Title

SEARCH FOR AN 80-ms SPONTANEOUS FISSION ACTIVITY IN BOMBARDMENTS OF 249Bk WITH 15N

Author

Nitschke, J.M.

Publication Date

2012-02-17

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Submitted to Nuclear Physics A

SEARCH FOR AN 80-ms SPONTANEOUS FISSION ACTIVITY IN BOMBARDMENTS OF ^{249}Bk WITH ^{15}N

J. M. Nitschke, M. Fowler, A. Ghiorso, R. E. Leber,
M. J. Nurmia, L. P. Somerville, K. E. Williams, E. K. Hulet,
J. H. Landrum, R. W. Lougheed, J. F. Wild, C. E. Bemis, Jr.,
R. J. Silva, and P. Eskola

January 1980

Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

SEARCH FOR AN 80-ms SPONTANEOUS FISSION ACTIVITY IN BOMBARDMENTS OF ^{249}Bk WITH ^{15}N

J. M. Nitschke, M. Fowler,* A. Ghiorso, R. E. Leber, M. J. Nurmia, L. P. Somerville, K. E. Williams*

Nuclear Science Division, Lawrence Berkeley Laboratory, 1 Cyclotron Road, Berkeley, CA 94720†

E. K. Hulet, J. H. Landrum, R. W. Lougheed, J. F. Wild,

Nuclear Chemistry Division, Lawrence Livermore Laboratory, P. O. Box 808, Livermore, CA 94550

C. E. Bemis, Jr., R. J. Silva,**

Chemistry Division, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830

P. Eskola,

Department of Theoretical Physics, University of Helsinki, Siltavuorenpenger 20, SF-00170, Helsinki 17, Finland

---

* Present address: Los Alamos Scientific Laboratory, University of California, Los Alamos, NM 87545.

** Present address: Lawrence Berkeley Laboratory, 1 Cyclotron Road, Berkeley, CA 94720.

† This work was supported by the Division of High Energy and Nuclear Physics Division of the U. S. Department of Energy under contracts No. W-7405-ENG-48 and W-7405-ENG-26.

Abstract: A rotating drum system was used to search for an 80-ms spontaneous fission (sf) activity in the reaction of ^{15}N with ^{249}Bk. No such activity was found beyond a cross section limit of 0.3 ± 0.3 nb. A sf activity with a half-life of about 20 ms and a formation cross section of 12 nb at 82 MeV was observed. The identity of this activity has not been determined.

NUCLEAR REACTION: ^{249}Bk (^{15}N, X) sf; E = 78, 82, 86, 88, and 100 MeV, observed sf emitters, $T_{1/2}$ = 20 ms, 13 ms, and 2.6 h, measured $\sigma(E)$, comments on nonobservation of 80-ms sf activity.

I. Introduction

Claims by the Joint Institute for Nuclear Research (JINR) at Dubna for the discovery of element 104 are based on their early observation of a 0.3-s spontaneous fission (sf) activity, which was assigned to the nuclide $^{260}104$.[1,11] Subsequent experiments by JINR caused a revision in the half-life, first to 0.1 s[2] and most recently to 80 ms.[3,4] JINR reported observing this same activity in a variety of bombardments with heavy ions in which $^{260}104$ could be expected to be a product of compound nucleus reactions. Peak cross sections for the formation of this activity increased progressively from ~0.5 to 8 nb for the reactions $^{242}Pu(^{22}Ne, 4n)$, (0.5 nb),[12] $^{246}Cm(^{18}O,4n)$, (1.5 nb),[3] $^{248}Cm(^{16}O,4n)$, (~1 nb),[3] and $^{249}Bk(^{15}N, 4n)$, (8 ± 2 nb),[4] respectively. However, Ghiorso et al.[5] were unable to observe this decay period in the sf activities produced from ~92-MeV ^{18}O and ~94-MeV ^{16}O bombardments of ^{246}Cm and ^{248}Cm. Furthermore, they determined that the sensitivity of their experiments was a factor of 4 to 6 times more than adequate to detect the 80-ms activity if the results of JINR were correct. Thus, contradictory results have come from similar experiments at Lawrence Berkeley Laboratory (LBL) and JINR.

It appeared that resolution of this question required that further experiments be performed to determine whether an 80-ms sf activity could be assigned to $^{260}104$. Accordingly, beginning in 1976, we carried out a series of bombardments of ^{249}Bk with ^{15}N ions at five different ion energies ranging from 78 to 100 MeV.*) It is important to note that this target-projectile combination was chosen because both theoretical estimates[6]) and comparative experimental data indicate that the reaction ^{249}Bk (^{15}N, 4n) would have a higher yield than reactions making use of targets with lower atomic number. We expected that ^{260}104 would be produced with a peak cross section of approximately 14 nb with 82-MeV ^{15}N ions.

---

* A preliminary report of these experiments was given at the 3rd International Conference on Nuclei far from Stability, Cargèse, Corsica (France), 1976.

2. Experiment

A ^{15}N beam from the LBL 88-in. cyclotron was used to bombard a 4.8-mm-diameter target containing 85 μg of ^{249}Bk. The ^{249}Bk, chemically separated from its ^{249}Cf daughter just three days prior to the first bombardments, was sublimed as BkF_3 onto a 0.012-mm-thick substrate of beryllium.[7]) The ^{249}Bk target, facing away from the incoming beam, was edge-cooled by contact with a water-cooled collimating assembly containing beryllium degrading foils which reduced the energy of the ^{15}N ions by the desired amount. The energy of the ^{15}N ions entering the target assembly was determined by the frequency and the magnetic field of the cyclotron. This method was checked independently at one point with a magnetic analyzer. The target assembly together with the detector system is shown schematically in fig. 1.

The technique for detecting spontaneous fission events from the decay of $^{260}104$ entailed the collection of recoil atoms on the rim surface of a large rotating drum.[8]) Surrounding and facing the drum surface (0.8-mm gap) were stationary strips of muscovite mica which recorded fragment tracks. The recoil atoms were caught and carried on the surface of the drum until they decayed and left tracks in the mica. After the mica was etched in 48% hydrofluoric acid at 60°C for about 50 minutes, these tracks were readily identified in a microscope at 100X magnification. The tracks were counted independently by two different scanners. The track counting efficiency as measured with ^{252}Cf sources was 95%.

In these experiments, we used a water-cooled, 254-mm diameter drum rotating at a preset, electronically controlled speed (either 60 or 48 rpm). Accurate rotation rates were determined from the frequency of interruption of a light beam by a perforated disc mounted on the drum shaft. To reduce the density of long-lived nuclei such as ^{256}Md or ^{256}Fm which contributed a substantial sf background, we moved the 360-mm-long drum axially at a rate of 7.7 mm/s. The direction of this axial motion was reversed after each full passage so that any long-lived activity was continually distributed over the full drum surface. By taking only fission tracks in a narrow 10.6-mm-wide beam-centered path in the mica (fig. 2), corresponding to the location of ~75% of the products from compound nucleus reactions as determined by the $^{241}Am(^{15}N,4n)^{252}No$ reaction, we reduced the registration of long-lived activities by a factor of 34 compared to a fixed, rotating drum.

The bombardments varied in length from 4.5 to 18 hours. At the end of each run, the mica strips were recovered, etched, and scanned for characteristic fission tracks. Half-lives were determined from the exponential decay of the number of fission events with circumferential distance from the target, the time base being provided by calculating the surface velocity of the drum from the frequency of rotation. The multicomponent decay curves were resolved through least-mean-square (LMS) fitting by several computer programs. As a further precaution against bias, representatives from each of our laboratories independently analyzed the raw track-counting data.

Virtually identical results were obtained. In these analyses, the many tracks observed in the leading ~7 mm of the first mica strip (corresponding to the first 9 ms at 60 rpm drum speed) were not used because of the difficulty of identifying fission tracks in the area heavily damaged by scattered beam particles.

3. Results and Discussion

Bombardments with ^{15}N were made at 78, 82, 86, 88 and 100 MeV to insure that the maximum in the reaction cross section was not missed. We were able to observe a short-lived and a long-lived sf activity at every ion energy when the drum was rotating at either 60 or 48 rpm. The long-lived activity, evenly distributed over the entire drum surface, presented an approximately flat background. This activity was identified in separate recoil-catcher experiments as being due to the growth and decay of 2.6-hour ^{256}Fm fed by the electron capture decay of 77-minute ^{256}Md.[9] Its production cross sections calculated from both the drum and the foil-collection experiments are given in table 1.

The short-lived activities exhibited half-lives of 13 to 20 ms, depending on the bombarding energy, and an apparent cross section that has a peak value of 12 nb at 82 MeV. The decay curve and the LMS analyses allowing for one component plus a constant ^{256}Md-^{256}Fm background are shown in fig. 3. The cross sections and half-lives obtained at various energies are shown in Table 1 and fig. 4. Further information about the short-lived activity was obtained by means of the density profile of the fission tracks on the first mica strip taken at right angles to the rotation of the drum (fig. 2). The relatively narrow distribution suggests that this activity might be produced in a compound nucleus-like reaction.

These data allow us to reach the following conclusions:

1. No statistically significant amount of sf activity attributable to an 80-ms half-life was produced at any of the

bombarding energies used. This result was confirmed by using four different LMS computer programs to search the data for a component with an 80 ms half-life in addition to the 13 to 20 ms activity and a long-lived background. The results varied from negative intensities at 78 and 82 MeV bombarding energies to a maximum positive intensity of about 4% of the short-lived component in the three higher energy bombardments.

2. A sf activity with a half-life of about 20 ms and a formation cross section that peaks at about 12 nb at a ^{15}N energy of 82 MeV is clearly observed. This is probably the same activity that was discovered earlier in bombardments of ^{248}Cm with 94-MeV ^{16}O ions.[5] On the other hand, it remained undetected in irradiations of either ^{246}Cm with ^{16}O ions or ^{248}Cm with ^{15}N ions.[10] Because $^{260}104$ cannot be made in the latter reaction and would have an undetectable small yield in the former, but can be made from $^{248}Cm(^{16}O,4n)$ and $^{249}Bk(^{15}N,4n)$ reactions, the results of these cross bombardments would be consistent with the assignment of this activity to $^{260}104$. Although this assignment would be further supported by the absolute magnitude of the cross section, the narrow width of the peak of the excitation curve and the forward-peaked angular distribution, we feel that we cannot at present eliminate other possibilities without additional experiments. This activity might still be due to a transfer reaction or charged particle evaporation producing a nuclide somewhat lower in Z than 104.

3. There is evidence for another short-lived sf activity with a half-life of about 13 ms. It apparently has a broad excitation function that extends to our highest bombarding energies. This activity conceivably could be due to 13.7-ms $^{242sf}Am$ produced in a transfer reaction. The broadness of the excitation function could imply that the half-life of the major "20-ms" component might be a few milliseconds longer than 20 ms from an admixture of the 13-ms component.

We conclude that an 80-ms sf activity was not observed in these bombardments covering a broad range of ^{15}N energies. The maximum cross section allowed by our results for the formation of an 80-ms sf activity is 0.3±0.3nb. This cross section must be compared to the 8±2 nb reported by Druin and co-workers[4]) and the 14 nb calculated by Alonso[6]) for the $^{249}Bk(^{15}N,4n)\ ^{260}104$ reaction. A calculation using the same parameters correctly reproduces the measured cross-section of the quite similar reaction, $^{249}Cf(^{15}N,4n),\ ^{260}105$. We can only conclude that the nuclide $^{260}104$ does not decay by spontaneous fission with a half-life in the range of 80-300 ms as claimed by the JINR investigators. We suggest that their activity is either produced from some common impurity in their targets or is due to an experimental artifact.

The authors wish to express their gratitude to the following colleagues for their support and able assistance: P. A. Baisden, I. Binder, D. M. Lee, D. J. Morrissey, R. J. Otto, G. T. Seaborg, and K. E. Thomas. We also wish to thank the staff of the 88 in. cyclotron for many hours of excellent beams.

Table 1. Half-lives and formation cross sections observed for the short and long-lived components at each bombardment energy. The last column lists formation cross sections obtained for $^{256}Md/^{256}Fm$ in short duration experiments where the recoil products were collected on thin Al foils.

N^{15} Energy (MeV)	Integrated flux $x10^{-17}$	$t_{1/2}$ short component(ms)	σ short component(nb)	σ ^{256}Md (nb)	σ ^{256}Md Foil Exp. (nb)
77.9	1.17	19.9 ± 2.0	6.8 ± 2.4	–	28.6 ± 9.5
81.6	1.51	20.1 ± 1.7	11.7 ± 4.1	–	272 ± 32
85.7	0.29	17.9 ± 2.5	5.4 ± 1.9	240 ± 90	–
88	1.31	12.7 ± 2.0	4.2 ± 1.5	280 ± 100	408 ± 51
100	1.18	12.8 ± 3.0	3.6 ± 1.3	–	208 ± 42

References

1) G. N. Flerov, Yu. Ts. Oganesyan, Yu. V. Lobanov, V. I. Kuznetsov, V. A. Druin, V. P. Perelygin, K. A. Gavrilov, S. P. Tret'yakova, and V. M. Plotko, At. Energ. (USSR) 17 (1964) p. 310 (translation: Soviet At. Energy. 17 (1964) p. 1046.

2) Yu. Ts. Oganesyan, Yu. V. Lobanov, S. P. Tret'yakova, Yu. A. Lazarev, I. V. Kolesov, K. A. Gavrilov, V. M. Plotko and Yu. V. Poluboyarinov, At. Energ. (USSR) 28 (1970) p. 393 (translation: Soviet At. Energ. 28 (1970) p. 502.

3) V. A. Durin, Yu. S. Korotkin, Yu. V. Lobanov, Yu. V. Poluboyarinov, R. N. Sagaidak, G. M. Solov'eva, S. P. Tret'yakova, Yu. P. Kharitonov, Yad. Fiz. 24 (1976) p. 254.

4) V. A. Druin, B. Bochev, Yu. P. Kharitonov, Yu. S. Korotkin, V. N. Kosyakov, Yu. V. Lobanov, E. A. Minin, Yu. V. Poluboyarinov, A. G. Rykov, R. N. Sagaidak, S. P. Tretyakova, Joint Institute for Nuclear Research, Dubna, USSR, Report E7-10499 (1977).

5) A. Ghiorso, Proc. of the R. A. Welch Foundation Conf. on Transuranium Elements, Houston, (1970) ed. W. O. Milligan, p. 107.

6) J. R. Alonso, in Gmelin Handbuch der Anorganischen Chemie, Ergänzungswerk, ed. K. V. Baczko et al., (Vol. 7b, Part A1, II), (Springer, Berlin, 1974), p. 28.

7) R. W. Lougheed and E. K. Hulet, Nuclear Instruments and Methods (in press), UCRL-75782.

8) A. Ghiorso, P. A. Baisden, I. Binder, R. E. Leber, D. Lee, D. Morrissey, J. M. Nitschke, M. J. Nurmia, R. J. Otto, L. P. Sommerville, K. Thomas, K. E. Williams, E. K. Hulet, R. W. Lougheed, J. F. Wild, J. H. Landrum, R. J. Silva, C. E. Bemis, M. Fowler, Lawrence Berkeley Lab., Annual Report of the Nuclear Science Division, LBL 5075 (1975) p. 40.

9) R. W. Hoff, E. K. Hulet, R. J. Dupzyk, R. W. Lougheed, and J. E. Evans, Nucl. Phys. A169 (1971) p. 641.

10) K. E. Williams and L. P. Somerville, Lawrence Berkeley Laboratory, unpublished data (1976).

11) G. N. Flerov, Yu. Ts. Oganesyan, Yu. V. Lobanov, V. I. Kuznetsov, V. A. Druin, V. P. Perelygin, K. A. Gavrilov, S. P. Tret'yakova and V. M. Plotko, Phys. Lett. 13 (1964) p. 73.

12) Yu. Ts. Oganesyan, Yu. V. Lobanov, S. P. Tret'yakova, Yu. A. Lasarev, I. V. Kolesov, K. A. Gavrilov, V. M. Plotko, Yu. V. Poluboyarinov, Joint Institute for Nuclear Research, Dubna, USSR, Preprint JINR-P7-4797 (1969).

Figure Captions

Fig. 1. Isometric view of the drum system.

Fig. 2. Fission track distribution on the micas perpendicular to the rotation of the drum.

Fig. 3. Decay curves and LMS analysis of the spontaneous fission events (beam energy: 82 MeV, beam integral: $1.51 \cdot 10^{17}$ ions).

Fig. 4. Dependence of the cross section for the short-lived sf component on the energy of the ^{15}N projectile.

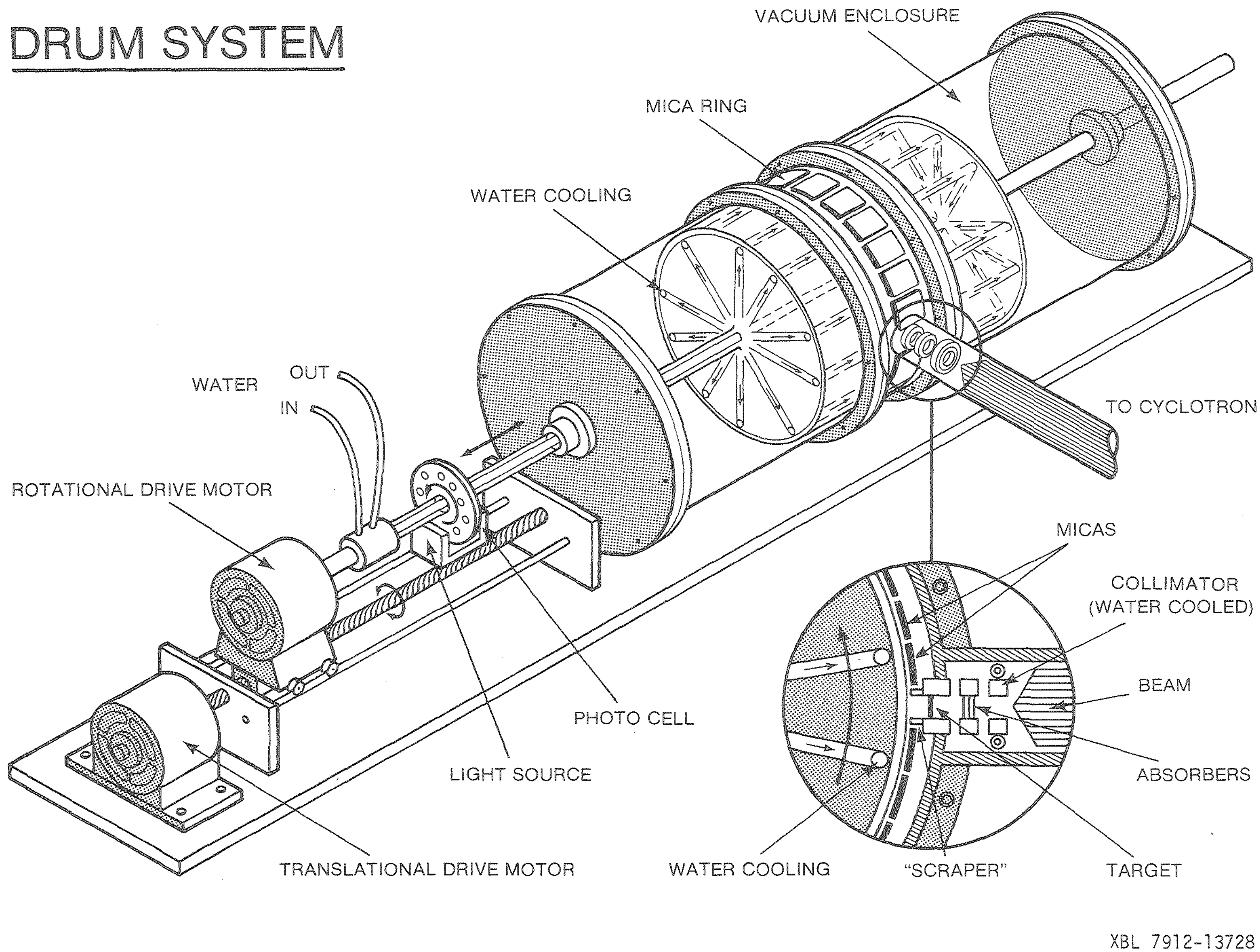
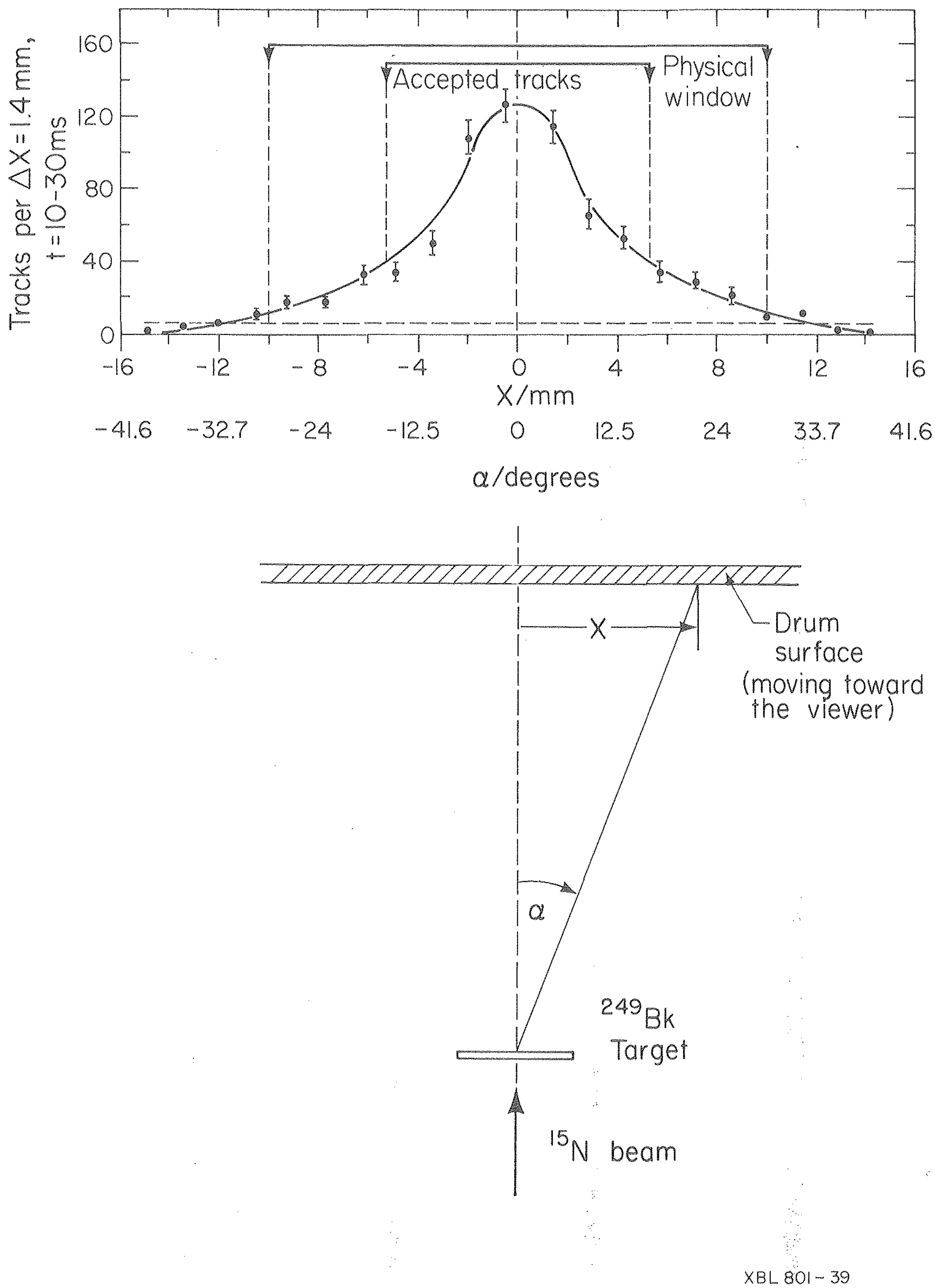
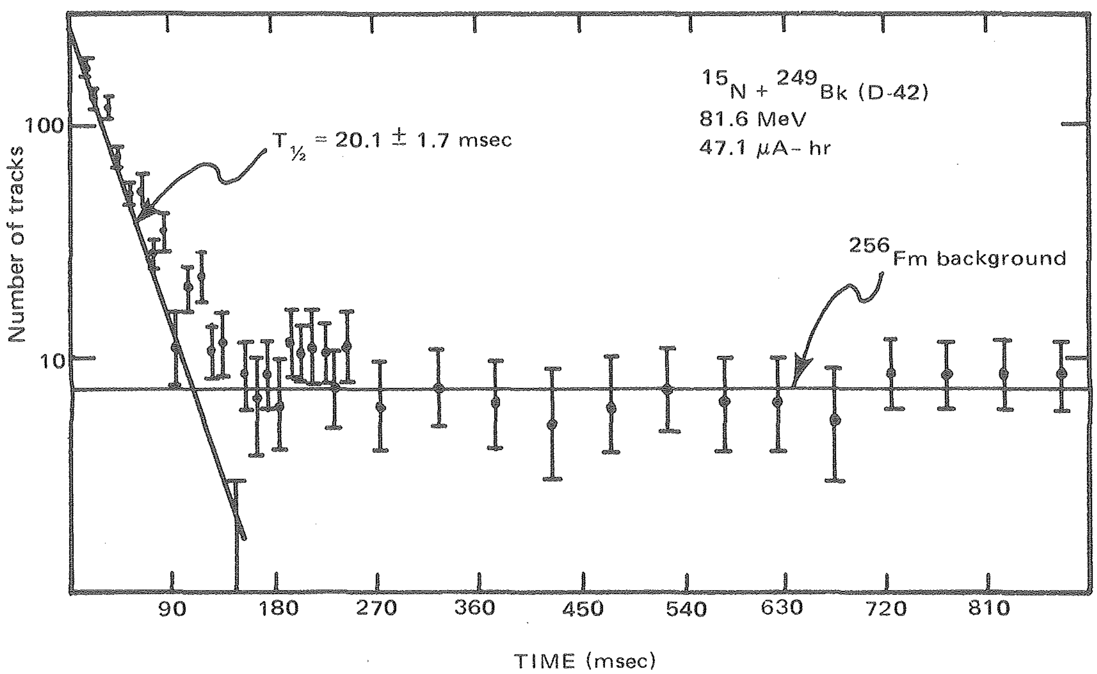

DRUM SYSTEM
VACUUM ENCLOSURE
MICA RING
WATER COOLING
WATER
OUT
IN
TO CYCLOTRON
ROTATIONAL DRIVE MOTOR
MICAS
COLLIMATOR
(WATER COOLED)
BEAM
PHOTO CELL
LIGHT SOURCE
ABSORBERS
TRANSLATIONAL DRIVE MOTOR
WATER COOLING
"SCRAPER"
TARGET
XBL 7912-13728

Fig. 1



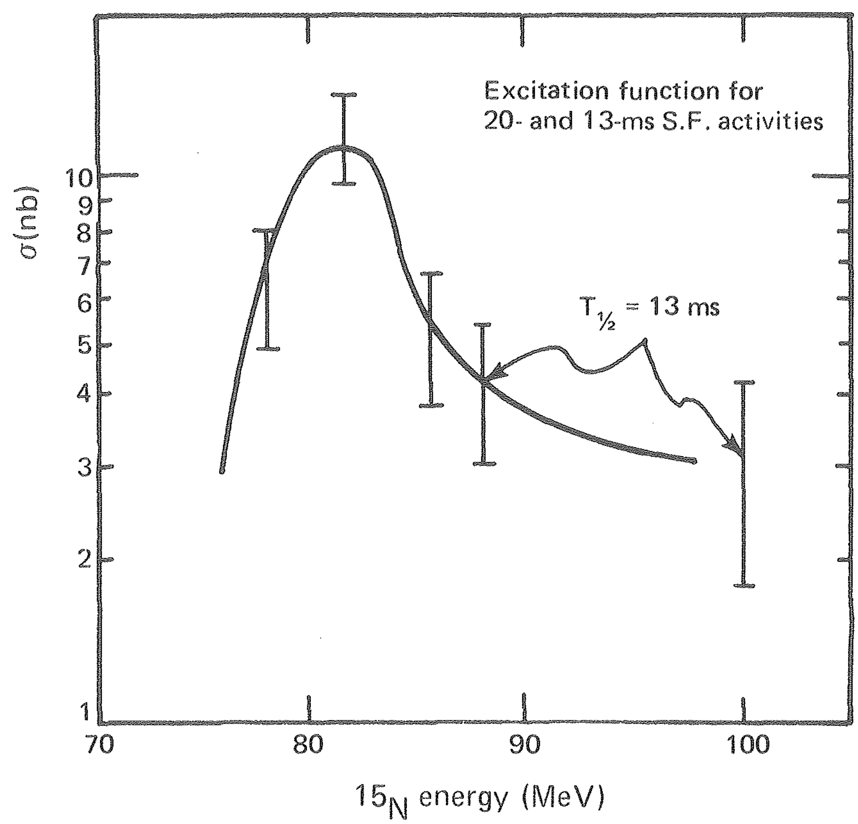
Tracks per ΔX=1.4mm, t=10-30ms
160
120
80
40
0
Accepted tracks
Physical window
-16
-12
-8
-4
0
4
8
12
16
X/mm
-41.6
-32.7
-24
-12.5
0
12.5
24
33.7
41.6
α/degrees
X
Drum surface (moving toward the viewer)
α
249Bk Target
15N beam
XBL 801-39

Fig. 2



XBL766- 8243

Fig. 3



XBL766 - 8245

Fig. 4