Title
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Authors
McCaslin, Joseph B.
Stephens, Lloyd D.

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University of California

Ernest O. Lawrence Radiation Laboratory

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ABSTRACT

Correct evaluation of radiation exposures received by individuals working near high-energy accelerators depends upon measurement of the radiation field involved. The measurements of the separate components and the determination of their energy spectrum are basic to this evaluation. No single instrument or detector can make these measurements; therefore, a variety of detectors must be employed.

A detector we describe for the measurement of very-high-energy particles utilizes the spallation of mercury to ^{149}Tb, an alpha emitter with a 4.12-h half-life. This reaction has a threshold between 500 and 600 MeV, and extends the use of threshold detectors into another energy decade.

The Hg → ^{149}Tb reaction has been successfully used to measure relative neutron production from targets as a function of angle for energies up to 26 GeV.

*This work was done under the auspices of the U. S. Atomic Energy Commission.

INTRODUCTION

A primary responsibility of the Health Physics Department is measuring the stray radiation fields produced by high-energy particle accelerators. In the past, we have developed and reported on a series of detectors for this purpose. They have been used in part to help determine the energy spectrum of radiation fields. Each previously reported detector was required to have enough sensitivity to measure the low fluxes typically permitted in regions of personnel occupancy.

We now have designed and built a detector that uses the spallation of elemental mercury for detecting small fluxes of high-energy particles with energies above 0.5 GeV. Preliminary results show that elemental mercury can be used as a threshold detector. Spallation of the mercury produces ^{149}Tb, which emits 3.95-MeV alpha particles with a 4.1-h half-life. The ^{149}Tb can easily be extracted for counting. The sensitivity of this detector is limited only by the amount of mercury that can be processed conveniently.

DETECTOR CRITERIA

Threshold detectors and other detectors for health physics use at accelerators have been described in the literature.[1] It is sufficient to say here that these detectors should be sensitive enough to permit measurements of very low fluxes because such measurements are often made when the detectors are separated from the primary source of radiation by many feet of shielding material.

Gold has been suggested as a proton-beam monitor with high threshold energy.[2-4] Unfortunately, small gold foils cannot yield the sensitivity needed for health physics use, and we do not know of an easy method to extract ^{149}Tb from large quantities of gold. An extraction must be made in order to achieve high sensitivity, because the maximum range of ^{149}Tb alphas in gold is only about 11 mg/cm^2.[5]

Simplifications in the extraction process seemed possible if a liquid were used instead of a solid. This approach was rewarded in an unforseen way when it was found that ^{149}Tb floats to the top of the irradiated mercury. But gravity separation is too slow to be useful, taking several half-lives. The obvious solution, and the one which we use, is to centrifuge the mercury and then lift a large portion of the ^{149}Tb from the mercury surface by using pressure-sensitive cellulose acetate tape.[6] The ^{149}Tb has been identified on the tape samples both by half life and by energy. Production of ^{149}Tb from mercury was shown to be due to high-energy interactions by exposing a mercury compound first to 300-MeV protons and then to 700-MeV protons of the circulating beam at the 184-Inch Cyclotron. Figures 1 through 4 show that ^{149}Tb is formed at 700 MeV, but not at 300 MeV. Gold samples were included in these tests.

EXPERIMENTAL PROCEDURE

An experiment was designed to yield the following information:

1. The efficiency of ^{149}Tb extraction from mercury.

2. Whether centrifuging separates the ^{149}Tb from the total volume of mercury or from only a small volume of mercury near the surface.

3. The specific activity of ^{149}Tb from mercury relative to gold.

4. The sensitivity of a practical mercury threshold-detector system.

Gold and mercury samples were placed in the 6.2-GeV external proton beam of the Bevatron to determine the sensitivity of the mercury threshold-detector system relative to gold. Figure 5 shows that a number of thin gold foils were placed directly in front of the mercury sample, and thick gold foils were placed in front and in back of the mercury. The thin foils were used as beam monitors so that the incident proton beam intensity could be determined. The thick foils were used to determine beam loss through the mercury sample. Losses due to beam absorption, scattering, and misalignment were indicated by a reduction in induced activity of the rear foil.

After irradiation, a 100-g aliquot was withdrawn from the center of the 481-g sample of irradiated mercury by means of a hypodermic syringe and placed in a separate polyethylene container. Unirradiated mercury was then added to each sample to bring both to equal level and weight. An equal ratio of the weights and activities for these two samples would indicate that terbium is separated from the entire volume of the sample rather than from just a volume near the surface.

Pressure-sensitive adhesive tapes were cut to the size of the mouths of the plastic containers and placed adhesive side down on top of the Hg. Care was taken to prevent air pockets from being trapped between the tape and the surface of the mercury. An aluminum disk was then placed over each tape, and the caps were replaced on the containers (see Fig. 6). The plastic containers were placed in tool-steel holders, which were then attached to the centrifuge (see Fig. 7).[7, 8] The samples were centrifuged for 1 hour at 1700 G. Although we have not determined the minimum centrifugation time for a given G force, it appears that 1 hour at 1700 G assures maximum separation. Figure 8 shows G force plotted vs centrifuge speed.

After centrifuging, the tapes were removed and counted on a methane-flow gas proportional counter (Fig. 9). Successive tape samples were taken on the 381-g mercury sample (A-1), and their counting rates were corrected to zero time and then summed. The ratio of the corrected first-tape activity to the corrected sum activity is called the extraction efficiency. This process was repeated for the 100-g sample (A-2). The corrected sums of A-1 and A-2, after correction for counting efficiency, constitute the total activity induced in the irradiated 481-g sample. The total activity per gram was then compared with that of a thin Au foil.

EXPERIMENTAL RESULTS

A. Incident-Proton Flux Density

The activity of the thin gold foils is plotted vs foil weight in Fig. 10. The 1.58 mg/cm^2 foil was used for the beam-intensity measurement because self-absorption of the ^{149}Tb alphas is negligible for such a thin foil. An incident proton flux density of $2.43 \times 10^9/\text{cm}^2$-sec is indicated by this foil. A reaction cross section of 1.1×10^{-27} barns was used.

B. Effective Mercury Weight

Activity in the front and rear thick gold foils showed that the incident proton beam was attenuated through the mercury sample and that a correction for beam loss would be necessary. This correction was applied to the mercury sample in such a way as to imply a reduction in the mass of the mercury sample. This correction is necessary in order to directly compare the terbium activity per gram of mercury with the activity per gram of gold for the same incident-proton flux density. Obviously the proton flux density at the rear of the mercury sample is less than at the front.

The ratio of activities of the back and front thick gold foils is 0.639. The average beam through the mercury is therefore 82% of the incident beam, or the effective weight of the mercury sample is 394 g instead of 481 g.

C. Extraction Efficiency

Figure 11 shows the counting rate of successive tape samples corrected to zero time and saturation. The summation of all A-1 tapes, including extrapolated values for tapes 5 through 8, yields a counting rate of 3.32×10^7 counts per minute (c/m), and the extraction efficiency is then

$$\frac{1.95 \times 10^7 \text{ c/m (first A-1 tape)}}{3.32 \times 10^7 \text{ c/m (all A-1 tapes)}} = 58.6\%.$$

For the A-2 sample we have

$$\frac{5.00 \times 10^6 \text{ c/m (first A-2 tape)}}{8.64 \times 10^6 \text{ c/m (all A-2 tapes)}} = 57.9\%.$$

Tape samples should be counted between 100 and 1500 minutes after the end of the irradiation so as to avoid counting α-particles from emitters that have half-lives shorter or longer than that of ^{149}Tb. Analysis of the gamma spectrum of the mercury and tape samples with a Ge (Li) detector does not indicate that there is a suitable gamma ray which can be used as an alternative method of detection.[9]

D. Volume Separation

Experimental results show that ^{149}Tb is concentrated from the volume of the mercury sample because each sample A-1 and A-2 bears the same relationship of activity to weight:

$$\text{Weight ratio} = \frac{381\ \text{g (A-1)}}{100\ \text{g (A-2)}} = 3.81, \text{ and}$$

$$\text{Activity ratio} = \frac{3.32\times 10^{7}\ \text{(all A-1 tapes)}}{8.64\times 10^{6}\ \text{(all A-2 tapes)}} = 3.84\ .$$

E. ^{149}Tb Activity Per Gram of Mercury Relative to Gold

The specific activities of mercury and gold are seen to be nearly identical:

$$\text{Sp. act. (Hg)} = \frac{4.19\times 10^{7}\ \text{c/m (A1 + A2)}}{394\ \text{g (effective)}} = 1.06\times 10^{5}\ \text{c/m-g}$$

$$\text{Sp. act. (Au)} = \frac{8.96\times 10^{2}\ \text{c/m}}{(1.58\ \text{mg/cm}^2)\times(5\ \text{cm}^2)} = 1.13\times 10^{5}\ \text{c/m-g.}$$

F. Sensitivity of a Practical Detection System

The sensitivity depends on the amount of mercury used and the ability of the centrifuge to process it. However, if we assume (a) negligible beam removal by the mercury sample, (b) negligible build up of secondaries with energy greater than the threshold energy, and (c) a windowless foil counter that detects 100% of the alphas emitted in a 2π solid angle, then the zero-time saturated counting rate at 58.6% extraction efficiency is 5.9×10^{-5} c/m-g per proton/cm^2-sec. For 500-g mercury samples this becomes 3.0×10^{-2} c/m per proton/cm^2-sec.

CONCLUSIONS

The amount of ^{149}Tb activity produced in a gram of mercury is approximately the same as that in gold, but because about 58% of the Tb can be extracted effectively from large volumes of mercury, use of the latter system offers an enormous increase in sensitivity. In this experiment the increase amounted to a factor of more than 10^4 compared to a 5-cm^2 gold foil which was infinitely thick compared to the range of the ^{149}Tb alphas.

A sensitivity of about 3×10^{-2} c/m per proton/cm^2-sec can be obtained with 500-g mercury samples, although an additional correction may be necessary for mercury self-shielding during the irradiation. A tape sample from a 500-g capsule irradiated to saturation in a field of 3.3 protons/cm^2-sec would yield an initial counting rate of 0.1 c/m. This corresponds to an easily attainable background of 0.1 c/m for a methane-flow counter.

It is assumed that neutrons and protons are equally effective in producing ^{149}Tb from mercury, so that this method is also applicable to neutron detection.

APPENDIX

Figures 12 and 13 show gross α activity from one of the tape samples extending over a period greater than 10 half-lives.

Results of two experimental exposures made at the Proton Synchrotron at the CERN Laboratory in Geneva, Switzerland are shown in Figs. 14 and 15.[10] The actual flux values are plotted vs the angle from the target. The ^{149}Tb data indicate a very peaked yield toward zero degrees, i.e., for particle energies greater than 500 MeV. The ^{11}C points for the same runs indicate a higher flux at lower-energy particles, greater than 20 MeV. The ^{149}Tb data taken at the 25-cm-radius points have been corrected for inverse-square dependence. It is not possible to apply a similar correction to ^{11}C, S, or Al data, indicating a significant contribution from sources other than the primary target.

Figure 14 data was from an experimental run at 26.4 GeV/c. Figure 15 data was from an experimental run at 14.64 GeV/c.

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4. R. B. Duffield and G. Friedlander, unpublished results reported in Brookhaven National Laboratory Report BNL-303, 1954, p. 27.
5. L. Winsberg, Phys. Rev. 135, B1105 (1964).
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7. International Equipment Company, Needham Heights, Massachusetts, Model 428 clinical centrifuge using modified trunnion rings.
8. International Equipment Company, Needham Heights, Massachusetts, tachometer Model 748.
9. Alan R. Smith, Lawrence Radiation Laboratory, private communication.
10. Shielding Experiment at the CERN-PS, CERN/LRL/RHEL Collaboration, 1966 (in preparation).

FIGURE LEGENDS

Fig. 1. Counting rate vs decay time for gold foil irradiated with 300-MeV protons.

Fig. 2. Counting rate vs decay time for gold foil irradiated with 700-MeV protons.

Fig. 3. Counting rate vs decay time for HgF_2 (1.35 g), irradiated with 300-MeV protons.

Fig. 4. Counting rate vs decay time for HgF_2 (1.45 g), irradiated with 700-MeV protons.

Fig. 5. Detector array for sensitivity determination.

Fig. 6. Centrifuge trunnion and modified trunnion rings containing special tool-steel containers.

Fig. 7. (a) Centrifuge trunnion in place containing one polyethylene mercury container.

(b) Centrifuge and tachometer used in this experiment.

Fig. 8. Plot of rotor speed (in rpm) vs g force at 10-cm radius.

Fig. 9. Windowless methane-gas proportional counter.

Fig. 10. Thin-gold-foil counting rates vs thickness.

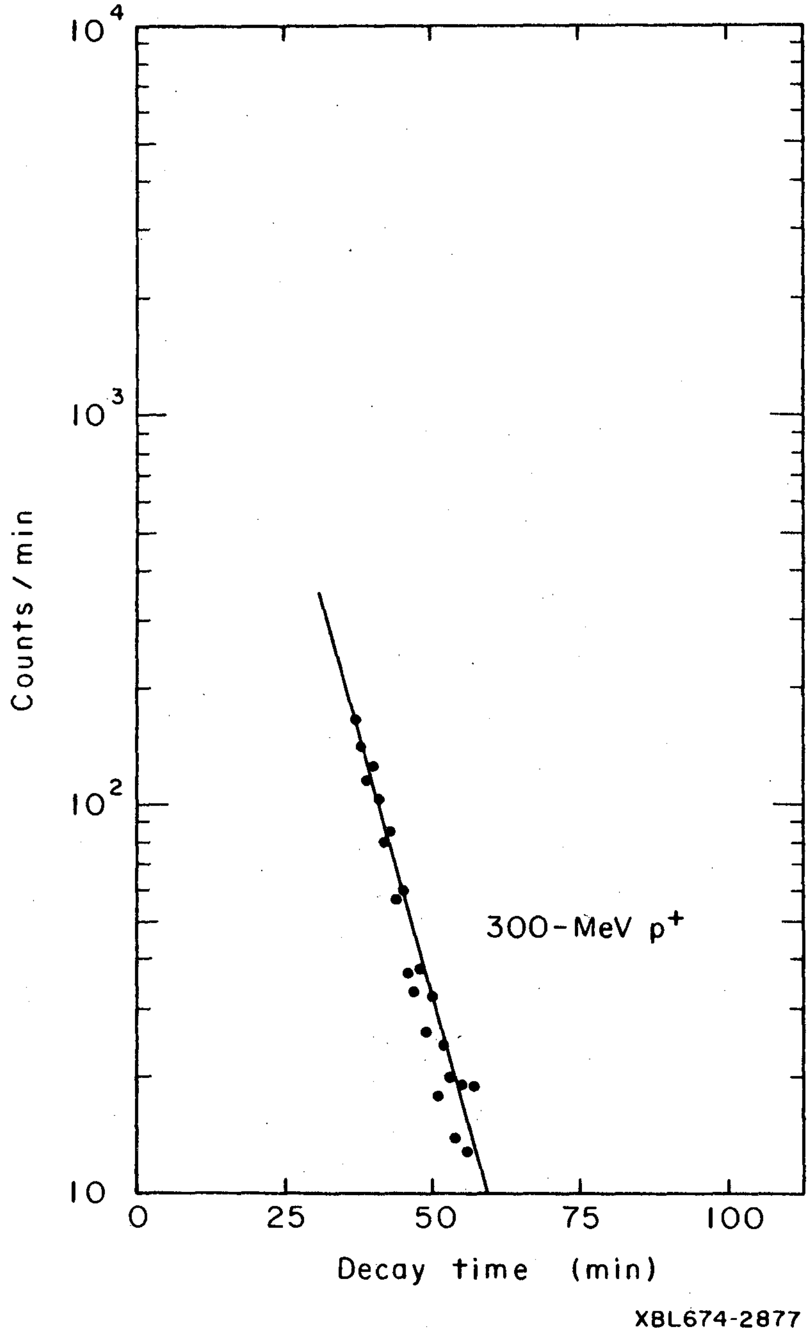
Fig. 11. Counting rates of successive tape samples corrected to zero time and for saturation.

Fig. 12. Gross α activity from a tape sample vs decay time.

Fig. 13. Gross α activity from a tape sample vs decay time.

Fig. 14. Experimental results taken at the CERN Proton Synchrotron; particle flux vs target angle for both Hg → Tb and $^{12}C \rightarrow {}^{11}C$, taken at 26.4 GeV/c.

Fig. 15. Experimental results taken at the CERN Proton Synchrotron; particle flux vs target angle for both Hg → Tb and $^{12}C\,(n, 2n)\,^{11}C$, taken at 14.64 GeV/c.



Counts / min
10⁴
10³
10²
10
0
25
50
75
100
Decay time (min)
300-MeV p⁺
XBL674-2877

Fig. 1

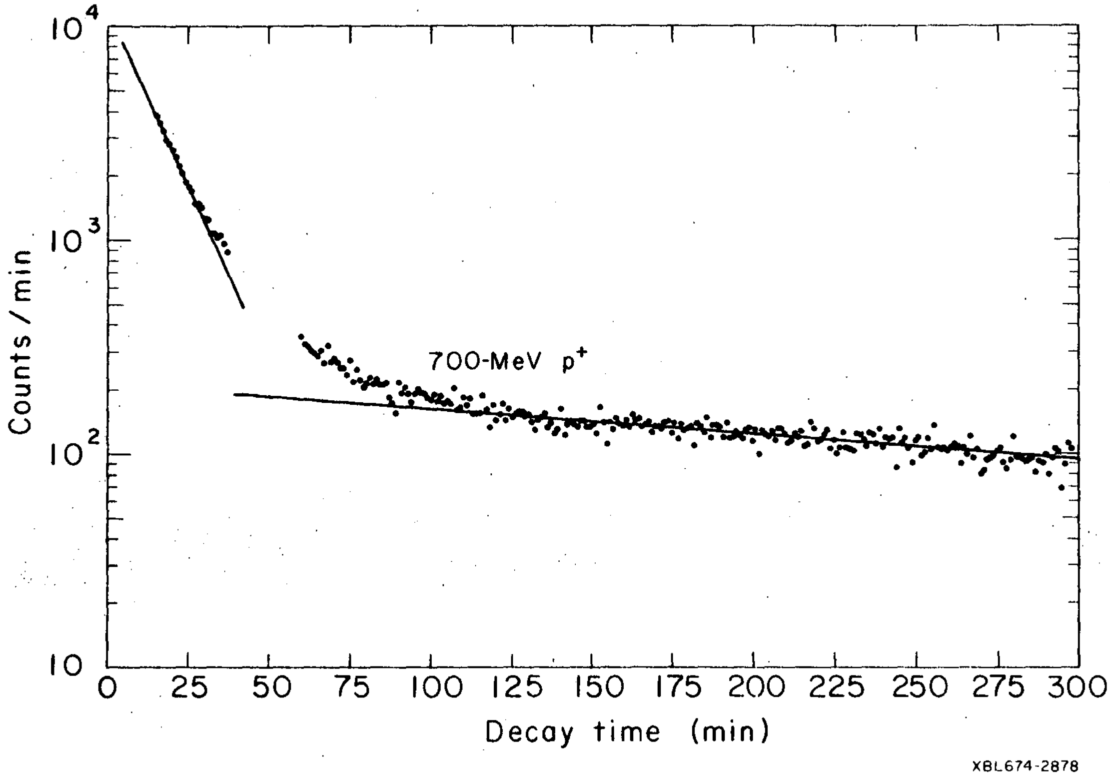
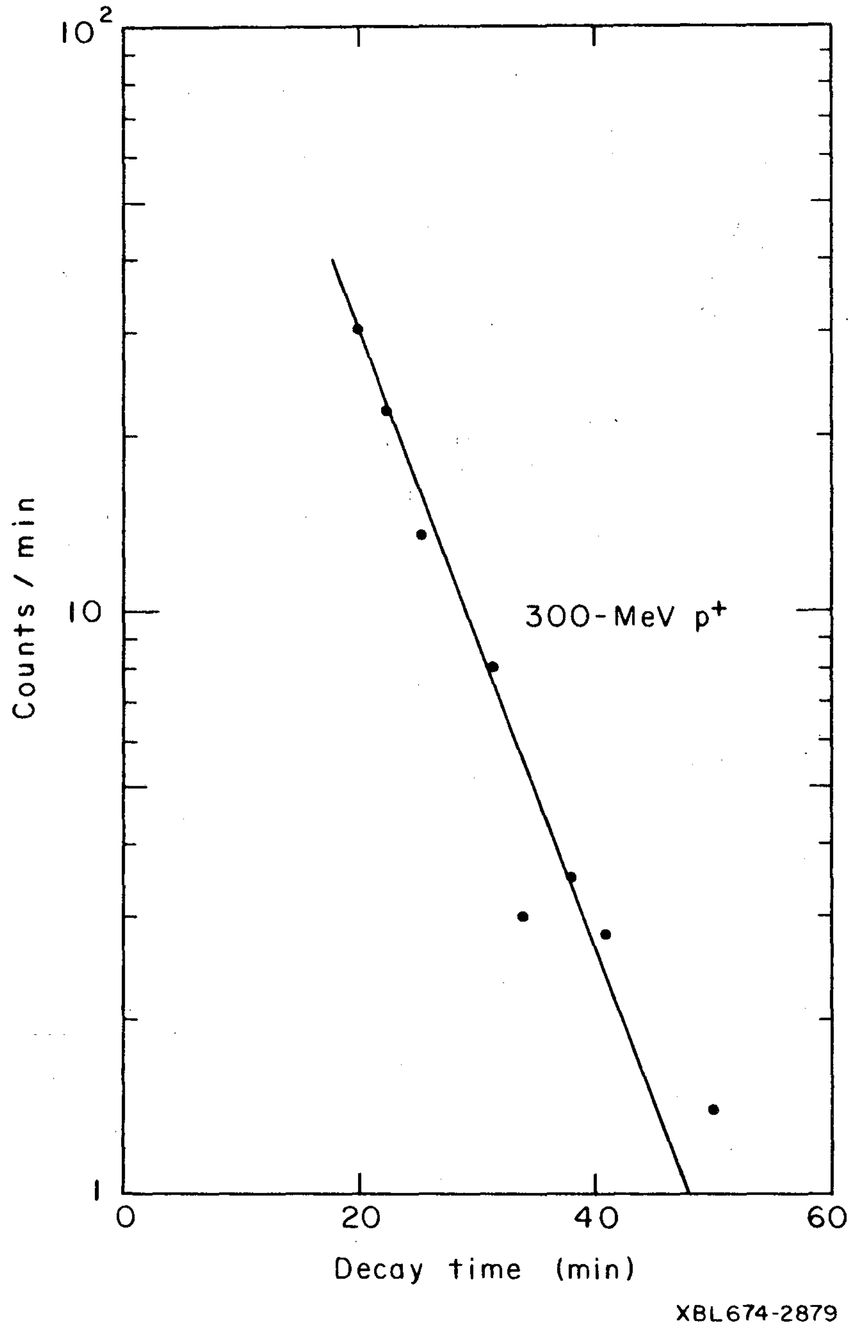

10^4
10^3
10^2
10
Counts / min
700-MeV p+
0
25
50
75
100
125
150
175
200
225
250
275
300
Decay time (min)
XBL674-2878

Fig. 2



10²
10
1
Counts / min
300-MeV p⁺
0
20
40
60
Decay time (min)
XBL674-2879

Fig. 3

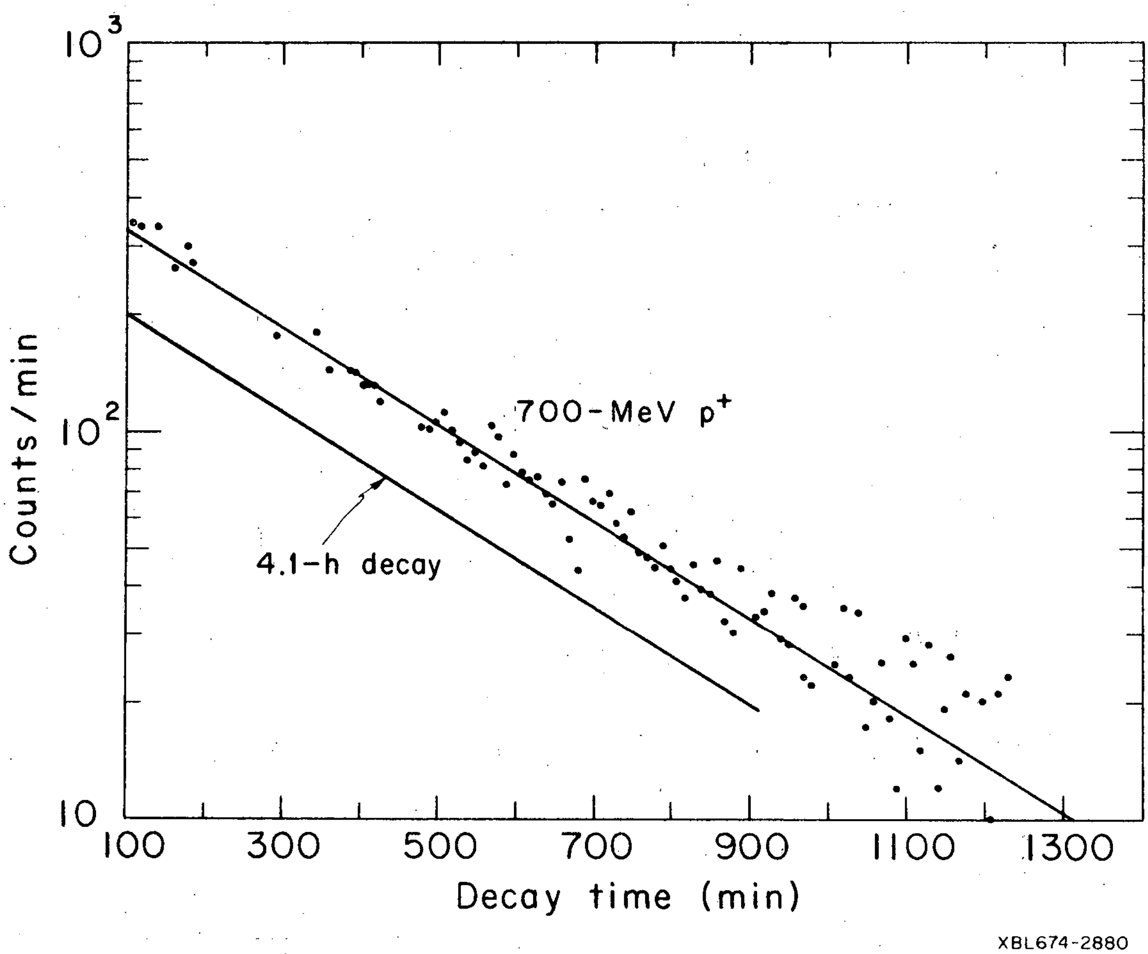
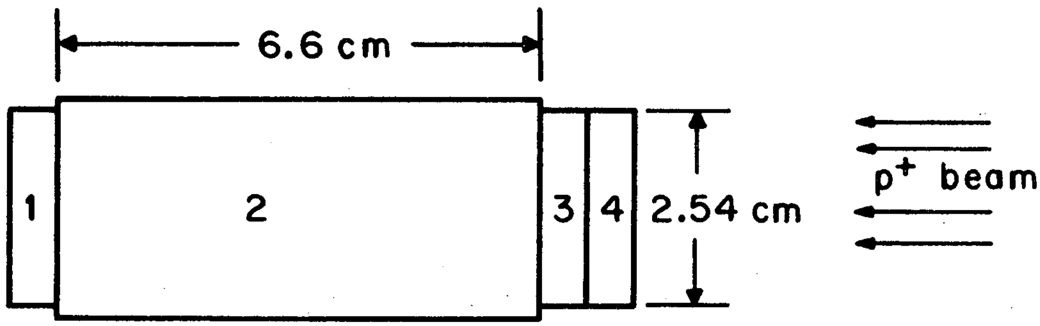

10^3
10^2
10
Counts / min
700-MeV p^+
4.1-h decay
100
300
500
700
900
1100
1300
Decay time (min)
XBL674-2880

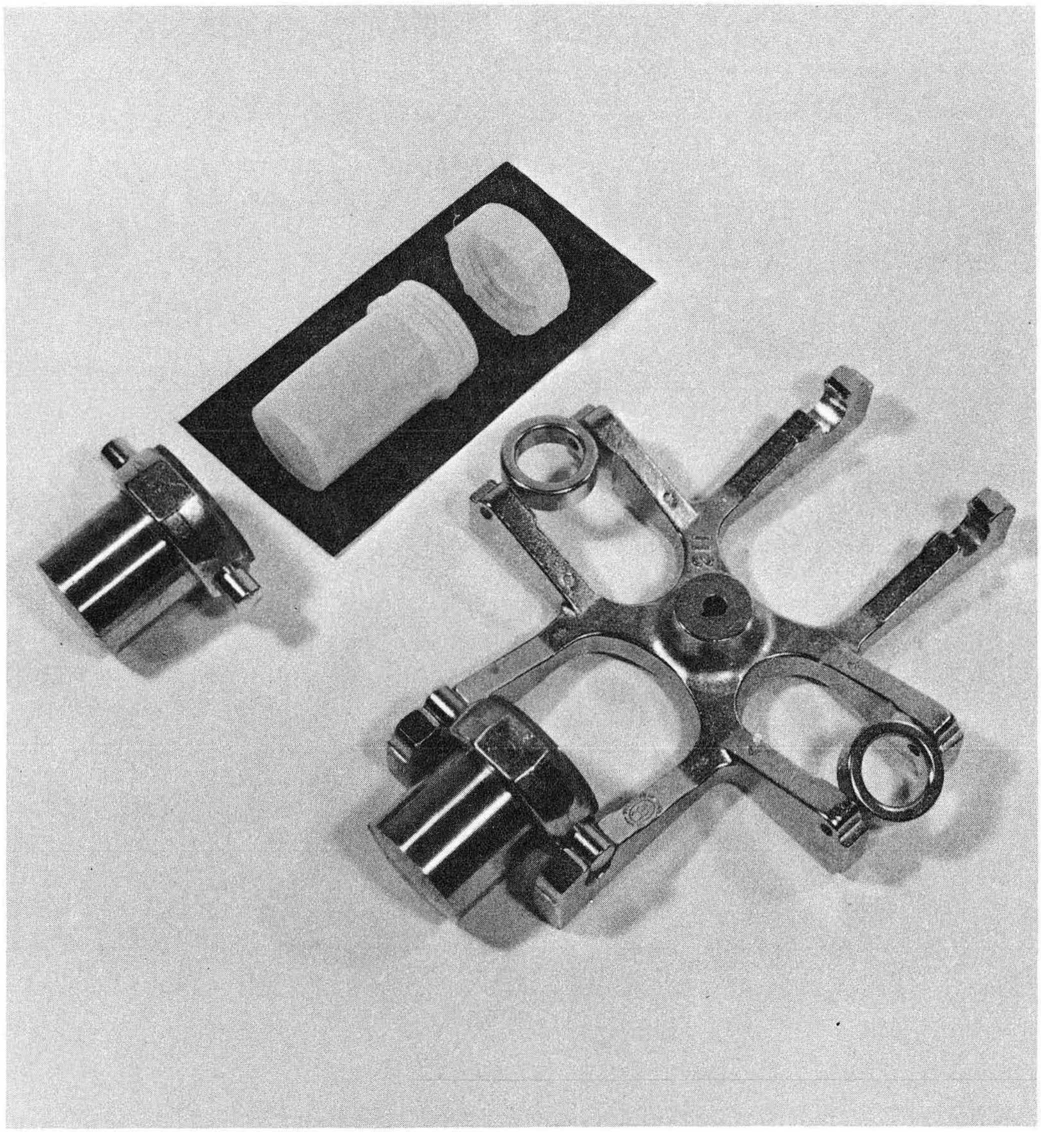
Fig. 4



1. 95.6-mg/cm^2-thick Au foil
2. 481 g mercury
3. 96.3-mg/cm^2-thick Au foil
4. Seven thin Au foils

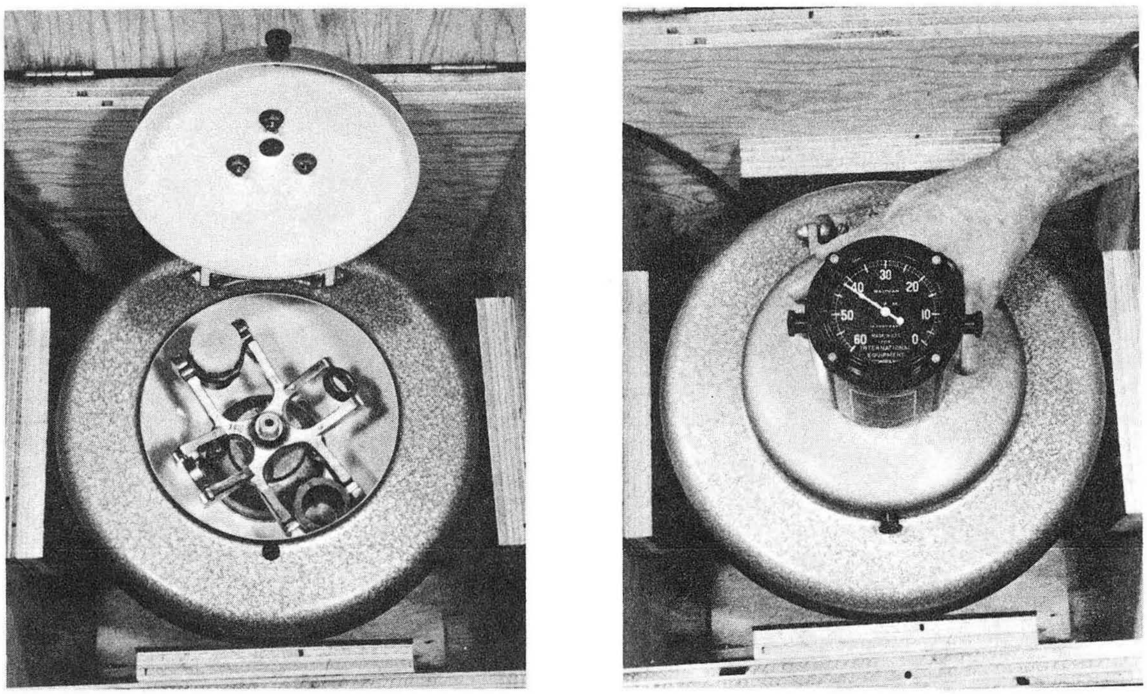
XBL674-2881

Fig. 5



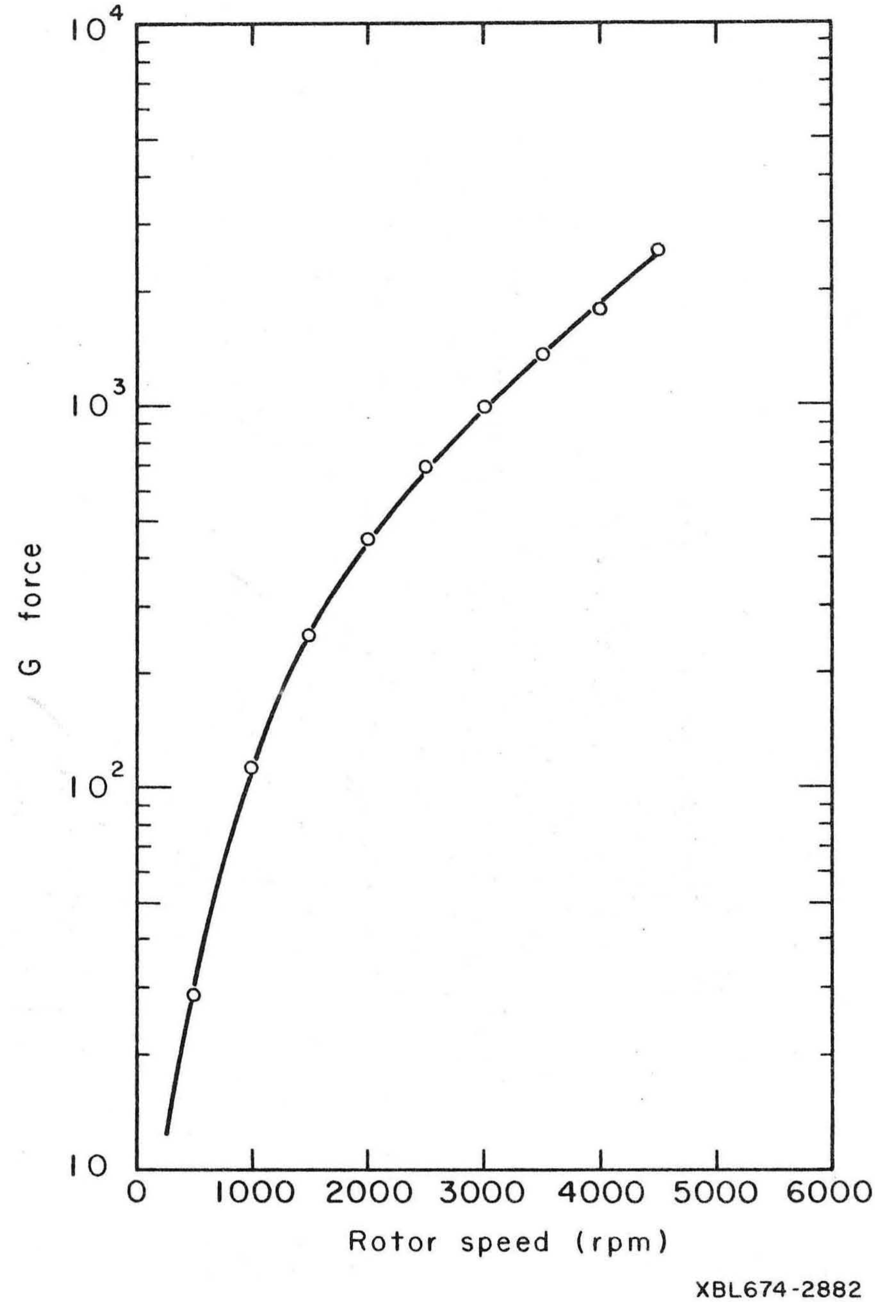
XBB 674-2203

Fig. 6



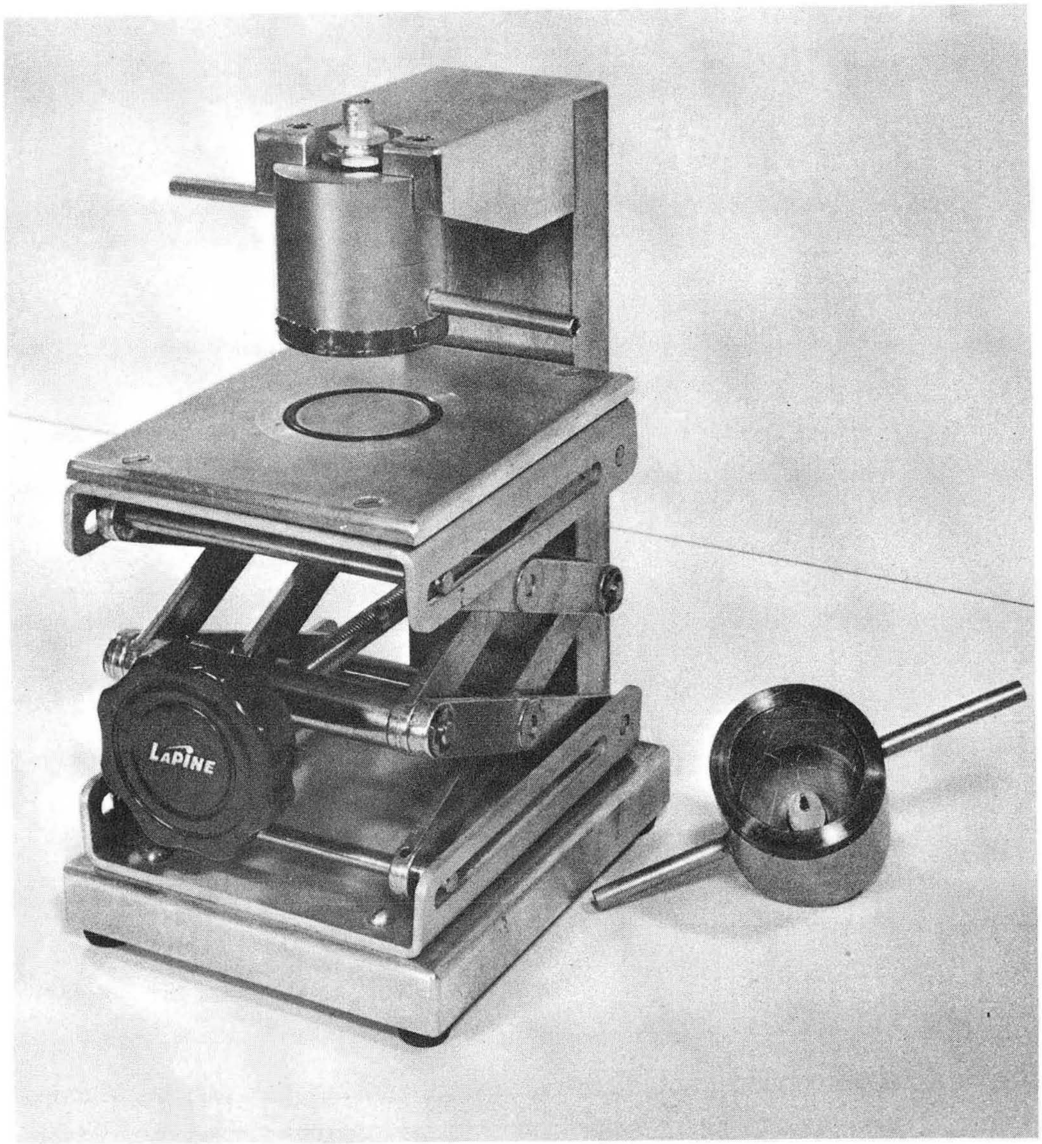
(a) (b) XBB 675-2614

Fig. 7



10^4
10^3
10^2
10
G force
0
1000
2000
3000
4000
5000
6000
Rotor speed (rpm)
XBL674-2882

Fig. 8



XBB 674-2202

Fig. 9

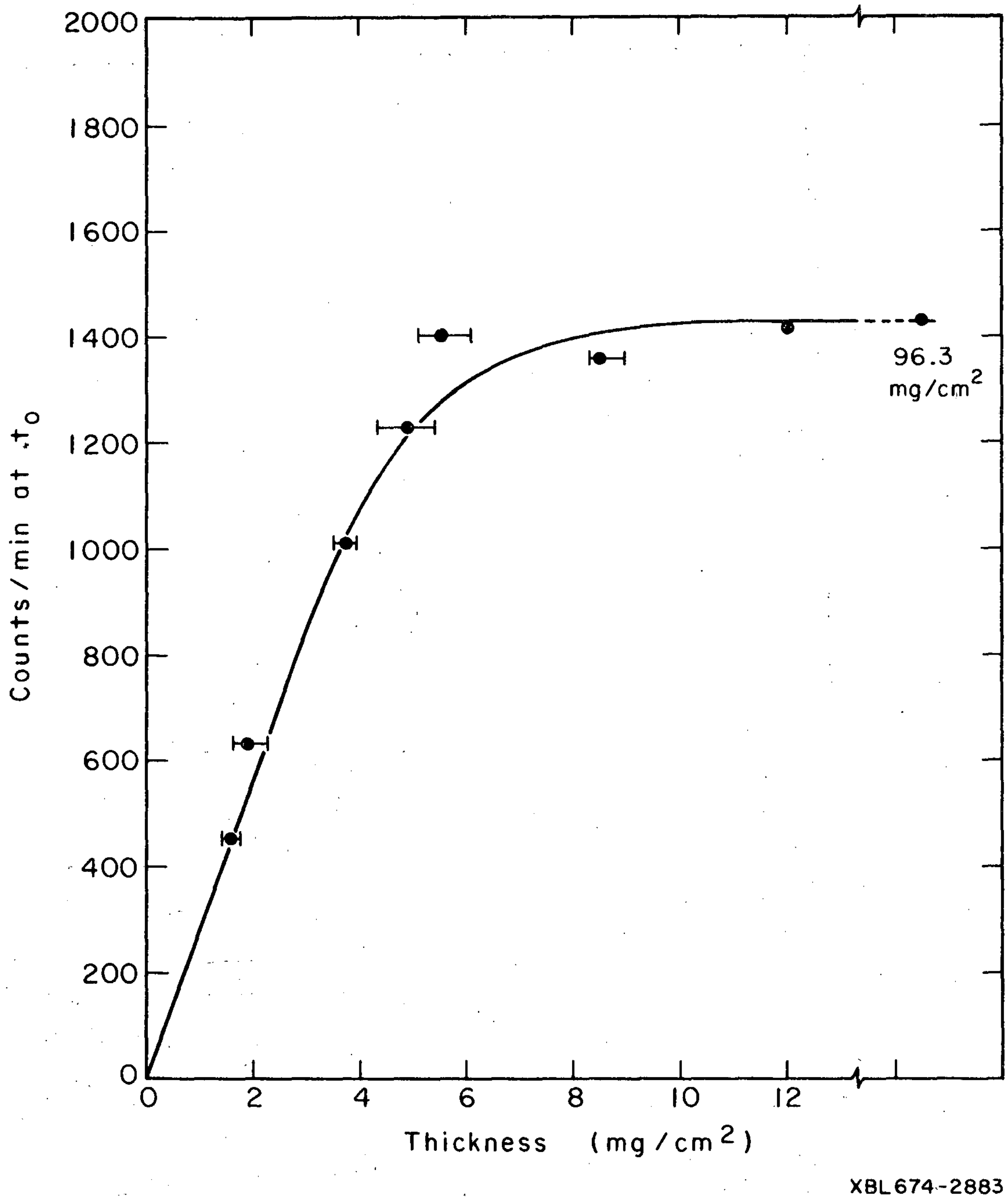

2000
1800
1600
1400
1200
1000
800
600
400
200
0
Counts/min at t_0
0
2
4
6
8
10
12
Thickness (mg/cm^2)
96.3
mg/cm^2
XBL674-2883

Fig. 10

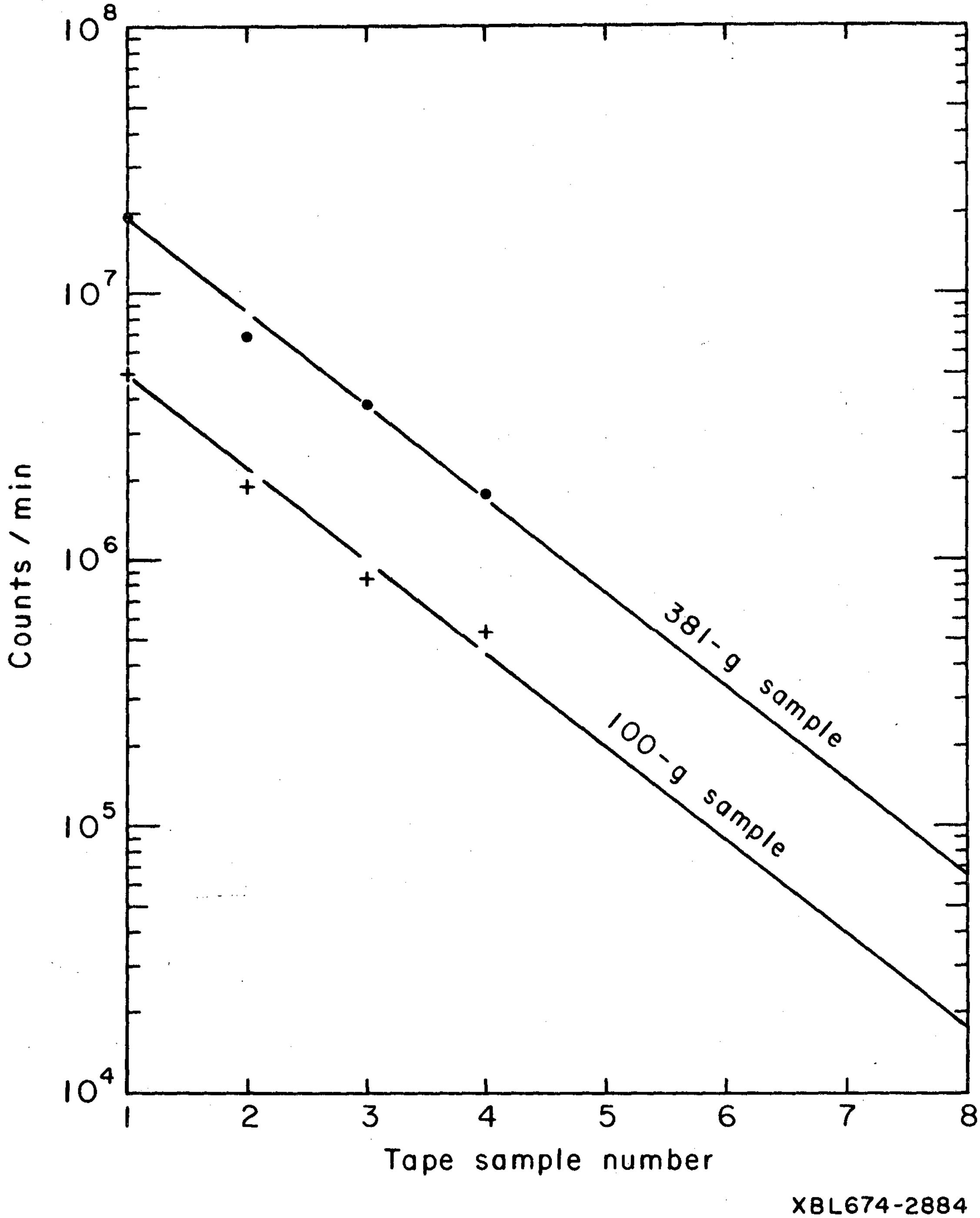
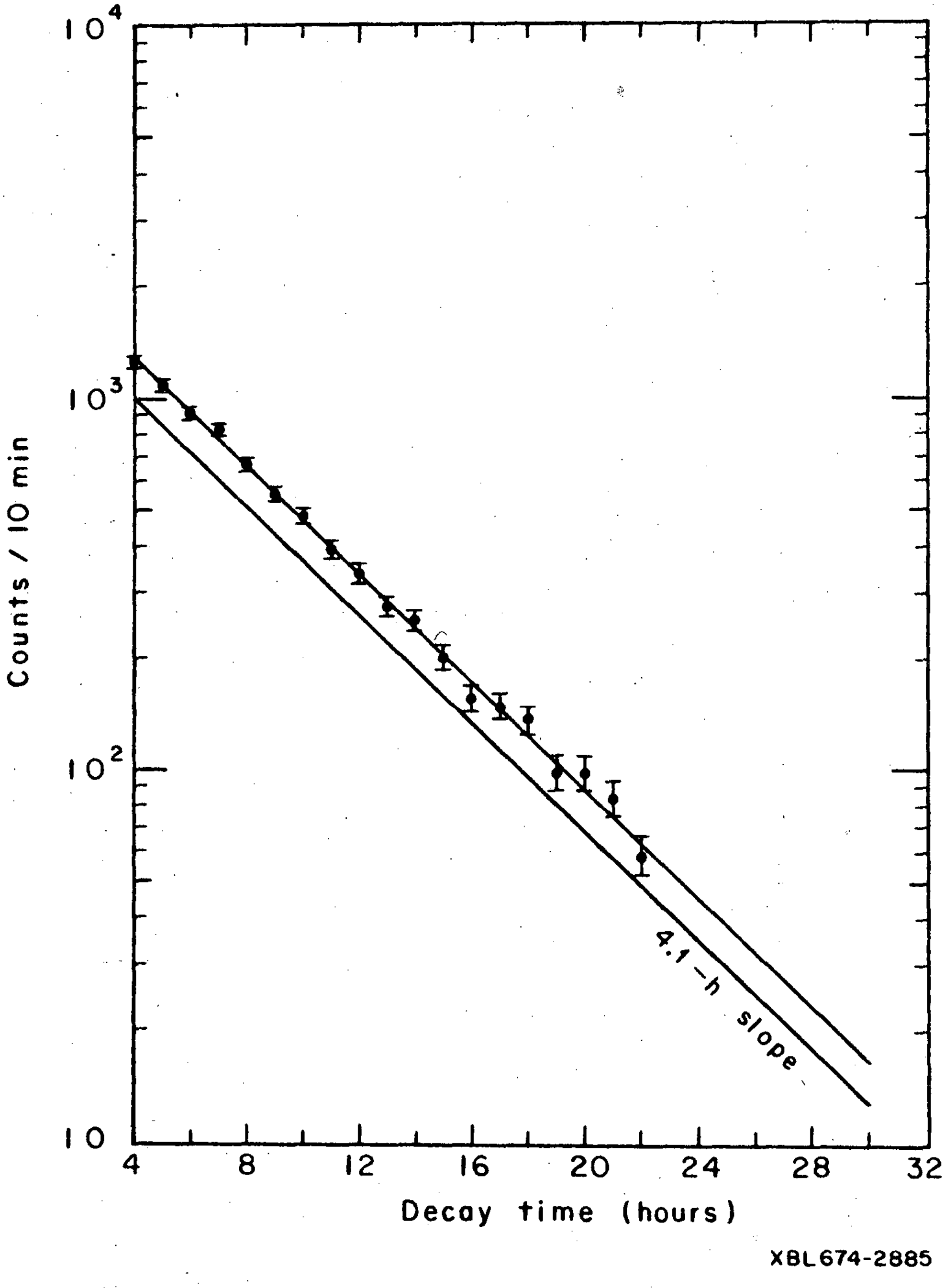
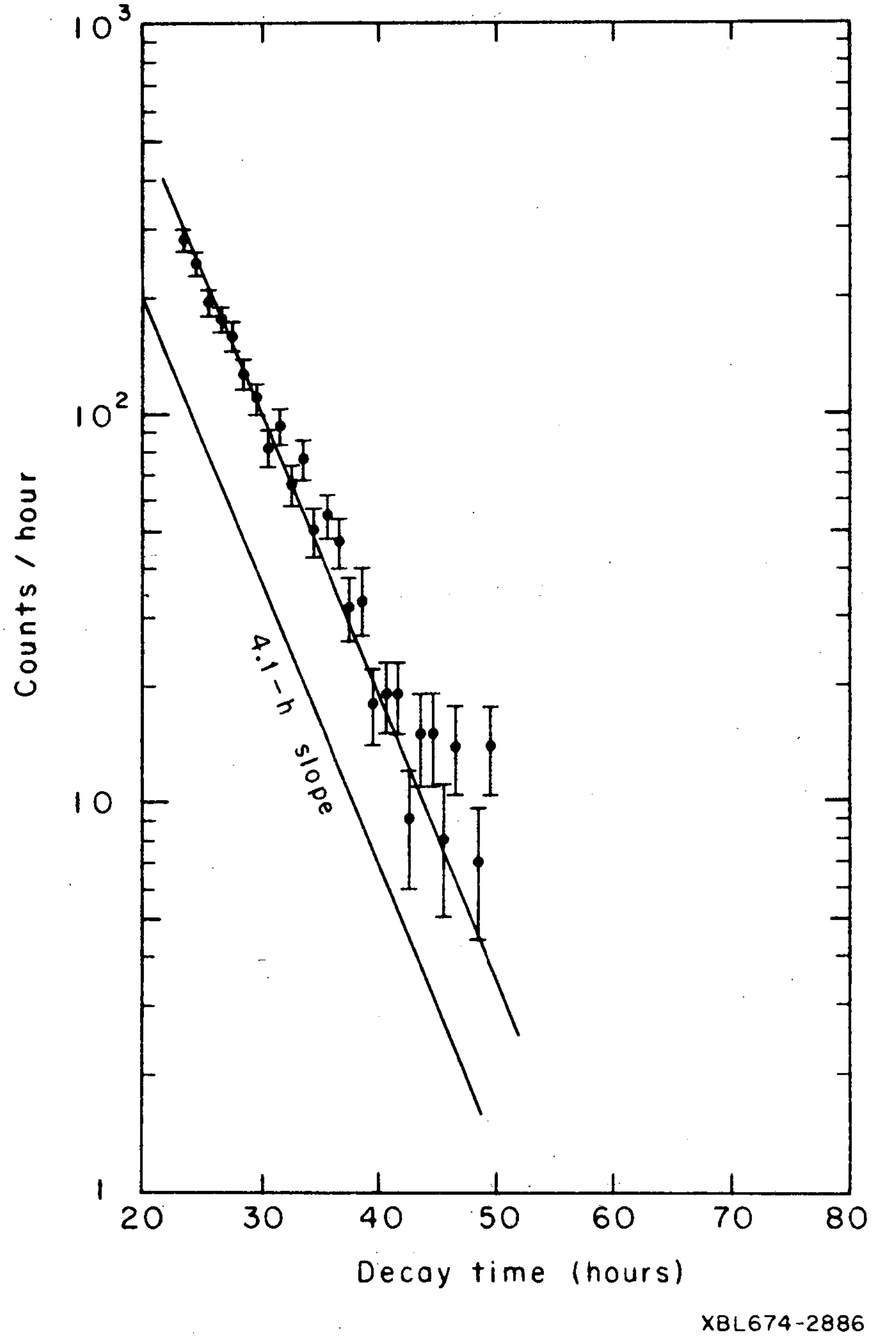

Counts / min
10^8
10^7
10^6
10^5
10^4
1
2
3
4
5
6
7
8
Tape sample number
381-g sample
100-g sample
XBL674-2884

Fig. 11



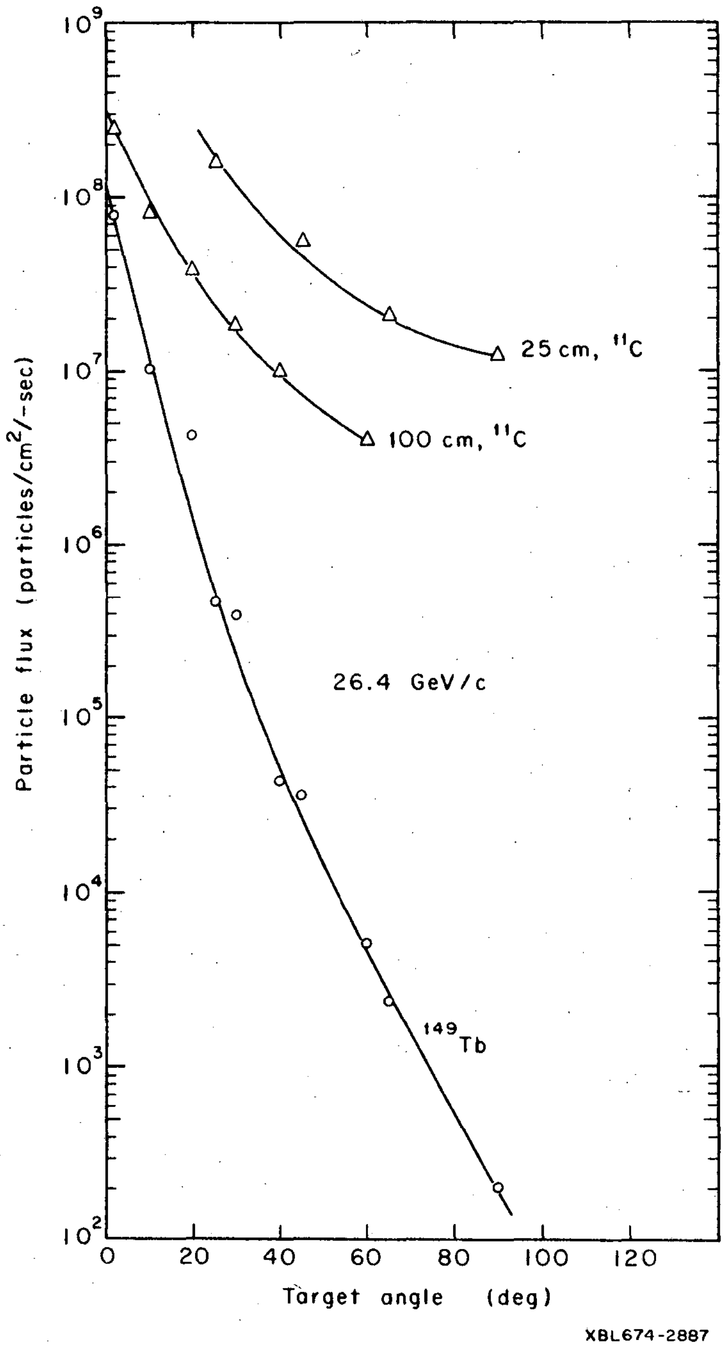
10⁴
10³
10²
10
Counts / 10 min
4
8
12
16
20
24
28
32
Decay time (hours)
4.1-h slope
XBL674-2885

Fig. 12



10^3
10^2
10
1
Counts / hour
4.1-h slope
20
30
40
50
60
70
80
Decay time (hours)
XBL674-2886

Fig. 13



10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
Particle flux (particles/cm²/-sec)
25 cm, ¹¹C
100 cm, ¹¹C
26.4 GeV/c
149Tb
0
20
40
60
80
100
120
Target angle (deg)
XBL674-2887

Fig. 14

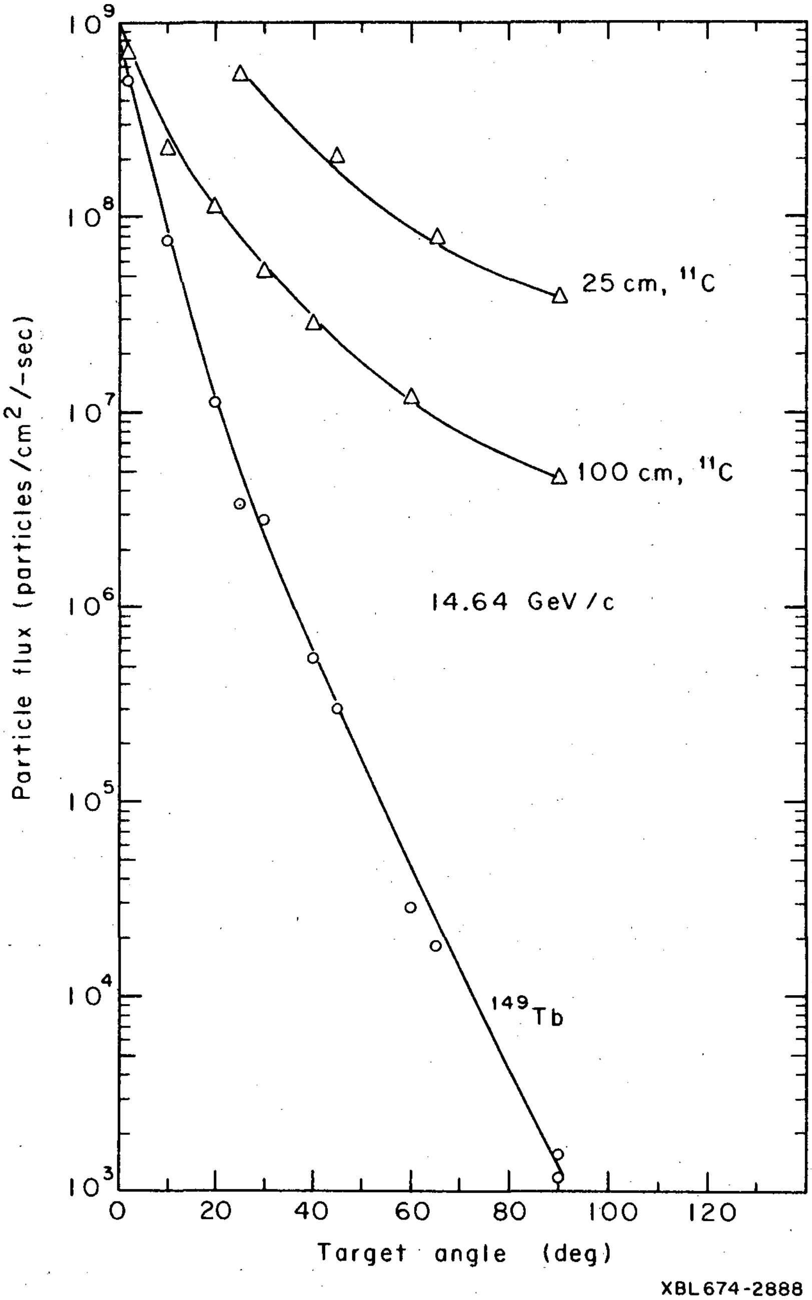

Particle flux (particles/cm²/-sec)
10⁹
10⁸
10⁷
10⁶
10⁵
10⁴
10³
0
20
40
60
80
100
120
Target angle (deg)
25 cm, ¹¹C
100 cm, ¹¹C
14.64 GeV/c
¹⁴⁹Tb
XBL674-2888

Fig. 15