

PUERTO RICO NUCLEAR CENTER

A SIMPLE DEVICE FOR HALF-LIFE MEASUREMENTS
OF HIGH GAMMA RAYS EMITTERS



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"A Simple Device for Half-Life Measurements of High Gamma Rays Emitters"

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Work performed at Puerto Rico Nuclear Center,
Mayaguez, P. R., under U. S. Atomic Energy
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ABSTRACT

The basic principle involved is the disintegration of deuterium by high energy gamma photons to produce fast neutrons, which are moderated by an optimum thickness of paraffin to achieve the best balance between moderation and capture. The moderated neutrons are detected by a BF_3 counter.

The detector is made of two concentric polyethylene bottles with a 2 mm. gap filled with D₂O. The outer bottle has a diameter of 12.4 cm. The inner bottle is filled with paraffin and a 1.8 cm. hole drilled to locate a BF_3 counter.

The half-lives of Na^{24} , S^{37} , Ca^{49} , Mn^{56} , As^{76} , and fission products from the P.R.N.C. fuel elements have been measured.

^{*}Operated by the University of Puerto Rico for the Atomic Energy Commission.

INTRODUCTION

The results of other workers^(1,2,3,4,5,e) with neutrons produced in the photo disintegration of deuterium, indicate the possibility of studying some nuclide properties, using those neutrons. For this it is of interest to develop a simple γ neutron conversion device.

Two types of interaction contribute to the cross section of the $D(\gamma,n)p$ reaction: photoelectric and photomagnetic interactions. In both cases the incident γ ray interacts with only the proton. Fig. 1 shows the cross section for the disintegration of D by electric dipole absorption. The maximum cross section of 2.23 mb. occurs at $E = 4.46$ mev.

Monoenergetic γ rays produce a monoenergetic group of neutrons. It can be shown that the energy E_n (in mev) of the emitted neutrons from a deuterium target is approximately equal to:

$$E_n = 1/2 [E_\gamma - E_t] \dots \dots \dots (1)$$

where, E_t = is the threshold energy (in mev) for the (γ,n) reaction and E_γ is the photon energy up to 10 mev.

Since for the $D(\gamma,n)H$ reaction, $E_t = 2.23$ mev., a rough estimate of the photon-neutron energy in terms of the energy of the γ rays emitted from various radioisotopes can be obtained from eq. (1). Although most of yields from $D(\gamma,n)p$ have been obtained^(4,5) these will change with the geometry and distance of the source from the detector.

1. E. Bell and G. Elliot, Phys. Rev. 79,282 (1950)
2. R. Mobley and R. Laubenstein, Phys. Rev. 80,309 (1950)
3. R. Sher J. Halpern and AkMann., Phys. Rev. 84,387 (1951)
4. A. Wattenberg, Phys. Rev. 71,497 (1947)
5. S. Russel, D. Sachs., A.Wattenberg and R.Fields: Phys. Rev. 73,545 (1958)
6. S. Bernstein, N.S. Preston, G.Wolf and R.Slattery Phys. Rev. 71,545 (1947)

EXPERIMENTAL

A. DESIGN OF THRESHOLD DETECTOR: Since neutrons coming from the photodisintegration of D have energies of the order of a fraction of an mev., it is necessary to thermalize them prior to their detection with a BF_3 counter.

An experiment was performed to estimate the optimum thickness of paraffin required for maximum number of neutrons detected. Fig. 2 shows a diagram of the experimental set up. Fig. 3 shows the experimental results and it was concluded that a maximum count rate was obtained with a paraffin thickness of about 7 cm. Using a paraffin thickness of 5.5 cm. a good balance between moderation and capture was achieved.

Fig. 4 shows the details of construction of the threshold detector. It is made of two concentric polyethylene bottles with a 2 mm. gap filled with D_2O . The outer bottle has a diameter of 12.4 cm. The inner bottle is filled with paraffin and a 1.8 cm. hole drilled at the center to locate a BF_3 counter.

B. IRRADIATIONS: The γ -ray emitting isotopes were activated in the PRNC reactor operated at the power level of 1MW. and at a flux of $2 \times 10^{12} \text{ n cm}^{-2} \text{ sec}^{-1}$. Either the swimming pool or the pneumatic system facilities were used. The activities obtained were up to 0.5 C.

C. MATERIALS: The irradiated materials were analytical or reagent grade NaHCO_3 , Mn powder, CaO, S, Al_2O_3 , Se and $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$.

D. COUNTING PROCEDURES: Depending upon the γ source, each experiment was performed as follows:

1. Determination of the background in the absence of the source.
2. Determination of any ionization in the BF_3 counter due to the γ radiation. It was noticed that at 150v.

above the plateau, γ -rays affected the BF_3 counter.

3. A system similar to the threshold detector, but without D_2O was placed near the source. If the source was not fission products, the count rate due to photodisintegration in paraffin was negligible.

If the gamma source was the fission products a waiting time was required for the decay of all delayed neutrons.

RESULTS AND DISCUSSION

Significant response of the detector to γ rays, indicates that such photons are above the threshold of 2.23 mev. In such a case, the detector can be used to measured accurately half-lives of individual or mixtures of γ emitters. Furthermore, this technique permits the investigation of new high energy γ rays.

a) Na^{24} and Mn^{56} : The decay rate of these radionuclides was measured together. Both sources were placed in such a way that each radionuclide contributed to about the same count rate.

Fig. (5) shows the decay curve resolved into its components. The half-lives measured, 15 hr. for Na^{24} and 2.58 hr. for Mn^{56} agreed with those measured by standard procedures⁽⁷⁾.

b) Ca^{48} : This isotope was chosen to confirm the existence of its high energy γ ray recently reported⁽⁸⁾. Fig. (6) shows the

7. D. Struminger, J. M. Hollander and G. T. Seaborg. Rev. of Mod. Phys., 30, 585 (1958)

8. G. D. O'Kelley, N. H. Lazar, and E. Eichler Phys. Rev. 101, 1059 (1956).

decay curve. The half-life of 8.8 min. measured, agreed with that obtained by standard procedures.

c) A_s^{76} : Three γ rays over 2.23 mev. are reported for $As^{76(9)}$

The time of irradiation was 4 hr, and the amount 1.5 g. The count rate was followed for 72 hr. Fig. (7) shows the decay curve; the half-life measured was 26.8 hr., which agreed with that reported⁽⁷⁾ for As^{76} .

d) S^{37} : A single experiment was performed irradiating sulphur.

Since the cross section and isotopic concentration of S^{36} are very low, the count rate after 55 min. irradiation was also low. Fig. (8) shows the decay curve. The half-life of 5.1 min. measured, agreed with other reported data⁽¹⁰⁾.

e) Se^{83} : Several irradiations were performed, using up 10 g. of natural Se. No evidence was found of the existence of the γ ray of 2.29 mev⁽¹¹⁾.

FISSION PRODUCTS DECAY

Three experiments were performed to study the fission products decay rate:

1. With the reactor shut down, a survey was made with the detector in the pool to find a position where the background was about 100 c/min., then a fuel element from the reactor was placed at about 2 ft. from the detector. Fig. 9 shows the gross

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9. R. Girgis, R.A. Ricci and R.V. Lieshart, Nucl. Phys. 13,461 (1959)
 10. J. G. Elliot and F.C. Young, Nucl. Sc. and Eng. 5,55 (1959)
 11. R. G. Cochran and W.W. Reatt, Phys. Rev. 113,852 (1959)

activity and the four components resolved from the curve. The half-lives of these components were 54 hr., 4.4 hr., 1.6 hr., and 27 min.

2. One gram of uranyl nitrate was irradiated for 1 min. Measurements were made 2 min. after irradiation so that the decay of the delayed neutrons would be negligible with respect to the longer life photoneutrons. Fig. 10 shows the gross activity and the three components resolved from the curve. The half-lives of these components were 27.5 min., 7.3 min, and 2.25 min. This experiment confirmed the existence of a 27 min. activity and showed another activity much shorter than 2.25 min. The shorter ones were difficult to resolve because of the presence of delayed neutrons.

To assign a nuclide to each measured half-life, the following characteristics of fission products were considered:

1. Half-lives taken from the references^(7,12) were compared.

12. W. Seelmann - Eggebert et al. - Nuklidkarte mit Erläuterungen
-2 Auflage -Karlsruhe (1961).

J. Blomeke and M. Todd - ORNL 2127 (1957)

G. N. Walton: Atomic Energy Waste (Edited by E. G. Lueckauf)

3-97 - Butterworths (1961).

2. The radioisotopes with γ rays over 2.23 Mev were chosen.
3. In some cases, comprehensive data of the characteristics of the radioisotope is not yet available. In such cases those nuclides with total disintegration energy over 2.23 Mev⁽¹³⁾ were chosen.
4. The fission yield of the isotope, was also taken into account⁽¹²⁾.

$T_{1/2} = 1.3$ min: No nuclide with this half-life was reported. However, Rb^{90} and Kr^{88} with $T_{1/2} = 2.9$ and 2.8 min., and γ rays over 2.23 Mev are known⁽¹²⁾. It can be assumed that these nuclides are measured together with other of shorter half-lives. It should be kept in mind, that this half-life was difficult to resolve, due to the delayed neutrons activity.

$T_{1/2} = 7.3$ min: It agrees with that reported for Sr^{93} and Pm^{152} . However, for these isotopes, γ rays over 2.23 Mev are unknown. On the other hand, the total disintegration energy of these are 5.23 Mev and 4.16 Mev.

$T_{1/2} = 27.5$ min: This can attributed to a mixture of Cs^{138} (32 min.) and Y^{94} (20 min.).

$T_{1/2} = 16$ hr.: Also to a mixture of Kr^{87} and La^{142} .

$T_{1/2} = 4.4$ hr.: This agreed with those reported for Ru^{105} and Sb^{129} . However, they have no γ rays over 2.23 Mev. reported.

$T_{1/2} = 54$ hr.: The only radioisotope with high γ ray over 2.23 Mev., and with half-life in the neighborhood of 54 hr., is La^{140} , with $T_{1/2} = 40$ hr. The 54 hr. measured half-life, can be

13. A. G. Cameron, A revised semi-empirical Atomic Mass formula-
CRP 690 (1958).

explained for this isotope, only on the basis of an equilibrium with Ba^{140} in the fission chain. In such a case, however, the La^{140} activity obtained in fission would be 5 times greater than that for ^{140}Ba , which is not easy to accept. Therefore, this half-life should be attributed to a new fission product, with γ over 2.23 Mev. or an equilibrium with a daughter of these characteristics.

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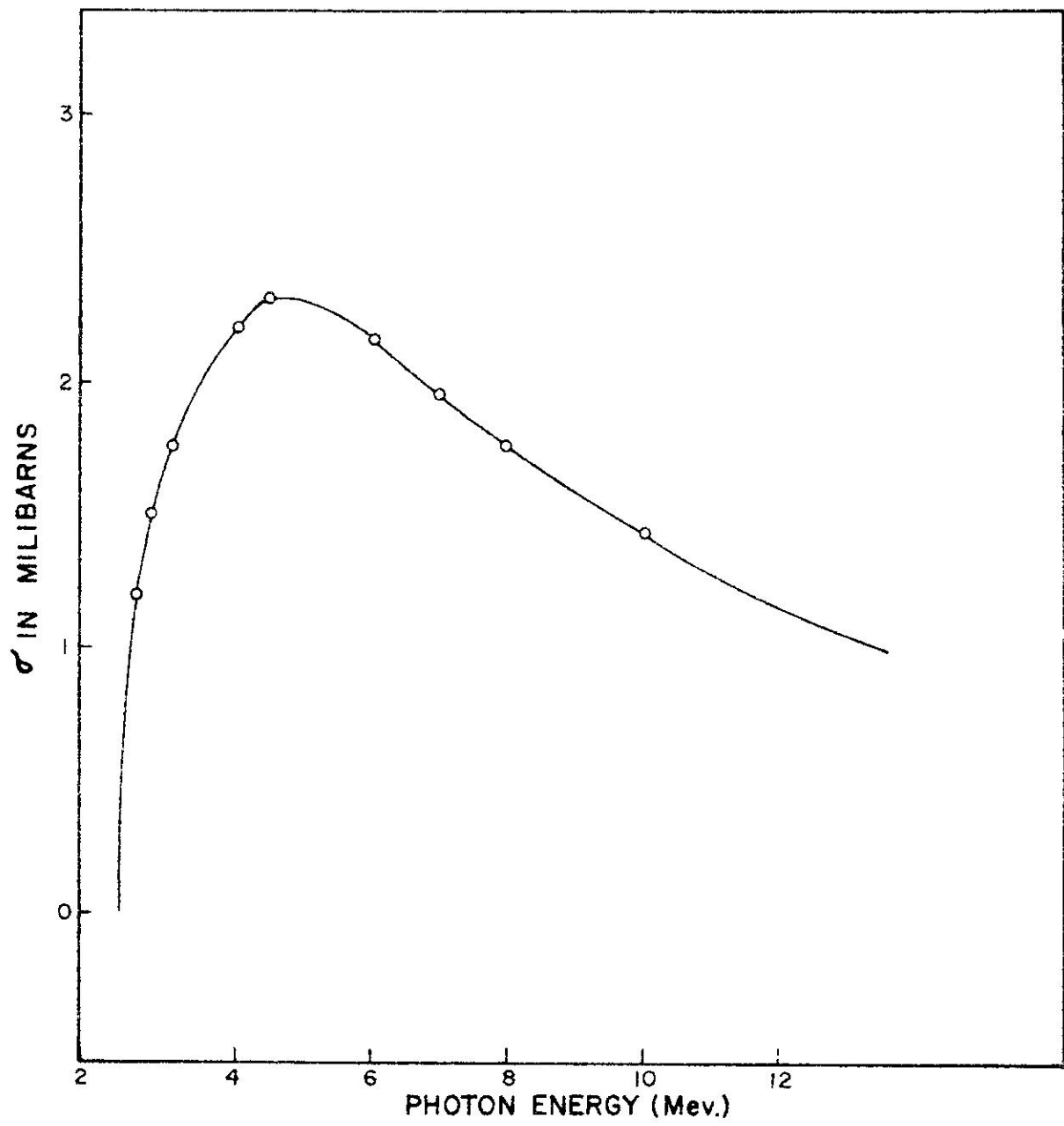


Fig. 1 Photo-electric cross section of the deuteron.

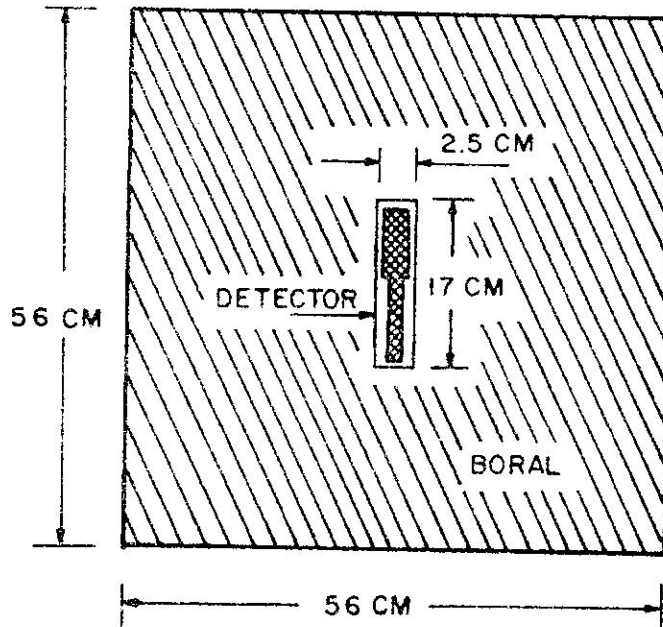
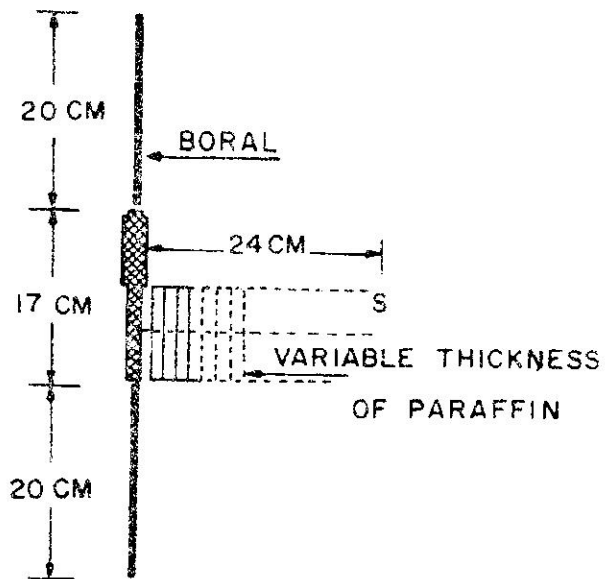


Fig. 2 Experimental set-up to obtain the optimum paraffin thickness.

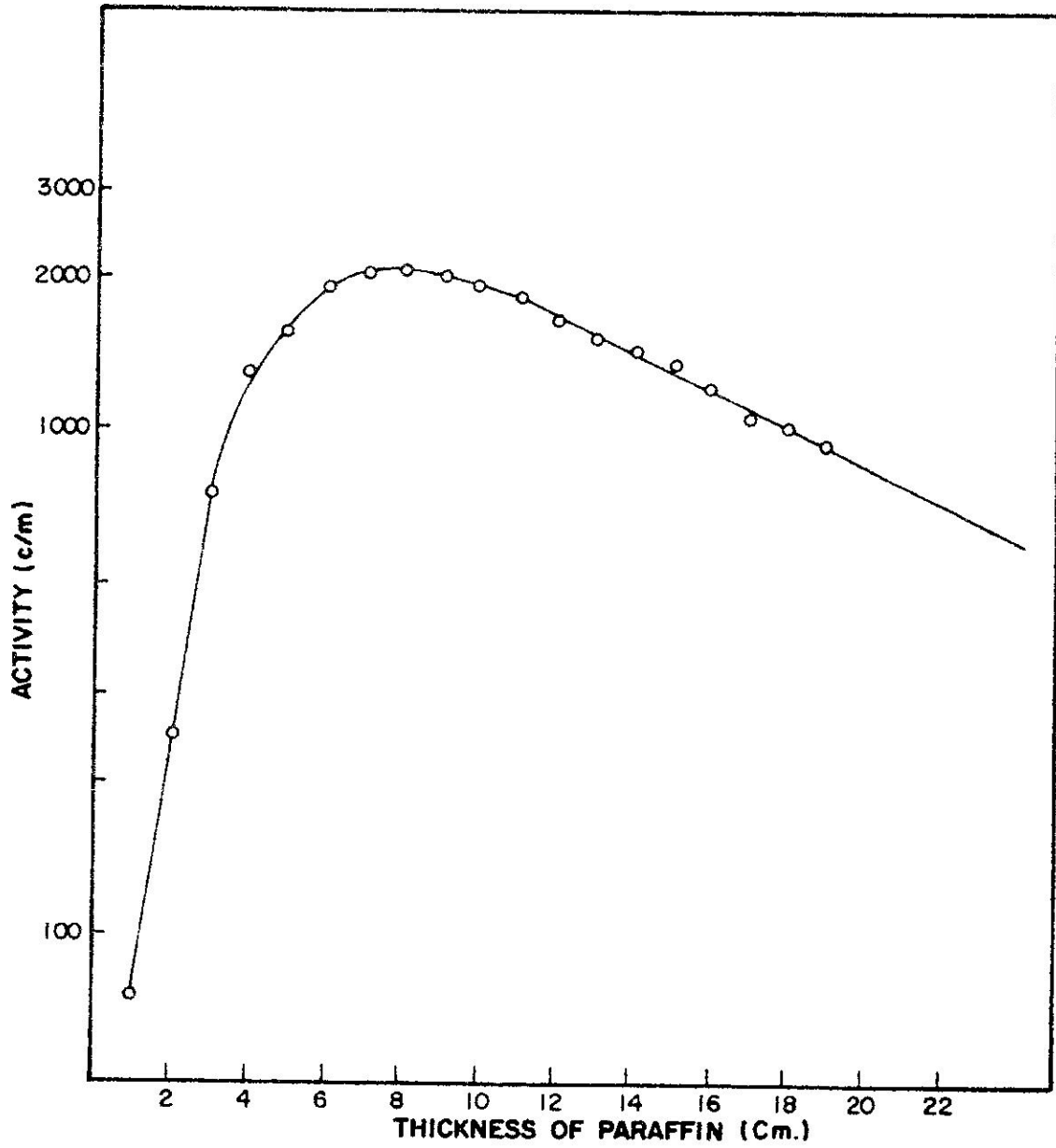


Fig. 3 Neutron attenuation curve for obtaining the maximum paraffin thickness.

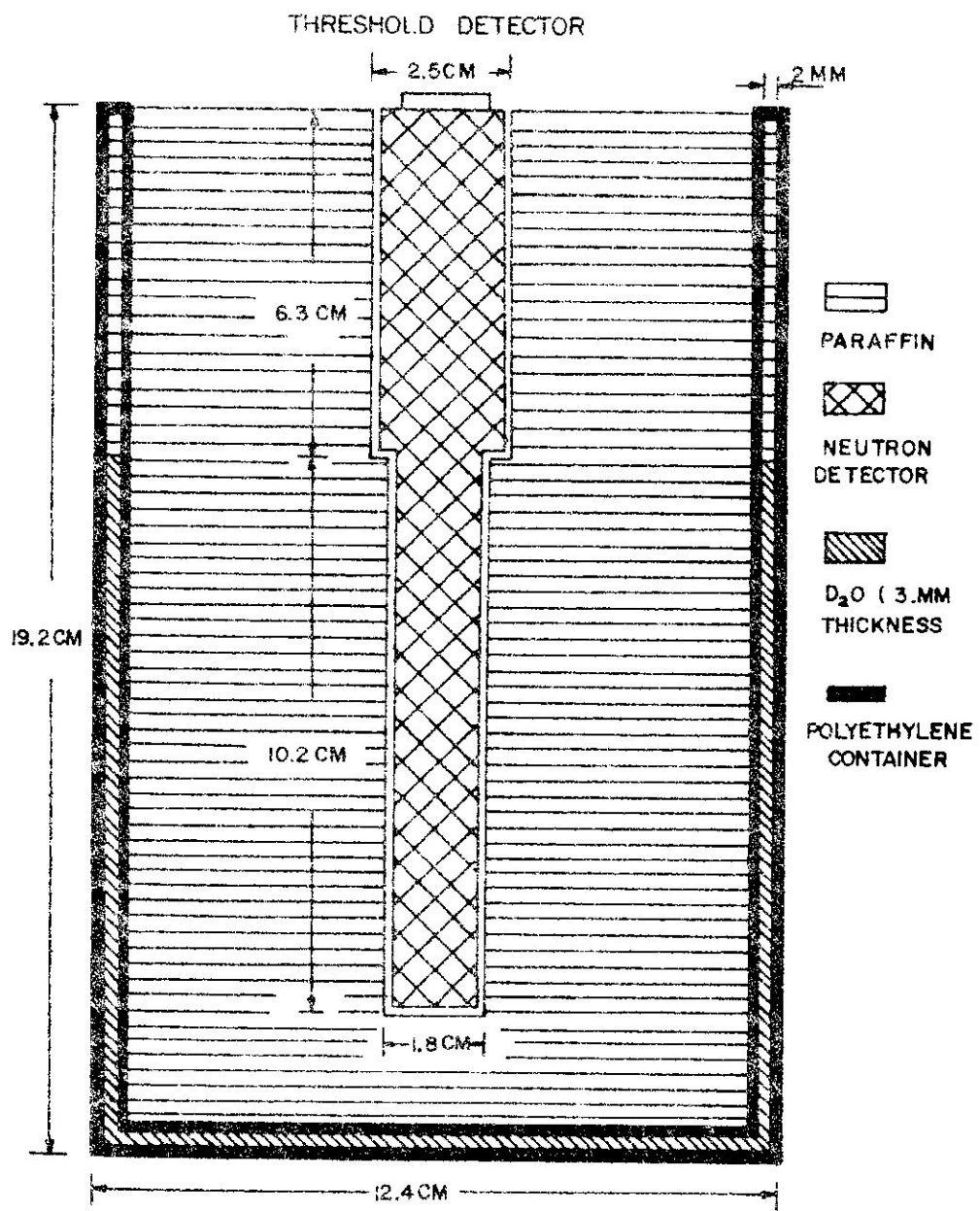


Fig. 4 Schematic diagram of the detector.

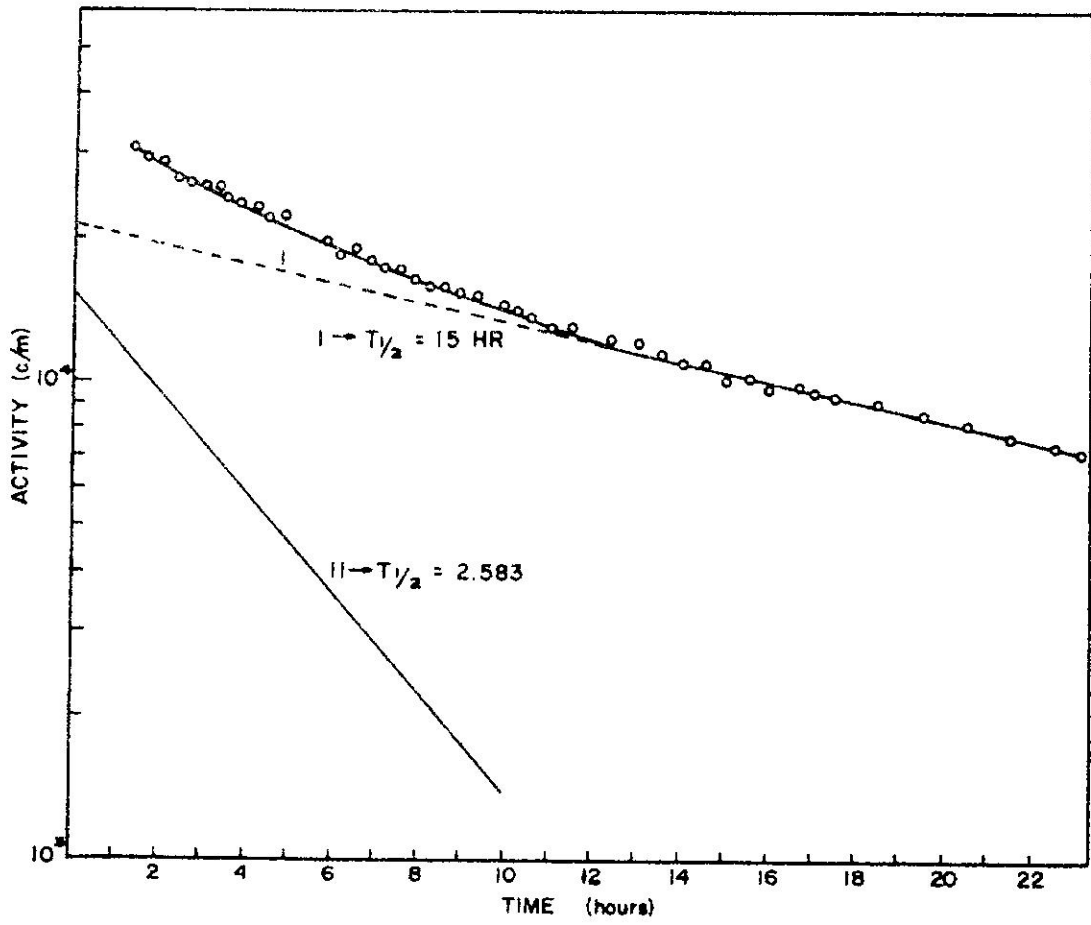


Fig. 5 Typical decay curve of Na^{24} and Mn^{56} .

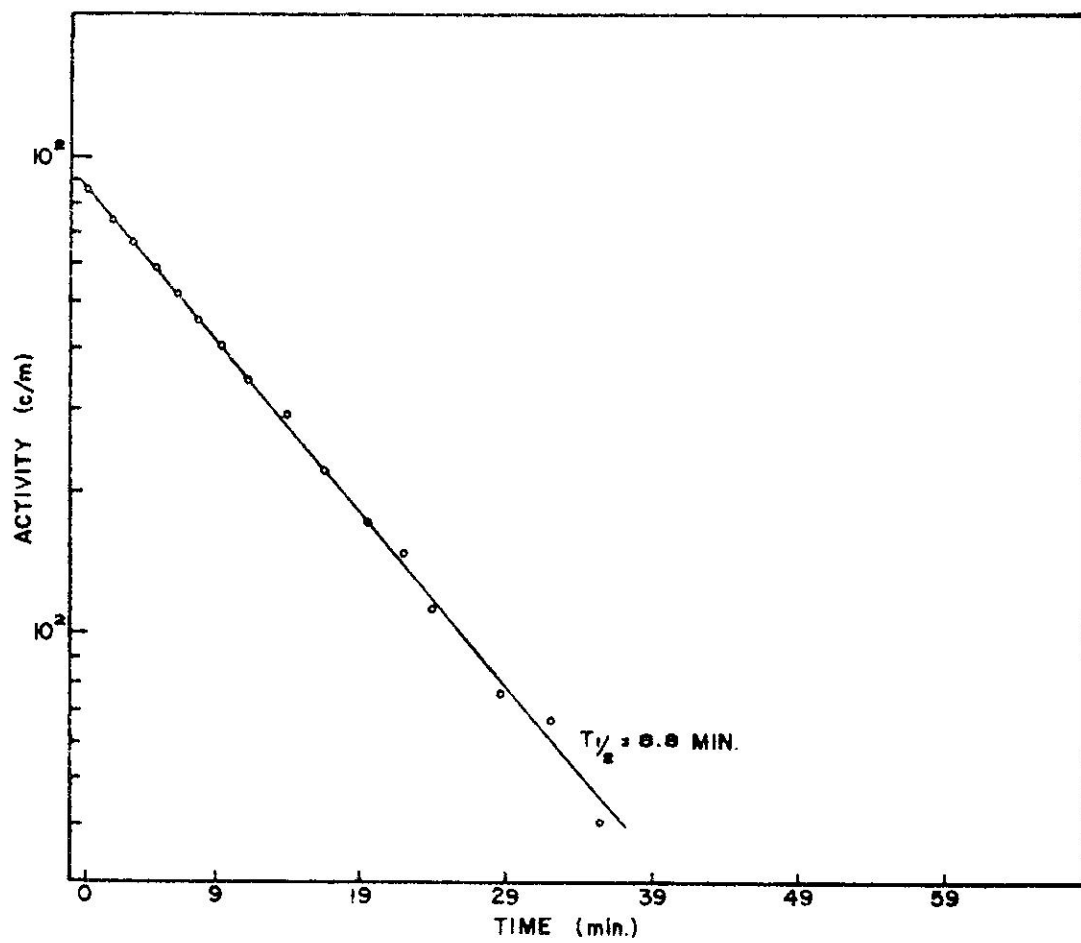


Fig. 6 Decay curve of Ca^{49} .

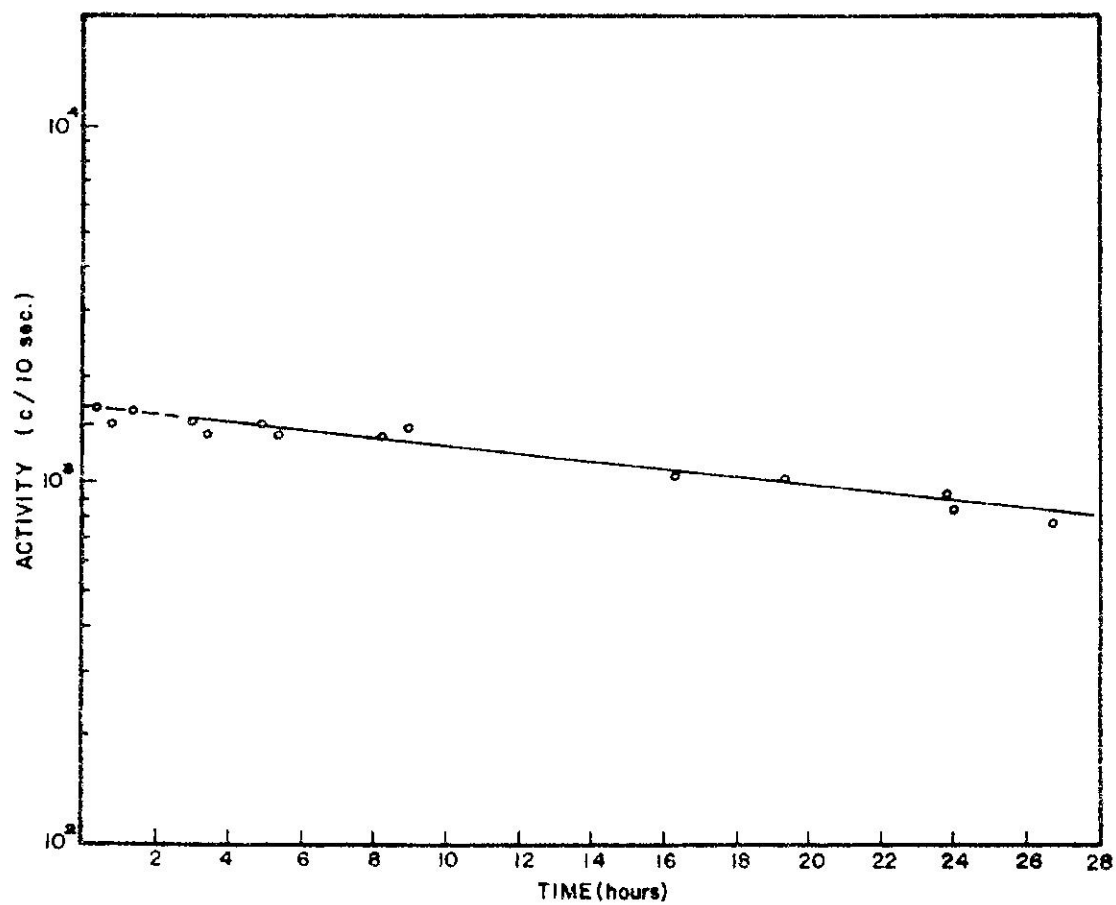


Fig. 7 Decay curve of As^{76} .

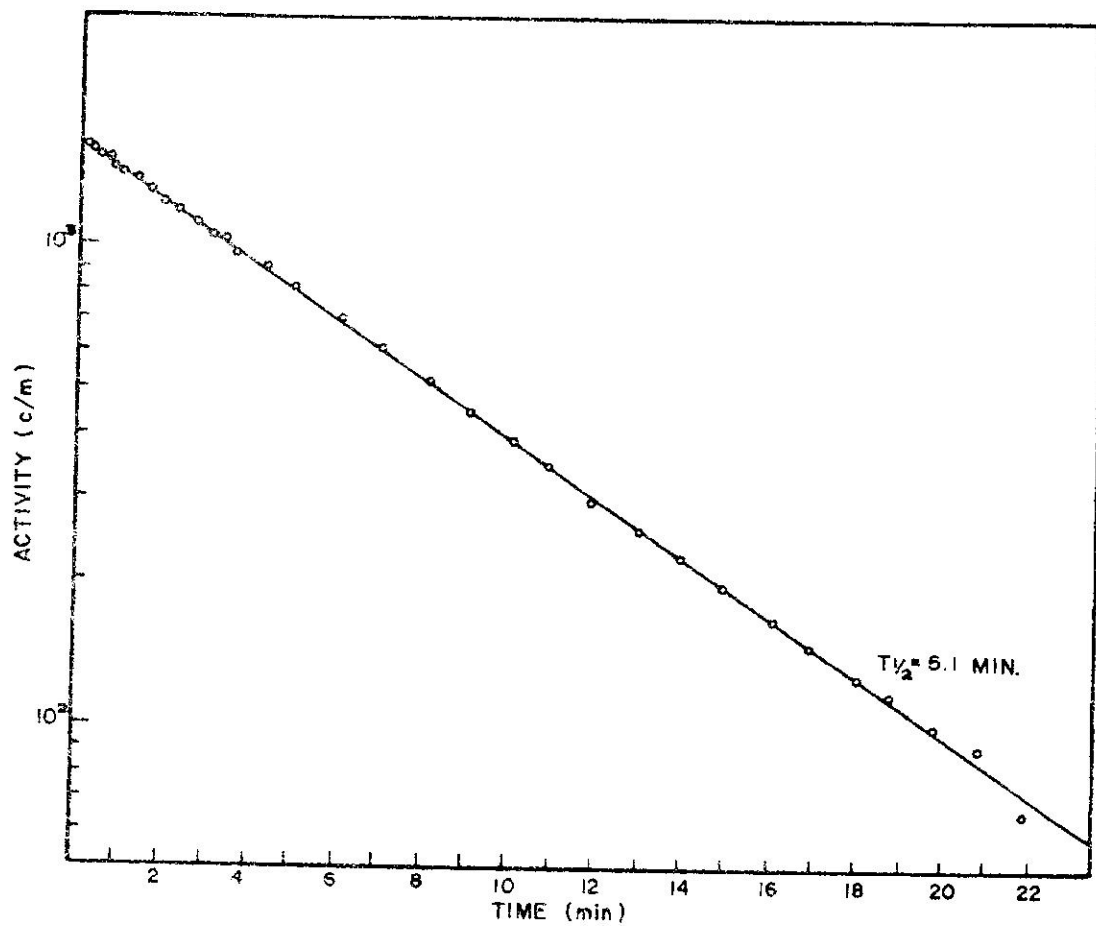


Fig. 8 Decay curve of S^{38} .

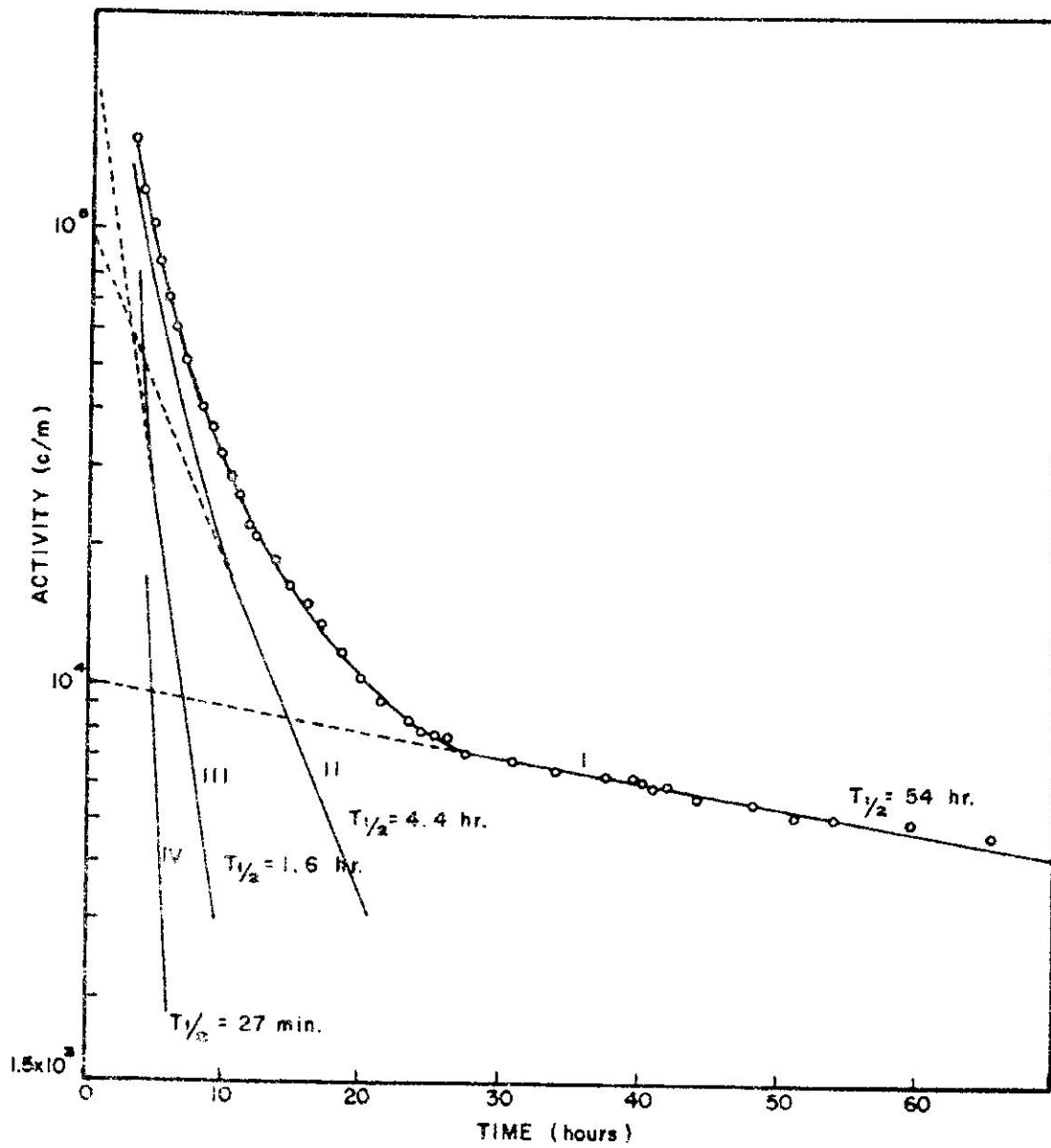


Fig. 9 Fuel elements decay curve.

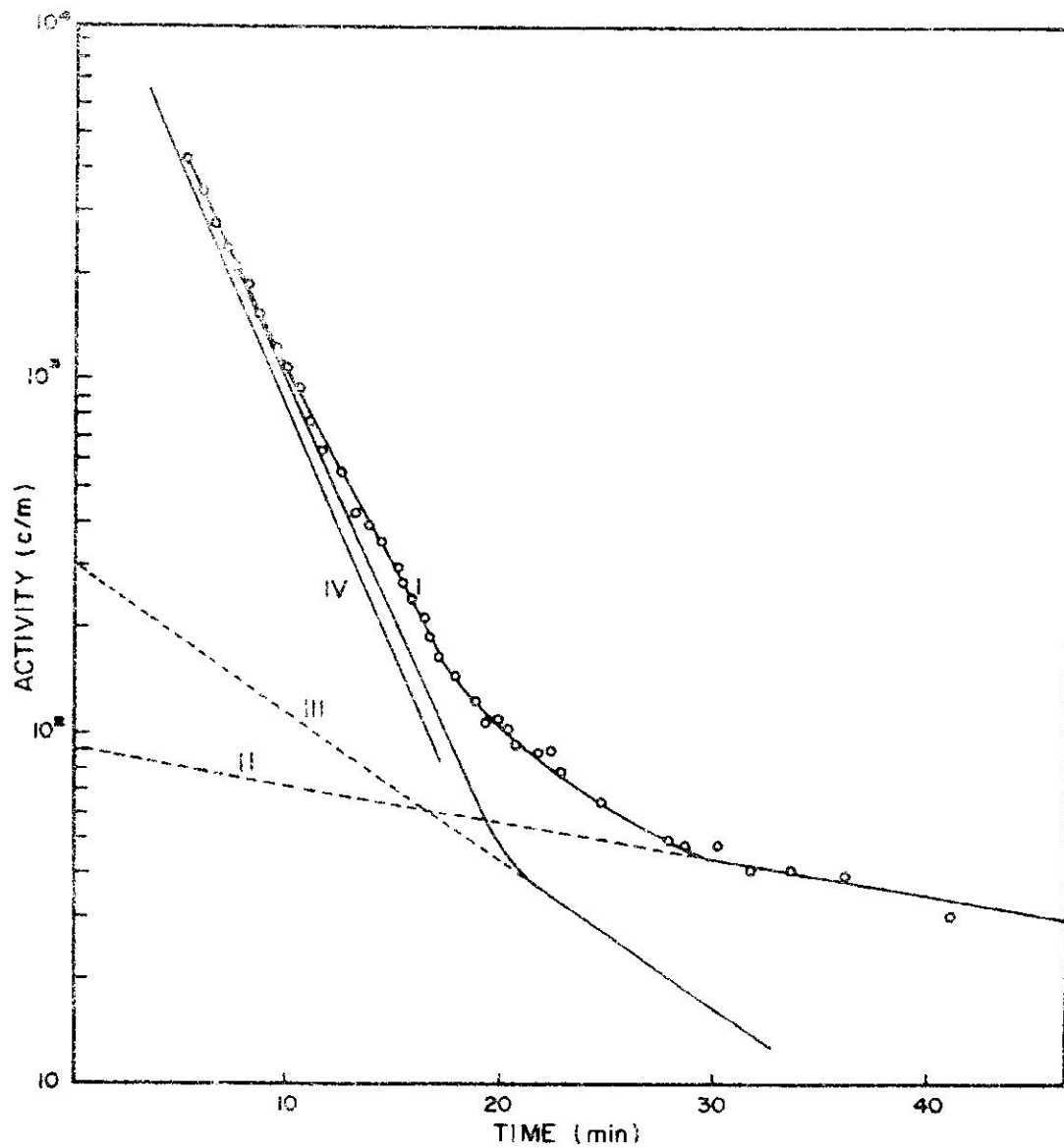


Fig. 10 Decay of the 1 min. exposure of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$. - Curve I: total activity; II: extrapolated activity of $T_{1/2} = 27.7$ min.; III: extrapolated activity of $T_{1/2} = 7.8$ min; curve IV : activity of 2.3 min.