

## The Half-Life of Thallium-204†

GARMAN HARBOTTE

*Chemistry Department, Brookhaven National Laboratory, Upton, New York*

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A determination of the half-life of thallium-204 has given the value  $4.0 \pm 0.1$  years.

THE half-life of the long-lived isotope of thallium formed by neutron capture has been reported to be 2.7 years<sup>1</sup> and 3.5 years.<sup>2</sup> In this note we wish to report an additional determination of the half-life.

A sample of active thallium was obtained as thallos nitrate which had been irradiated with neutrons in the Oak Ridge Reactor, and had cooled for 2.2 years. The thallos nitrate was dissolved, scavenged twice with ferric hydroxide, oxidized to thallic hydroxide, washed and dissolved in perchloric acid. The trivalent thallium was finally extracted from a perchlorate-chloride solution into ether.

A few drops of the ethereal solution were evaporated to dryness in the center of an aluminum plate; the radioactive deposit was covered with a coating of "Glyptal" which was baked on when dry, and the

varnish was covered with a layer of "Aquadag" to prevent charging effects.

The resultant source, which had a highly reproducible counting rate of about 19 000 counts per minute, was counted at frequent intervals in a "Nucleometer" (methane-flow proportional counter) over a period of about 2.9 years. The source was oriented reproducibly in the counting chamber, a million or more counts were usually recorded, and a standard was counted each day as a check on the operation of the "Nucleometer." At all counting rates employed in this experiment the coincidence loss was well under 1 percent and consequently no correction for coincidence loss was made.

A least-squares fit of the results, plotted as  $\log(TI^{204})$  counting rate corrected for background) versus time, yields a half-life of  $4.02 \pm 0.12$  years. This half-life is not far from the earlier value of 3.5 years but is entirely inconsistent with the later values of 2.7 years.

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<sup>1</sup>A. F. Voigt, private communication listed in "Table of Isotopes" by Hollander, Perlman and Seaborg, December, 1952 [Revs. Modern Phys. 25, 469 (1953)]; and E. E. Lockett and R. H. Thomas, *Nucleonics* 11, No. 3, 14 (1953).

<sup>2</sup>K. Fajans and A. F. Voigt, *Phys. Rev.* 60, 619 (1941).

## $\pi^-$ and $\mu^-$ Meson-Induced Fission\*

W. JOHN AND W. F. FRY†

*Radiation Laboratory, Department of Physics, University of California, Berkeley, California, and  
Department of Physics, University of Chicago, Chicago, Illinois*

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Meson-induced fission has been studied in uranium-loaded Ilford C-2 emulsions exposed to the  $\pi^-$  and  $\mu^-$  meson beams of the University of Chicago cyclotron. 3762  $\pi^-$  endings were studied including 16 fission events. 3573  $\mu^-$  endings were studied including 7 fission events. The probability of a  $\pi^-$  meson ending in fission is found to be  $(4.3 \pm 1.5) \times 10^{-3}$  in C-2 emulsion containing 0.052 g cm<sup>-3</sup> of uranium. The corresponding probability for  $\mu^-$  mesons is found to be  $(1.7 \pm 0.7) \times 10^{-3}$ . Assuming that the emulsion is a homogeneous mixture and that the atomic capture probability is proportional to  $Z$ , it is found that  $(37 \pm 13)$  percent of  $\pi^-$  absorptions in uranium produce fission and  $(15 \pm 6)$  percent of  $\mu^-$  absorptions produce fission. If the emulsion is considered to be composed of AgBr and gelatin with the uranium in the gelatin, the corresponding percentages are found to be  $(18 \pm 6)$  percent and  $(7 \pm 3)$  percent.

### INTRODUCTION

THE probability that a  $\pi^-$  meson stopping in uranium-loaded emulsion will cause a fission event is the product of the probability that the meson

will be absorbed in an uranium nucleus and the probability that a  $\pi^-$  meson absorption by the uranium nucleus causes fission. Experimental data on  $\pi^-$  absorption in uranium-loaded emulsion can be used to calculate these probabilities if certain assumptions are made.  $\pi^-$  absorption in uranium-loaded emulsions was previously studied by Al-Salam.<sup>1</sup> The present investigation takes

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† Present address, Department of Physics, University of Wisconsin, Madison, Wisconsin.

<sup>1</sup>S. G. Al-Salam, *Phys. Rev.* 84, 254 (1951).

TABLE I. Measurements of  $\pi^-$  induced fissions.

| Event number   | Total range of fission track in microns | Ratio of ranges of fission fragments |
|----------------|---|--------------------------------------|
| 1 <sup>a</sup> | 29                                      | 1.1                                  |
| 2              | 26                                      | 1.0                                  |
| 3              | 30                                      | 1.4                                  |
| 4              | 19                                      | 1.0                                  |
| 5              | 26                                      | 1.2                                  |
| 6              | 26                                      | 1.1                                  |
| 7              | 24                                      | 1.0                                  |
| 8              | 23                                      | 1.0                                  |
| 9              | 27                                      | ~1.1                                 |
| 10             | 29                                      | 1.3                                  |
| 11             | 24                                      | 1.1                                  |
| 12             | 24                                      | 1.6                                  |
| 13             | 24                                      | 1.1                                  |
| 14             | 24                                      | 1.3                                  |
| 15             | 28                                      | 1.1                                  |
| 16             | 27                                      | 1.3                                  |

<sup>a</sup> Third particle emitted from the fission.

advantage of the much improved conditions of observation made possible by the use of the meson beam at Chicago. The present study also includes the study of  $\mu^-$  absorption in uranium-loaded emulsions.

#### EXPERIMENTAL DETAILS

200-micron Ilford C-2 emulsions were loaded with uranium. The plates were presoaked in water for about 30 minutes, then soaked in a solution of uranium acetate for an hour and a half. The plates were allowed to dry for three hours and then exposed for one hour to slow  $\pi^-$  mesons from the external meson beam of the University of Chicago cyclotron. The plates were soaked in developer at 8°C and developed at 29°C. Total elapsed time from the uranium solution soaking to fixing was about seven hours. The plates for the study of  $\mu^-$  capture were treated as above except that they were exposed to the slow  $\mu^-$  beam of the cyclotron.<sup>2</sup>

The emulsions were scanned at a magnification of 270 $\times$ . Fission events were studied at 1400 $\times$ . Fields of view were taken sufficiently far apart to eliminate the possibility of overlap. The concentration of uranium in the emulsion was determined by counting the uranium alphas in a given volume. Allowance was made for the gradation of uranium density with depth. Only the top  $\frac{2}{3}$  of the emulsion was scanned. The effective concentration of uranium in the  $\pi^-$  plates was found to be 0.052 g cm<sup>-3</sup> corresponding to 4.63 $\times 10^6$  alphas hr<sup>-1</sup> cm<sup>-3</sup> of dry emulsion. The uranium concentration in the  $\mu^-$  plates was 1.15 times higher.

#### APPEARANCE OF EVENTS

The plates were relatively free of background tracks except for uranium alphas. A fission event appears as a "hammer" track at the meson ending. The two prongs are collinear and approximately equal in length. The fission track is about 26 $\mu$  long and somewhat heavier than the uranium alpha tracks which are about 22 $\mu$

long. The fission events are fairly characteristic in appearance, however, in each case the event was carefully checked in detail at high magnification.

#### RESULTS OF $\pi^-$ INDUCED FISSION

A total of 3762  $\pi^-$  endings were studied. Sixteen fission events were seen. One fission was accompanied by the ejection of a third particle at 27° to the fission track. This third track lay completely in the emulsion with a total range of 309 $\mu$  and was identified as a proton from the grain count. The range corresponds to that of a 7-Mev proton, making the third particle a possible knock-on proton. The measurements of the fission events are given in Table I. The probability of a  $\pi^-$  meson ending in fission is found to be  $(4.3 \pm 1.5) \times 10^{-3}$  in C-2 emulsion containing 0.052 g cm<sup>-3</sup> of uranium before development. The error is estimated from the statistics and the uncertainty in uranium concentration.

#### RESULTS OF $\mu^-$ INDUCED FISSION

A total of 3573  $\mu^-$  endings were studied, including 7 fission events. Measurements of the fission events are given in Table II. The probability of a  $\mu^-$  meson ending in fission in C-2 emulsion containing 0.052 g cm<sup>-3</sup> of uranium before development is found to be  $(1.7 \pm 0.7) \times 10^{-3}$ .

#### DISCUSSION OF $\pi^-$ INDUCED FISSION

Let us denote by  $P_a$  the probability that a  $\pi^-$  meson stopping in uranium-loaded emulsion will be absorbed in a uranium nucleus and by  $P_f$  the probability that a  $\pi^-$  meson absorption by the uranium nucleus causes fission. Then the probability  $P$  that a  $\pi^-$  meson stopping in uranium-loaded emulsion will cause a fission event is given by  $P = P_a P_f$ . Unfortunately, neither  $P_a$  nor  $P_f$  is known from experiment.

We have calculated  $P_a$  in two different ways. First it was assumed that the emulsion was a homogeneous mixture. The atomic capture probability, that is, the probability that a  $\pi^-$  meson is captured into an orbit about a nucleus, was taken to be proportional to the atomic number of the capturing nucleus in accordance with the calculation by Fermi and Teller.<sup>3</sup> Then, using the known composition of Ilford C-2 emulsion, the percentage weight/atomic weight for each element was

TABLE II. Measurements of  $\mu^-$  induced fissions.

| Event number | Total range of fission track in microns | Ratio of ranges of fission fragments |
|--------------|---|--------------------------------------|
| 1            | 29                                      | 1.4                                  |
| 2            | 25                                      | 1.1                                  |
| 3            | 24                                      | 1.3                                  |
| 4            | 23                                      | 1.1                                  |
| 5            | 27                                      | ~1.1                                 |
| 6            | 27                                      | 1.1                                  |
| 7            | 25                                      | 1.2                                  |

<sup>2</sup> W. F. Fry, Phys. Rev. 85, 676 (1952).

<sup>3</sup> E. Fermi and E. Teller, Phys. Rev. 72, 399 (1947).

weighted by  $Z$  to find  $P_a$ . Hydrogen was omitted from the calculation in accordance with the observation by Panofsky *et al.*<sup>4</sup> that  $\pi^-$  mesons are rarely absorbed by hydrogen in compounds. However, the omission of hydrogen has little effect on the calculation.  $P_a$  is found to be  $11.5 \times 10^{-3}$ . Taking  $P$  from the experimental data, one finds  $P_f$  is  $(0.37 \pm 0.13)$ .

$P_a$  was also calculated under the assumption that the emulsion consists of grains of AgBr imbedded in gelatin and that the uranium is dissolved in the gelatin fraction only. Fry<sup>2</sup> has estimated from  $\mu^-$ -electron decay that 39 percent of the slow  $\mu^-$  mesons stopping in emulsion stop in the gelatin and 61 percent in AgBr. This is also in agreement with a rough calculation based on stopping powers.  $P_a$  is then given by 0.39 times the probability that a meson ending in the gelatin is absorbed by an uranium nucleus. The latter probability was found by taking the percentage weight/atomic weight of the elements of the gelatin including uranium but omitting hydrogen and weighting by  $Z$ .  $P_a$  is found to be  $23.8 \times 10^{-3}$ , and finally  $P_f$  is  $0.18 \pm 0.06$ . The assumptions of this calculation seem more applicable than the homogeneous mixture assumption, although the resulting  $P_f$  seems low considering observations of photofission and fast neutron fission.

We have also calculated the  $Z$  dependence of  $P_a$  assuming that  $P_f=1$ . The percentage weight/atomic weight of the various elements (omitting hydrogen) were weighted by  $Z^n$ . Both the homogeneous mixture and the AgBr-gelatin assumptions give about the same results. It is found that  $n=0.1$  to  $0.5$  agrees with our data. Our experimental result for the number of fissions per  $\pi^-$  meson ending is about one-half of the figure found by Al-Salam.<sup>1</sup> However, the values overlap when the probable errors are considered. Al-Salam<sup>1</sup> concluded that essentially all  $\pi^-$  mesons entering uranium cause fission on the basis of a homogeneous mixture calculation. It should be pointed out that, on the AgBr-gelatin assumption, the  $P_f$  calculated from Al-Salam's data is somewhat less than  $0.5$ .

The  $\mu^-$  contamination on the  $\pi^-$  plates is estimated to be less than 1 percent.

#### DISCUSSION OF $\mu^-$ INDUCED FISSION

The prong distribution for 1516  $\mu^-$  endings is given in Table III. The distribution is in agreement with that obtained by Morinaga and Fry<sup>5</sup> in a study of  $\mu^-$  stars on plates exposed under similar conditions. Morinaga and Fry estimate that this prong distribution corresponds to a  $\pi^-$  contamination of less than 3  $\pi^-$  per 1000 meson endings. One would expect about 0.02 fission per 1000 endings from this  $\pi^-$  contamination. We observed about 100 times this number of fissions per 1000

TABLE III. Prong distribution of 1516 stars on  $\mu^-$  plates.

| Number of prongs | Number of events |
|------------------|------------------|
| 0                | 1459             |
| 1                | 43               |
| 2                | 6                |
| 3                | 6                |
| 4                | 2                |

endings. Hence we conclude that most of the fission events on the  $\mu^-$  plates were induced by  $\mu^-$  mesons.

The study of  $\mu^-$  stars in emulsions by Morinaga and Fry<sup>5</sup> showed that  $\mu^-$  capture frequently gives rise to a nuclear excitation of 25 Mev. The calculations of Tiomno and Wheeler<sup>6</sup> gave nuclear excitations of the order of 15 Mev. The compound nucleus formed when a  $\mu^-$  meson is captured by  ${}_{92}\text{U}^{238}$  is  ${}_{91}\text{Pa}^{238}$ . In estimating the fission threshold of  ${}_{91}\text{Pa}^{238}$ , we note that the fission parameter  $Z^2/A$  for  ${}_{91}\text{Pa}^{238}$  has nearly the same value as that for  ${}_{90}\text{Th}^{234}$ . The photofission threshold for  ${}_{90}\text{Th}^{234}$  has been measured and found to be 5.4 Mev.<sup>7</sup> In addition, the  $\mu^-$  meson must supply the mass difference between  ${}_{91}\text{Pa}^{238}$  and  ${}_{92}\text{U}^{238}$ , which is about 2 Mev.

If the atomic capture probability is taken to be proportional to  $Z$ , the homogeneous-mixture calculation gives  $0.15 \pm 0.06$  as the probability of a  $\mu^-$  meson absorption in the uranium nucleus causing fission. The AgBr-gelatin calculation gives  $0.07 \pm 0.03$ . Both of the results are in agreement with the upper limit of 0.25 set by Galbraith and Whitehouse.<sup>8</sup>

The observed number of fissions per  $\mu^-$  ending in emulsion of a given uranium loading appears to be about one-half the number of fissions per  $\pi^-$  ending, although the uncertainties are large.  $\pi^-$  capture would give an excitation energy of the compound nucleus of about 140 Mev. As the nucleus evaporates particles it loses excitation; after each evaporation there is a certain probability that fission will take place, until finally the excitation of the nucleus is below the fission threshold.  $\mu^-$  capture leaves the compound nucleus with a much lower excitation than in the case of  $\pi^-$  capture, perhaps of the order of 15 Mev. Fewer particles need to be evaporated before the excitation is below the fission threshold. Hence in this interpretation the overall probability of fission in the case of  $\mu^-$  capture would be smaller.

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<sup>6</sup> J. Tiomno and J. A. Wheeler, *Revs. Modern Phys.* **21**, 153 (1949).

<sup>7</sup> Koch, McElhinney, and Gasteiger, *Phys. Rev.* **77**, 329 (1950).

<sup>8</sup> W. Galbraith and W. J. Whitehouse, *Phil. Mag.* **44**, 77 (1953).

<sup>4</sup> Panofsky, Aamodt, and Hadley, *Phys. Rev.* **81**, 565 (1951).

<sup>5</sup> H. Morinaga and W. F. Fry, *Nuovo cimento* **10**, 308 (1953).