RESEARCH ON HEAVY ELEMENTS USING THE JYFL GAS–FILLED RECOIL SEPARATOR RITU*†

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A gas-filled recoil separator for studies of heavy elements produced in heavy-ion-induced fusion reactions has been constructed. New neutron-deficient isotopes with $Z = 85 \div 90$ have been identified through their alpha decay.

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1. Introduction

During the last fifteen years, high efficiency recoil separators for heavy-ion-induced fusion products have provided a wealth of new information on heavy elements (defined in this work as elements with $Z > 82$). In these devices, the fusion products recoiling out of a thin target are collected in

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the focal plane detector while the primary beam is kinematically separated in-flight using electric and/or magnetic fields [1]. The separation time is on the order of 1 μs and the efficiency is typically 10–30 percent in the case of fairly symmetric reactions. Suppression of the full energy accelerator beam is usually excellent while mass resolution is often moderate. The identification of nuclides is normally based on decay chains leading to known isotopes. The region of heavy elements where production rates are small, half-lives are short, and alpha decay is an important decay mode, is a natural working ground for heavy ion recoil separators.

Perhaps the most well-known recoil separator is the velocity filter SHIP [2] at GSI, Darmstadt. Development of target preparation [3] and focal plane and time-of-flight detector techniques has made possible the identification of the three known heaviest elements as well as many other discoveries, mainly in the heavy element region [4]. Another device which has been productive in this field is the JINR, Dubna, electrostatic separator VASSILISSA [5].

A relatively inexpensive method to achieve high transmission is to use a dipole magnet with the field region filled with dilute gas such as helium to collect all ionic charge states of the fusion products. In the following, work done using the gas-filled recoil separator RITU [6, 7] at the Accelerator Laboratory of the Department of Physics, University of Jyväskylä (JYFL) is described. The separator was designed specifically for work in the heavy element region. In the first experiments, new neutron-deficient isotopes with $Z = 85$–90 have been synthesized. Experiments in the transuranium region have also been performed. Preliminary results from these studies will be presented here.

2. Gas-filled separators in the study of heavy elements

The idea of using a gas-filled separator magnet was realized by Cohen and Fulmer in studies of fission products [8]. Later, the method was developed by Karnaukhov *et al.* [9] and Armbruster *et al.* [10]. The first gas-filled separator used in the study of heavy elements produced in heavy ion reactions [11] was SASSY [12] constructed at LBL, Berkeley. It was used for example to search for superheavy elements produced in the reaction ^{48}Ca + ^{248}Cm [13]. Other gas-filled separators used in studies of transuranium elements are the GSI post-separator NASE [14] and the JINR, Dubna separator [15].

In a heavy ion fusion reaction, products recoiling out of the target have a wide charge state distribution and normally only part of these products can be collected after magnetic separation. If the gap of the separator's dispersive element is filled with low pressure gas so that the recoiling ions will suffer a few thousand collisions with gas atoms while traversing the field region, they will follow a trajectory determined by the average ionic charge q_{ave} in the gas. This will lead to a high transmission. The trajectory of the primary beam particles will normally be sufficiently different from that of the wanted products so that very good full energy primary beam suppression on the order of $10^{-12} - 10^{-15}$ is achieved [12]. Since the average charge state in the gas is to first order directly proportional to the velocity of the ions, velocity focussing is also achieved. The important properties of the separator can be combined into the following formula for the magnetic rigidity $B\rho$:

$$B\rho = \frac{mv}{q_{\rm ave}} = \frac{mv}{[(v/v_0)eZ^{1/3}]} = \frac{0.0227 \cdot A}{Z^{1/3}}\mathrm{Tm}, \qquad (1)$$

where v_0 is the Bohr velocity $= 2.19 \cdot 10^6$ m/s and A and Z are the mass and proton number of the ion, respectively. The expression $q_{\rm ave} = (v/v_0)eZ^{1/3}$ above results from using the Thomas–Fermi model of the atom. It can be seen from (1) that a gas-filled separator is essentially a mass separator. The mass resolution is low, typically on the order of 10 percent.

2.1. RITU, the JYFL gas-filled separator system

An important factor in the performance of a gas-filled separator is multiple scattering in the gas. As the pressure of the filling gas is varied, an optimum pressure can be found at which the focal plane image size is minimized. This is due to the interplay between charge state focussing on the one hand and multiple scattering on the other hand.

Normally, gas-filled separators have been designed so that separation of the primary beam takes place immediately behind the target to reduce widening of the primary beam. This has lead to the simple DQQ design of SASSY [12], the GSI gas-filled separator [14], and the Dubna separator [15]. In the design of RITU, a short strong quadrupole was added in front of the dipole for improved matching with the acceptance of the dipole. A hexapole was added to correct some of the aberrations especially in vacuum mode. The scheme of RITU is shown in Fig. 1. Some of the design parameters are given in Table I.

TABLE I

RITU parameter values

Magnetic configuration	$Q_1DQ_2Q_3$	Dipole exit angle	$-25°$
Maximum beam rigidity	2.2 Tm	Pole gap (D)	100 mm
Bending radius	1.85 m	Q_1 effective length	350 mm
Acceptance	10 msr	Q_1 maximum gradient	13.5 T/m
Dispersion	10 mm/%	Q_1 aperture diameter	105 mm
Mass resolving power (vacuum mode)	100	$Q_{2,3}$ maximum gradient	6.0 T/m
Dipole bending angle	25°	$Q_{2,3}$ effective length	600 mm
Dipole entrance angle	0°	$Q_{2,3}$ aperture diameter	200 mm
		Total weight	17500 kg
		Total length	4.8 m

To remove gaseous impurities from the separator field region, continuous helium gas flow is made use of. Gas pressure inside the dipole chamber is monitored and the pumping of the helium out of the separator is regulated accordingly. Pressures used are typically 0.5 – 3 mbar. The high vacuum of the accelerator beam tube is separated from the helium atmosphere by a 0.5 mg/cm^2 Ni foil. The targets are mounted on a manually operated rotating wheel just downstream from the gas window.

At the focal plane there is a sixteen-strip PIPS detector with active area 80 mm (hor.) x 35 mm (vert.). Each 5 mm wide strip is position sensitive in the vertical direction. The position resolution, as measured through observed nucleus–alpha particle correlations, is 500 μm (FWHM). Signals from the preamplifiers are processed through two amplification branches, one suitable for alpha decays and the other for fission and signals from impinging recoil nuclei.

3. Detection of time- and position-correlated event chains

In all experiments so far performed using RITU, the study of reaction products has been based on alpha decay chains where the known daughter, and in some cases the granddaughter, provides the identification. This procedure, sometimes referred to as the genetic correlation method, relies on the observation of position- and time-correlated event chains and on the estimation of their significance [16]. In the present work, event chains of the type nucleus–alpha–alpha have normally been searched for. Here, nucleus refers to the observation of a possible evaporation residue, as determined on the basis of pulse height only. Alpha refers to the observation of a possible alpha decay event, again specified on the basis of pulse height. Maximum

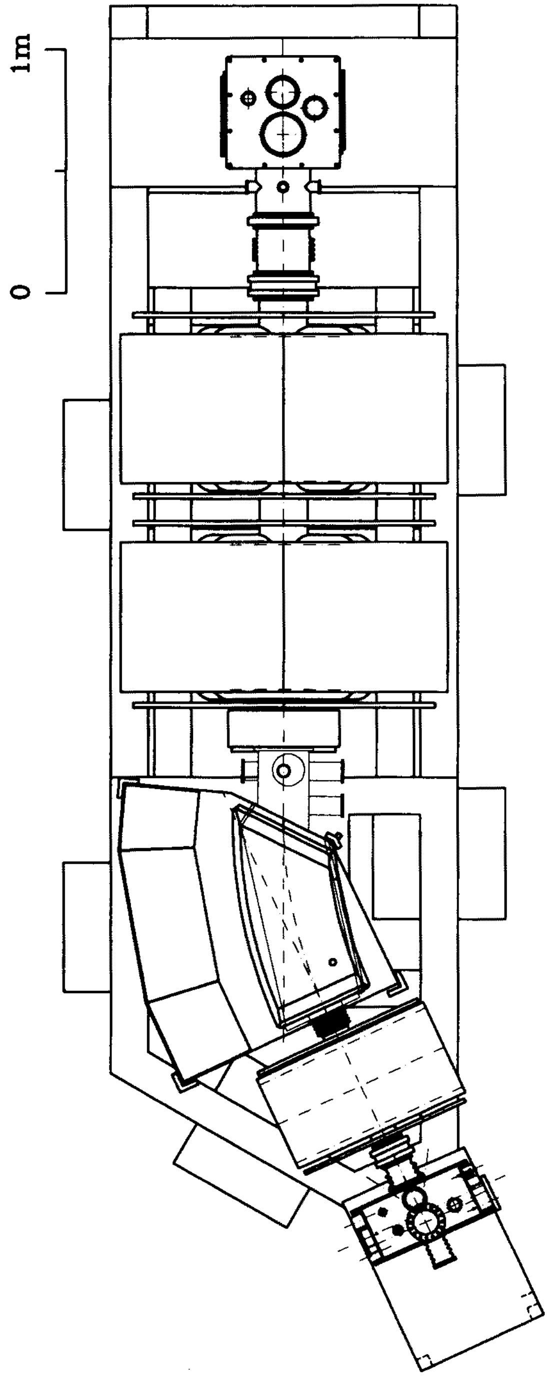

Fig.1 Scheme of the gas-filled recoil separator RITU

search times for the pairs nucleus–first alpha and first alpha–second alpha, respectively, were chosen on the basis of the corresponding half-lives. The expected number of accidental nucleus–alpha–alpha chains was estimated on the basis of the counting rates of events of type nucleus and type alpha.

4. Decay properties of neutron-deficient nuclides in the At–Th region

In this work, neutron-deficient isotopes of elements ($85 \leq Z \leq 90$) At–Th have been studied. Typically, these nuclides have some 20 neutrons less than corresponding isotopes at the beta stability line. Alpha decay is their dominating decay mode and half-lives are shorter than 1 s.

Alpha decay is a favorable decay mode in the study of exotic nuclei in the sense that detector systems have essentially 100 % efficiency for detecting the decays. (Typically, slightly more than 50 % of the decays of implanted nuclei are observed with the full energy and the rest with the alpha particle escaping and leaving only part of its energy in the crystal.) The energy resolution is good and background is low. Normally only very limited information on the excited states can be collected since in many cases only one alpha line is observed. With high enough production rates, it has on the other hand been possible to observe fine structure in the alpha decay of nuclei in this region [17]. Another application has been the study of shell model intruder states in Tl and Bi isotopes [18]. In some cases [18], alpha decay provides the only way to study these states because the M4 transitions between the intruder and ground state are strongly hindered (*cf.* Sect. 5.3.). Often, however, the information gained is restricted to the half-life of the level and an estimate of the ground state mass.

5. Studies on alpha decay of neutron-deficient isotopes with $85 \leq Z \leq 90$

A large number of neutron-deficient nuclei in the At–Th region were studied quite long ago using the he-jet method [19] at the LBL HILAC accelerator [20–23]. Later on, progress in this region was made through the use of on-line isotope separators [24, 25] and recoil separators [26–28] in combination with heavy ion fusion reactions.

5.1. The experiments

The primary ion beams were produced with the laboratory's K = 130 MeV heavy ion cyclotron equipped with a room temperature ECR ion source. The iron beam was produced using the MIVOC method [29] developed at JYFL. The intensity of the primary beam varied between 10^{11}

and 10^{12} particles/s. Stationary targets were used in all reactions. Target thickness was normally 300–350 $\mu g/cm^2$ and the targets were prepared by rolling or by evaporation on 40 $\mu g/cm^2$ carbon backings. The Pr target used in the reaction $^{56}Fe + ^{141}Pr$ had a thickness of 200 $\mu g/cm^2$ and the material was deposited on a 40 $\mu g/cm^2$ carbon backing using the off-line isotope separator at the Accelerator laboratory of the University of Helsinki. The expected average ionic charge q_{ave} (Eq. (1)) of evaporation products was calculated as described in [12].

Alpha energy calibration was based on activities produced in the bombardments and imbedded in the PIPS detector. Alpha energy resolution varied between 28 and 35 keV. Excitation functions were normally measured by using a set of nickel or Havar foils to reduce the bombarding energy.

5.2. Results

We have identified new At, Rn, Fr, Ra, Ac, and Th isotopes. The results concerning Ac isotopes have been published elsewhere [30]. In the following, preliminary results from our alpha decay studies concerning At, Ra, and Th will be presented.

5.2.1. At isotopes

We have produced new At isotopes in the reaction $^{56}Fe + ^{141}Pr$. For calibration purposes, $^{40}Ar + ^{165}Ho$ and $^{28}Si + ^{175}Lu$ irradiations leading to the production of At isotopes were also performed. In the present work, the previously unpublished isotopes $^{193,194,195}At$ were identified.

Treytl and Valli identified light At isotopes down to ^{196}At [20]. We were able to reproduce their data for ^{196}At, produced in the present work in the 1n evaporation channel. In [20] it was deduced that $^{194,195}At$ were also produced but their alpha decay could not be observed due to the short half-lives. Later on, these nuclides were produced in the $^{56}Fe + ^{141}Pr$ reaction at LBL and separated using the SASSY gas-filled separator [12]. The half-life and alpha energy of ^{194}At were determined to be 180±80 ms and 7.20±0.02 MeV, respectively [31]. In ^{195}At, two alpha decaying states were observed [32]. The half-lives and alpha energies were measured to be 150±30 ms and 7.12±0.02 MeV and 140±50 ms and 7.19±0.03 MeV, respectively. The 7.19 MeV decays of ^{195}At were found to be correlated with decays of the 12 s ground state of ^{191}Bi [18].

In the present work, we were able to essentially confirm the LBL results. We observed additional alpha activities identified as belonging to $^{194,195}At$. Alpha–alpha correlations between states in ^{194}At and states previously identified in ^{190}Bi [33] were observed (Fig. 2) at a bombarding energy of 256 MeV. The 150 ms isomeric state in ^{191}Bi identified in [11] was found to be populated by 6950±30 keV alpha decays from a state in ^{195}At with a half-life of $0.63^{+0.32}_{-0.16}$ s. The ground state decays of ^{191}Bi were observed to be correlated with a complex alpha decay pattern with $E_\alpha \sim 7070 - 7220$ keV. Studies of the decay of ^{195}At were performed at a bombarding energy of 249 MeV.

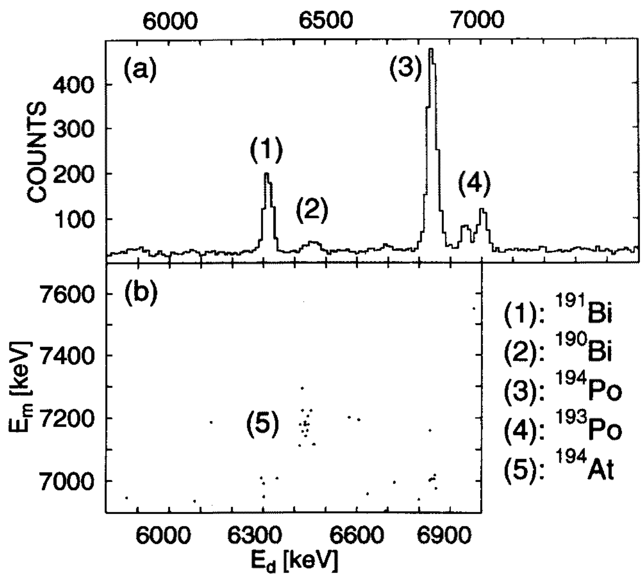

Fig. 2. Mother and daughter alpha particle energies for all chains of type recoil nucleus–alpha–alpha observed in the reaction 256 MeV ^{56}Fe + ^{141}Pr. Maximum search times were 1.0 s for the pair recoil nucleus–alpha and 20 s for the pair alpha–alpha.

At 265 MeV bombarding energy, five chains were observed in which the daughter decay could be identified as belonging to ^{189}Bi. The mother activity is the new isotope ^{193}At. A relatively wide distribution of alpha energies was recorded for this nuclide, so also in this case there is evidence for two decaying states.

5.2.2. Ra isotopes

The lightest Ra isotope investigated by Valli *et al.* [22] was ^{206}Ra. Heßberger *et al.* [28] provided improved data on the decay of ^{206}Ra and discovered the new alpha emitters ^{207m}Ra and ^{205}Ra.

In the present work, Ra isotopes were produced in the reactions ^{40}Ar + ^{170,171}Yb and ^{35}Cl + ^{175}Lu. The isotopic composition of the ^{171}Yb targets was 90.4 % ^{171}Yb, 7.3 % ^{172}Yb, and ^{168,170,173,174,176}Yb < 1 % each. The composition of the ^{170}Yb targets was 72 % ^{170}Yb with other abundances unknown. We studied activities produced in the reaction ^{40}Ar + ^{171}Yb at five laboratory bombarding energies of 187 – 223 MeV. In the reaction ^{40}Ar + ^{170}Yb, the energies were 214 MeV and 223 MeV and in the reaction ^{35}Cl + ^{175}Lu 172, 180, 187, and 194 MeV.

We were able to confirm the results of [28] concerning ^{207m}Ra and ^{206}Ra. Our results are presented in Table II. We observed the two activities identified as ^{205g}Ra and ^{205m}Ra in [28]. They were identified on the basis of alpha–alpha correlations to ^{201g}Rn and ^{201m}Rn. In contrast to the findings of Heßberger *et al.*, the correlations can be explained simply in terms of two separate decay chains connecting ^{205g}Ra to ^{201g}Rn and ^{205m}Ra to ^{201m}Rn, respectively.

TABLE II

The half-lives and alpha energies E_α of At, Ra, and Th isotopes measured in the present work and corresponding earlier data when available. The literature values for ^{196}At are from Ref. [20] and those for $^{205-207}$Ra from Ref. [28]. In [28], one common value for the alpha energy and half-life of ^{205m,g}Ra were given. More details on the decay of astatine isotopes ^{194,195}At can be found in the text.

Nuclide	Half-life (ms) (present work)	Half-life (ms) (literature value)	E_α (keV) (present work)	E_α (keV) (literature value)
^{196}At	300^{+800}_{-200}	300 ± 100	7040 ± 25	7057 ± 7
^{195}At	630^{+320}_{-160}	—	6950 ± 20	—
^{207m}Ra	63 ± 16	55 ± 10	7330 ± 15	7320 ± 10
^{206}Ra	250 ± 50	240 ± 20	7268 ± 10	7270 ± 10
^{205g}Ra	210^{+55}_{-35}	220 ± 60	7350 ± 25	7360 ± 20
^{205m}Ra	190^{+50}_{-30}	220 ± 60	7375 ± 25	7360 ± 20
^{204}Ra	72^{+24}_{-14}	—	7488 ± 25	—
^{211}Th	37^{+28}_{-11}	—	7792 ± 20	—
^{210}Th	10^{+50}_{-5}	—	7896 ± 25	—

In three of the irradiations, 214 MeV ^{40}Ar + ^{170}Yb, 223 MeV ^{40}Ar + ^{170}Yb, and 194 MeV ^{35}Cl + ^{175}Lu, a new alpha line with 7.49 MeV energy was observed. The correlation search showed this alpha decay to be followed by a 6900±10 keV decay with a half- life of $0.98^{+0.40}_{-0.22}$ s. The daughter activity can be identified as ^{200}Rn [34]. The 7.49 MeV alpha decay thus belongs to the new isotope ^{204}Ra. In Fig. 3, all nucleus–alpha–alpha chains observed in the 214 MeV ^{40}Ar + ^{170}Yb irradiation are shown. Seven chains represent the decay of ^{204}Ra. The expected number of accidental triple chains of type nucleus–alpha$_1$–alpha$_2$ with 7400 keV $\leq E_{\alpha 1} \leq$ 7600 keV and 6870 keV $\leq E_{\alpha 2} \leq$ 6930 keV is 0.2. Maximum search times were 1 s and 3 s for the mother decay and the daughter decay, respectively. Altogether 16 ^{204}Ra chains were seen in the three irradiations. Table II shows the combined results for the decay properties of this isotope together with those for $^{205-7}$Ra.

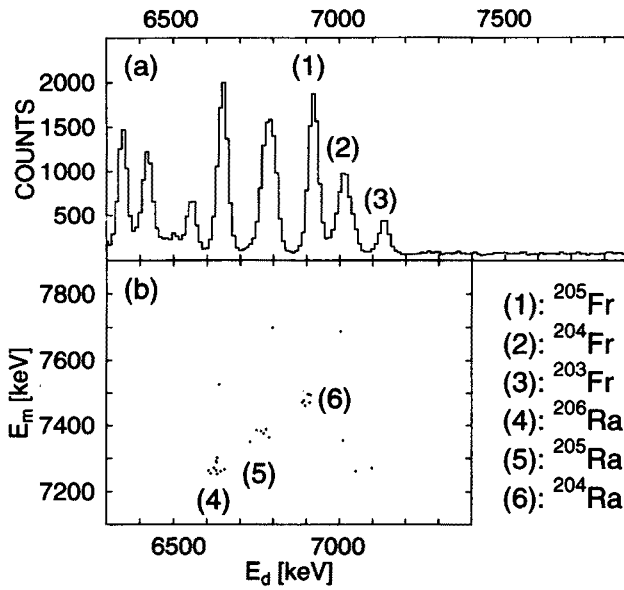

Fig. 3. Mother and daughter alpha particle energies for all chains of type recoil nucleus–alpha–alpha observed in the reaction 214 MeV ^{40}Ar + ^{170}Yb. Maximum search times were 1.0 s for the pair recoil nucleus–alpha and 3.0 s for the pair alpha–alpha.

5.2.3. Th isotopes

Neutron-deficient Th isotopes have been studied by Valli and Hyde ($A = 213 - 217$) [23] and by Vermeulen *et al.* (^{212}Th) [35]. The decay properties of pairs of isotopes such as ^{213,214}Th are often similar in this region of the chart of nuclei. The alpha energy and half-life of the new isotope ^{211}Th were expected to be almost identical to those of ^{212}Th.

In the present work, Th isotopes were produced in the reaction ^{35}Cl + ^{181}Ta. The target had a thickness of 350 μg/cm^2 and it was prepared by rolling. At 186 MeV and 193 MeV bombarding energies, a total of five three-alpha decay chains identified as originating from the new isotope ^{211}Th were observed. The measured daughter alpha particle energy, 7136±15 keV, and half-life, $1.13^{+0.86}_{-0.34}$ s, are compatible with decay properties of both ^{207}Ra and ^{208}Ra. Identification of the isotope as ^{211}Th is based on the measured alpha particle energy, 6499 ± 15 keV, and half-life, ~ 50 s of the granddaughter, which can unambiguously be assigned to ^{203}Rn. The alpha decay energy and half-life of ^{211}Th, 7792 ± 20 keV and 37^{+28}_{-11} ms, respectively, can be compared with the corresponding data for ^{212}Th [35], 7802 ± 10 keV and 30^{+20}_{-10} ms.

At 193 MeV bombarding energy, one decay chain assigned to the new isotope ^{210}Th was observed. The identification was based on the measured alpha decay energy, 7263 ± 20 keV, and life time, 0.99 s of the daughter decay which are compatible with the data for ^{206}Ra (Table II). Our data for new Th isotopes are shown in Table II.

5.3. Discussion

Alpha decay of both odd and odd-odd Bi isotopes has been studied extensively by the Leuven group [18, 33, 36, 37]. The present work provides new data for the systematic study of intruder states in At and Bi isotopes. In the decay of odd Bi isotopes down to ^{187}Bi, produced in heavy ion fusion reactions, the alpha spectra are dominated by unhindered transitions between the $1/2^+$ intruder state in Bi and the $1/2^+$ ground state of Tl and between the $9/2^-$ ground state of Bi and the $9/2^-$ intruder state in Tl [18]. Also in odd-odd Bi isotopes down to ^{190}Bi [33, 36], unhindered decays are seen between two pairs of states, with probable spin and parity assignments of 3^+ and 10^-, respectively.

The new isotopes $^{193-195}$At have been unambiguously identified in the present work. Alpha decay of ^{193}At populating the 0.68 s state in ^{189}Bi with probable $9/2^-$ assignment was observed. Due to the small number (5) of decay chains it can only be stated that probably two alpha transitions with $T_{1/2} \sim 40$ ms and $E_\alpha \sim 7.34 - 7.40$ MeV were seen. The low-spin (probable 3^+) state in ^{190}Bi was observed to be populated by ^{194}At decays but again with a complex decay pattern (Fig. 2). Two alpha transitions with $E_\alpha \sim 7.14$ MeV and ~ 7.19 MeV and $T_{1/2}$ correspondingly ~ 40 ms and ~ 250 ms were observed. In the decay of ^{195}At, the $1/2^+$ intruder state in ^{191}Bi was seen to be populated by alpha decays presumably from the $1/2^+$ state. Correspondingly, alpha decays populating the $9/2^-$ ground state of ^{191}Bi were observed. Also in this case, a number of different mother alpha transitions were observed with $E_\alpha \sim 7.07$ MeV ($T_{1/2} \sim 160$ ms) and ~ 7.14 MeV (~ 160 ms), and ~ 7.22 MeV (~ 80 ms).

The above results are tentative and more information is required for an understanding of the decays of ^{193,194,195}At. One possible reason, in principle, for apparently complex alpha decay schemes is the summing up of conversion electron and alpha particle energies in the detector. This possibility, although improbable in the present case, needs to be considered.

The interpretation of the decay data of the Th and Ra isotopes produced in this work is straightforward. Systematic trends expected on the basis of heavier isotopes are followed. Our results are presented in Table II. A detail apparent from an overview of the alpha decay energies of nuclei with $84 \leq Z \leq 90$ is that the occurrence of isotope pairs with similar alpha energies and half-lives comes to an end at $N = 117$ for all elements up to Ra. At least in the case of ^{206}Ac, the verification of the persistence of this behaviour for even heavier elements seems possible in the near future.

6. Studies in the transuranium region

In extreme cases in the transuranium region, large irradiation doses on the order of 10^{18-19} particles lead to the observation of the decay of only a few atoms of the wanted species. In these experiments, very low background is necessary. In order to study the separation capabilities of RITU in this field, we have performed tests using the reactions ^{22}Ne + ^{209}Bi, ^{40}Ar + ^{208}Pb, and ^{28}Si + ^{232}Th. Results from the Ne + Bi irradiation will be given below. The analysis of the other studies is in progress.

6.1. The reaction ^{22}Ne + ^{209}Bi

The reaction ^{22}Ne + ^{209}Bi was used in the study [38], where the new isotopes ^{226,227}Np were identified. The isotopes $^{225-227}$Np were also studied in [39, 40]. In the present work, ^{226}Np was identified on the basis of alpha–alpha correlations between decays of ^{226}Np and ^{214}Fr. In the Ne + Bi irradiation, slow pulsing of the cyclotron beam was not yet available. So, in this asymmetric reaction, signals from evaporation residues were indistinguishable from those from alpha decays. Nevertheless, the resulting triple or quadruple decay chain data were completely free from background. All chains could be identified, often with pile-up decays due to the short half-lives of the $N = 128, 129$ nuclides forming part of the chain. Three chains where the ^{226}Np life time was measured were seen. The resulting half-life, 58^{+70}_{-20} ms, is compatible with the value of 31 ± 8 ms measured by Ninov *et al.* [38].

The early work on the decay of ^{194,195}At at LBL was performed in collaboration with A. Ghiorso and S. Yashita.

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