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A. Guglielmetti, R. Bonetti, G. Poli, R. Collatz, Z. Hu, R. Kirchner, E. Roeckl, N. Gunn,
P.B. Price, B.A. Weaver, A. Westphal, J. Szerypo

Gesellschaft für Schwerionenforschung mbH
Planckstraße 1 • D-64291 Darmstadt • Germany
Postfach 11 05 52 • D-64220 Darmstadt • Germany

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A. Guglielmetti, R. Bonetti, G. Poli
Istituto di Fisica Generale Applicata dell'Universitá di Milano and Istituto Nazionale di Fisica Nucleare, Sezione di Milano, I-20133 Milano, Italy
R. Collatz, Z. Hu, R. Kirchner, E. Roeckl
Gesellschaft für Schwerionenforschung, D-64220 Darmstadt, Germany
N. Gunn, P.B. Price, B.A. Weaver, A. Westphal
Physics Department and Lawrence Berkeley Laboratory, University of California at Berkeley, Berkeley, California 94720, USA
J. Szerypo
Institute of Experimental Physics, Warsaw University, PL-00681 Warsaw, Poland

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Abstract

By means of the on-line mass separator at Gesellschaft für Schwerionenforschung, Darmstadt, we produced ^{114}Ba through the ^{58}Ni(^{58}Ni,2n) reaction, separated it as a ^{114}Ba^{19}F^{+} beam, and implanted it into a stopper foil positioned in the center of an array of track detectors, which were used to search for ^{12}C radioactivity of ^{114}Ba. A total number of $(5.4\pm1.7)\cdot10^{4}$ ^{114}Ba atoms were implanted. No ^{12}C event was found after a total exposure time of 116 hours, corresponding to a ^{58}Ni beam dose of $1.3\cdot10^{17}$. The resulting upper limit of $3.4\cdot10^{-5}$ (84% c. l.) for the branching ratio for ^{12}C decay of ^{114}Ba is considerably lower than the limits obtained in previous experiments, which represents an inconsistency at levels of more than 90%. A semiempirical estimate of 19.3 MeV for the upper limit of the Q-value for ^{12}C decay of ^{114}Ba is derived.

1 Introduction

Recently, intense experimental and theoretical research has been focused on investigating the properties of nuclei far from the valley of β-stability, particularly in the region around the doubly-magic nucleus ^{100}Sn. This research included measurements (and predictions) of masses, decay modes and their branching ratios, and recently culminated in the identification [1, 2] and mass measurement [3] of ^{100}Sn. It was motivated by a fundamental interest in the interplay between microscopic (shell) and macroscopic (liquid-drop) effects in a so far rather unexplored region of nuclei, and was made possible by recent developments in the technology of production of secondary (exotic) ion beams.

The experiments on ^{12}C decay of ^{114}Ba recently performed at Dubna (Dubna94) [4] and GSI (GSI95) [5] lie within this framework. We recall briefly that in both experiments ^{114}Ba was produced by the ^{58}Ni(^{58}Ni,2n) reaction, and ^{12}C clusters were searched for by means of arrays of solid-state nuclear track detectors. However, whereas in Dubna94 the detectors were in direct view of the production target, and were therefore subjected to a high background of neutron recoils, in GSI95 ^{114}Ba atoms were ionized in a chemically selective way, mass-selected by means of the GSI on-line mass separator, subsequently implanted in a stoppper foil and studied far away from the production site. Both Dubna94 and GSI95 advocated some evidence for ^{12}C cluster decay of ^{114}Ba, even though cautiously declaring to yield only upper limits for the branching ratio of ^{12}C decay of ^{114}Ba (b(^{12}C)) of 10^{-4} and $3.8{\cdot}10^{-4}$, respectively. The premature claim of observation of ^{12}C emission from ^{114}Ba in an earlier conference contribution on GSI95 [6] had to be withdrawn after the final data evaluation [5]. Among the 20 claimed cases of spontaneous emission of clusters heavier than α-particles but lighter than fission fragments [7], that of ^{12}C from ^{114}Ba was the only one not involving the trans-lead region. Predictions of cluster radioactivity above ^{100}Sn were published as early as 1989 [8]. More recently, stimulated by Dubna94 and GSI95, several groups [9, 10, 11, 12] have addressed the problem of extrapolating predictions, that have generally been quite successful in reproducing the experimental decay rates in the trans-lead region, to a completely different region of rather poorly investigated nuclei.

In view of the unsatisfactory experimental situation, we decided to repeat GSI95 with improvements concerning the control of the ^{114}Ba beam intensity and the total dose of ^{114}Ba atoms. A further aim was to ensure that the track detector array is free of any background from energetic ions with $Z>2$. The experimental conditions of this experiment (GSI 97) are described in the next section, followed by a discussion of the results in section 3 and an outlook in section 4.

2 Experimental details

The experimental set-up is schematically shown in Fig. 1. The ^{114}Ba particles were produced at the GSI on-line mass separator [13] using the reaction ^{58}Ni(^{58}Ni,2n). A 50 particle·nA ^{58}Ni beam of 255 MeV from the UNILAC accelerator and a 2.6 mg/cm^2 thick target of highly enriched ^{58}Ni were used. The target could be exchanged by one of several others placed on a target wheel. This exchange was performed by remote control when the beam monitor, discussed below, showed signs of target degradation. After passage through thin heat shields and the window, the recoiling reaction products were stopped inside the ion source in two tantalum foils of ~3 mg/cm^2 each. At the ion source temperature of around 2400 K the barium recoils are swiftly released from the thin catchers as thermalized particles.

As ion source we used exclusively the high-temperature cavity source with on-line fluorination by CF_4 addition. Compared to the previously used standard cavity, this fluorination source has in the meantime reached an equally good ionization efficiency, a higher degree of reliability and reproducibility, and - above all - it ionizes in the fluoride sideband selectively BaF, all contaminations including CsF being reduced to levels [14] well below 10^{-5}. The CF_4 was introduced from a test leak with a flow of $2.2 \cdot 10^{-6}$ std·cm^3/s. This ensures efficient fluorination as indicated by the continuously controlled BaF^+/Ba^+ ratio of 12±4, the variation reflecting the scatter of the 5 ion sources used. The ions were accelerated to 55 keV, mass-separated and alternatively focussed onto two detection arrays, installed at different beam lines of the mass separator: First a set-up of two silicon telescopes with (17±2)% detection efficiency each, for determining and monitoring the ^{114}BaF beam intensity via the β-delayed proton (βp) decay of ^{114}Ba or neighbouring barium isotopes, and second a $\sim 4\pi$ track detector array for the determination of ^{12}C cluster emission from ^{114}Ba.

Due to the barium selectivity of the fluorination source, we were able to measure the ^{114}BaF beam intensity any time via the previously determined [15] ratio of 25±7 implanted ^{114}BaF ions in the carbon stopper foils in front of the telescopes per detected βp event. The track detector set-up, however, is on-line a blind detector in the sense that it does not allow for a simultaneous collection of ^{114}BaF and control of its beam intensity. Fortunately, losses in ^{12}C-cluster counting statistics by alternating measurements were avoided by using the spatially separated ^{115}BaF beam for control purposes. ^{115}Ba is a perfect monitor for the ^{114}BaF beam: It is produced in the same target via the 1n reaction channel, has practically the same halflife and a sufficiently high βp decay rate. The latter was determined in a separate measurement to be (0.80±0.12) times that of ^{114}Ba in agreement with the production cross-sections and the βp branching ratios for ^{114,115}Ba given in [15].

The ^{115}Ba monitor excludes the two hardly verifiable and thus neglected different-target/different-halflife uncertainties in GSI95, which could have led to an eventual systematic over-estimation of the ^{114}Ba beam intensity in that study [5], where most of the ion sources used during the measurement did not discriminate against cesium. The control of the ^{114}Ba beam intensity was at that time only possible via the βp decay of ^{117}Ba (the only light barium isotope without βp competition from the much more strongly produced isobaric cesium isotope) which required a different production target, actually the copper part of a combined ^{63}Cu/^{58}Ni target. Even though not very probable, the functioning of one target does not guarantee the intactness of the other, and also the separation efficiency of a shorter-lived isotope might decrease faster within the lifetime of a catcher ion-source system than that of a longer-lived isotope.

In parallel with the βp monitoring of ^{115}Ba, the ^{114}BaF^{+} ions were implanted into a thin stopper of carbon in the center of the track detector array, where the ~500 ions/h of ^{114}BaF were of course unmeasurable. To ensure, however, their definite implantation into the stopper during the week-long measurement, they were continuously monitored by means of a pilot beam of the stable isobar ^{133}Cs (typically on the nA-level), intensity variations of which would immediately cause alarm. The stopper foil was only 20 μg/cm^2 thick in order to keep the energy correction for eventually detected ^{12}C tracks small. During tests performed prior to GSI97, the carbon foils had developed visible marks under the ^{133}Cs bombardment, the consequences of which could not be judged in situ. Since a degradation leading to an eventual transparency for the ^{114}BaF ions could not be excluded, the stopper was backed in closest distance by an identical foil. The absence of marks on the back-up foil was taken as guarantee for the functioning of the stopper.

In GSI95 [5], the phosphate glass plates (BP-1) used as track detectors contained a background of stored tracks of low-Z ions due to both natural radioactivity and 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. In order to eliminate such background in the present experiment, we heated the glass plates in a vacuum oven for 32 h at 250 °C just before assembling them into the track detector array for the irradiation with the ^{114}BaF beam. Calibrations done both at Berkeley and at Milano showed that such an annealing completely removed latent tracks of ions with $Z \leq 6$, reduced by a factor 8 the etch rate of latent tracks of higher Z ions, and produced no detectable shift in the response of the glass that was subsequently irradiated with carbon ions with energy comparable to that expected in ^{114}Ba decay, i. e. 15-17 MeV. This feature was checked by irradiating annealed and unannealed samples of BP-1 glass with light heavy-ions at different energies and comparing their etch rates.

Immediately after their heat treatment, the glass plates were installed on the inner surface of a metallic sphere. The glass surface amounted to 210 cm^2, corresponding to a geometrical efficiency of the array of (94±5)% of 4π. After the end of the irradiation with the ^{114}BaF beam at GSI, the glass plates were taken to Milano, avoiding air transport, and then etched in 50% HBF_4 at 65 °C for 34 hours. This time was chosen on the basis of previous calibrations of BP-1 glasses performed at the XTU Tandem of Legnaro (Italy) with ^{12}C beams of 15 MeV - the energy deduced from the track range of the three events measured in GSI95 - in order to develop the latent tracks almost until the end of their range, thus maximizing the optical contrast. The glasses were then manually scanned under an optical microscope in Milano, re-etched for additional 5 h under the same etching conditions and then sent to Berkeley for a second, independent manual scanning. We decided to perform this second analysis at a different etching time in order to be able to detect ^{12}C ions with higher energy, approximately up to 20 MeV, as predicted by some mass formulas [16] for ^{12}C decay of ^{114}Ba.

3 Results and discussion

In 116 hours of integrated exposure time, with a total ^{58}Ni beam dose of $1.3\cdot10^{17}$, we registered 1700 βp events from ^{115}Ba decay in the silicon telescopes. On the basis of the above-mentioned normalization factors of 25±7 and $1/(0.80\pm0.12)$, this corresponds to $(5.4\pm1.7)\cdot10^{4}$ ^{114}BaF ions collected in the carbon foil at the center of the track detector array. In the whole analyzed glass surface, each of the two independent scanners found one and only track. Upon comparison of coordinates, it proved to be the same track. That event failed the two criteria for a ^{12}C emission from ^{114}Ba. Upon comparison with the calibration curves in Fig. 2, its charge was found to be $4\leq Z\leq5$, and its trajectory, measured with a stereomicroscopic method in conjunction with a Scanning Electron Microscope, was found not to have originated in the stopper foil. Most probably it resulted from cosmic ray spallation that occurred during the one-week time interval after the track detectors were annealed and before they were etched. No evidence for ^{12}C events was found, resulting in an upper limit of $3.4\cdot10^{-5}$ (84% c. l.) for b(^{12}C). This corresponds, on the basis of the known total halflife of $0.43^{+0.30}_{-0.15}$ s [5] of ^{114}Ba, to a lower limit of $1.2\cdot10^{4}$ s (84% c. l.) for the partial halflife of this nucleus for ^{12}C decay. We recall that Dubna94 and GSI95 obtained lower limits of $4.3 \cdot 10^3$ and $1.1 \cdot 10^3$ s, respectively, both based on the above total halflife.

We now address the question whether the null result from GSI97 is statistically compatible with the carbon events previously reported by Dubna94 and GSI95 as actually being due to cluster emission from ^{114}Ba. Such a comparison has of course to take "relative figures of merit" for the expected detection rate of ^{12}C clusters into account, the relevant experimental parameters being compiled in Table 1. Of three ^{12}C tracks that were recorded during GSI95 [5], one had a direction which was not compatible with an origin in the stopper foil, thus suggesting that this, and possibly all three events, were due to cosmic ray spallation reactions within the detector itself. For the present analysis, we consider two carbon events from GSI95, the experimental figure of merit relative to GSI97 being 0.33 (see Table 1).

In case of Dubna94, ref. [4] mentions that "some ten tracks were found to have a length exceeding that of most of the background tracks produced by ^{12}C recoils". For our analysis we assume that in Dubna94 actually 20 carbon events can be distinguished from background as can be seen from Fig. 3 of ref. [4]. The figure of merit for Dubna94 is 1.57 relative to GSI97 (see Table 1) under the assumption that the excitation function of the production cross-section for ^{58}Ni(^{58}Ni,2n)^{114}Ba is quantitatively covered in both experiments. For Dubna94 this is evident, in spite of the rather high incident energy of 280 MeV, due to the thickness of the strongly inclined 3.5 mg/cm^2 thick $^{\mathrm{nat}}$Ni target. In the case of GSI97, with 255 MeV incident energy, the 2.6 mg/cm^2 ^{58}Ni target covers twice the value of $\sim$30 MeV calculated for the FWHM of the excitation function [17], the correct location of which was controlled by variation of the ^{58}Ni beam energy to 269 and 246 MeV, respectively. In both cases the βp-rate of ^{114}Ba decreased, for the higher energy actually by a factor of more than 2.

Our quantitative statistical analysis of the consistency of the results reported by Dubna94, GSI95, and GSI97 is based on the work of Gehrels [18]. He computed the confidence limits on the ratio of observed counting rates for two independent experiments with a given expected ratio of counting rates, which we will take to be the ratio of figures of merit. Using Gehrels' method, we conclude that the results obtained by GSI95 and GSI97 are inconsistent with ^{12}C radioactivity at a confidence level of >90%, in the sense that a random observation of the event ratio would lie with >90% probability above the measured ratio of 0:2. In the case of the null result from GSI97 and the data from Dubna94, one would even with >99.93% probability expect to find a ratio above the measured one of 0:20. The conservative interpretation of these low consistency levels is that the results from both Dubna94 and GSI95 were due to a background unrelated to ^{12}C radioactivity.

The known upper limit of $3.7 \cdot 10^{-3}$ for the α decay branching ratio [5] can easily be used to deduce a "semiempirical" upper limit for the ^{12}C decay energy (Q(^{12}C)). For this purpose we calculate the α decay rate for ^{114}Ba with the model of Ref. [19], in which the penetrability is figured by using an elastic scattering nuclear potential and the spectroscopic factor S is either taken from the systematics of α emitters in the trans-lead region ($S=6.3 \cdot 10^{-3}$) or extracted from the known α decay properties of ^{106}Te [20] ($S=3.3 \cdot 10^{-2}$). The resulting upper limits for the α decay energy of ^{114}Ba are 3.6 and 3.7 MeV, respectively. Such values, combined with the known α decay energies of ^{106}Te and ^{110}Xe [20], yield an upper limit of 17.2-17.3 MeV for the ^{12}C energy, corresponding to an upper Q(^{12}C) limit of 19.2-19.3 MeV.

This result, although not entirely of experimental origin, is important for two reasons. First, it shows that it is rather unlikely that the (unknown) ^{12}C kinetic energy was so high that events due to cluster decay had escaped our measurement which was optimized to detect 15-20 MeV ^{12}C ions. Second, it suggests that the figure of merit of future experiments has to be increased by several orders of magnitude in order to test the theoretical predictions. This is easily understood by considering that for a Q-value of 19.3 MeV the most widely used theoretical models [11, 21, 22, 23] predict halflives for ^{12}C decay of ^{114}Ba which range between $5 \cdot 10^{7}$ and $5 \cdot 10^{9}$ s, therefore much longer than the presently obtained lower limit. It has indeed been shown, by taking into account the well-known strong dependence of the cluster decay rate on the Q-value, that only by invoking a considerably higher Q(^{12}C) value of 21-22 MeV could a partial halflife of the order of 10^{4} s [10, 11, 23] be reproduced with standard parameters. The fact that the above-mentioned estimate of Q(^{12}C) is based on a semiempirical procedure should not be of much concern, considering the relative stability typical of tunneling phenomena: indeed, a one order-of-magnitude variation of the theoretical decay rate would imply a variation of only 150 keV in Q(^{12}C) [19].

4 Summary and conclusion

Our experiment GSI97 has considerably lowered the upper limit for the branching ratio of a possible ^{12}C cluster decay of ^{114}Ba and, moreover, leaves little room for a claim that this decay mode had actually been observed in previous experiments. Concerning possible improvements of future experiments on this exotic disintegration mode, the now available experience in handling of the BaF ion source would enable one to collect approximately $1.5 \cdot 10^4$ isotopically separated ^{114}Ba atoms per day, hence a ^{114}Ba dose of $1.5 \cdot 10^5$ in a 10 day experiment. On the one hand, such a tripling of the ^{114}Ba dose compared to GSI97 does not seem to us to be a sufficiently strong argument for another search for ^{12}C decay of this isotope. On the other hand, the value of $1.5 \cdot 10^5$ would represent an improvement by a factor of 30 compared to our previous search for α decay of ^{114}Ba [5]. Such a measurement appears attractive as it would either yield evidence for this decay and thus, combined with the known α data of ^{106}Te and ^{110}Xe [20], the ^{12}C decay energy, or at least allow one to deduce an improved semiempirical estimate of this quantity.

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Table 1: Figures of merit (i.e. product of ^{58}Ni beam dose, ^{58}Ni content of target and ^{12}C overall detection efficiency) for the number of ^{12}C-clusters to be detected in GSI97 (taken as reference), GSI95 [5] and Dubna94 [4], respectively.

Experiment	GSI 97	GSI 95	Dubna 94
Total ^{58}Ni beam dose	$1.25 \cdot 10^{17}$	$4.4 \cdot 10^{16}$	$3.5 \cdot 10^{17}$
^{58}Ni content of target	0.999	0.998	0.68
Overall ^{12}C-cluster detection efficiency	0.085 [a]	$\lesssim$0.08 [b]	0.07
Relative figure of merit	1	$\lesssim$0.33 [b]	1.57
"^{12}C cluster decay" events seen	$0^{+1.84}_{-0}$	$2^{+2.6}_{-1.3}$	$20^{+5.5}_{-4.4}$
Inconsistency with GSI 97 (Cf. level) [c]	-	$> 90\%$	$> 99.93\%$

[a] Product of (94±5)% solid angle of our track detector array and (9±3)% overall separation efficiency for ^{114}BaF [15, 14], averaged over the 116 hours of measurement.
[b] Efficiency averaged over 54 hours of measurement. The "$\lesssim$" sign considers the ^{58}Ni target thickness of only 2 mg/cm^2 and the neglected uncertainties concerning the ^{114}Ba beam intensity, mentioned in section 2; however, deviations from the stated value are not expected to be substantial.
[c] For definition see text. The values include the uncertainties of the respective figures of merit. Since no uncertainties are given in the publication concerning Dubna94 [4], an uncertainty of ±32% as for GSI95 and GSI97 is considered here. However, even conceding to Dubna94 a factor-of-two uncertainty, leaves an inconsistency of $> 99.5\%$.

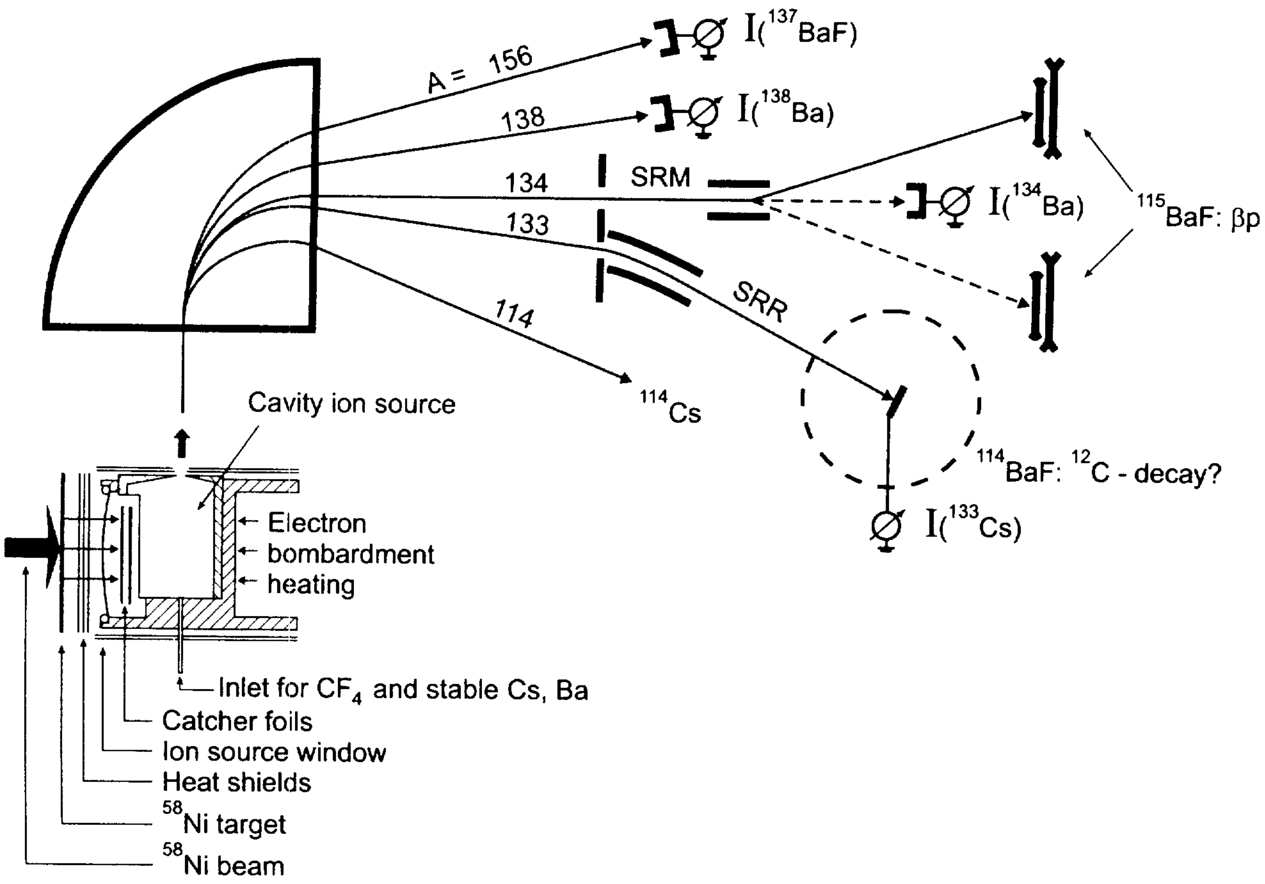

Fig 1. Schematic experimental set-up. The neutron-deficient barium isotopes, produced in the ^{58}Ni(^{58}Ni,xn) reaction, were stopped in thin tantalum catchers inside a high-temperature cavity ion source, re-evaporated, converted into BaF$^+$ ions by reaction with CF$_4$-vapour, and thermo-ionization. Four mass-separated ion beams were *permanently* monitored: Mass 156 and 138 in Faraday cups in the focal plane chamber for control of the effectiveness of the on-line fluorination of barium; mass 134 in the central beam line SRM to control via the βp decay of ^{115}BaF the on-line production of short-lived barium isotopes (occasional position control via ^{134}Ba); mass 133 in the right beam line SRR. The mass-133 beam (permanent control of presence and position via a pilot beam of ^{133}Cs) was stopped in a thin carbon foil in the center of the track detector array in order to register ^{12}C-clusters emitted from ^{114}Ba. The beam of ^{114}Cs with its high βp rate could, if needed, be sent into SRM for fast-checks of the ^{58}Ni-targets and/or the performance of the silicon telescopes. Note that ^{133}Cs and $^{134-138}$Ba are stable isotopes, and that cesium is only suppressed in the fluoride sideband, but well-present as metallic ion.

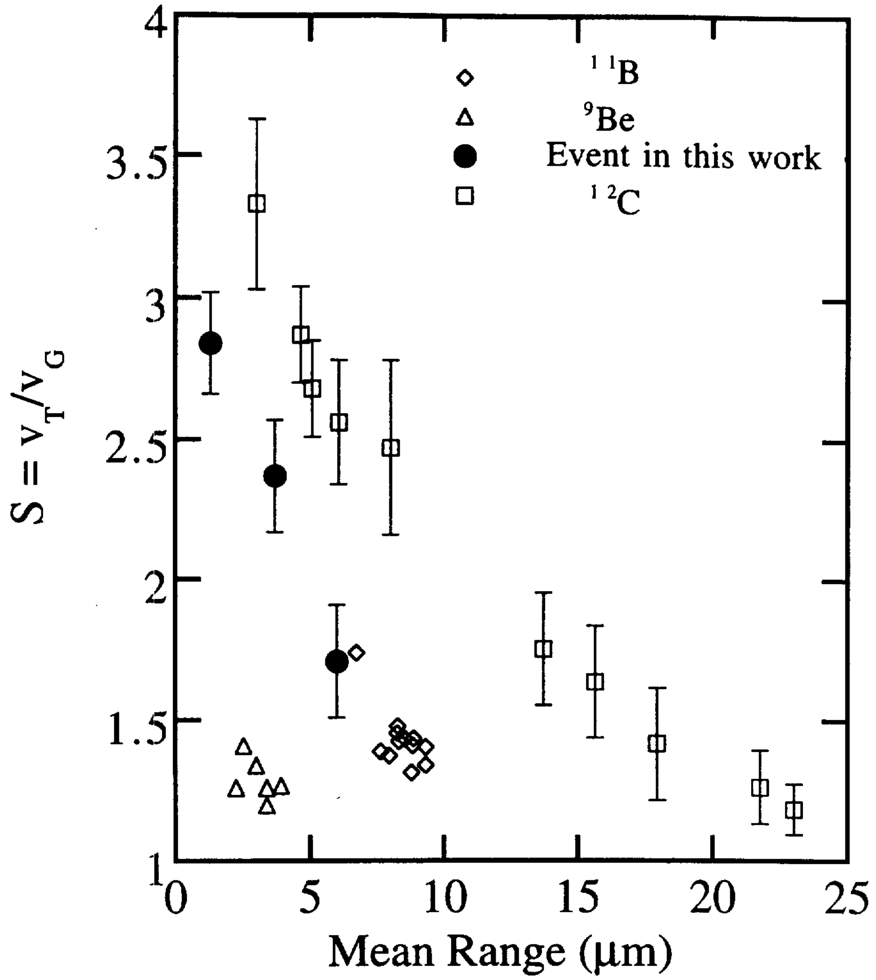

Fig. 2. Comparison of the track detector sensitivity S for ions of ^{12}C, ^{11}B, ^{9}Be, and the one event found in this experiment. S is defined as the ratio of track etching velocity v_T and general etching velocity v_G. Related values of S and the mean range were measured at different points along the particle range. Three such results were obtained for the one event observed in this work (filled circles). As far as the ^{12}C data (squares) are concerned, the five data points at low range were obtained for a beam energy of 14.72 MeV, whereas those at larger range are for higher energy.