E_{out}),$$

where A_4 is the heavy fragment and $E_{c.m.}$ the bombarding energy in the center of mass system.

^{45}Sc, and ^{50}Ti with ^{238}U at near-barrier energies.[10]

In the following we discuss two alternatives for forming neutron-rich heavy fragments with high survivability in $^{48}Ca+^{248}Cm$ reactions near the barrier. The low excitation energies of heavy fragments are reproduced if one assumes a relatively compact scission shape for the separating fragments (no prolate deformation). Table II summarizes calculated heavy fragment excitation energies which were obtained under the assumptions that: (1) the outgoing fragments are spherical and separate at a scission radius given by $r=1.16(A_3^{1/3}+A_4^{1/3}+2)$ fm, where A_3 and A_4 are the mass numbers of the separating nuclei; (2) angular momentum effects can be neglected because of the near barrier energy;[12] and (3) the total excitation energy is shared between A_3 and A_4 in proportion to the fragment masses. As Table II shows, the calculated excitation energies are in good agreement with those estimated from Fig. 2. An interesting aspect of such reaction energetics is the low excitation energy and extreme neutron-richness of the complementary light fragments. Complementary to the most probable Rn, Ra, Ac, and Th isotopes shown in Fig. 1, are exotic nuclides such as ^{79}Zn, ^{73}Ni, ^{69}Co, ^{66}Fe, and ^{60}Cr. Due to the large width of the isotopic distributions (5 to 6 mass units FWHM) even more neutron-rich isotopes might be formed. In the case of the assumed compact scission shapes, the excitation energies would be very low, i.e., close to or even below the neutron separation energies in the light fragments. However, one should keep in mind that the estimates of excitation energies mentioned above are model dependent.

As an alternative assumption, prolate deformations of the separating fragments[14,15] may be assumed. Then, for equal temperatures in both fragments this would give rise to some additional 50 MeV of excitation in the heavy fragment. It is difficult to see how these highly excited fissionable fragments could ever survive. But there is evidence that they do, both in our data and those of Töke *et al.*[10] However, since fissionlike energies are consistent with the $^{48}Ca+^{238}U$ experiment for fragments from quasifission processes, it is the second assumption that might be erroneous, i.e., the assumption of a thermal equilibrium between both reaction products. In order to produce cold and neutron-rich heavy fragments by the observed flow of mass from target to projectile, the complementary light fragments would be highly excited. This would be in line with wall-and-window friction[16] occurring mostly in the light fragment because the mass flow is extremely one-directional. If we assume 15–20 MeV of excitation energy in the Rn through Th isotopes, then the remaining excitation energy in the complementary light isotopes of Zn through Cr would be between 60 and 90 MeV. This would produce light evaporation residues with neutron numbers close to the stability line. Nonequilibrium division of the available excitation energy between target and projectile has already been observed in several heavy ion induced reactions.[17,18] For example, for the system $^{58}Fe+^{238}U$ at 8.5 MeV/u and total kinetic energy losses up to about 70 MeV, it is found[18] that the total excitation energy divides about equally between heavy (h) and light (l) fragments, i.e., $E_h^*/E_l^*\simeq 1$. Our interpretation, however, goes far beyond such an assumption because we would have to assume $E_h^*/E_l^*\simeq 0.2-0.3$. Such an extreme distribution of the total excitation energy onto one fragment has so far only been observed in few nucleon exchange reactions with light ions such as, e.g., in the reaction $^{14}N+^{164}Gd$.[19]

IV. SUMMARY

High and essentially constant yields for the below-curium elements Rn through Pu were observed. A com-

parison between predicted primary and measured mass numbers of the targetlike products gives evidence that surviving reaction products are produced with very low excitation energy. We have suggested that this low excitation energy may be due to compact scission shapes. In such events the production of cold, extremely neutron-rich Zn through Cr isotopes might be expected as complementary fragments. On the other hand, evidence contained in the binary coincidence data[9,10] for the reaction $^{48}Ca+^{238}U$ suggests that deformed scission configurations, i.e., much higher excitation energies, are obtained. Then, the high survivability of the targetlike fragments with respect to sequential fission seems to indicate mass flow from target to projectile which would produce a light fragment that is more highly excited than its heavy complement. In this case neutrons would probably be evaporated from the highly excited light fragment and extremely neutron-rich isotopes of light elements would not result. Both possibilities discussed suggest novel and interesting consequences which should be checked experimentally.

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*Present address: Eidgenössisches Institut für Reaktorforschung, Würenlingen, Switzerland.

†Also at Gesellschaft für Schwerionenforschung, Darmstadt, Federal Republic of Germany.

‡Present address: Lawrence Livermore National Laboratory, Livermore, CA 94550.

§Present address: Lawrence Berkeley Laboratory, Berkeley, CA 94720.

**Present address: Radiochemisches Laboratorium, Universität Bern, Bern, Switzerland.

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