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Identification of the new isotope ^{114}Ba and search for its α and cluster radioactivity

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Using the on-line mass separator at the Gesellschaft für Schwerionenforschung Unilac we produced ^{114}Ba through the ^{58}Ni(^{58}Ni,$2n$)^{114}Ba reaction and measured its production cross section to be $0.20^{+0.13}_{-0.09}$ μb. The new isotope ^{114}Ba represents the heaviest $N = Z + 2$ nucleus known to date. With ΔE-E telescopes we measured the total (β-decay) half-life to be $T_\beta = 0.43^{+0.30}_{-0.15}$ s and the partial α-decay half-life to be $T_\alpha \geq 1.2 \times 10^2$ s (1 MeV$\leq E_\alpha \leq$ 4 MeV) for ^{114}Ba. With track detectors we found a half-life for spontaneous ^{12}C emission $T_C \geq 1.1 \times 10^3$ s based on three carbon events.

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Cluster radioactivity is now a well established, although rare, decay mode of heavy nuclei. Intense experimental research in the last decade has led to the detection of 20 cases of spontaneous emission of clusters ranging from ^{14}C to ^{32}Si from trans-lead nuclei, with branching ratios relative to α decay from 10^{-9} down to 10^{-16} and partial half-lives from 10^{11} up to 10^{28} s [1]. The general features of this new radioactive decay mode have been established; above all, its strong dependence on the barrier penetration factor and consequently on the Q value, which must be large in order to compensate for the small preformation factors typical of such complex clusters [2]. For this reason, all heavy residual nuclei resulting from cluster emission have been found so far to differ from the doubly magic ^{208}Pb by three nucleons at most. Analogously, a new island of cluster radioactivity having residual nuclei close to doubly magic ^{100}Sn has been predicted by Greiner *et al.* and Poenaru *et al.* [3,4]. They found the most favorable case to be ^{12}C emission from ^{114}Ba and calculated the decay rate using various mass predictions [5] for the then unknown ^{114}Ba nucleus. The results of their predictions strongly depend on the adopted Q value: a 2-MeV spread in the Q values results in a factor $\sim 10^5$ spread in the partial half-lives. More recently others [6–10] have taken up the challenge of calculating the decay rate for ^{12}C emission from ^{114}Ba; their results for the same choice of Q value span more than eight orders of magnitude. This situation is in complete contrast with what is found in the trans-lead region of cluster radioactivity, where Q values are generally known and all models predict half-lives for the same decay mode that typically agree within an order of magnitude [11].

It is clear that the possible island of cluster radioactivity in the trans-tin region would be interesting to explore. Apart from the possibility of discriminating among different theoretical approaches, the mere detection and measurement of the partial ^{12}C decay rate of ^{114}Ba could shed light on the behavior of cluster radioactivity in this region of the Chart of the Nuclides far below ^{208}Pb, possibly providing information on the interplay with α decay and on the role of the nearly double shell closure in a nucleus such as ^{102}Sn far from the stability line.

The first searches for ^{114}Ba$\rightarrow$^{12}C+^{102}Sn decay were performed by Oganessian *et al.* in 1992 and 1993 [12]. They bombarded a nickel target with the internal ^{58}Ni beam of the U-400 cyclotron and, by using a theoretical estimate of ~ 1 μb for the cross section for the reaction ^{58}Ni(^{58}Ni,$2n$)^{114}Ba, they deduced a total collection of $\sim 3 \times 10^6$ ^{114}Ba atoms in a very high neutron background ($\sim 10^{13}$ neutrons). In a polycarbonate film that surrounded the target they detected $\sim 10^3$ tracks. All of them were attributed to recoils of fast-neutron-induced reactions with nuclei in the plastic film except for ~ 10 tracks that were found to have a range long enough to be compatible with ^{12}C spontaneously emitted by ^{114}Ba. They presented data showing a continuum of measured ranges of particles which they interpreted to be carbon ions without showing data on the charge distribution or charge resolution. From their range cut they determined an upper limit of 10^{-4} for the branching ratio for carbon emission. In subsequent work they reduced the neutron background by transporting the ^{58}Ni+^{58}Ni reaction products far from the target with a gas jet. Nevertheless, the neutron background was still too high to permit them to improve their limit on the branching ratio.

The purpose of the present experiment was to search for ^{12}C emission from ^{114}Ba under cleaner experimental conditions and without making any assumption about the ^{114}Ba half-life or the ^{58}Ni(^{58}Ni,$2n$)^{114}Ba reaction

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cross section. We used the on-line mass separator at the Gesellschaft für Schwerionenforschung Unilac to produce the four new neutron-deficient barium isotopes ^{114}Ba, ^{115}Ba, ^{116}Ba, and ^{118}Ba, and to study their decays. In this paper we restrict ourselves to ^{114}Ba and describe its production, identification through β, and β-decayed proton activities, and a search for α and ^{12}C cluster radioactivity (in the latter case, using glass detectors calibrated with ^{12}C, ^{11}B, and ^{9}Be ions).

A stack target of ^{63}Cu and ^{58}Ni foils each of 2 mg/cm^2 thickness was bombarded with a ^{58}Ni beam from the Unilac with an average intensity of 40 particle nA. The beam energy was 4.9 MeV/nucleon on the ^{63}Cu and 4.2 MeV/nucleon on the ^{58}Ni target. The reason for using the stack target was to produce and measure simultaneously ^{114}Ba from the ^{58}Ni target and ^{117}Ba from the ^{63}Cu target in order to monitor the long-term stability of the mass separator while collecting ^{114}Ba for the search for cluster radioactivity. Reaction products were stopped in a hot catcher inside the separator ion source, released via solid state diffusion and surface desorption, ionized to charge-state 1+, accelerated to 55 keV and finally separated unambiguously, according to mass, into three beam lines.

We used two thermoionization sources of the cavity type [13]: a 2300-K cavity with a gas line for CF$_4$ addition (subsequently referred to as a fluorination source) and our conventional high-temperature cavity operating at 2700 K. The advantage of the former is that it suppresses all isobaric contaminants to below a level of 10^{-6} by separating barium as BaF$^+$ ions [14]. The advantage of the latter was its approximately two times higher separation efficiency for short-lived barium isotopes during the first part of the experiment, with the drawback, however, of not discriminating against cesium. This is no problem in the search for cluster emission, since barium is the only candidate in the A = 114 chain. The fluorination source was used to investigate the isotopes $^{114-116,118}$Ba and, in particular, to determine the ^{114}Ba source strength by β-counting as well as the ratio of the delayed-proton rates following the β decays of ^{114}Ba and ^{117}Ba. The latter ratio, together with the delayed-proton emission probability measured for ^{114}Ba [16], was used to determine the ^{114}Ba source strength with the high-temperature cavity, where the strong ^{114}Cs contamination prevents a direct measurement. The implicit assumption that the ratio of the separation efficiencies for the two ion sources does not depend on the half-life was experimentally confirmed by separate measurements for barium in the half-life range from 0.1 to 5 s [15].

The half-life of ^{114}Ba and its source strength were determined by studying the β decay of ^{114}Ba. This was accomplished by implanting ^{114}BaF$^+$ molecules into a tape in front of a β telescope composed of a silicon ΔE detector followed by a plastic scintillator [positron detection efficiency 25(3)%]. To avoid excessive buildup of daughter activity, the tape removed the source from the detector every 2 s. A fit to the resulting grow-in curve yielded a half-life of $0.43^{+0.30}_{-0.15}$ s and an intensity of 0.12(4) s^{-1} for the ^{114}BaF$^+$ beam. A more detailed account of these measurements, together with the data for $^{115-118}$Ba, will be given in [16]. From the measured intensity of the ^{114}BaF$^+$ beam and its total separation efficiency of $11^{+4}_{-3}\%$ (for the determination, see [15]), we were able to deduce the ^{58}Ni(^{58}Ni,2n)^{114}Ba cross section. The result averaged over ^{58}Ni beam energies between 3.4 and 4.2 MeV/nucleon is $0.20^{+0.13}_{-0.09}$ μb.

In an earlier measurement we collected for ~ 16 h ^{114}BaF$^+$ (from a fluorination source which at that time had a separation efficiency of about 6%) and for ~ 38 h ^{114}Ba$^+$ (from the at that time more efficient high-temperature cavity) on a 0.8-μm-thick aluminum foil placed in the center of a sphere of 9.2 cm diameter, whose inner surface was covered with barium-phosphate glass plates acting as nuclear track detectors [17]. The geometrical efficiency of this detector device was 94 ± 5%. The total number of collected ^{114}Ba atoms was calculated to be 17 000 ± 5500. We etched the glass plates in 48% HBF$_4$ at 50 °C for 24 h, a time just sufficient to develop ^{12}C tracks of ~ 17 MeV to the end of their range as determined from a calibration with accelerated carbon ions. We then scanned the plates both manually and with an automated system.

In previous experiments on cluster radioactivities the track detectors contained a huge background of very short etch pits due to nuclei in the detector elastically scattered after collisions with α particles. By contrast, the plates (comprising a total area of ~ 210 cm^2) exposed to the collected activity (^{114}Ba, ^{114}Cs and other decay daughters) contained only ~ 30 tracks clearly recognizable as background because of their short range or large zenith angle, which meant that they could not be assigned to particles emitted from the collector. Some of these tracks were evidently due to spallation recoils produced by cosmic rays that passed through the glass either during its air transport or while in the laboratory for more than one year before the experiment was done.

Figure 1 shows data for all events found on the front

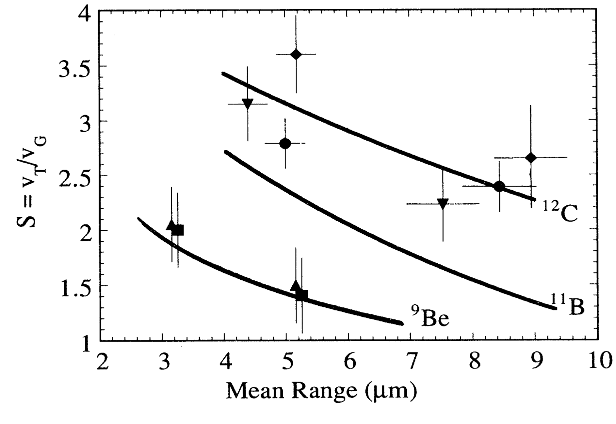

FIG. 1. Sensitivity vs residual range for events found on the glass detectors exposed at GSI. Each event is represented by two S values corresponding to short and long residual range, respectively. Pairs of correlated values are marked by identical symbols. Curves are based on calibrations with ^{12}C ions (by the Milano group at Legnaro) and with ^{11}B and ^{9}Be (at Lawrence Berkeley Laboratory [20]).

surfaces of glass and that passed simple cuts on angle of entry ($\theta < 40^\circ$) and range ($> 5\ \mu$m). For each event the sensitivity S, defined as the ratio of track-etching velocity v_T to general etching velocity v_G, was determined at two points along the particle's range [18]. This double measurement allowed the charge to be determined (resolution $\sigma_Z = 0.3$ charge unit). Two of the events on the front surface were similar to those found on the back surface and on plates stored in a freezer. Those events were identified as having $Z \leq 4$ and range $\leq 9\ \mu$m. We argue that the two events on the front surfaces did not result from ^{114}Ba decay: the presence of occasional similar events on the back surface and on plates stored in a freezer indicates that a background activity unrelated to ^{114}Ba can lead to production of such tracks.

The remaining three events on the front surface were identified as carbon, with energy 13.2 ± 0.9, 14.8 ± 1.4, and 15.3 ± 1.1 MeV, i.e., slightly lower than the calibration energy, as shown in Fig. 2. Two of these events had trajectories compatible with an origin in the foil collector, while the direction of the third event differed from the radial direction by 43°. We cannot rule out the possibility that one or more of the three observed carbon tracks are due to cosmic ray spallation in the glass, with two of them fortuitously having the right trajectories. We thus report only an upper limit of 3.8×10^{-4} for the branching ratio for ^{12}C emission. This 1σ limit includes the uncertainty of the three observed events ($^{+97\%}_{-53\%}$), of the detection efficiency ($\pm 5\%$), and of the number of implanted ^{114}Ba atoms ($\pm 32\%$). This result, together with the measured β decay half-life, yields a lower limit of $T_C \geq 1.1 \times 10^3$ s for the partial half-life for ^{12}C emission. It is worth pointing out, however, that all three carbon tracks were found on the front sides of the glass plates and had about the same energies, and that we found no events with $Z = 5$ or with $Z > 6$, such as would be expected for a continuous charge distribution of cosmic ray recoils. It is interesting to note that the average ^{12}C energy of 15.3 ± 1.1 MeV, obtained after correction for energy loss in the collector foil, corresponds to a Q value of 17.1 ± 1.1 MeV for ^{12}C decay. This result, together with the known Q_α values for ^{106}Te and ^{110}Xe [19], yields a surprisingly low Q_α value of 1.6 ± 1.1 MeV for ^{114}Ba.

For a total time of 27 h we implanted ^{114}BaF$^+$ on a 40-μg/cm^2-thick carbon catcher to search for α decay using the same ΔE-E telescope we used to count β-delayed protons ($\varepsilon \approx 17\%$). We collected $(5.2 \pm 1.6) \times 10^3$ ^{114}Ba atoms but did not find any indication for α lines in the ΔE spectrum measured in anticoincidence with the E detector. The few events found in this spectrum did not differ from what was observed due to room background. We were able to infer an upper limit of $b = \lambda_\alpha/\lambda_{\text{tot}} \leq 3.7 \times 10^{-3}$ for the branching ratio for α decay of ^{114}Ba. This 1σ limit includes the uncertainties of (i) one event ($^{+230\%}_{-83\%}$) per 50-keV interval for an α-energy range of the ΔE-anticoincidence spectrum between 1 and 4 MeV, (ii) the number of β-delayed protons observed simultaneously in the ΔE-E telescope ($\pm 6\%$), and (iii) the measured [16] delayed-proton emission probability of ^{114}Ba and its decay daughters ($\pm 30\%$). The corresponding lower limit on the partial half-life for α decay is 1.2×10^2 s. Table I summarizes the results of our measurements.

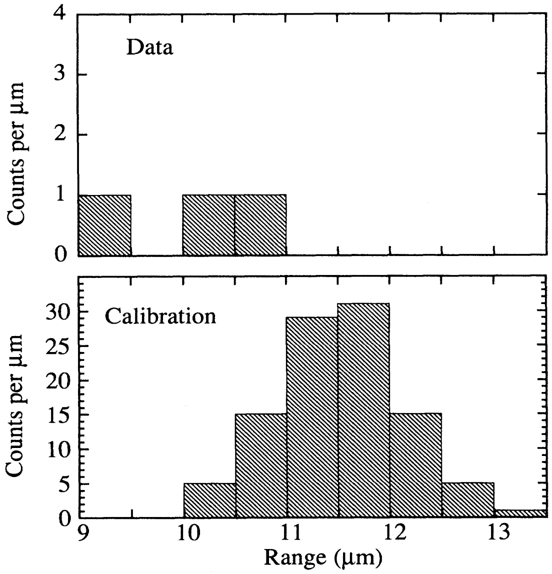

FIG. 2. Total range for the three carbon events (upper histogram) and for the 17 MeV ^{12}C calibration ions.

In contrast to the work of Oganessian *et al.* [12], our observation of carbon ions yields the first limit for clus-

TABLE I. ^{114}Ba measurements. Beam: 4.2 MeV/nucleon ^{58}Ni; target: 2 mg cm^{-2} ^{58}Ni. Ion sources: thermoionization without fluorination at 2700 K for barium + cesium beams; thermoionization with fluorination for pure barium beams, source temperature initially 2300 K, later 2400 K. η_S = total separation efficiency; η_D = detector efficiency; SB = surface barrier.

Measured radiation	Time (h)	$A = 114$ beam	η_S (%)	^{114}Ba atoms collected	Detectors	η_D (%)	Results
β	24	Ba	5-15	~ 7500	SB-plastic telescope	25 ± 3	$T_{1/2} = 0.43^{+0.30}_{-0.15}$ s
β	6	Ba	$11^{+4}{}_{-3}$	2600 ± 900	SB-plastic telescope	25 ± 3	$I(^{114}\text{Ba}) = 0.12 \pm 0.04\ \text{s}^{-1}$, $\sigma = 0.20^{+0.13}_{-0.09}\ \mu$b
^{12}C	16	Ba	5.5 ± 2	17100 ± 5500	Track detectors	94 ± 5	≤ 3 events
	38	Ba+Cs	~ 10				
α	27	Ba	5-11	5200 ± 1600	SB-telescope	17 ± 2	no α line observed (≤ 1)

ter radioactivity from mass-separated $A = 114$ sources. It is not inconsistent with the limits reported in (12). It is interesting to note that an assignment of the three observed carbon tracks to cluster radioactivity of ^{114}Ba with the above-mentioned decay rate and decay energy would mean that theoretical predictions severely underestimate the decay rate, ranging from a factor $\sim 10^5$ for the model of Kadmenski *et al.* [6] to $\sim 10^{12}$ for the model of Poenaru *et al.* [4].

The fluorination method combined with on-line mass separation, used successfully in this work to identify the heaviest known $N = Z + 2$ nucleus ^{114}Ba and to measure both its half-life and its production cross section, offers the chance to realize several improvements. Firstly, the measured ^{58}Ni(^{58}Ni,$2n$) cross section, which is one of the very few available for the $2n$ channel far from β stability, allows one to deduce *absolute* decay branching ratios for charged particle emission from ^{114}Ba even if, as in [12], an experiment is unselective with respect to nuclear charge or mass number. Secondly, *absolute* partial half-lives can be calculated from such branching ratios by using the β-decay half-life of ^{114}Ba determined in this work. A direct comparison with theoretical predictions of partial half-lives is thus possible. Thirdly, to improve the statistics and eliminate all background tracks due to cosmic ray interactions and natural radioactivity, we plan a new experiment using glass that will be heated to remove all background tracks just before the experiment. If cluster radioactivity could be firmly established at approximately the rate and the decay energy indicated by the three carbon events, ^{114}Ba would be the only case where cluster radioactivity dominates over α decay, as the α-decay mode would be strongly hindered due to its small Q value.

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[1] R. Bonetti and A. Guglielmetti, in *Nuclear Decay Modes*, edited by D. N. Poenaru and W. Greiner (IOP, Bristol, England, in press).

[2] D. N. Poenaru, in *Proceedings of the 6th International Conference on Nuclear Reaction Mechanisms*, Varenna, 1991, edited by E. Gadioli (Ricerca Scientifica ed Educazione Permanente, Milano, 1991).

[3] W. Greiner, M. Ivascu, D. N. Poenaru, and A. Sandulescu, in *Treatise on Heavy Ion Science*, edited by D. A. Bromley (Plenum, New York, 1989), Vol. 8, p. 641.

[4] D. N. Poenaru *et al.*, At. Data Nucl. Data Tables **48**, 231 (1991).

[5] P. E. Haustein, At. Data Nucl. Data Tables **39**, 185 (1988).

[6] S. G. Kadmenski *et al.*, Izv. Akad. Nauk Rossii **57**, 12 (1993).

[7] R. Blendowske and H. Walliser, Phys. Rev. Lett. **61**, 1930 (1988); H. Walliser (private communication).

[8] F. Barranco (private communication).

[9] B. Buck, A. C. Merchant, S. M. Perez, and P. Tripe, J. Phys. G **20**, 351 (1994).

[10] D. S. Delion, A. Insolia, and R. J. Liotta, J. Phys. G. **20**, 1483 (1994).

[11] P. B. Price, in *Clustering Phenomena in Atoms and Nuclei*, edited by M. Brenner, T. Lonnroth, and F. B. Malik (Springer Verlag, Berlin, 1992).

[12] Yu. Ts. Oganessian *et al.*, Z. Phys. A **349**, 341 (1994); V. L. Mikheev *et al.*, in *Proceedings of the 7th International Conference on Nuclear Reaction Mechanisms*, Varenna, 1994, edited by E. Gadioli (Ricerca Scientifica ed Educazione Permanente, Milano, 1994).

[13] R. Kirchner, Nucl. Instrum. Methods Phys. Res. Sect. A **292**, 203 (1990).

[14] H. L. Ravn, S. Sundell, and L. Westgaard, J. Inorg. Nucl. Chem. **37**, 383 (1975).

[15] R. Kirchner, GSI Science Report No. GSI-95-1, 1994, p. 287.

[16] Z. Janas *et al.*, Phys. Scr. **T56**, 262 (1995).

[17] S. Wang, S. W. Barwick, D. Ifft, P. B. Price, and A. J. Westphal, Nucl. Instrum. Methods Phys. Res. Sect. B **35**, 43 (1988).

[18] P. B. Price *et al.*, Phys. Rev. C **43**, 1781 (1991).

[19] G. Audi and A. H. Wapstra, Nucl. Phys. **A565**, 66 (1993).

[20] P. B. Price, A. Richards, and A. J. Westphal (unpublished).