Brief Reports

Brief Reports are short papers which report on completed research or are addenda to papers previously published in the **Physical Review**. *A Brief Report may be no longer than 3½ printed pages and must be accompanied by an abstract.*

Search for ^{34}Si ions in ^{241}Am decay

M. Paul,* I. Ahmad, and W. Kutschera
Argonne National Laboratory, Argonne, Illinois 60439
(Received 12 May 1986)

An experiment has been performed to measure the spontaneous fission branch of ^{241}Am and to search for decay events by emission of ^{34}Si nuclei. Lexan polycarbonate foils were used for the detection of decay fragments. Tracks produced in the foils by 81 MeV ^{28}Si ions from an accelerator and fission fragments from a ^{248}Cm source were compared with those produced by particles emitted from a 0.5 mCi ^{241}Am source. We identified 33 fission fragments corresponding to a fission/alpha branching ratio of $(2.4\pm0.5)\times10^{-12}$, in fair agreement with previous measurements. No ^{34}Si tracks were observed which results in an upper limit of 4.2×10^{-13} for the ^{34}Si/alpha ratio at the 90% confidence level. Two theoretical calculations predict values of 4.0×10^{-13} and 1.1×10^{-15} for this branching.

It is now well established that certain heavy nuclides have a very weak decay branch in which nuclei emit particles heavier than alpha particles but lighter than fission fragments. The most thoroughly studied case[1–5] of such a radioactivity is that of ^{223}Ra, emitting ^{14}C nuclei with a ^{14}C/alpha branching ratio of 5.9×10^{-10}. Since the discovery of this new form of radioactivity by Rose and Jones,[1] several other cases of cluster decay[4,6–8] have been observed. These decays are limited, at present, to the emission of ^{14}C and ^{24}Ne nuclei. It was pointed out by Barwick *et al.*[6] that there are several weak spontaneous fission branches known in heavy elements whose strength is comparable to expected cluster decay branches. Some of these fission measurements including the one on ^{241}Am did not discriminate between the fission fragment and cluster particle and therefore it is possible that the observed events belonged to cluster decay.

Different techniques have been utilized for the identification of the emitted particles. Rose and Jones[1] and Alexandrov *et al.*[3] have used telescopes of surface barrier detectors to identify the emitted particles by energy loss (ΔE) and total energy (E) measurements. A more elegant method, used by Gales *et al.*[2] and Kutschera *et al.*,[5] utilizes a magnetic device to separate the heavy particle from the large amount of accompanying α particles. The emitted fragments are then identified by magnetic analysis and ΔE-E measurements. However, these devices have limited solid angles and hence cannot be effectively used for very small branching ratios. Much larger solid angles (as large as 2π) can be achieved by using solid state track detectors.[9] Furthermore, these detectors are insensitive to α particles. Several cases of cluster decays[4,6–8] have been studied using this technique.

Simple barrier penetration calculations indicate that the rate of decay by cluster emission depends very strongly on the decay energy. This was the criterion used by Rose and Jones to select ^{223}Ra for their investigation.[1] Such calculations indicate that, among transplutonium elements, the emission of ^{34}Si ions by ^{241}Am (Q value $=93.84$ MeV) is a favorable case. Theoretical calculations, based on a model of highly asymmetric fission, give ^{34}Si/alpha branching ratios of 4.0×10^{-13} (Ref. 10) and 1.1×10^{-15} (Ref. 11). On the other hand, spontaneous fission branching in the range of 10^{-12} has been observed[12,13] in ^{241}Am. Since in these experiments no attempt was made to distinguish between fission and ^{34}Si events, it was interesting to search specifically for the cluster decay. While this work was in progress, the results by Hourani *et al.*[14] were published. They used a magnetic solenoid spectrometer to search for ^{34}Si ions emitted by a 7 mCi ^{241}Am source. They did not observe any ^{34}Si particle and obtained an upper limit of 3×10^{-12} for the ^{34}Si/alpha branching ratio. Their experiment was not designed for the detection of fission fragments. In the present work we have made a definite identification of the fission fragments and obtain a fission branching ratio in fair agreement with the previously measured values. We did not observe any ^{34}Si ion and obtain an upper limit an order of magnitude lower than the value reported by Hourani *et al.*[14]

In order to identify the emitted particles, we used Lexan polycarbonate foils. It has been shown by Price *et al.*[4] that by measuring the detailed dimensions and shapes of the tracks in different materials one can determine charge, mass, and energy of the particles. For the calibration of tracks heavy-ion beams from accelerators can be used. In our experiment, a beam of 93.5 MeV ^{28}Si particles from the Argonne National Laboratory FN tandem accelerator was used to calibrate 250 μm thick Lexan foils for the detection of Si ions. Foils exposed to 81 MeV Si ions, which were obtained from the scattering of 93.5 MeV ^{28}Si ions from a gold target, were placed in the dark for a day or more and then etched with a 6.25N KOH solution at 55° C for 20 to 60 min. Part of the foils were exposed also to a ^{248}Cm source and, using the same etching procedure, it was observed that tracks from ^{248}Cm spontaneous fission fragments are easily differentiated from ^{28}Si tracks by their length and shape [Fig. 1(a)]. The foils were also exposed to an intense dose of α particles from a ^{249}Cf source and it was found that doses of up to about 10^{12} α particles/cm^2 can be tolerated. The detection and identification of fragments is ultimately limited by a background of short tracks produced by low energy nuclei recoiling by elastic scattering of α particles.

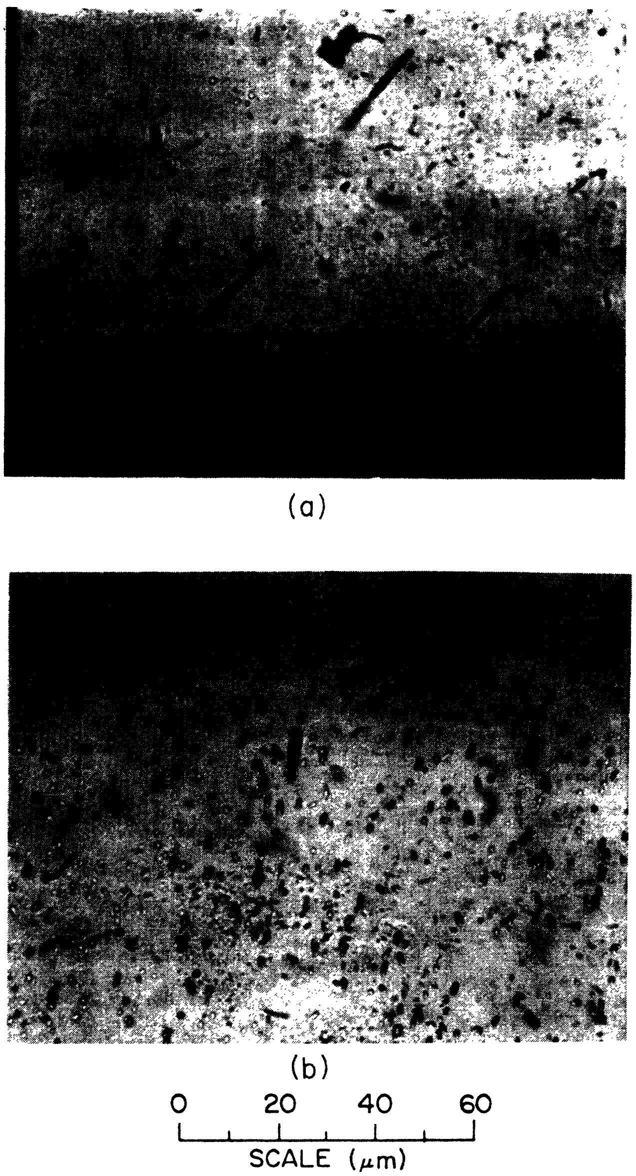

FIG. 1. Photographs of particle tracks in Lexan polycarbonate foils. Part (a) shows three long tracks due to 81 MeV ^{28}Si ions and three short tracks produced by fission fragments from ^{248}Cm. The two large irregular spots are due to accidental foil impurity. Part (b) shows a single fission track due to ^{241}Am. The area shown in (b) is approximately 1.3×10^{-4} cm^2 and was exposed to a total of 5×10^7 α particles. The background made of short tracks is attributed to nuclei of the foil material recoiling by the scattering of impinging α particles.

The ^{241}Am source used in the present experiment was prepared by electrodeposition on a 125 μm thick Pt disk covering an area of about 2 cm^2. The source was of extremely high purity; mass spectrometric analysis and alpha and gamma measurements showed that the source had no heavy element impurity which could interfere with the present measurement. The total amount of ^{241}Am in the source was determined by α and γ spectrometry. The γ-ray spectrum of the source was measured with a 5 cm$^2\times$1 cm Ge detector; the source to detector distance was 75 cm. The activity in this source was determined by comparing its spectrum (59.5 keV γ ray) with that from a calibrated ^{241}Am source. The gamma counting yielded a value of (0.49 ± 0.01) mCi. The alpha spectrum of the source was measured with a 25 mm^2 Si detector; the source was placed at a distance of 12 cm from the detector with a 0.5 mm diameter aperture. The detector efficiency was measured with a calibrated ^{249}Cf source. This method gave the activity of the source as (0.48 ± 0.01) mCi. The average of the two measurements is (0.485 ± 0.008) mCi.

A Lexan polycarbonate foil was shaped into a cylinder of 38 mm diameter and 30 mm height and was placed around the ^{241}Am source. The top part of the cylinder was covered with a cone-shaped Lexan foil. The ^{241}Am source was covered with a 180 μg/cm^2 Ni foil in order to stop nuclei recoiling from alpha decay. The source along with the foils was placed in a vacuum chamber at a pressure of $\sim10^{-2}$ Torr. The foil was exposed to the source for a period of 38 d. Exposed foils were etched as described above and one half of the exposed area (corresponding to a solid angle of approximately 1.5 sr) was scanned under a microscope. We found 33 tracks whose lengths and shapes were consistent with fission fragments, and no candidate for a decay by ^{34}Si emission was observed (Fig. 2). The overall efficiency (geometrical and

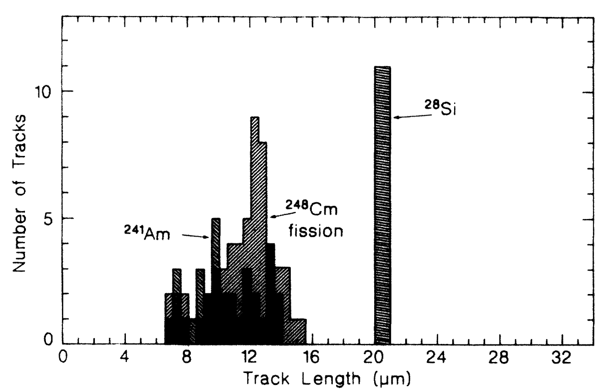

FIG. 2. Length spectrum of tracks produced by 81 MeV ^{28}Si ions, spontaneous fission fragments from a ^{248}Cm source, and fragments emitted from the ^{241}Am source. All tracks produced by the ^{241}Am source are consistent with fission tracks. No tracks attributable to decay by ^{34}Si emission were observed.

optical) of the measurement was determined by exposing a Lexan foil in the same setup to a calibrated ^{248}Cm source. We obtain for the spontaneous fission to alpha branching ratio of ^{241}Am a value of $(2.4\pm0.5)\times10^{-12}$. This branching ratio is in agreement with the value of $(1.9\pm0.7)\times10^{-12}$ measured by Druin, Mikheev, and Skobelev[12] but slightly lower than the ratio of $(3.77+0.08)\times10^{-12}$ obtained by Gold, Armani, and Roberts.[13] The present measurements also give an upper limit of 4.2×10^{-13} for the ^{34}Si/alpha branching at the 90% confidence level. Theoretical values for this branching ratio are 4.0×10^{-13} (Ref. 10) and 1.1×10^{-15} (Ref. 11).

In conclusion, our experiment confirms the spontaneous fission branch of ^{241}Am observed previously and establishes that the ^{34}Si emission is at least an order of magnitude smaller than the fission branch. It would, however, be interesting to pursue the search for a case where the three hadronic decay modes (alpha, heavy fragment, and fission) coexist.

The authors wish to thank Dr. R. J. Armani for providing us with the ^{241}Am source and Dr. W. G. Oltman for the Lexan foils. This research was supported by the U.S. Department of Energy under Contract W-31-109-Eng-38.

Note added. After the submission of our article, we learned about two other measurements on the ^{34}Si ion emission from ^{241}Am. These measurements give an upper limit of 5×10^{-14} (Ref. 15) and 3×10^{-15} (Ref. 16).

*Permanent address: Racah Institute of Physics, Hebrew University, Jerusalem, Israel.

[1]H. J. Rose and G. A. Jones, Nature (London) **307**, 245 (1984).

[2]S. Gales, E. Hourani, M. Hussonnois, J. P. Schapira, L. Stab, and M. Vergnes, Phys. Rev. Lett. **53**, 759 (1984).

[3]D. V. Alexandrov, A. F. Belyatsky, Yu. A. Glukhov, E. Yu. Nikolsky, A. A. Ogloblin, and D. N. Stepanov, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 152 (1984) [JETP Lett. **40**, 909 (1985)].

[4]P. B. Price, J. D. Stevenson, S. W. Barwick, and H. L. Ravn, Phys. Rev. Lett. **54**, 297 (1985).

[5]W. Kutschera, I. Ahmad, S. G. Armato III, A. M. Friedman, J. E. Gindler, W. Henning, T. Ishii, M. Paul, and K. E. Rehm, Phys. Rev. C **32**, 2036 (1985).

[6]S. W. Barwick, P. B. Price, and J. D. Stevenson, Phys. Rev. C **31**, 1984 (1985).

[7]A. Sandulescu, Yu. S. Zamyatnin, I. A. Lebedev, B. F. Myasoedov, S. P. Tretyakova, and D. Hasegan, Joint Institute for Nuclear Research Rapid Communication 5-84, 1984.

[8]S. P. Tretyakova, A. Sandulescu, Yu. S. Zamyatnin, Yu. S. Korotkin, and V. L. Mikheev, Joint Institute for Nuclear Research Rapid Communication 7-85, 1985.

[9]R. L. Fleischer, P. B. Price, and R. M. Walker, *Nuclear Tracks in Solids: Principles and Applications* (University of California, Berkeley, 1975).

[10]D. N. Poenaru, M. Ivascu, A. Sandulescu, and W. Greiner, J. Phys. G **10**, L183 (1984); D. N. Poenaru, W. Greiner, M. Ivascu, D. Mazilu, and A. Sandulescu, Gesellschaft für Schwerionenforschung Report GSI-85-21, 1985.

[11]Yi-Jin Shi and W. J. Swiatecki, Nucl. Phys. **A438**, 450 (1985); and private communication.

[12]V. A. Druin, V. L. Mikheev, and N. K. Skobelev, Zh. Eksp. Teor. Fiz. **40**, 1261 (1961) [Sov. Phys. JETP 13, 889 (1961)].

[13]R. Gold, R. J. Armani, and J. H. Roberts, Phys. Rev. C **1**, 738 (1970).

[14]E. Hourani, M. Hussonnois, L. Stab, L. Brillard, S. Gales, and J. P. Schapira, Phys. Lett. **160B**, 375 (1985).

[15]P. B. Price, Phys. Bull. **36**, 589 (1985).

[16]S. P. Tretyakova *et al.*, Joint Institute of Nuclear Research Rapid Communication 13-85, 1985.

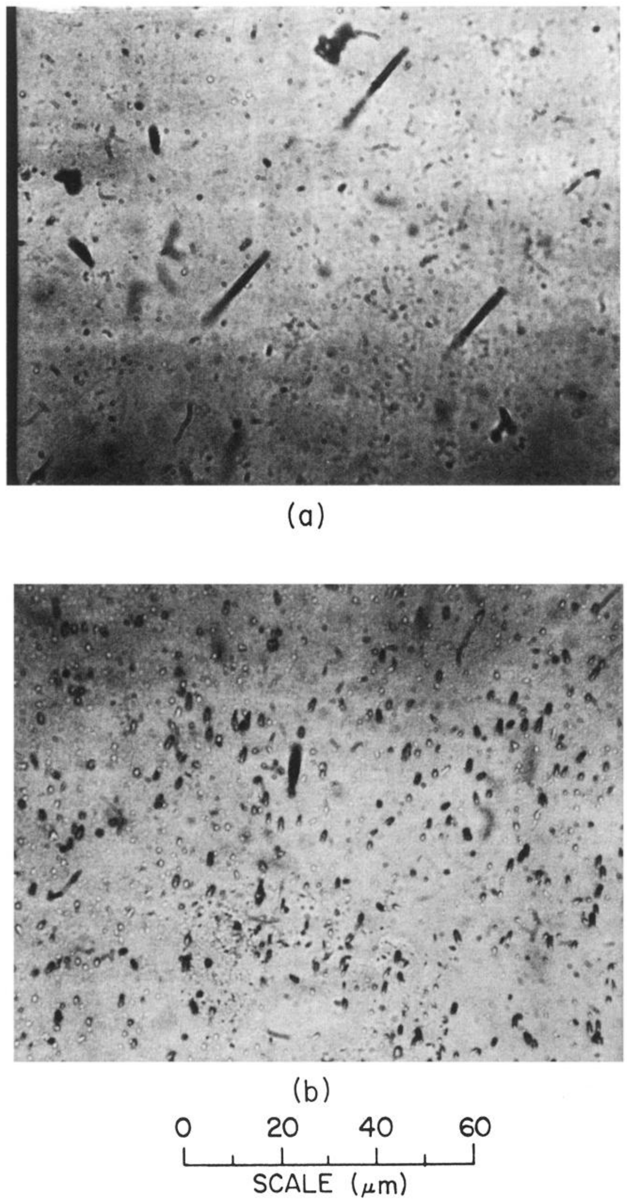

FIG. 1. Photographs of particle tracks in Lexan polycarbonate foils. Part (a) shows three long tracks due to 81 MeV ^{28}Si ions and three short tracks produced by fission fragments from ^{248}Cm. The two large irregular spots are due to accidental foil impurity. Part (b) shows a single fission track due to ^{241}Am. The area shown in (b) is approximately 1.3×10^{-4} cm^2 and was exposed to a total of 5×10^{7} α particles. The background made of short tracks is attributed to nuclei of the foil material recoiling by the scattering of impinging α particles.

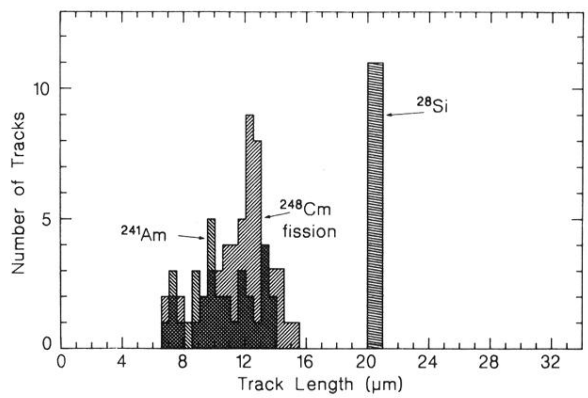

FIG. 2. Length spectrum of tracks produced by 81 MeV ^{28}Si ions, spontaneous fission fragments from a ^{248}Cm source, and fragments emitted from the ^{241}Am source. All tracks produced by the ^{241}Am source are consistent with fission tracks. No tracks attributable to decay by ^{34}Si emission were observed.