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DISCOVERY OF FERMIUM-259*

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Abstract

A 1.5-sec spontaneous fission activity has been produced by irradiation of ^{257}Fm with 16-MeV tritons at the Los Alamos Tandem Van de Graaff Facility. On the basis of cross section and fission systematics, this 1.5-sec activity has been attributed to ^{259}Fm formed by the reaction, ^{257}Fm(t,p)^{259}Fm. ^{259}Fm is the heaviest known isotope of fermium and has more neutrons than any other isotope so far identified. The mass distribution of the ^{259}Fm is highly symmetric and the average total kinetic energy is very high, 230 ± 2 MeV. These features continue the trend toward more symmetric mass distributions and higher total kinetic energies observed previously for the fermium isotopes as the number of neutrons is increased.

1. Introduction

At the first International Conference on the Propert s of Nuclei Far from the Region of Beta-Stability held in Lysekil, Sweden in 1966, we reported[1)] on the production of heavy element isotopes in underground nuclear tests. At that time, we were extremely puzzled at our failure to detect any evidence for the presence of the mass 259 chain although we felt it should have been produced. The spontaneous fission (SF) half life of ^{257}Fm was known to be 125 yr and estimates indicated that the SF half life for ^{259}Fm should have been around a month, but we were able to set limits of ≤5 h or ≥7.5 years for the SF half life of ^{259}Fm, assuming initial production with an abundance 1/30th that of the 257 chain and that the beta-decay chain had not been terminated by spontaneous fission before reaching ^{259}Fm. However, with the discovery in 1971 by Hulet and co-workers[2)] that the half life of ^{258}Fm was only 380 μsec, it appeared that a "disaster" in spontaneous fission half lives had occurred after N = 157. The half life of ^{259}Fm then might easily be very much less than the 5-h upper limit we had set earlier. Assuming the reduction in half life for the addition of 2 neutrons to ^{257}Fm was the same as that of 4×10^{-8} between ^{256}Fm and ^{258}Fm, a half life of 2-3 min could be estimated for ^{259}Fm. However, calculations of Randrup et al.[3] indicated that disappearance of the second fission barrier at ^{258}Fm could account for its very short half life. Consequently, ^{258}Fm and ^{259}Fm should have about the same half life except for the special hindrance associated with the odd nucleon and the ^{259}Fm half life might then be only a few-tenths of a second. Since ^{258}Fm was the only nuclide known with more than 157 neutrons, it was of considerable importance in the estimation of unknown spontaneous fission half lives to determine the effect of adding still another neutron to fermium.

Even though only about 10^9 atoms of ^{259}Fm could be obtained, the production and detection of ^{259}Fm by the (t,p) reaction on ^{257}Fm appeared feasible because the cross section was expected to be of the order of 10 mb[4] and 10-μA beams of 16-MeV tritons could be obtained at the Tandem Van de Graaff at Los Alamos Scientific Laboratory. In this way we have succeeded in producing and making on-line measurements of a 1.5-sec spontaneous fission activity which we have attributed[5] to ^{259}Fm, thus solving the mystery of the missing mass 259 chain.

2. Experimental Methods

The targets were prepared by electrodeposition of ^{257}Fm, together with 10 to 50 μg of ^{209}Bi, in a 2.2-mm diameter circle on 0.013-mm beryllium foil. The targets contained from 1.3×10^9 to 0.9×10^9 atoms of ^{257}Fm. The α-disintegration rate of ^{211}Po, formed by the (t,p) reaction on the ^{211}Bi in the target, was used to monitor the relative recoil efficiency of the target during the course of the irradiation. Targets containing only ^{209}Bi were prepared in the same manner and used for background measurements.

The targets, which were irradiated with 16-MeV tritons at an average intensity of 10 μA, were held in a water-cooled copper block. Nuclei recoiling from the target were caught on 30-μg/cm^2 carbon foils mounted on the circumference of a vertical wheel. The wheel was rotated by a high-speed stepping motor so that the carbon foils could be moved stepwise from the collection point in front of the target into selectable positions between stationary pairs of silicon surface-barrier detectors. After one complete rotation of the wheel, a given foil would again be in collection position. The apparatus is shown in Figure 1. In these experiments either 2, 3 or 5 detector pairs were used. The entire system was attached to the accelerator beam line and evacuated. The irradiation and counting sequences were continuous although the beam was deflected while the foils were being moved. Bombardment and counting times were the same, but were varied between 1 and 6 sec. The time from collection to counting, during which the wheel moved, was varied from 0.15 to 0.30 sec. Pulse-height and time information for coincident fission fragments was recorded using an on-line SDS-930 computer system. A ^{252}Cf source, temporarily mounted in position on the wheel, was used in energy calibration of the detectors.

3. Results

Fission data were taken at five different time intervals with three different detector configurations during ten days of nearly continuous bombardments with $\sim$ 10 μA of 16-MeV tritons. The fission rate of 10 to 23 coincident SF/h was proportional to the amount of ^{257}Fm in the target and to the beam current. Background checks with the ^{209}Bi targets showed that < 1% of the observed fissions were due to background effects or detector noise. Analysis of the number and time after irradiation of the coincident fission fragments recorded by each detector pair showed at least 2 components, one with a half life of about 1.5 sec and the other much longer. (See Figure 2.) Events with total kinetic energy greater than 230 MeV showed a much smaller proportion of long-lived activity. A 2-component least squares analysis of these events gave a half life of 1.5 ± 0.2 sec for the shorter activity when the other half life was fixed at 7 to 12 hours. The cross section for formation of the 1.5-sec activity was estimated to be about 10 mb assuming a recoil efficiency of 40%.

The long-lived SF activity built up to around 20% in 10-hour or longer bombardments. The fermium fraction from chemical processing of a set of recoil foils and separation of the heavy actinides by elution from a cation exchange column with α-hydroxyisobutyrate showed the presence of 2.6-h ^{256}Fm (92% SF decay). It was presumably formed by beta decay of 25-min ^{256}Es produced by the (t,α) reaction. There was also some evidence for SF activity in the einsteinium fraction separated 1.5 h after the end of irradiation. This activity appeared to decay with an 8 to 12-h half life. We postulated that a longer-lived isomer of ^{256}Es might have been formed which also β-decayed to ^{256}Fm. This was later confirmed[6)] in bombardments of ^{254}Es with 16-MeV tritons in which a 7.6-h, high-spin isomer of ^{256}Es was identified.

From our measurements of the energies of 497 coincident fission fragment pairs, an average total kinetic energy (TKE) of 222.7 ± 1.3 MeV was obtained. (See Figure 3.) The distribution is very broad (σ = 29 MeV), probably indicative of the presence of more than one SF species. The TKE data as a function of time are summarized in Table I and show some decrease with time after irradiation, but even at late times the TKE is still higher than that of 198 MeV measured[7,8] for ^{256}Fm. In order to determine whether there was an energy bias in our system we prepared ^{256}Fm via the following reaction:

$$^{254}\text{Es}(t,p)^{256}\text{Es} \xrightarrow{\beta^-} {}^{256}\text{Fm}.$$

The recoils were caught in the same manner as in the ^{257}Fm bombardments and energy measurements of the ^{256}Fm fission fragments were made with the same detector arrangement as before, both with the beam on and off. A TKE of 197.2 MeV was obtained for the ^{256}Fm, in good agreement with the values reported in the literature, but significantly lower than the TKE's shown in Table I for the later times. Even though there are very few events in these later time intervals, the differences of 7 to 20 MeV seem to be large enough to indicate the presence of another relatively long SF activity in addition to ^{256}Fm.

A provisional mass distribution was derived from the energy data for these events. It is shown in Figure 4 together with those obtained for ^{252}Cf and ^{256}Fm in our system. Even though a considerable number of longer-lived events are included, the mass distribution is very symmetric with a half-width at half-maximum of only ~13 mass units. An amount of ^{256}Fm equivalent to the fraction of long-lived fission events relative to the 1.5-sec activity was subtracted from this distribution, and the resulting extremely narrow mass distribution shown in Figure 5 was obtained. The mass distributions as a function of time were also obtained and although they broaden somewhat with time, they still appear symmetric and do not match those obtained for ^{256}Fm. Again, another long-lived component in addition to ^{256}Fm may be indicated.

4. Discussion

We have assigned the 1.5-sec spontaneous fission activity to ^{259}Fm on the basis of the following points:

1. The relatively high production cross section of ∿10 mb is typical[4,6] of (t,p) reactions in this region. The production rate for the (t,n) reaction was shown to be only 0.06 of that for the (t,p) in our 16-MeV triton bombardments[6] of ^{254}Es.

2. Although an isomer of ^{258}Md is a possible assignment since the (t,2n) cross section for ^{254}Es was 0.5 of that for (t,p), the half life of such an isomer (-1) of ^{258}Md would be expected to be much longer than 1.5 sec since ^{258}Ms is so close to stability and the decay energy is probably quite small. In addition, part of the cross section must be taken up in the formation of the known 54-d ^{258}Md.

3. Assignment to ^{257}Es produced by a (t,^{3}He) reaction is not credible because of the small cross section and expected long SF half life for ^{257}Es.

4. The other fermium isotopes which might conceivably be produced are already known. The very high TKE and narrowly symmetric mass distribution of the 1.5-sec activity continue the trends toward higher TKE and more symmetric mass distributions observed previously for the fermium isotopes as the number of neutrons increases and seem most consistent with assignment to mass 259.

5. The half life is consistent with systematic trends in the SF half lives of the fermium isotopes.[2]

There is some evidence for another relatively long-lived SF activity in addition to ^{256}Fm formed via the (t,α) reaction. The TKE and mass distributions observed at late times do not match those for pure ^{256}Fm. Further, if the ratio of the (t,α) to (t,p) production rates of 0.5 observed for ^{254}Es holds for ^{257}Fm, the calculated amount of ^{256}Fm produced would not account for all the long-lived SF activity detected. Admittedly, the statistics are poor, but if such a SF activity is postulated to be due to an isomer (1-) of ^{258}Md, it would decay by either electron capture or beta decay to short-lived SF activities. ^{258}Md could be produced by the (α,n) reaction on ^{255}Es and ^{259}Fm would not be produced in such a bombardment. We hope to be able to perform this experiment, both to identify another isotope of ^{258}Md and to preclude the possibility that the 1.5-sec activity is due to ^{258}Md. Production of such an isomer of ^{258}Md would also make it possible to study the mass and energy distributions from the SF of the resulting ^{258}No or ^{258}Fm. The SF characteristics of ^{258}No would be of particular interest for comparison with the trends observed in the fermium isotopes.

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Figure 1. Apparatus for collection and counting of recoil products. The wheel carrying the carbon foils was rotated periodically to bring each foil between the pairs of surface-barrier detectors. (One set of detectors is in position behind the carbon foils; the other set with its support has been removed to show the foils.) The top foil is in collection position in front of the target.

Figure 2. Decay curves for the spontaneous fission activities produced in the bombardment of ^{257}Fm with 16-MeV tritons: (a) all coincident fissions summed from runs in which the foils were advanced every 3, 4, or 6 sec; (b) coincident fissions with total kinetic-energies exceeding 230 MeV for runs with a 4 sec/station advance rate. The short-lived component assigned to ^{259}Fm was resolved by least-mean-squares fitting of the curves.

Figure 3. Total kinetic energy for 497 events observed from 0-14 sec after bombardment of ^{257}Fm with 16-MeV tritons.

Figure 4. Provisional mass distribution for 497 events observed from 0-14 sec after bombardment of ^{257}Fm with 16-MeV tritons.

Figure 5. Mass distribution for ^{259}Fm obtained after subtraction of long-lived SF, due to ^{256}Fm.

↑ Top

Fig. 1

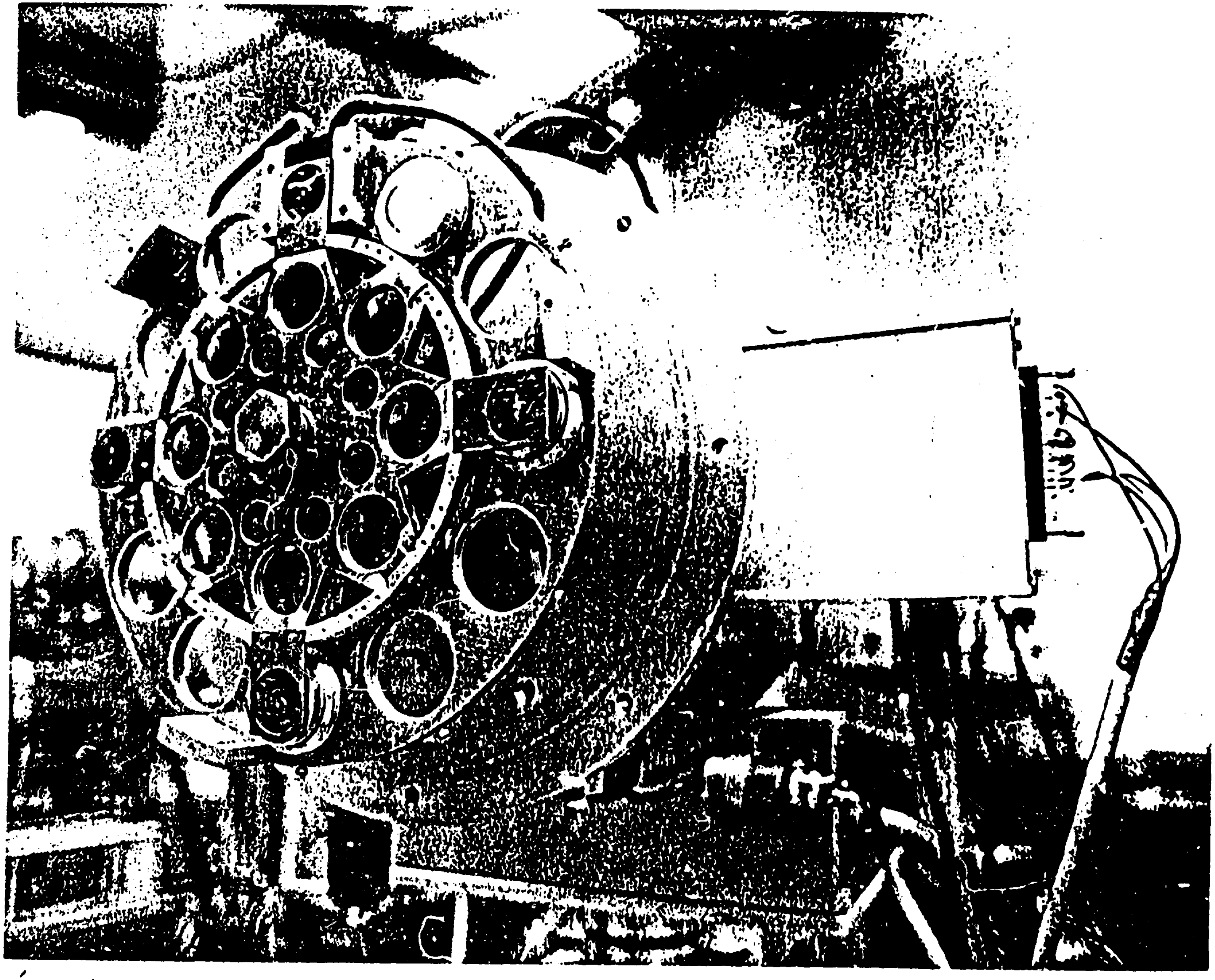
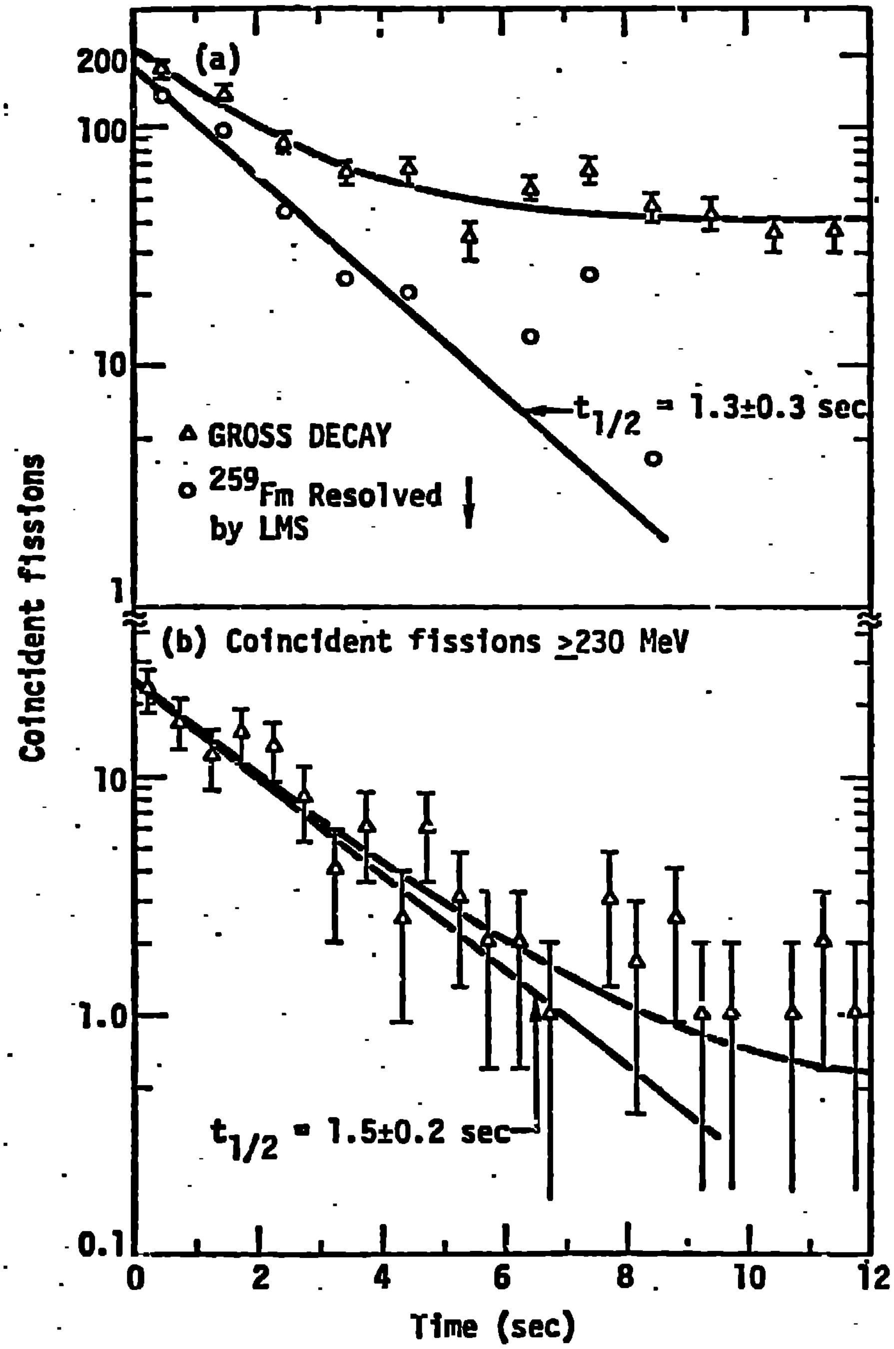
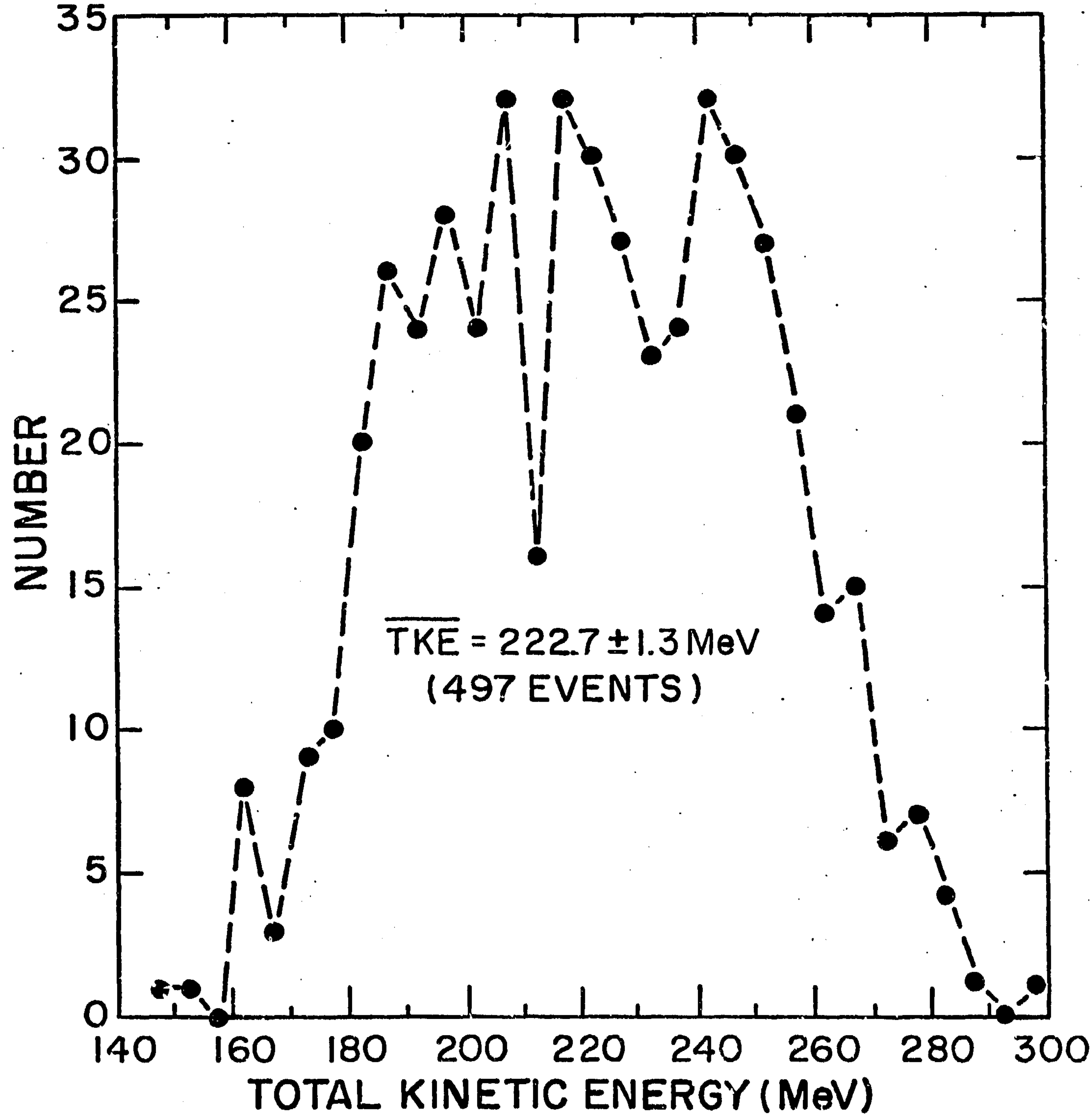


Fig 2

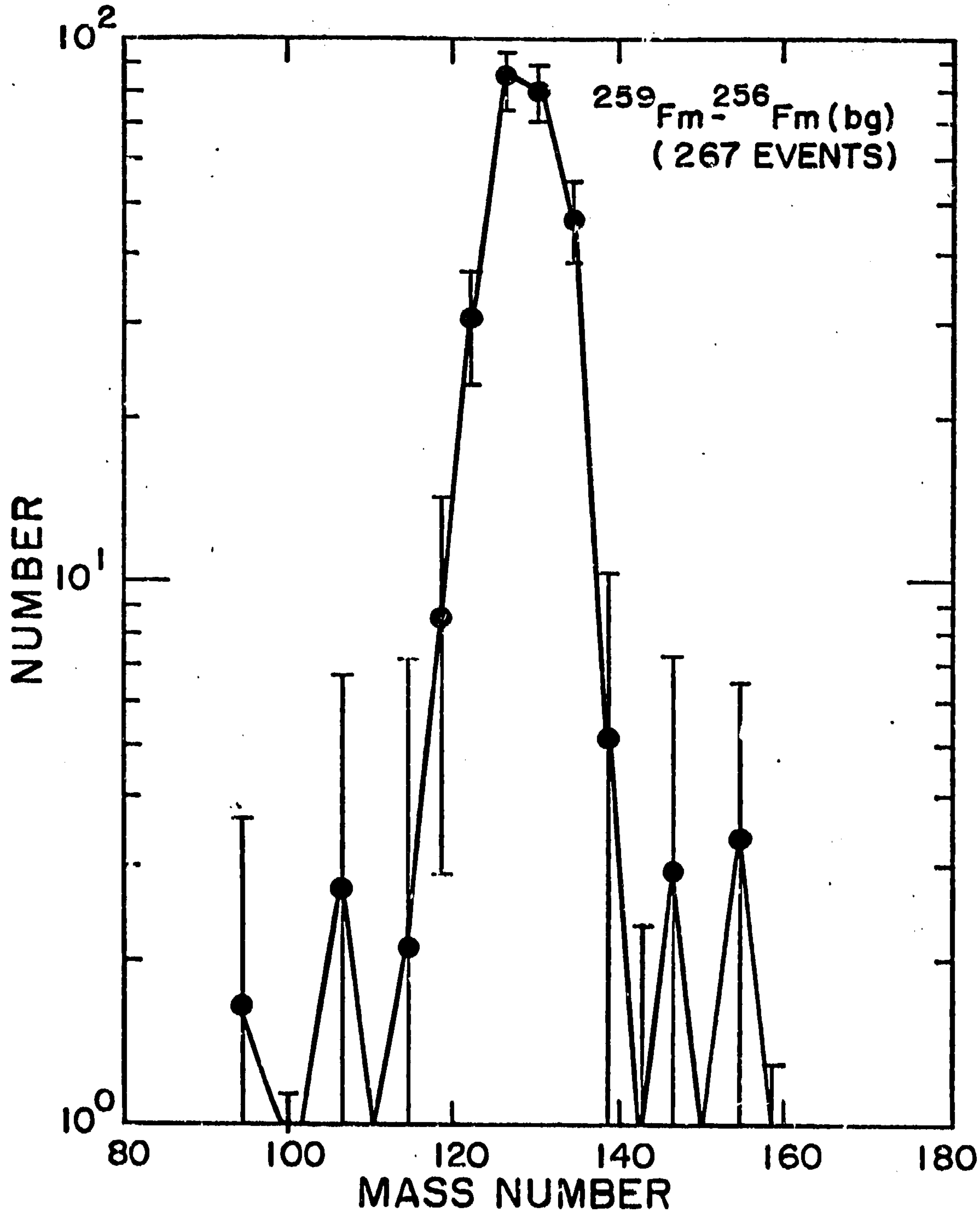


(a)
GROSS DECAY
259Fm Resolved by LMS
t1/2 = 1.3±0.3 sec
(b) Coincident fissions ≥230 MeV
t1/2 = 1.5±0.2 sec
Coincident fissions
Time (sec)
200
100
10
1
1.0
0.1
0
2
4
6
8
10
12

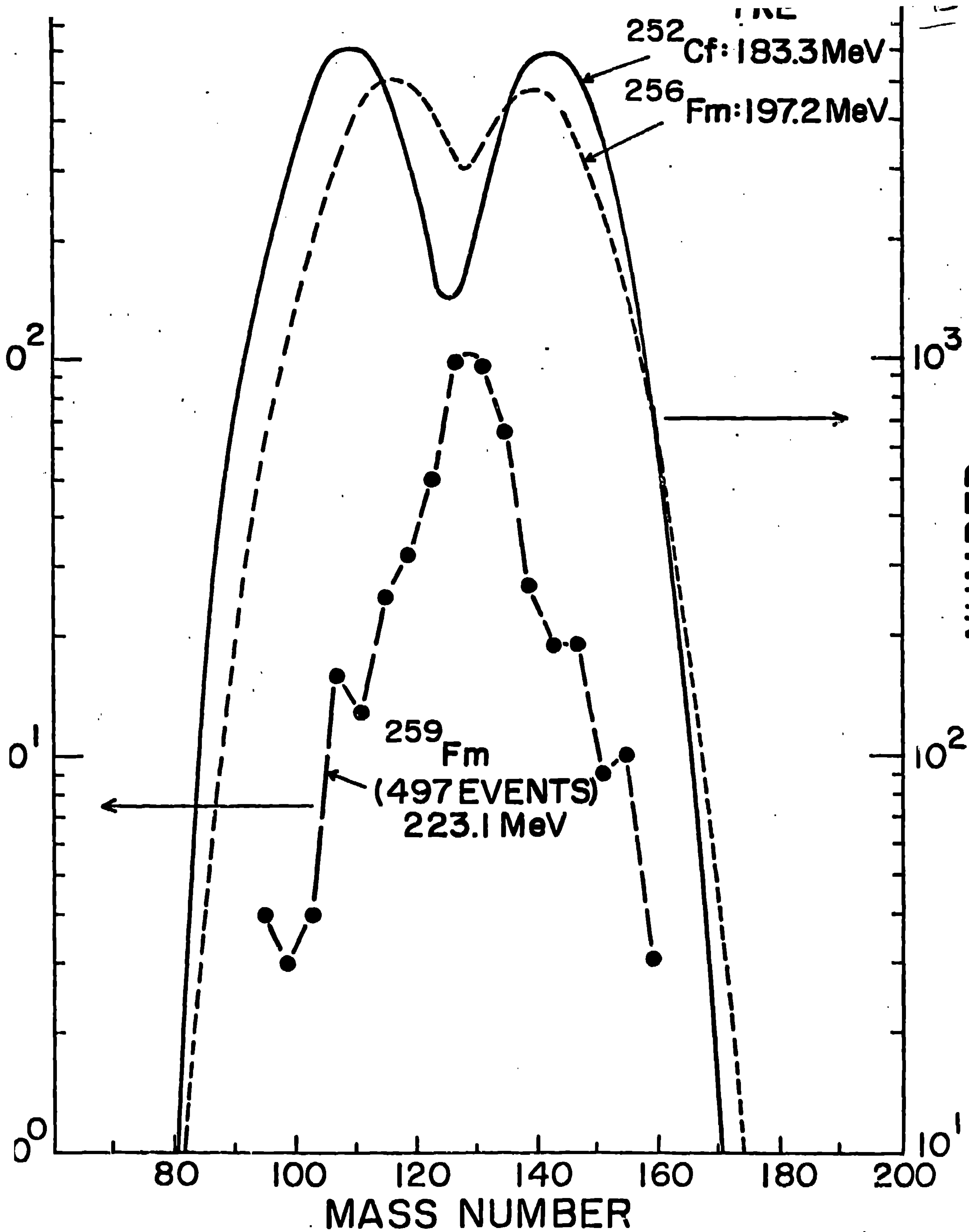


35
30
25
20
15
10
5
0
NUMBER
140
160
180
200
220
240
260
280
300
TOTAL KINETIC ENERGY (MeV)
$\overline{TKE}$ = 222.7 ± 1.3 MeV
(497 EVENTS)

Fig. 4



$^{259}Fm - ^{256}Fm$ (bg)
(267 EVENTS)
NUMBER
10^2
10^1
10^0
80
100
120
140
160
180
MASS NUMBER



252Cf: 183.3 MeV
256Fm: 197.2 MeV
259Fm
(497 EVENTS)
223.1 MeV
80
100
120
140
160
180
200
MASS NUMBER

TABLE I.

VARIATION IN AVERAGE TOTAL KINETIC ENERGY WITH TIME.

Time Interval (seconds)	Average Total Kinetic Energy[a] (MeV)	Fission Events	σ[b]
0-2	229.7 ± 1.8	246	27.9
2-5	221.1 ± 2.6	142	30.9
5-8	212.5 ± 3.0	64	24.0
8-11	204.6 ± 4.5	32	25.5
11-14	218.1 ± 5.4	13	19.3
0-14	222.7 ± 1.3	497	29.0

[a]The quoted error is the standard deviation for each average total kinetic energy.

[b]σ is the standard deviation of the population, i.e., the square root of the second central moment or variance.