Detection of Long-Range Fragments from Decay of Cf^{252}†

JOSEPH B. NATOWITZ,* ARASTOO KHODAI-JOOPARI,‡ AND JOHN M. ALEXANDER§
Department of Chemistry, State University of New York at Stony Brook, Stony Brook, New York

AND

T. DARRAH THOMAS§
Department of Chemistry, Princeton University, Princeton, New Jersey
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Long-range products from the decay of Cf^{252} have been observed by range discrimination and detection of tracks in mica and lexan. Integrated yields (events per binary fission) of $\gtrsim 18\times10^{-6}$ and kinetic energies of 1.5–4 MeV/amu are indicated for fragments of B, C, N, and O. Integrated yields of $\gtrsim 3\times10^{-6}$ and kinetic energies of 1.5–2.5 MeV/amu are indicated for fragments of oxygen or heavier nuclei.

I. INTRODUCTION

EXPERIMENTS with both photographic emulsions and semiconductor detectors have established the existence of light fragments ejected in low abundance during the spontaneous fission decay of Cf^{252}.[1] In addition to H and He isotopes, Cosper, Cerny, and Gatti[2] observed Li ions with an abundance of $\approx 4\times10^{-6}$ per binary fission,[3] and Be ions with an abundance of $\approx 5\times10^{-6}$. No "significant intensity" of B or C ions was observed, but they could have been detected only for energies ≥ 33 and ≥ 43 MeV, respectively. Further counter experiments by Raisbeck and Thomas have given evidence for some production of B and C.[4] Fragments of ≈ 50 amu with abundance of $\geq 1\times10^{-6}$ have been observed in the ternary-fission experiments of Muga.[5]

The kinetic energies of these low-yield fragments are such that their ranges in matter are greater than the ranges of normal binary-fission fragments. Thus it is possible to employ range-discrimination techniques to separate the light fragments from the ordinary fission fragments. In the experiments reported here, we have used H_2 gas as an absorber to stop the ordinary fission fragments; then we have detected long-range products by observing tracks in lexan and diamond-shaped pits in mica.[6] As employed here, the lexan registers fragments of $Z\geq 5$ (B^{11}, C^{12}, etc.) and the mica registers fragments of $Z\geq 9$ (F^{19}, Ne^{20}, etc.). Range measurements and detector thresholds delineate a region in energy-mass space (described later in Fig. 5) for the fragments observed.

II. EXPERIMENTAL METHOD

We have performed a series of integral-range experiments in which mica and lexan track detectors were employed to observe the yield (number of events per binary fission) of fragments emitted from Cf^{252} as a function of range in H_2. These experiments were carried out in the chamber depicted in Fig. 1. Fragments emitted from a Cf^{252} source of $\approx 1.5\times10^8$ fissions per min passed through a Ni source cover of 0.52 mg/cm², a Mylar isolation window of 1.5 mg/cm² (consisting of three 0.5-mg/cm² Mylar disks), and H_2 gas at pressures of 0.75–2.0 atm. At the end of their paths the fragments impinged on 1.3×9-cm strips of mica and lexan. The angle of incidence was always $\geq 15°$ and

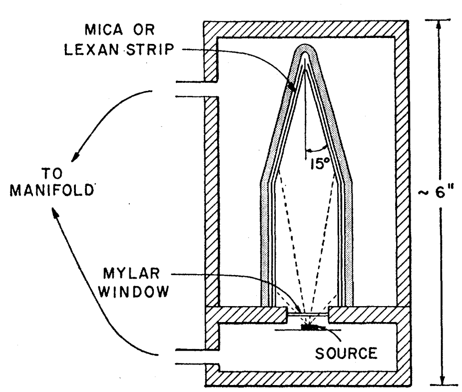

FIG. 1. Schematic diagram of the detection chamber. The Cf^{252} source was covered by a Ni foil of 0.52 mg/cm². Fragments impinged on the track detectors at angles of 40°–15°. The first 7 mm of the track detectors were shadowed from the source by the window mount.

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* Present address: Department of Chemistry, Texas A & M University, College Station, Tex.
‡ Present address: Chemistry Department, Pahlavi University, Shiraz, Iran.
§ A. P. Sloan Fellow.
[1] E. K. Hyde, *The Nuclear Properties of the Heavy Elements* (Prenctice-Hall, Inc., Englewood Cliffs, N. J., 1964), Vol. III; S. L. Whetstone, Jr., and T. D. Thomas, Phys. Rev. Letters **7**, 298 (1965); Phys. Rev. **154**, 1174 (1967). See these papers for other references.
[2] S. W. Cosper, J. Cerny, and R. G. Gatti, Phys. Rev. **154**, 1193 (1967).
[3] In this paper we express all yields or abundances of light fragments as events per binary fission.
[4] G. M. Raisbeck and T. D. Thomas, 153rd Meeting of the American Chemical Society, Miami Beach, Fla., 1967 (unpublished). [*Note added in proof*. These workers now report yields of 0.7×10^{-6} (>25 MeV) and 1.4×10^{-6} (>33 MeV) for B and C, respectively. The apparent discrepancy is probably only a reflection of different detection thresholds.]
[5] M. L. Muga, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965* (International Atomic Energy Agency, Vienna, 1965), Vol. II, Paper SM-60/73, p. 409.
[6] R. L. Fleischer, P. B. Price, and R. M. Walker, Ann. Rev. Nucl. Sci. **15**, 1 (1965).

$\leq 40°$. Behind the primary detector strips, additional strips were placed to serve as blanks.

The mica strips were annealed at 600°C for 2 h and then pre-etched in 48% HF at ≈ 20°C for 6 h, prior to being used in the experiments. The pre-etching caused tracks from fission of U impurities to be developed into very large and easily identified diamonds. The lexan strips were not pretreated. Following irradiations of suitable duration, usually about 10 days, the mica strips were etched (usually for 2 h, see Sec. III) in 48% HF at room temperature. The lexan strips were etched (usually for 45 min, see Sec. III) in 6.2N NaOH at 63°C to develop tracks which would be observable with optical microscopes. A trace of wetting agent (Dow Benax) was added to the NaOH etching solution. As most of the kinetic energy was lost before the fragments reached the detector, it was found necessary to use these long etching times and also to employ phase-contrast microscopes especially for scanning of mica. With the etching conditions we observe in the mica shallow diamonds of varying size ($\leq 6\ \mu$ across). The depth of these diamond-shaped pits was observed to be less than $2\ \mu$ after a 37-min etch, and they were too faint for quantitative counting after a 15-min etch. In lexan, we observed conical tracks of varying length up to $11\ \mu$. Only tracks of length $\geq 2\ \mu$ were counted because low-energy α particles may give rise to small ($\approx 1\ \mu$) spots in the lexan.[6]

The registration properties of lexan and mica were studied by exposures to beams from the Heavy-Ion Linear Accelerator at Yale University. Beams of Li^7, B^{11}, C^{12}, N^{14}, O^{16}, F^{19} and Ne^{20} of 1 MeV/amu were employed. Aluminum degrader foils were sometimes used to lower the beam energy to about 0.5 or 0.1 MeV/amu. About 10^8–10^9 projectiles (measured by integral charge collection) impinged on the detector plates at an angle of incidence of 30°. The plates were then etched for various times, the nature of the tracks was noted, and the efficiencies were estimated.

An attempt was made to detect the long-range fission products by measurement of the energy spectrum of products which penetrate 18.6-cm H_2 (1 atm, 15°C). The chamber shown in Fig. 1 was used with a semiconductor detector mounted 9 cm from the source. It was not possible to identify the long-range fragments in these experiments, but it was possible to set upper limits on yield versus fragment energy. These limits are used for B, C, and N in Fig. 5, which will be discussed later.

III. RESULTS AND DISCUSSION

A. Registration Properties

In lexan which had been exposed to a beam of heavy ions, we typically observed a dense region of small conical tracks with a small admixture ($\sim 1/10^4$–10^6) of larger, more distinct cones. The small cones correspond to primary registration of the projectiles and the more distinct cones may be beam impurities or recoil atoms from reactions of the beam projectiles with atoms in the detector or degrader foils. In mica, the primary registration gave rise to shallow diamonds with a small admixture of distinct tracks, intepreted as recoil tracks. The sizes and shapes of the primary tracks, or diamonds, were observed in regions of low beam density at the edge of the major beam spot. Efficiencies of registration were estimated by counting recoil tracks in regions of high beam density and determining the ratio of primary tracks to recoil tracks in regions of low beam density. This method is considered to have a precision of about 50%. More precise efficiency studies are in progress, and photographs of many kinds of tracks and diamonds will be published in the near future.[7]

For mica we have found that F^{19} and Ne^{20} ions register as bright diamonds with 100% efficiency for etching times of 1 h or greater. Ions of O^{16}, N^{14}, C^{12}, and B^{11} of ≈ 0.1 MeV/amu register as diamonds of lesser brilliance (less depth) with efficiencies of $\approx 50\%$, $\approx 25\%$, $\approx 10\%$, and $\approx 1\%$, respectively. The diamonds from B^{11} and C^{12} are so faint after a 2-h etch that it is very doubtful that they would have been counted in the fission studies. Also, the efficiencies of these ions (B^{11},O^{16}) were 5–100 times lower at 1 MeV/amu than at 0.1 MeV/amu.

In a separate series of experiments,[7] we have found that Al^{27} and S^{32} beams of energy 0.2–0.6 MeV/amu leave 1–4-μ tracks in mica. These tracks assume diamond shapes after etching for 60 min. They are readily distinguishable from the very shallow diamonds previously mentioned, especially for etching times of 10–30 min. Our interpretation of the diamond shapes is that they correspond to a cross-sectional view of a track defined by the intersection of the etched region with the mica surface. The width of this diamond increases with etching time, but the depth of the track does not. Ions of $Z \leq 10$ do not leave tracks which we can etch out to their full length. They do, however, damage the mica sufficiently to give an observable diamond of depth less than $2\ \mu$. Ions of Al^{27} and S^{32} are capable of leaving tracks which can be etched to depths greater than $2\ \mu$.[7] Thus, we would have distinguished ions of $Z \geq 13$ [provided that they have enough energy (≈ 0.3 MeV/amu) to leave an etchable region of $> 2\ \mu$ in the mica] from ions of $Z = 8$–10. Ions of $Z \geq 13$ having energy less than ≈ 0.3 MeV would register as diamonds which would not have been distinguished from ions of $9 \leq Z \leq 12$.

From these registration data, we conclude that, in mica, fragments of $Z \geq 9$ (F, Ne, etc.) have been counted with high efficiency in the fission study. Oxygen fragments have possibly been counted, but with low efficiency ($< 50\%$), and fragments of $Z \leq 7$ probably make a negligible contribution to the yields recorded in mica.

[7] J. Mahony, J. M. Alexander, and A. Khodai-Joopari (unpublished data).

TABLE I. Yield[a] of tracks in lexan for various etching times.

Etching time, min	Degrader, cm H_2, and track length, μ: 12 cm H_2, $\geq 2\,\mu$	12 cm H_2, $\geq 4\,\mu$	12 cm H_2, $\geq 7\,\mu$	14.2 cm H_2, $\geq 2\,\mu$	14.2 cm H_2, $\geq 4\,\mu$	14.2 cm H_2, $\geq 7\,\mu$	16.6 cm H_2, $\geq 2\,\mu$	16.6 cm H_2, $\geq 4\,\mu$	16.6 cm H_2, $\geq 7\,\mu$	17.9 cm H_2, $\geq 2\,\mu$
40	12	1.8	0.2	8.5	1.4	0	2.2	0.27	0	1.1
80	15	5.6	1.8	12.5	4.5	0.2	4.3	0.65	0	3.6
120	17	7.8	2.5	21	11	1.5	7.5	2.0	0.3	8.5

[a] All yields are multiplied by 10^6.

The track lengths observed in lexan for ions of B, C, N, O, and F are shown in Fig. 2. We have found that ions of C^{12} and heavier register as cones with essentially 100% efficiency. No tracks were observed for Li^7. Each of the ions is expected to have a range of ≈ 11 and $\approx 5\,\mu$ for 1 and 0.5 MeV/amu, respectively. Apparently the complete range is being developed by a 45-min etch into a visible track for C^{12}, N^{14}, O^{16}, and F^{19} projectiles of 0.5 MeV/amu. However, for O^{16} and F^{19}, only after a 75-min etch is an 11-μ track developed for projectiles of 1 MeV/amu. For S^{32} of 1 MeV/amu an 11-μ track is completely developed by 45 min of etching.[7] Apparently the etching rate for the tracks decreases with increasing incident energy between 0.5 and 1.0 MeV/amu. Also, there is a marked drop in development of track length for B^{11} compared to C^{12} of 0.5 MeV/amu. By 45 min of etching the tracks from 0.5-MeV C^{12} are developed to $5\,\mu$, but tracks from 1.0-MeV C^{12} are only developed to $1\,\mu$. Only a few of the B tracks (0.5 or 1 MeV/amu) are developed to $2\,\mu$ or greater by 45 min of etching.

In Sec. B we describe fission experiments in which tracks in lexan of length $\geq 2\,\mu$, $\geq 4\,\mu$, and $\geq 7\,\mu$ were recorded as a function of etching time. From Fig. 2 we would conclude that ions of C and heavier of 0.5 MeV/amu would be developed to their full range by a 45-min etch. For a 45-min etch only part of the boron tracks would be counted as 2-μ tracks and by extrapolation Be probably would not be counted. An increase in the etching time from 45 to 105 min would develop most B tracks to a length greater than $2\,\mu$, but less than $4\,\mu$. Conversely F and Ne tracks would not develop appreciably in length from 45 to 105 min of etching. Tracks from C, N, and O would develop appreciably in length if their energy was ≈ 1.0 MeV/amu, but for energies of ≤ 0.5 MeV/amu, 45 min of etching would develop the entire track. Throughout this paper we assume that isotopes of the same element have similar registration properties.

B. Fission Studies

In Fig. 3 the yield of events recorded in mica during the fission experiment is presented as a function of range (in H_2 gas at 1 atm and 15°C). The yields of diamonds in mica remained constant as the etch times were increased from 2 to 4 h. In the lexan, however, increased etching time from 40 to 120 min resulted in large increases in the measured yields, as is indicated in Table I to be discussed later. Notwithstanding, most of the lexan scanning was for 45-min etches because longer etches tend to damage the lexan surface and make the counting more difficult. Therefore we present data in Fig. 4 for a 45-min etching of lexan. The yields in this figure should be regarded as lower limits, which nevertheless represent the basic features of the yield distribution.

In constructing Figs. 3 and 4 and Table I the ranges in Ni and Mylar have been converted to ranges in H_2 by using the following factors[8]:

$$R_{H_2}/R_{Mylar} = 3.7\ \text{cm}/(\text{mg/cm}^2)$$

and

$$R_{H_2}/R_{Ni} = 2.0\ \text{cm}/(\text{mg/cm}^2)\,.$$

These conversion factors have been taken from range data for α particles for a velocity corresponding to 2 MeV/amu. Use of the factors for 1 or 5 MeV/amu would change the calculated ranges by only ± 0.6 cm H_2.

As is noted in these figures, the yield decreases rapidly with range in H_2 up to ≈ 13 cm in H_2. Beyond that point, which occurs at a yield of $\approx 3\times 10^{-6}$ in mica and $\approx 2\times 10^{-5}$ in lexan, the yield falls off much more gradually with range, reaching a yield of $\approx 1.5\times 10^{-7}$ in both detectors at ≈ 19 cm H_2. This behavior strongly suggests the existence of long-range fragments emitted in low

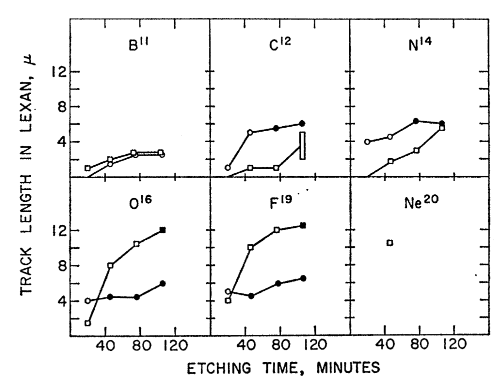

FIG. 2. Projected track length in lexan as a function of etching time in 6.2*N* NaOH at 63°C. Circles are for beams of 0.5 MeV/amu; squares for 1.0 MeV/amu. Open symbols indicate pointed cones; closed symbols indicate tracks with "rounded" ends. (This distinction cannot be made for B^{11} ions.) The incident angle was 30°.

[8] W. Whaling, in *Handbuch der Physik*, edited by S. Flügge, (Springer-Verlag, Berlin, 1958), Vol. 34, p. 193.

TABLE II. Possible sources of interference.

Possible source of spurious events	Reason for rejection
1. Neutron activation of U impurity in:	
(a) the Mylar window	Observed yields in mica would require impossibly large content of U in the window. Front-end shadowing rules out window as source of events.
(b) the track detectors	Blank tests and the reproducibility of the yield versus range curve at five different pressures rule this out.
(c) the chamber walls	Precluded by the reproducibility of the results at five different pressures.
2. Cf^{252} contamination inside the chamber	No detectable α radioactivity inside the chamber for the reported experiments. Source cover, window, and chamber were changed several times. Reproducibility of experiments.
3. Recoil of light atoms from the Ni or Mylar as a result of collision with the fission fragments.	Test for scattering from Mylar window. Kinematics rule out Ni. Ratio of number of lexan tracks to mica tracks.
4. Registration of decay α's or of <40 MeV fission α's either directly or by production of heavy recoils	Registration tests employing He^4 beams from the Yale Hilac and natural α source show that the registration efficiencies are too low to explain the observed yields.
5. Holes in Mylar window	The reproducibility as well as the absolute yields and shape of the yield versus range curve rule this out.
6. Existence of etchable tracks in the detectors prior to the experiment	Blank tests and reproducibility among the experiments preclude this as a source of the long-range events.

yield from the Cf^{252}, which can be observed beyond the edge of the integral range distribution of normal binary events. Naturally, such an interpretation is valid only if the various phenomena which might lead to similar tracks or diamonds can be ruled out.

The possible sources of these spurious events which we have considered appear on the left in Table II. Opposite each of those entries are indicated the observations which argue against those phenomena as being responsible for the long-range events. Some of these observations are discussed more thoroughly in the Appendix.

In summary, the data available to us are as follows: (a) Registration characteristics and thresholds for mica and lexan (see Sec. III A). (b) Integral range curves (Figs. 3 and 4). (c) Yield versus etching time for the lexan (Table I). (d) Maximum residual energies after penetrating 18.6 cm H_2. These data can be used to construct a picture of the energy and mass distributions of the long-range fragments. Figure 5 summarizes the results of such an analysis.

In Fig. 5, vertical lines have been drawn at the Z values corresponding to the detector thresholds, namely $Z=9$ for mica (with some possibility of detection of $Z=8$) and $Z=5$ for lexan, with some possibility of incomplete registration of boron. (Only tracks of $\geq 2\,\mu$ were counted.) The lowest energies which we could detect correspond to registration of a 2-μ track in mica or lexan after penetration of 13 cm H_2 (total range of 15 cm H_2 or equivalent to 4.8 mg/cm^2 Al). The corresponding energies (for products of equal neutron and proton numbers) have been taken from the range-energy curves of Northcliffe.[9] The high-energy limit for mica detection ($9\leq Z\leq 12$) is fixed by the fact that less than 10^{-7} fragments per fission penetrate more than 19.6 cm H_2 and then register a 2-μ diamond. (For O^{16} this range is 20.5 cm H_2, because the efficiency is $\lesssim 50\%$ for 0.1-MeV O^{16} ions.) The high-energy cutoff for mica ($Z\geq 13$) is fixed by the fact that no "tracks" are observed in the region of 14 cm H_2. Mica scanned after a 15-min etch revealed no tracks; and after a 37-min etch revealed only flat diamonds of $\leq$2-μ length. Low-energy fragments of $Z\geq 13$ may register as diamonds in the region of 14 cm H_2, but the absence of "tracks" eliminates the possibility of high-Z fragments (13, 14, 15, etc.) of 19 cm range in H_2.[7] This gives rise to the high-energy cutoff shown for $Z\geq 13$. The high-energy boundary for detection in lexan is not clear, because track-length development is not complete for B, C, and N of ≈ 1 MeV/amu. We turn to the data from semiconductor detectors to set an upper limit on the energies of B, C, and N fragments of yield $\leq 10^{-7}$. We have set an upper limit of 10^{-7} for the yield of fragments

[9] L. C. Northcliffe, Ann. Rev. Nucl. Sci. **13**, 67 (1963).

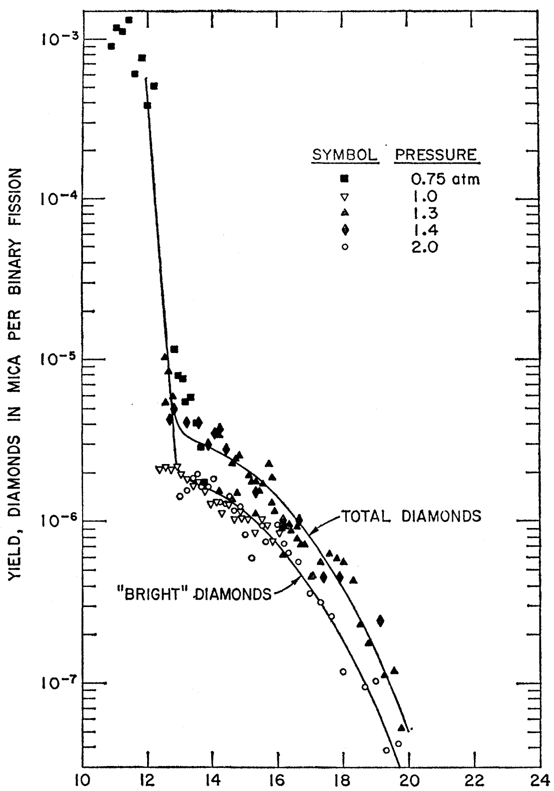

FIG. 3. Yield of fragments versus range in H_2, as detected in mica. Various symbols are for different exposures with the H_2 pressure as indicated. Open symbols correspond to "bright diamonds"; closed symbols to total diamonds. The thickness traversed in Ni and Mylar has been converted to cm of H_2 as described in the text. The break in the curves at 13 cm of H_2 indicates a group of long-range products. These are integral range curves.

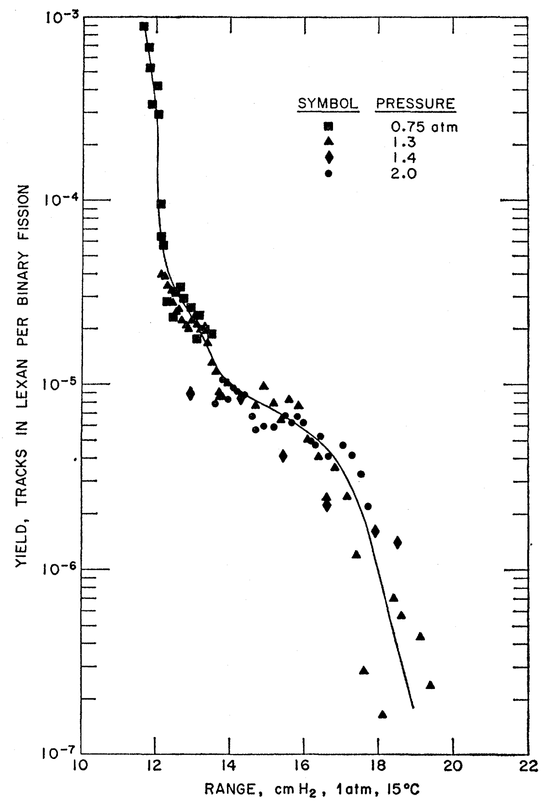

FIG. 4. Yield of fragments versus range in H_2, as detected in lexan (track length $\geq 2\mu$, 45-min etch; see text). Various symbols are for different exposures with the H_2 pressure as indicated. This is an integral range curve.

with energy greater than 25 MeV after penetrating 18.6 cm H_2. This corresponds to eneriges 40, 47, and 54 MeV for B, C, and N, respectively. The yield shown in Fig. 5 for the region of $Z \gtrsim 9$ is that observed in mica, namely 3×10^{-6}. The yield for $5 < Z \lesssim 8$ is obtained by subtracting the yield observed in mica from that observed in lexan, namely $21 \times 10^{-6} - 3 \times 10^{-6}$ or 18×10^{-6}. These assignments depend only on the detector thresholds.

By reference to Table I and Fig. 2 we can attempt a further breakdown of the yields observed in lexan. This further breakdown depends on the track-length observations in lexan versus etching time. These assignments make use of a more delicate feature of track registration. In Table I, we list yields of tracks of length $\geq 2\,\mu$, $\geq 4\,\mu$, and $\geq 7\,\mu$ as a function of degrader thickness and etching time. B^{11} ions of 0.5–1 MeV/amu give tracks which may grow over the 2-μ borderline, while C^{12}, N^{14}, and possibly O^{16} ions of ≈ 1 MeV/amu may grow from $< 4\,\mu$ to 4–7 μ. Tracks from O^{16} may develop across the 4- or 7-μ borderline; tracks from F^{19} and heavier ions are not expected to increase in length across either the 2- or 4-μ borders as etching times increase from 45–105 min. They may, however, grow over the 7-μ borderline. Therefore, we can assign to species of oxygen or heavier, those tracks of $\geq 7\,\mu$, namely

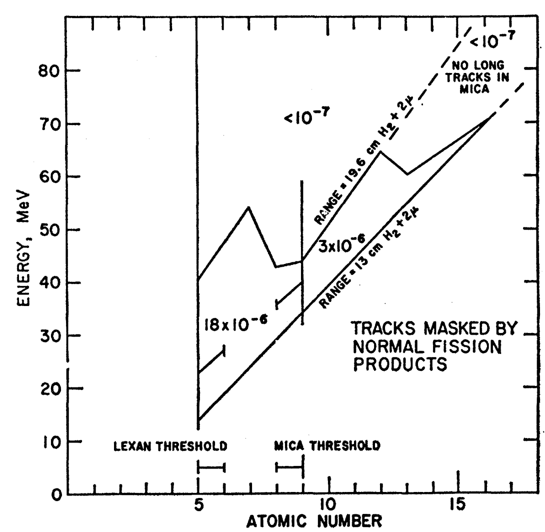

FIG. 5. Energy and atomic number limits for the long-range fragments as determined by ranges and registration properties. Yields are indicated for the energy-atomic number regions accessible in this study (see text).

2.5×10^{-6}. Also we can assign to C, N, and O those tracks which grow with etching time to 4–7 μ, namely 4–8×10^{-6}. Similarly we can assign mainly to B (with possible contribution from C of ≈1 MeV/amu) those tracks which grow from $<2\,\mu$ to 2–4 μ, namely 3–7.4×10^{-6}. Of the very-long-range events, observed at 17.9 cm H_2, 7.4×10^{-6} of the total 8.5×10^{-6} appear to be from boron. Of the events observed at 12–14.2 cm or just beyond the ordinary products, 4–8×10^{-6} of the total $\approx21\times10^{-6}$ appear to be from carbon, nitrogen, or oxygen, and 2.5×10^{-6} appear to be from O or heavier species. This analysis of track length in lexan versus etching time should be considered to be preliminary because of statistical limitations and the present lack of knowledge of track registration at energies other than 0.5 and 1 MeV/amu. Nevertheless, elemental yields of the order of a few in a million are indicated for fragments of B, C, N, and O. The mica detection does demand an integrated yield of $\gtrsim3\times10^{-6}$ for oxygen or heavier species, as mentioned before, which is consistent with this analysis of lexan detection. The absence of mica "tracks" excludes yields of $>10^{-7}$ for Al or heavier species with ranges of ≈19 cm H_2.

The over-all picture of light-fragment production from Cf^{252} decay is as follows: (a) Cosper *et al.*[2] report Li and Be yields of $\approx5\times10^{-6}$. (b) We report B, C, N, and possibly O with integrated yield of $\approx18\times10^{-6}$ probably divided rather evenly between B, C, and N. Raisbeck and Thomas also report observation of B and C.[4] (c) We report an integrated yield of $\gtrsim3\times10^{-6}$ for species of $8<Z<13$. (d) Muga[5] reports a yield of $>1\times10^{-6}$ for species of $Z\approx25$. These fragments would not be distinguishable in our work from the products cited in (b) and (c), because their ranges would be rather short (≈14 cm H_2).

It is interesting that Muga reports an additional group of fragments from U^{236} fission with mass about 32 and energy about 90 MeV. We have looked for this type of fragment in Cf^{252} decay, and we can set an upper limit of 10^{-7} for the yield. This finding is consistent with Muga's study of Cf^{252}. Currently we are studying U^{236} fission with the same techniques.

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APPENDIX

A. Front-End Shadowing

As is indicated in Fig. 1, the geometry of the track-detector experiments is such that fragments originating in the source or source cover are collimated so as to strike the track-detector strips ≥7 mm from the front edge. Fragments originating at the Mylar window could register at a position ≥3 mm from the front edge. Scans of the mica detectors at 5 mm from the front edge allow us to set a limit of 3×10^{-8} on the yield of events with their origin at the Mylar window. From this datum, we conclude that neither activation of U impurity in the Mylar window nor Cf^{252} contamination on the inside of the Mylar window could result in the distribution observed in mica at long ranges.

Although the mica exhibited a distinct front-end shadow as just described, the lexan revealed many short tracks (1–3 μ) in the shadowed region. These events probably result from C^{12} and O^{16} ions ejected from the window. The total number of these events corresponds to a yield of 3×10^{-5} scattered atoms per fission. This yield is comparable to the yield of long-range events shown in Fig. 4, but most of the scattered C^{12} and O^{16} atoms are expected to have low energies. Thus we expect that the number of tracks from scattered C^{12} and O^{16} will decrease with range in H_2 much more rapidly than shown by the data in Fig. 4. We have performed another kind of shadowing test for scattering to confirm this expectation. This test is described in Sec. C below.

B. Blank Tests

In each of the experiments, both the back surfaces of the primary track detectors and the second detectors placed behind the primary detectors served as blanks for comparison with the primary detector.

The "tracks" in lexan and the diamonds in mica from these blanks were randomly distributed and the number of events did not differ significantly from those of etched blanks which had not been used in any irradiations. This random distribution of marks can account for $\approx2\%$ of the events corresponding to a range of 14 cm H_2 and $\approx15\%$ of the events corresponding to a range of 18 cm H_2.

C. Scattering

We have considered the possibility that "knock-on" C, O, or Ni atoms (atoms ejected from the Ni source cover or Mylar window by a collision with the heavy fission fragments) could explain the long-range events which are observed. The maximum energy that a Ni atom can obtain from a 105-MeV fragment of 100 amu is 98 MeV, which corresponds to an average range of 12.5 cm H_2. This maximum range cannot be reconciled with the shape of the curve in Figs. 3 or 4.

The possibility of scattering from the window has been explored in a separate experiment in which a lexan strip of width $\frac{1}{16}$ in. and thickness 3 mg/cm^2 was placed on the top of the Mylar window. This strip passed through the center of the hole, directly over the Cf source. Under the ideal conditions of a point source this would have produced a shadowed region into which fragments emitted from the source could not pass. If, however, atoms of C or O scattered from the window were responsible for the observed long-range events, the yield of such events should not be reduced by more than 15% by the inclusion of the $\frac{1}{16}$-in. strip. Atoms scattered from the window would, of course, not be restricted to head-on collisions. In this experiment, the yields in the shadowed region of both the mica and the lexan were reduced by a factor of 6 lower than shown in Figs. 3 and 4. The appearance of some events in the shadowed region can be accounted for by the fact that the source is not a point source, but has extended itself into a somewhat larger area than the original deposition area of 0.1 cm^2.

The geometry of the scattering test does not allow us to exclude the possibility that the long-range events result from scattered O atoms originating in the source or source cover. However, the test excluded scattering of oxygen from the Mylar window which contains a much greater number of O atoms than the source or source cover. Thus it seems unlikely that scattered oxygen originating near the source can be responsible for the long-range events.

D. α-Registration Tests

We have performed several experiments to determine the registration efficiency of fission α particles (≤ 42 MeV, yield 3×10^{-3}) and α particles from radioactive decay (≤ 6 MeV, yield 3×10^{1}). For energies ≤ 6 MeV, natural α sources were employed. The Yale Hilac supplied He^4 beams for the studies at higher energies. The results of these registration experiments are summarized in Table III.

TABLE III. Effect of He ion registration on observed yields.

Detector	α energy (MeV)	Registration efficiency	Maximum contribution to yield in Cf^{252} experiment
Mica	42	$\leq 4\times10^{-6}$	$\leq 1.2\times10^{-8}$
	3	$\leq 4\times10^{-10}$	$\leq 1.2\times10^{-8}$
Lexan	42	$\leq 1.3\times10^{-5}$	$\leq 3.9\times10^{-8}$
	3	$\leq 8.3\times10^{-9}$	$\leq 2.5\times10^{-7}$

E. Hole Tests

The reproducibility of different experiments rules out the possibility that one or several small holes in the Mylar window could explain the abundance of observed long-range fragments. Such holes would be expected to lead to "hot spots" on the detectors. It might be considered that the Mylar is a mesh of small holes leading to a high transmission of fission fragments. In this respect, we note than an over-all yield of $\sim 10^{-6}$ as observed in the mica requires a transmission of $>10^{-2}$ through each of the three components of the Mylar window. However, since 0.5 mg/cm^2 of Mylar is equivalent to ≈ 2 cm of H_2, the random superposition of three such disks would lead to a yield curve quite different from that in Fig. 3. Finally, it should be pointed out that if all spurious effects other than holes are ruled out by the various tests and observations recorded here, then the difference in yields between mica and lexan rules out the possibility that the tracks in lexan could be the result of transmission of ordinary products through holes in the window.