

the differential cross section at 37° , the total cross section for inelastic scattering from the virtual state is $(5.0 \pm 1.0) \times 10^{-26}$ cm². The calculation, with ordinary forces, gave 4.5×10^{-26} cm², and it is not likely that the value for exchange forces would be widely different. It appears, therefore, that the neutron-well model

gives approximately the correct value for the product of width times height of the state, as defined by $|A_i|^2$.

The author is very grateful to the National Research Council for financial aid, and to Professor R. Marshak for suggesting and guiding the work reported.

Tc⁹² and Tc⁹³ by Relative Cross-Section Measurements

D. N. KUNDU* AND M. L. POOL
Ohio State University, Columbus, Ohio
(Received September 3, 1948)

Under 5-Mev proton and 10-Mev deuteron bombardments on natural Mo and on electromagnetically enriched Mo isotopes, activities of 2.7-hour, 47-minute, and 4.5-minute half-lives are found to be produced from Mo⁹². The relative cross sections for the production of these activities are 25:127:1, respectively. On comparing these figures with those for the (p, n) : (p, γ) reactions from Mo⁹⁵, the 47-minute activity is assigned to Tc⁹², and the 2.7-hour and the 4.5-minute activities to Tc⁹³. Tc⁹² decays by K -capture only attended with 1.5-Mev γ -rays. For the 2.7-hour Tc⁹³, the decay is 93 percent by K -capture and 7 percent by emission of positrons of maximum energy 0.83 Mev. There are also γ -rays of energy 2.0 Mev associated with this activity.

I. INTRODUCTION

A NUMBER of technicium activities are known to which no definite mass numbers have been assigned. The situation has remained inconclusive in spite of the recent use of electromagnetically enriched isotopes of Mo as target materials. The reason is that the usual method of assigning mass numbers to artificially produced radioisotopes by production through appropriate cross reactions from neighboring elements is not immediately available for deciding the issue due to the lack of suitable stable target isotopes. Relative cross-section measurements may be expected to be of use in such cases. In a previous paper,¹ enriched Mo isotopes were used to obtain the relative cross sections for (p, n) to (p, γ) reactions of Mo⁹⁵. These results have been utilized to assign mass numbers to three technicium activities of half-lives 2.7 hours, 47 minutes, and 4.5 minutes.

* Research Fellow of the National Institute of Health, Bethesda, Maryland.

¹ D. N. Kundu and M. L. Pool, Phys. Rev. **74**, 1574 (1948).

II. REVIEW OF Tc ACTIVITIES

2.7 Hours.—A technicium activity with a provisional value of a 2-hour half-life was reported² as a result of deuteron bombardments of Mo. This activity was later³ shown to be a 2.7-hour positron emitter obtained by bombarding Mo with protons. To correlate these facts with the observations by others⁴ that a similar activity is produced by alpha-bombardments of columbium, the 2.7-hour period was assigned⁵ to Tc⁹⁶. Recently, by using a deuteron beam on enriched Mo⁹², it has been shown⁶ that the 2.7-hour activity is produced from Mo⁹². However, no definite assignment of the activity to Tc⁹² or Tc⁹³ could be made. The decay is claimed to take place by emission of positrons of maximum energy 1.2 ± 0.2 Mev and a hard γ -ray of 2.4 ± 0.5 Mev.

² G. T. Seaborg and E. Segrè, Phys. Rev. **55**, 808 (1939).

³ L. A. Delsasso, L. N. Ridenour, R. Sherr, and M. G. White, Phys. Rev. **55**, 113 (1939).

⁴ L. D. P. King, W. J. Henderson, and J. R. Risser, Phys. Rev. **55**, 1118 (1939).

⁵ G. T. Seaborg, Rev. Mod. Phys. **16**, 1 (1944).

⁶ E. E. Motta and G. E. Boyd, Phys. Rev. **73**, 1470 (1948).

40–53 Minutes.—With proton bombardment of Mo, a 40 ± 5 -minute Tc activity was reported to presumably decay by positron emission.³ The value of the half-life given by others was 53 ± 3 minutes.⁷ Later observers⁸ confirmed the value of 53 minutes. The maximum energy of the positrons was found to be 2.45 Mev. Also observed were x-rays of Mo and Tc and five γ -rays of energies ranging from 0.380 to 2.74 Mev associated with low energy electrons due to internal conversion of the γ -rays. Probable assignment was made to numbers 92 or 94. Recent workers,⁹ using enriched Mo^{94} under a deuteron beam, have given the value of the half-life as 50 ± 2 minutes.

They have confirmed the energy of the positrons but have found only one γ -ray of energy 0.9 ± 0.1 Mev. The assignment of this activity to Tc^{94} was considered as probable. The same observers⁶ have also announced a 40 ± 5 -minute activity obtained with deuteron bombardment of Mo^{97} and Mo^{98} . The decay was by the emission of charged particles of 2.0 ± 0.5 Mev. No assignment of mass number was made.

To anticipate some results of the present paper, a 47-minute Tc activity appears which also has to be placed at positions 92 or 93.

4.5 Minutes.—A 4.5-minute Tc activity was found by the above group of workers⁶ when

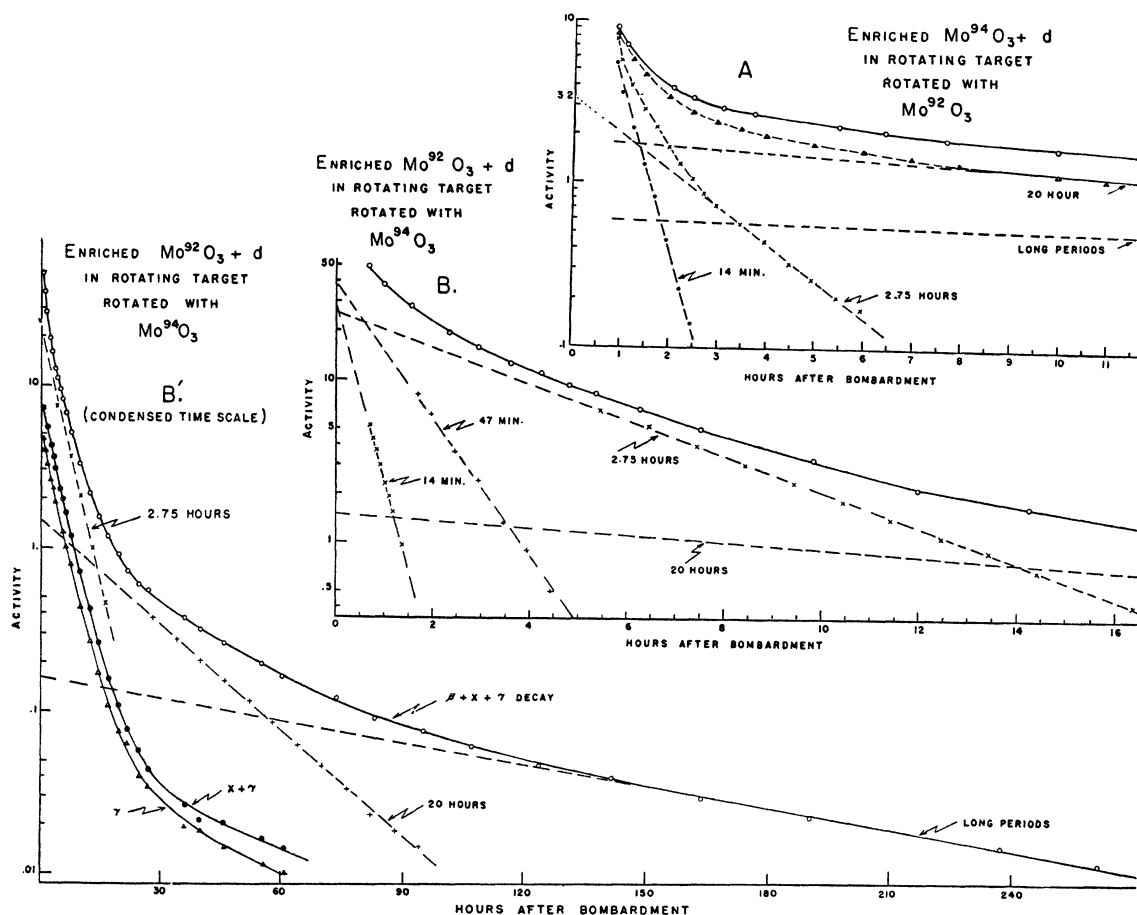


FIG. 1. Decay curves obtained by deuteron bombardment on MoO_3 enriched separately in Mo^{92} and Mo^{94} . The two enriched samples were placed on opposite sides of a rotating target and bombarded under the same beam. The 47-minute and the 2.75-hour activities are produced from Mo^{92} . The small amount of 2.75-hour activity seen in (A) is due to the small amount of Mo^{92} present in the enriched Mo^{94} sample. The $(x+\gamma)$ - and the γ -decay curves are also shown (B').

⁷ D. Ewing, T. Perry, and R. McCreary, *Phys. Rev.* **55**, 1136 (1939).

⁸ P. C. Gugelot, O. Huber, H. Medicus, P. Preiswerk, P. Scherrer and R. Steffen, *Helv. Phys. Acta.* **19**, 418 (1946); **20**, 240 (1947); O. Huber, P. Marmier, H. Medicus, P. Preiswerk, and R. Steffen, *Phys. Rev.* **73**, 1208 (1948).

⁹ E. E. Motta and G. E. Boyd, *Phys. Rev.* **74**, 220 (1948).

enriched Mo^{92} was bombarded with deuterons. The activity decays by emitting positrons of 4.3 ± 0.5 Mev and γ -rays of 1.3 ± 0.3 Mev. The assignment to either Tc^{92} or Tc^{93} was suggested.

It is thus apparent that the question as to which of these various activities are to be ascribed to Tc^{92} and Tc^{93} is of considerable interest. It is the purpose of this paper to show how the application of reaction cross-section measurements point toward reasonable mass assignments.

III. EXPERIMENTAL

Proton of 5 Mev and deuteron of 10 Mev were used to bombard natural molybdenum and three different samples of MoO_3 electromagnetically enriched in Mo^{92} , Mo^{94} , and Mo^{98} , respectively.** The methods of measurements of the charged particles, x-rays, and γ -rays are the same as described previously.¹ In order that the decay of

both the positive and the negative particles may be followed separately, the sample was placed between the pole faces of a large electromagnet and the particles of the desired sign bent circularly into a Geiger counter. The counts per minute were automatically recorded.¹⁰ To decide whether a particular activity produced was associated with one or the other of two target isotopes, the two enriched isotopes were placed on opposite sides of a rotating target holder and bombarded simultaneously under the same beam and the relative intensities compared. Chemistry was done after bombardments to identify the elements whenever necessary.

IV. RESULTS

2.7 Hours.—A $\text{Cb} + \alpha$ bombardment did not produce the 2.7-hour Tc activity, showing that this activity is not due to Tc^{96} as once supposed.⁵

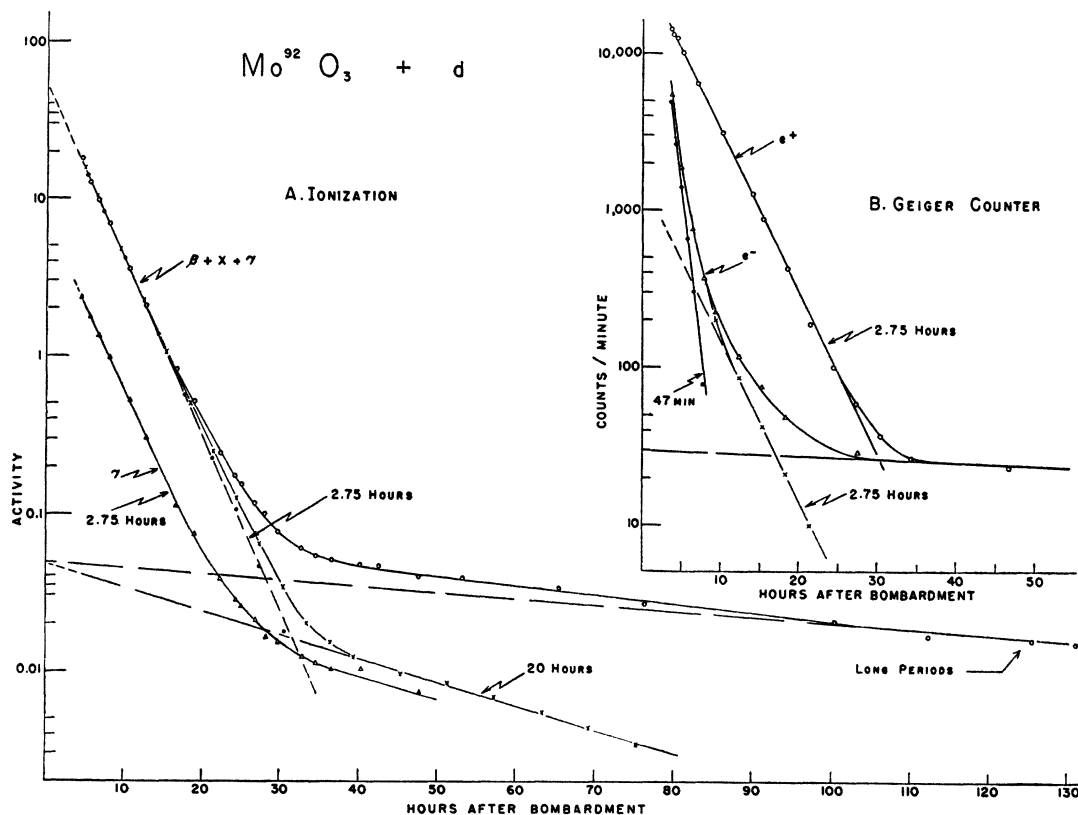


FIG. 2. Decay curves of enriched Mo^{92}O_3 d on electrometer showing 2.75-hour half-life and the associated γ -rays (A). The decay curves using Geiger counters on the positive and negative particles separately are shown in (B). The 47-minute activity is seen to emit negatively charged particles.

** Kindly supplied by the Y-12 Plant, Carbide and Carbon Chemicals Corporation, through the Isotope Division, United States Atomic Energy Commission, Oak Ridge, Tennessee.

¹⁰ L. L. Woodward and D. A. McCown, Rev. Sci. Inst. in press.

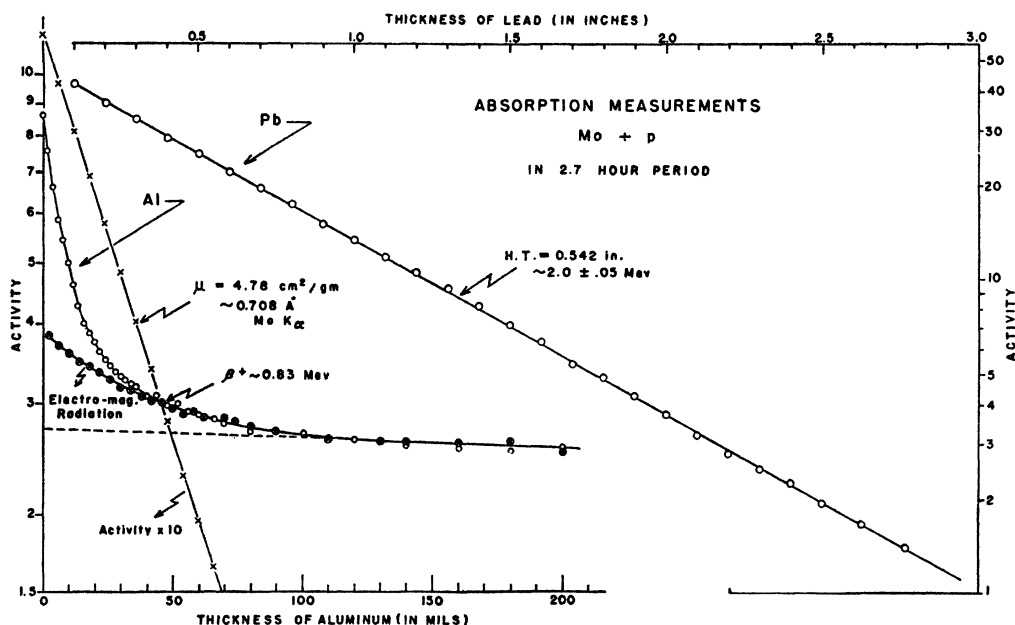


FIG. 3. Absorption curves in aluminum and head of $\text{Mo}+p$ in the 2.7-hour period showing x-rays corresponding to MoK_α line, positrons of energy 0.83 Mev and γ -rays of 2.00 Mev.

The activity, however, is produced under both proton and deuteron bombardment on natural Mo. By rotating enriched Mo^{92} with Mo^{94} under a deuteron beam, decay curves were obtained which are shown in Fig. 1. From the figure, the intensity of the 2.7-hour period in the Mo^{92} sample is seen to be more than eight times that in the Mo^{94} sample. In the Mo^{94} sample, all the Mo isotopes with mass numbers greater than 93 were present in a larger percent than in the Mo^{92} sample. This eliminates the assignment of any mass numbers greater than 93 to this activity. The 2.7-hour, therefore, belongs to either Tc^{92} or Tc^{93} . The decay curves followed separately on the positively and negatively charged particles are shown in Fig. 2. The charged particles emitted by the 2.7-hour activity are seen to be mostly positrons. The exact amount will be calculated later. Negatively charged particles of this period originating from conversion of the gamma-rays and Compton electrons are also observed to the extent of about five percent of the number of positrons. In the same figure, from an interval of over ten half-lives, a value of 2.75 ± 0.05 hours is obtained. As shown in Fig. 3, lead absorption measurements of the gamma-ray lead to an energy of 2.00 ± 0.05 Mev.

The absorption measurements with aluminum are of particular interest. Without the use of the electromagnet, the beta-end point might erroneously be taken to be about 100 mils, corresponding to an energy of approximately 1.4 Mev. On applying the magnetic field to remove the charged particles, it is observed that the part of the absorption curve beyond about 50 mils is due to electromagnetic radiation of wave-length 0.708 Å which is the K_α line of Mo. The end point for the positrons, as is clearly seen from Fig. 3, superimposed on the curve for x-rays gives the maximum energy as 0.83 Mev, contrary to the observations of others.⁶ This value is obtained after adding 2.68 mils to the end point as read from the curve, in order to correct for the thickness of air and window of the ionization chamber.

47 Minutes.—Separate bombardments of enriched Mo^{92} and Mo^{94} with deuterons show in Fig. 4 that a 47-minute activity is produced from Mo^{92} . The charged particles associated with the decay are all negative. The decay curve corresponding to $\text{Mo}^{94}+d$, however, does not show any 47-minute period. Also in the decay curve for the proton bombardment on Mo^{98} , there is no positron activity of 47-minute half-life. There is,

however, a very weak short period of the order of 55 minutes, which consists of negatively charged particles. The proton bombardments of Mo^{92} and Mo^{94} lead to the same observations with regard to this 47-minute period. In Fig. 5 the positron decay curve of the Tc from the $\text{Mo}^{94}+d$ bombardment also shows no 47-minute activity. The electrometer measurements substantiate this observation. In order to study more closely the connection of the 47-minute activity with Mo^{92} or Mo^{94} , we go back to Fig. 1, which shows the results obtained with the rotating target. The 47-minute period is obtained from $\text{Mo}^{92}+d$ sample and no observable amount is found in the decay curve of $\text{Mo}^{94}+d$.

Absorption measurements with aluminum foils show in Fig. 6 once again that without the use of the magnetic field, the beta-end point might wrongly be taken at about 150 mils, corresponding to 2.05 Mev. The removal of the charged particles by the magnet shows this part to be x-rays of wave-length 0.71A, corresponding to the K_{α}

radiation of Mo. The maximum energy of the negatively charged particles is 0.54 Mev after applying corrections for air and ion-chamber window. The energy of the γ -rays is found to be 1.50 ± 0.05 Mev from the same figure. The quantitative estimation of the x- and γ -rays as compared with the number of charged particles will be made in the next section. The decay takes place by K -capture alone.

4.5 Minutes.—No detailed study of this activity was made beyond noting that in $\text{Mo}+p$ bombardments of very short duration (15 minutes), there was an activity of the order of five minutes, in addition to the 14-minute activity of Tc^{101} . The x-rays, if any, and the γ -rays were too weak to be properly analyzed.

V. RELATIVE CROSS SECTIONS AND ASSIGNMENTS

From the activity values at the end of the bombardments, the saturation intensities for the electrons, x-rays, and γ -radiations are calculated

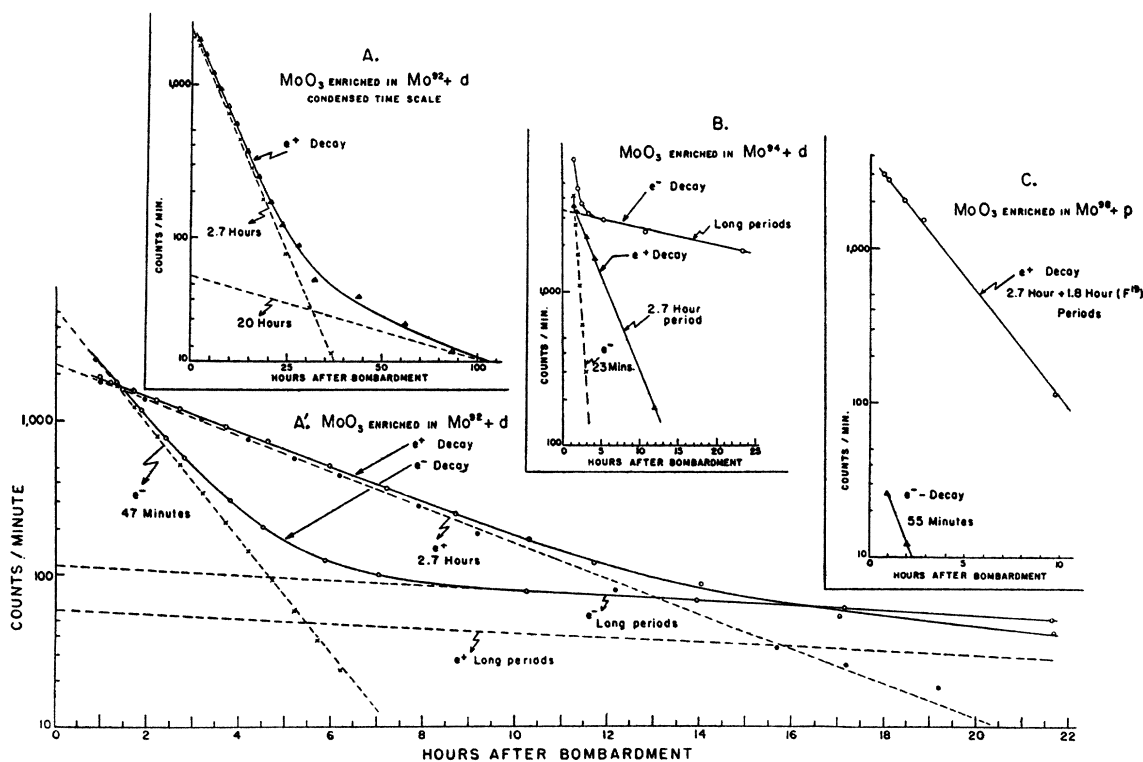


FIG. 4. Decay curves from separate bombardments of enriched Mo^{92} and Mo^{94} with deuterons and of enriched Mo^{98} with protons. No 47-minute activity is found in (B) $\text{Mo}^{94}+d$. The 47-minute activity is seen very clearly in (A') $\text{Mo}^{92}+d$ on the curve for negative particle emission. No 40-minute positron activity is observed in (C) $\text{Mo}^{98}+p$. The 2.7-hour activities seen in (B) and (C) are due to small amounts of Mo^{92} present in the enriched Mo^{94} and Mo^{98} samples.

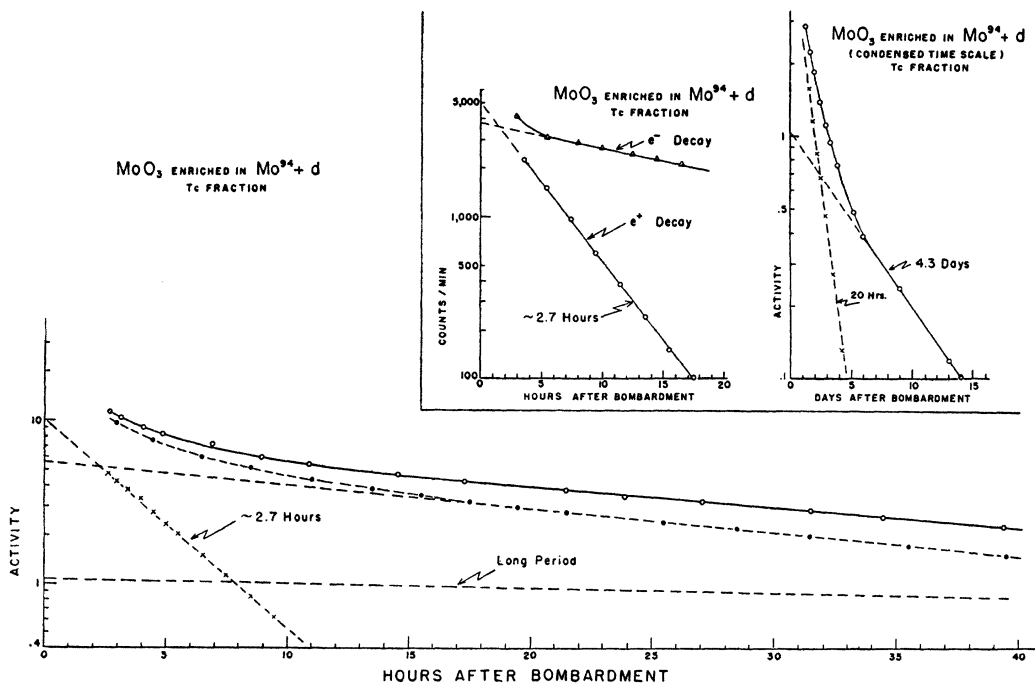


FIG. 5. Decay curves of the Tc fraction of enriched $Mo^{94}+d$. No 47-minute activity is seen either on the electrometer measurements or on the counter measurements for positive and negative particles. The 4.3-day activity is produced from Mo^{96} and Mo^{98} present in small amounts in the enriched Mo^{94} sample. The small amount of the 2.7-hour activity is similarly due to the presence of Mo^{94} in the enriched Mo^{94} sample.

to be 2.67, 0.52, and 0.605 ionization units, respectively, for the 2.7-hour activity and 3.34, 2.67, and 0.273 units, respectively, for the 47-minute activity. For the ionization chamber of the electrometer used, each electron of maximum energy of about 0.83 Mev on the average will produce 123 as much ionization as each x-quantum of 1A; the corresponding figure for an electron of

energy of 0.54 Mev will be 58.5. Thus there are about 24 x-quanta per positron emitted by the 2.7-hour activity and 50 x-quanta per electron observed in the 47-minute period. In the same chamber, each x-ray quantum and each γ -ray quantum produce ionization of the same order compared to the ionization due to charged particles. Therefore, in the 2.7-hour period, there

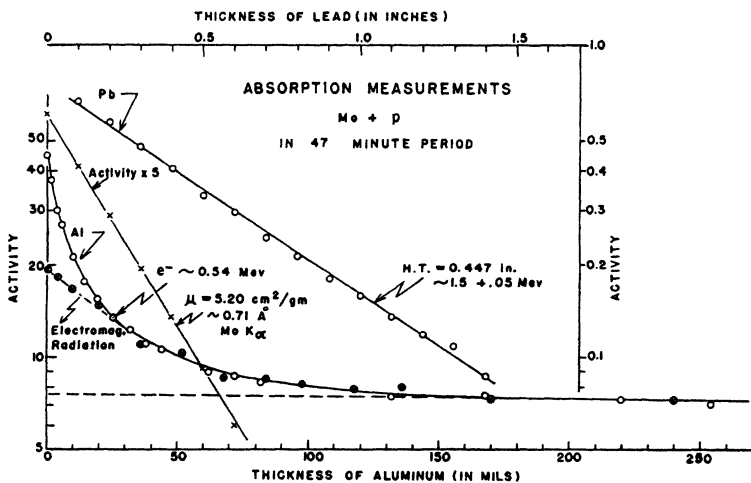


FIG. 6. Absorption curves in aluminum and lead of $Mo+p$ in the 47-minute period showing x-rays corresponding to MoK_{α} line, electrons of energy 0.54 Mev and γ -rays of 1.50 Mev.

are 0.9 x-quantum per γ -quantum and in the 47-minute period, 9.8 x-quantum per γ -quantum. Combining, we find that for the 2.7-hour activity, for each positron measured, there are 13.6 x-quantum and 15 γ -quantum which are being measured. For the 47-minute material, per electron measured, there are 21.4 x-quantum and 3.1 γ -quantum. These x-rays were found to be K_α line of Mo. Both these activities, therefore, decay by K -capture primarily. Each x-quantum is taken as due to one disintegration. For determining the relative cross sections, it would, thus, be enough to base the computation on the x-ray quanta alone.

The ratio of the cross sections σ_{92} (47 min.) / σ_{92} (2.7-hour) = $2.67/0.52 = 5.1$. The relative cross sections for the $(p, n)/(p, \gamma)$ reaction from Mo^{95} was found to be 6.5. On this basis, the 47-minute activity is produced by (p, n) and the 2.7-hour activity by (p, γ) reactions, respectively. The 47-minute activity is thus assigned to Tc^{92} and the 2.7-hour activity to Tc^{93} .

With regard to the 4.5-minute activity, the saturation intensity for positrons was 5.49 units, whereas in the same bombardment the saturation positron intensity of the 2.7-hour activity was 5.24 units. We saw that for the 2.7-hour period there are 25 disintegrations to each positron measured. Therefore, the cross section for the production of the 4.5-minute activity is about 1/25 of that for the production of the 2.7-hour activity, and, therefore, only about 1/127 of that for the 47-minute period. On the basis of this cross section consideration, the 4.5-minute activity is assigned to Tc^{93} as an isomer to the 2.7-hour activity.

VI. DISCUSSION

The assignment and relative cross sections are summarized in Fig. 7. The similarities in the

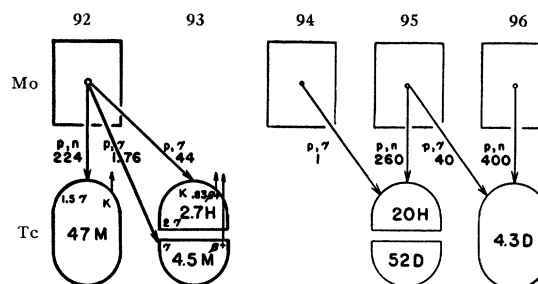


Fig. 7. Relative cross sections of proton reactions from Mo^{92} are compared with those from Mo^{94} , Mo^{95} , and Mo^{96} . Assignment of the 47-minute activity to Tc^{92} and of the 2.7-hour and 4.5-minute activities to Tc^{93} are made and the characteristic radiations are shown.

relative proton reactions from Tc^{92} and Tc^{95} are readily seen. These seem quite reasonable in view of the similarity of the decay characteristics of the 47-minute, 2.7-hour, 20-hour, and 4.3-day activities.

The characteristic radiations of the 47-minute activity studied in the present paper have been shown to be different from those of the 40- and 50-minute activities observed by others.^{6,9} It is, however, difficult to see how the 10-Mev deuterons failed to produce the 50-minute half-life reported as being produced by 16-Mev deuterons from Mo^{94} , especially when it was shown¹ that the (d, n) and $(d, 2n)$ reactions proceed with comparable readiness.

The help received from the Development Fund and the Graduate School of the Ohio State University is gratefully acknowledged. The chemical separations were ably done by H. L. Finston and R. M. Dyer. Thanks are also due the National Institute of Health, Bethesda, Maryland, for a Research Fellowship to one of the authors, and the Ghosh Board of the Calcutta University for facilities of traveling to this country.