PHYSICAL REVIEW C VOLUME 52, NUMBER 1 JULY 1995

α decay of the new isotopes ^{210}Th and ^{211}Th

J. Uusitalo, T. Enqvist, M. Leino, and W. H. Trzaska
Department of Physics, University of Jyväskylä, P.O. Box 35, 40351 Jyväskylä, Finland

K. Eskola
Department of Physics, University of Helsinki, 00014 Helsinki, Finland

P. Armbruster and V. Ninov
Gesellschaft für Schwerionenforschung, D-64220 Darmstadt, Germany
(Received 2 March 1995)

The new neutron-deficient isotopes ^{210}Th and ^{211}Th have been produced in fusion reactions with 5.2–5.7 MeV/nucleon ^{35}Cl ions on ^{181}Ta. The activities were identified on the basis of correlated α decay chains. Fusion evaporation products were separated in-flight using a gas-filled magnetic recoil separator. The measured α energies of ^{211}Th and ^{210}Th are (7792±14) and (7899±17) keV, respectively. The half-lives were found to be (37^{+28}_{-11}) ms (^{211}Th) and (9^{+17}_{-4}) ms (^{210}Th).

PACS number(s): 21.10.Dr, 23.60.+e, 25.70.−z, 27.80.+w

I. INTRODUCTION

The study of α decay properties of very neutron-deficient isotopes of heavy elements provides data on the mass surface close to the proton drip line. At present, the most efficient method of producing these nuclei is heavy-ion-induced fusion followed by neutron evaporation. Measurement of production cross sections of evaporation residues gives insight into the role of fission in the deexcitation process.

The present study is part of a program aimed at improving our understanding of heavy nuclei in the region $Z > 82, N < 126$ [1–3]. The half-lives of the most neutron-deficient isotopes of each element in this region are much shorter than 1 s and the production cross sections are smaller than 1 μb. Consequently, efficient and fast experimental methods are needed. During the past years, the standard apparatus for research in this field has been a high-acceptance recoil separator combined with a position-sensitive focal plane detector [2,4–6]. At the University of Jyväskylä, the gas-filled recoil separator RITU [2] has been constructed for the study of heavy elements produced in fusion. It combines high transmission, typically 10–20 % for reactions studied in this work, with separation times on the order of 1 μs.

Neutron-deficient isotopes of thorium were studied down to ^{213}Th by Valli and Hyde [7] using the ^{16}O + ^{206}Pb reaction and the helium-jet technique. Although the speed of the He-jet apparatus should still be adequate for observing even lighter Th isotopes, decreasing yields make the application of this technique difficult. Vermeulen *et al.* used the velocity filter SHIP [4] to identify ^{212}Th produced in the reaction ^{40}Ar + ^{176}Hf [8]. The method used in Ref. [8] as well as in the present work was to observe correlated α decay chains. This is a simple and reliable way to identify new α activities provided the effect of accidental correlations is thoroughly investigated. Under sufficiently low background conditions even one observed decay chain may be sufficient for the identification of a new nuclide as shown in recent work in the region of man-made elements [9].

Preliminary results from our work on the decay of ^{210,211}Th have been presented in Refs. [2,3]. New data measured under improved experimental conditions have been included in the present work.

II. EXPERIMENT

The heavy-ion-induced complete fusion evaporation reaction ^{181}Ta(^{35}Cl,xn)$^{216-x}$Th was used to synthesize neutron-deficient isotopes of thorium. The ^{35}Cl beam was produced by the K = 130 MeV cyclotron of the Accelerator Laboratory at the Department of Physics of the University of Jyväskylä (JYFL). The metal ions from volatile compounds (MIVOC) method [10] developed at JYFL was applied to produce the chlorine beam from an electron-cyclotron-resonance (ECR) ion source. The beam intensity and total dose were determined by regularly measuring the beam current with a Faraday cup in front of the target. The beam energy as determined by the cyclotron frequency was checked by measuring the field in the 90° bending magnet which delivered the beam into the separator cave. The bombarding energy was adjusted for excitation function measurements using a set of nickel degrader foils placed in front of the targets. The energy loss in the degrader foils was calculated using the TRIM program [11]. The bombarding energies were 182, 191, and 199 MeV. Typical beam intensities were 4×10^{11} particles/s and the total doses were $(1\text{–}2)\times10^{16}$ particles at each bombarding energy. Targets of rolled tantalum foils of thickness 350–390 μg/cm^2 were provided by the GSI (Gesellschaft für Schwerionenforschung) target laboratory.

The fusion evaporation products were separated in-flight from the primary beam particles using the gas-filled recoil separator RITU [2]. In a gas-filled recoil separator the fusion products which emerge from the thin target with a wide ionic charge state distribution are focussed into the focal plane independently of the original charge states. High transmission can thus be achieved. The transmission of RITU in the ^{175}Lu(^{40}Ar,4–5n)^{210,211}Ac reaction was determined to be approximately 25%.

At the focal plane of RITU there is a position sensitive PIPS detector made by Canberra Semiconductor, Belgium, for detecting impinging evaporation residues and their subsequent α decays. The detector is 35 mm high and 80 mm wide and it is divided into eight individual pads. The α energy resolution of the pads is typically 30 keV full width at half maximum (FWHM) in on-line conditions. Energy and position signals from the detector are recorded simultaneously with two different amplifications, one optimized for heavy recoil nuclei and the other for α particles. Position resolution (vertical) has been determined to be < 0.5 mm (FWHM). In addition, times of occurrence are recorded with 0.1 ms precision for all events observed in the detector. Events are stored in list mode for off-line analysis.

In the energy domain of the α particles there is a continuous background caused by beam-related events such as multiply scattered beam particles and targetlike recoils which have been slowed down. One could suppress these events by requiring anticoincidence with a transmission detector placed upstream from the focal plane detector [12]. However, such a detector operating in helium would reduce the transmission due to scattering in the detector windows. In some of the measurements in the present work, the accelerator beam was pulsed to provide clean α spectra during the beam pause. The pulsing electronics also provided the signal to the data acquisition system to tag each event according to whether it was in pulse or in pause.

The pressure of the helium filling gas in RITU was typically 100 Pa. A continuous flow of gas was introduced close to the target, and a pressure-controlled electric butterfly valve was used to regulate the pumping of the helium from the focal plane detector chamber to maintain a constant pressure. The helium atmosphere of RITU was separated from the high vacuum of the cyclotron beam line by a 0.45 mg/cm^2 nickel foil.

The proper field in the separator dipole magnet was chosen on the basis of previously measured data [12] and fine tuned according to the measured horizontal distribution of evaporation residues in the focal plane detector.

The α particle energy calibration was based on the decay of known activities produced in the bombardments and imbedded in the detector material. In some cases there is a slight uncertainty of the calibration energy due to the production of pairs of isotopes with nearly identical decay properties, a feature characteristic of this region of the chart of nuclei. The following data were used for the energy calibration: (6260±4) keV (^{205,206}Rn); (6416±3) keV (^{204}Rn); (7133±5) keV (^{207,208}Ra); (7470±15) keV (^{210,211}Ac).

III. RESULTS

As an example of singles α spectra, the low energy part of the pause spectrum from the 199 MeV ^{35}Cl + ^{181}Ta bombardment is shown in Fig. 1. The spectrum is dominated by activities arising from charged particle evaporation. To identify neutron-deficient thorium isotopes, the method of searching for correlated events [13] has been used in the analysis of the list mode data. Two events are taken as correlated if they occur in the same detector pad within a position window determined by the position resolution and within the specified time window. The significance of a correlation can be estimated on the basis of average counting rates. In the present work, events are classified as evaporation residues (ER) or α particles (α) depending on their energy. The evaporation residues are slowed down in the target, in the filling gas, and in the detector window. Their implantation energy signals are further reduced by the pulse height defect. At 199 MeV bombarding energy, the initial energy of the evaporation residues was 31 MeV and the nominal mean implantation energy was determined to be approximately 22 MeV. In the correlation search, an energy window of 14–30 MeV was used for the evaporation residues. In the following, the α particle energies of previously known isotopes are given according to Rytz [14] whenever possible.

The exceptional regularity of α decay energies and half-lives of neutron-deficient Th and Ra isotopes leads one to expect that the decay properties of ^{212}Th and ^{211}Th are almost identical. Since the same is true for ^{208}Ra and ^{207}Ra, an unambiguous identification of ^{211}Th and ^{212}Th by using the correlation method requires the detection of decay chains including the decays of the Rn granddaughters. In irradiations with 182 MeV and 191 MeV ^{35}Cl ions altogether five chains of the type ER-α_1-α_2-α_3 were observed in which the last decay could be assigned to ^{203}Rn on the basis of its unique decay characteristics [15]. No ER-α_1-α_2-α_3 chains originating from ^{212}Th were

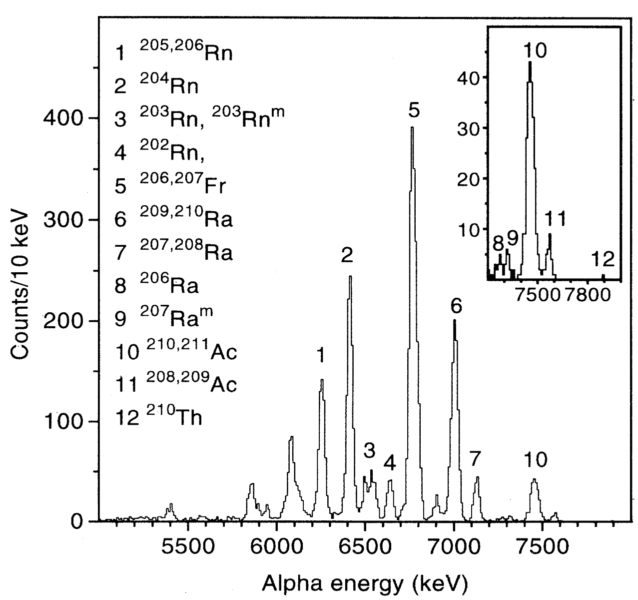

FIG. 1. The total (sum over all detector pads) singles pause α spectrum measured in the 199 MeV ^{35}Cl + ^{181}Ta irradiation.

observed at these energies. This is partly due to the long half-life, 1.24 min, and an appreciable β branch, 27%, of ^{204}Rn [16]. The measured α energy and half-life of the daughter activity were (7136±12) keV and $(1.1^{+0.9}_{-0.3})$ s, respectively. These data are compatible with the known decay properties of ^{207}Ra [17], $T_{1/2}$ = (1.3±0.2) s, and E_α = (7133±5) keV. The α particle energies of all observed mother-daughter event pairs from the 191 MeV and 199 MeV ^{35}Cl + ^{181}Ta irradiations are shown in Fig. 2. The measured α energy of the granddaughter activity was (6499±10) keV and the half-life was ~ 50 s. These properties are compatible with the decay of the (45±3) s ^{203}Rn with an α energy of (6498±5) keV. The error probability of the assignment is estimated to be 10^{-4}. By error probability we mean here the probability that the correlation is due to chance events [18]. This was calculated on the basis of the observed number of ER-α_1-α_2 chains identified as possible ^{211,212}Th-^{207,208}Ra decays and from the average counting rate of α-particle-like events within the energy range of 6470–6530 keV. In the 199 MeV ^{35}Cl irradiation during which the beam was pulsed, no events assigned to either ^{211}Th or ^{212}Th were observed.

Altogether two decay chains assigned to the new isotope ^{210}Th were observed, one at the 191 MeV bombarding energy and the other at 199 MeV. In the 191 MeV irradiation with a continuous accelerator beam, one correlated decay chain with two α events was observed in which the daughter activity could be identified as ^{206}Ra [19] (Fig. 2). The error probability was 2×10^{-4}. In the 199 MeV irradiation in which the beam was pulsed with a frequency of 25 Hz and a duty factor of 25%, one correlated three-α chain was found with all α particles occurring in the 30 ms long pauses. The daughter and granddaughter decays could be assigned to ^{206}Ra and ^{202}Rn [20], respectively. Due to the small counting rate of α-like events in the pause (Fig. 1), the error probability for this chain was 2×10^{-12}. The measured α energy and half-life of the daughter activity, (7262±15) keV and $(0.40^{+0.70}_{-0.16})$ s, respectively, are compatible with those of ^{206}Ra [18] for which an energy of (7272±5) keV and a half-life of (240±20) ms have been reported. Thus the two mother decays are assigned to the new isotope ^{210}Th. Our measured decay data for ^{210,211}Th are given in Table I.

TABLE I. α particle energies (E_α) and half-lives ($T_{1/2}$) of isotopes ^{210}Th and ^{211}Th. Reduced α decay widths δ^2 and half-lives normalized to ^{212}Po, calculated according to Ref. [21], are also given.

Nuclide	E_α (keV)	$T_{1/2}$ (ms)	δ^2 (keV)	Calc. $T_{1/2}$ (ms)
^{211}Th	7792±14	37^{+28}_{-11}	56	25
^{210}Th	7899±17	9^{+17}_{-4}	111	12

Production cross sections of 30 nb and 3 nb were determined for ^{211}Th and ^{210}Th, respectively. These are approximate values since the estimation of the transmission for reactions in which a large number of neutrons is evaporated from the compound nucleus suffers from lack of knowledge concerning details of the nucleon emission.

IV. DISCUSSION

There are no previously published data on the isotopes ^{210,211}Th. On the other hand, the previously known isotope ^{212}Th could not be observed in our measurements. The evidence for the identification of the new isotopes is provided by correlated decay chains leading to known

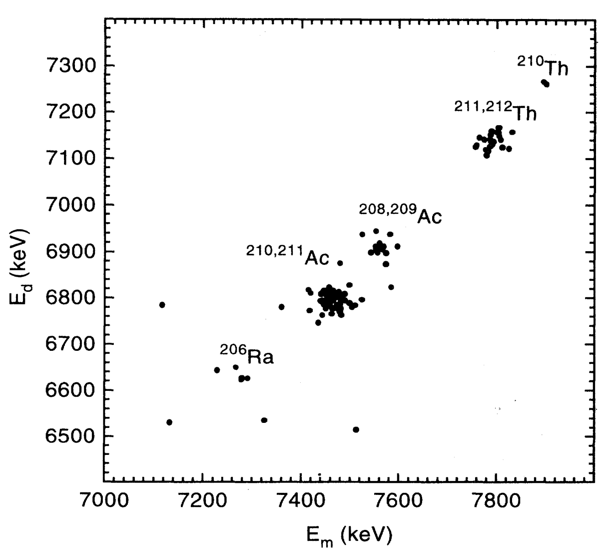

FIG. 2. Mother (E_m) and daughter (E_d) α particle energies for all chains of the type ER-α_1-α_2 observed in the 191 MeV and 199 MeV ^{35}Cl + ^{181}Ta irradiations. Maximum search times were 200 ms for the ER-α_1 pair and 10 s for the α_1-α_2 pair.

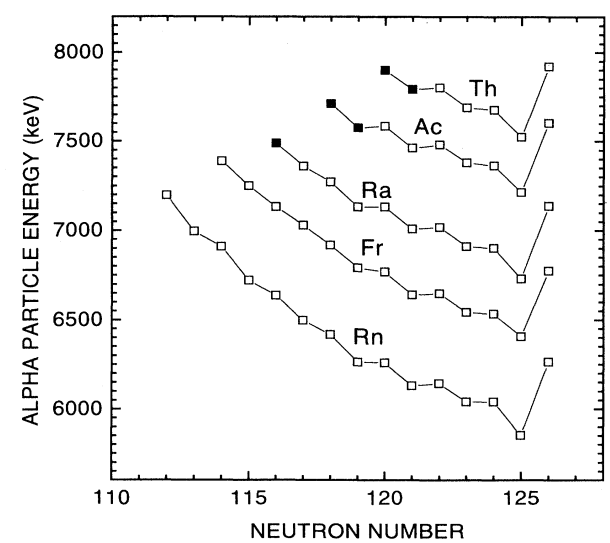

FIG. 3. α particle energies of ground state to ground state transitions for neutron-deficient Rn, Fr, Ra, Ac, and Th isotopes. Solid squares refer to values measured using RITU. A preliminary value for the α energy of ^{204}Ra from Ref. [3] is included. The rest of the data are from the present work, from Ref. [1] (^{207,208}Ac), and from the appropriate Nuclear Data Sheets.

nuclides. α energy calibration is determined on the basis of $z\alpha ypxn$ evaporation products observed in the irradiations. The lack of calibration peaks with well-established energies above 7133 keV and the small yields are reflected in the uncertainties given here for the α particle energies (Table I).

The measured α particle energies and half-lives fit well into the systematics of very neutron-deficient nuclei in this region. In particular, the decay properties of ^{212}Th and ^{211}Th are similar. The α particle energies of ^{212}Th and ^{211}Th are (7802$\pm$10) keV [8] and (7792$\pm$14) keV (present work), respectively. The corresponding half-lives are (30^{+20}_{-10}) ms [8] and (37^{+28}_{-11}) ms (present work). Half-lives calculated with the formalism of Rasmussen [21] and normalized to ^{212}Po are given in Table I. Experimental α particle energies of neutron-deficient Rn, Fr, Ra, Ac, and Th isotopes are shown in Fig. 3.

All decay chains starting from ^{211}Th were observed to proceed via the 45 s ^{203}Rn. No evidence for the feeding of the 55 ms isomer in ^{207}Ra [22] was observed.

The low yield of the activities does not allow precise determination of optimum bombarding energies but according to a rough estimate, the peaks of the excitation functions correspond to an excitation energy of 66 MeV and 77 MeV for ^{211}Th and ^{210}Th, respectively.

The low background conditions together with ever shorter half-lives which are beneficial to the correlation search would make the synthesis of even lighter Th isotopes feasible. Ground state proton decay is not expected to be a dominant decay mode until further beyond the proton drip line. However, increasingly longer beam times required make the continuation beyond ^{210}Th questionable with present maximum beam currents as dictated by the use of stationary targets.

ACKNOWLEDGMENT

This work was supported by the Academy of Finland.

[1] M. Leino, J. Uusitalo, T. Enqvist, K. Eskola, A. Jokinen, K. Loberg, W. H. Trzaska, and J. Äystö, Z. Phys. A **348**, 151 (1994).

[2] M. Leino *et al.*, in Proceedings of the Thirteenth International Conference on the Application of Accelerators in Research and Industry, Denton, Texas, 1994, edited by J. L. Duggan and I. L. Morgan [Nucl. Instrum. Methods B (to be published)].

[3] M. Leino *et al.*, in Proceedings of the XXIX Zakopane School of Physics, Zakopane, Poland, 1994, edited by R. Broda [Acta Phys. Pol. B **26**, 309 (1995)].

[4] G. Münzenberg, W. Faust, S. Hofmann, P. Armbruster, K. Güttner, and H. Ewald, Nucl. Instrum. Methods **161**, 65 (1979).

[5] A. V. Yeremin *et al.*, Nucl. Instrum. Methods A **274**, 528 (1989).

[6] Yu. A. Lazarev *et al.*, in *Proceedings of the International School-Seminar on Heavy Ion Physics*, Dubna, Russia, 1993, edited by Yu. Ts. Oganessian, Yu. E. Penionzhkevich, and R. Kalpakchieva (Joint Institute for Nuclear Research, Dubna, 1993), Vol. 2, p. 497.

[7] K. Valli and E. K. Hyde, Phys. Rev. **176**, 1377 (1968).

[8] D. Vermeulen, H.-G. Clerc, W. Lang, K.-H. Schmidt, and G. Münzenberg, Z. Phys. A **294**, 149 (1980).

[9] G. Münzenberg, Rep. Prog. Phys. **51**, 57 (1988).

[10] H. Koivisto, J. Ärje, and M. Nurmia, Nucl. Instrum. Methods B **94**, 291 (1994).

[11] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon Press, New York, 1985).

[12] A. Ghiorso, S. Yashita, M. E. Leino, L. Frank, J. Kalnins, P. Armbruster, J.-P. Dufour, and P. K. Lemmertz, Nucl. Instrum. Methods A **269**, 192 (1988).

[13] K.-H. Schmidt, W. Faust, G. Münzenberg, H.-G. Clerc, W. Lang, K. Pielenz, D. Vermeulen, H. Wohlfarth, H. Ewald, and K. Güttner, Nucl. Phys. **A318**, 253 (1979).

[14] A. Rytz, At. Data Nucl. Data Tables **47**, 205 (1991).

[15] M. R. Schmorak, Nucl. Data Sheets **46**, 287 (1985).

[16] M. R. Schmorak, Nucl. Data Sheets **72**, 409 (1994).

[17] M. R. Schmorak, Nucl. Data Sheets **43**, 383 (1984).

[18] K.-H. Schmidt, C.-C. Sahm, K. Pielenz, and H.-G. Clerc, Z. Phys. A **316**, 19 (1984).

[19] R. G. Helmer and M. A. Lee, Nucl. Data Sheets **61**, 93 (1990).

[20] M. R. Schmorak, Nucl. Data Sheets **50**, 669 (1987).

[21] J. O. Rasmussen, Phys. Rev. **113**, 1593 (1959).

[22] F. P. Heßberger, S. Hofmann, G. Münzenberg, A. B. Quint, K. Sümmerer, and P. Armbruster, Europhys. Lett. **3**, 895 (1987).