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Radioactive Isotopes of Ru and Tc

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A new isotope of Ru and one of Tc were found when Mo was bombarded with alpha-particles. The Ru activity decayed with a half-life of 1.65±0.05 hours. Positrons, x-rays, and gamma-rays were emitted with energies of 1.1 Mev, 0.7A, and 0.95 Mev, respectively. The Ru activity also was produced by fast neutron bombardment of Ru. The Tc activity decayed with a half-life of 20.0±0.5 hours and emitted x-rays of 0.7A and gamma-rays of 0.78 Mev. The Tc activity also was made by proton bombardment of Mo. Electromagnetically separated Mo^{92} of 92 percent abundance was bombarded with alpha-particles producing the 1.65-hour and 20-hour activities. Electromagnetically separated Mo^{94} of 75 percent abundance under alpha-particle bombardment made a 2.8-day activity. Thus the 1.65-hour, 20-hour, and 2.8-day activities can be assigned to Ru^{95}, Tc^{95}, and Ru^{97}, respectively.

I. INTRODUCTION

It has been reported that a 2.8-day Ru activity produced by neutron[1,2] and deuteron[1] bombardments of Ru decays by K-electron capture into the 90-day daughter isotope, Tc^{97}. The 2.8-day activity emitted x-rays, 0.2-Mev electrons, and 0.23-Mev gamma-rays; the mass assignment was made to the isotope Ru^{97}. A 45-day Ru activity resulting from neutron[3] and deuteron[4] bombardment of Ru and also from U-fission[5] has been found to decay by emitting 0.2-Mev beta-particles and 0.56-Mev gamma-rays. A half-life of less than 90 minutes was reported following an alpha-bombardment of molybdenum.[6] A 52-day Tc activity which decays by K-electron capture has been produced by proton and deuteron bombardments of molybdenum.[7]

The above Ru and Tc activities have been essentially confirmed in this laboratory; while doing so, however, two new radioactive substances have been found, one in Ru and one in Tc. It is the purpose of this paper to report on their characteristic radiations, and to make mass assignments.

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1 W. H. Sullivan, N. R. Sleight, and E. M. Gladrow, Phys. Rev. **70**, 778 (1946).
2 E. E. Motta, G. E. Boyd, and A. R. Brosi, Phys. Rev. **71**, 210 (1947).
3 K. Senma and H. Yamasaki, Phys. Rev. **59**, 402 (1941).
4 E. Bohr and N. Hole, Arkiv. Math. Astro. Fys. **A32**, No. 15, 8 (1946).
5 W. E. Grummitt and G. Wilkinson, Nature **158**, 163 (1946).
6 L. D. P. King, W. J. Henderson, and J. R. Resser, Phys. Rev. **55**, 1118 (1939).
7 J. E. Edwards and M. L. Pool, Phys. Rev. **72**, 384 (1947).

II. Ru FRACTION

Molybdenum sheet metal was bombarded by 20-Mev alpha-particles and 5-Mev protons. Immediately after the bombardments the Mo was dissolved in HCl and the active Ru and Tc were distilled as RuO_4 and $TcCl_3$ with an inactive Ru carrier into a concentrated solution of KOH containing a Re salt as carrier for the Tc. The Ru fraction was precipitated with alcohol and filtered. The supernate containing the active Tc was made $6N$ with H_2SO_4 and the Tc fraction was precipitated with H_2S.

The decay of the Ru fraction from a $Mo+\alpha$ bombardment is plotted in Fig. 1. Curve C in the insert represents a decay of the total radiation taken over a period of five months. When the intensity of the 45-day half-life material is subtracted from curve C, activities with half-lives of 2.8 days, 20 hours, and 1.65 hours are found. Curve A, representing the total radiation, is plotted over the first 40 hours of the decay. Successive subtractions show the relative ionization produced by the 1.65-hour, 20-hour, and 2.8-day activities in this sample. The time required to make the chemical separation was about 1.5 hours. Curve B shows the decay of the gamma-rays when the beta-particles and the x-rays were removed. It is observed that each of the above activities emits gamma-rays.

Lead absorption measurements taken during the 1.65-hour activity disclosed two gamma-rays

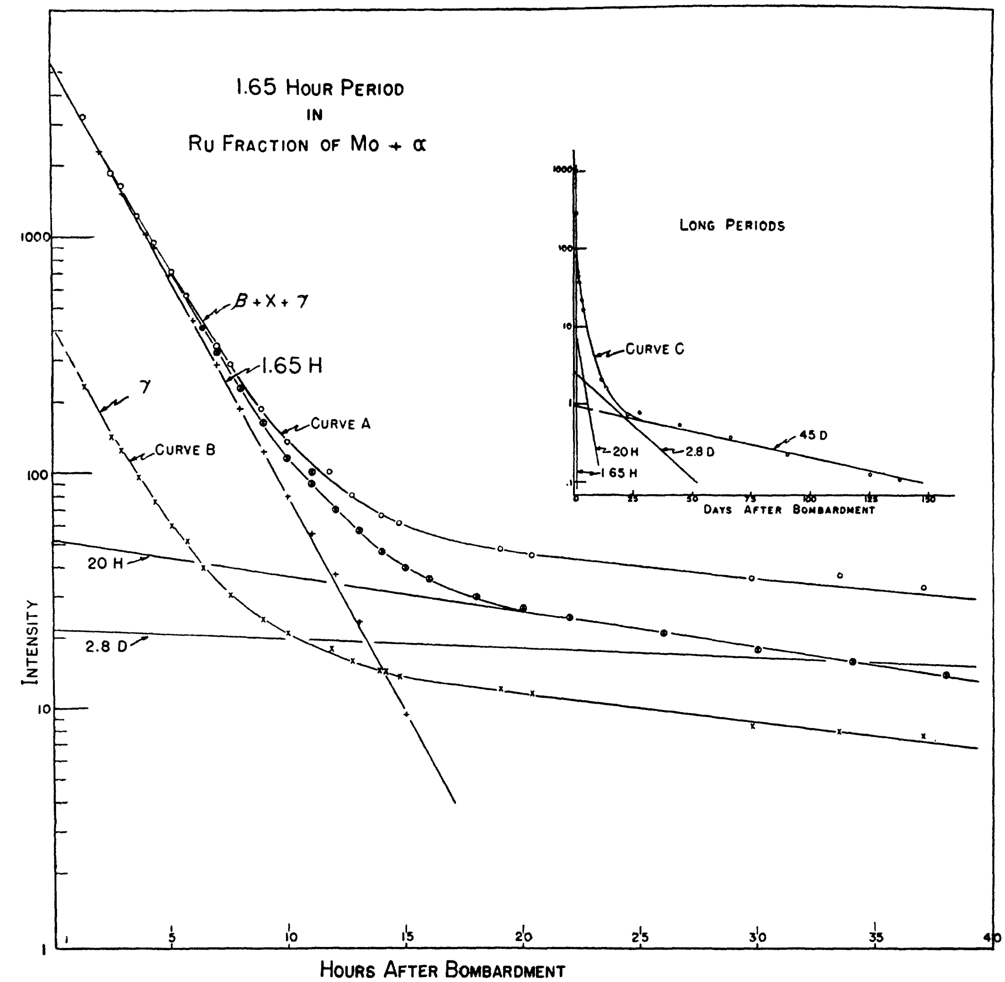

FIG. 1. Decay curves for (*A*) and (*C*) total radiation and (*B*) gamma-radiation of the Ru fraction of a Mo+α bombardment. The 1.65-hour Ru^{95}, 20-hour Tc^{95}, 2.8-day Ru^{97}, and 45-day Ru^{103} are shown with their relative ionization intensities.

with half-value thicknesses of 0.41 cm and 0.86 cm corresponding to energies of 0.52 Mev and 0.95 Mev, respectively. From Wilson cloud chamber observations, it is seen that the 1.65-hour substance decays by positron emission. Aluminum absorption measurements taken when the activity was mainly in the 1.65-hour period are plotted in Fig. 2. The curve shows that the positrons have a maximum range of 440 mg/cm^2 or a maximum energy of 1.1 Mev.[8] When the positrons are deflected from the ionization chamber by a strong magnetic field, it is possible to show by aluminum absorption measurements that x-rays are emitted. Such measurements are shown also in Fig. 2. The absorption coefficient obtained was about 5 cm^2/g and therefore indicates x-rays of wave-length approximately 0.7A. K_α x-rays characteristic of the elements in the Tc region have wave-lengths near this value.

Decay measurements may be taken using the magnet as above. These measurements indicate the presence of a small amount of ionization caused by the x-ray radiation in the 1.65-hour activity; therefore, *K*-electron capture takes place as well as positron emission. The probability of decay by means of *K*-electron capture is of the same order of magnitude as that by positron emission.

In addition to the alpha-bombardment of

[8] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).

molybdenum, ruthenium was bombarded by fast and slow neutrons. The fast neutrons produced the 1.65-hour, 2.8-day, and 45-day Ru activities. However, the slow neutron bombardment made only the 2.8-day and 45-day activities and not the 1.65-hour activity.

III. Tc FRACTION

The decay curves for the Tc fraction from a $Mo+\alpha$ bombardment are plotted in Fig. 3. The insert shows the decay curve of the total radiation taken over a period of five months. The long period denoted as the 52-day activity serves as the base line for curve A. Curve A shows the decay of the total radiation during the first nine days after bombardment.

Based upon measurements carried through ten half-lives, a value of 20.0 ± 0.5 hours was found. Curve B shows the decay due to the total electromagnetic radiation only. In subtracting curve B from curve A, it is seen that the ionizing intensity caused by the negatively charged particles emitted during the decay of the 20-hour period is very small compared to that of the electromagnetic radiation. Aluminum absorption measurements were made on an electrometer particularly designed for low energy particles. A maximum energy of 200 kev was recorded.

Curve C shows the decay due to the electromagnetic radiation after passing through one-quarter inch of aluminum. This curve is caused primarily by gamma-rays and shows the presence of a gamma-ray in the 20-hour period. Lead absorption measurements give a half-value thickness of 0.69 cm, which indicates the energy of the gamma-ray is approximately 0.78 Mev. By taking the difference between curve B and curve C, it is observed that the intensity of the resulting x-rays decays with a half-life of 20 hours. Aluminum absorption measurements made on the electromagnetic radiation are plotted in Fig. 4. These measurements show a measured half-value thickness of 0.132 g/cm^2 corresponding to an x-ray wave-length of 0.72A.

It was pointed out in the Ru section that the Ru fraction from the $Mo+\alpha$ bombardment also contained a 20-hour activity. An additional observation is now presented that the relative amount of 20-hour material in the Ru fraction is dependent on the period of time between the end of the bombardment and the chemical separation of the Ru and Tc fractions. In a sample where the chemical separation was made 15 hours after bombardment, the 20-hour activity was almost absent in the Ru fraction and was correspondingly more abundant in the Tc fraction. It is therefore plausible to conclude that the 20-hour substance is the daughter product of the 1.65-hour Ru activity. The 20-hour Tc activity was also produced by a proton bombardment of molybdenum.

IV. ELECTROMAGNETICALLY SEPARATED ISOTOPES

Samples of $Mo^{92}O_3$ and $Mo^{94}O_3$** were bombarded with alpha-particles. The Mo^{92} was enriched to 92 percent, electromagnetically, and the Mo^{94} enriched to 75 percent. Equal masses of the two enhanced isotopes were bombarded under nearly identical conditions by rotating a target, having the two samples on opposite sides. The decay curves of these respective activities are shown in Fig. 5.

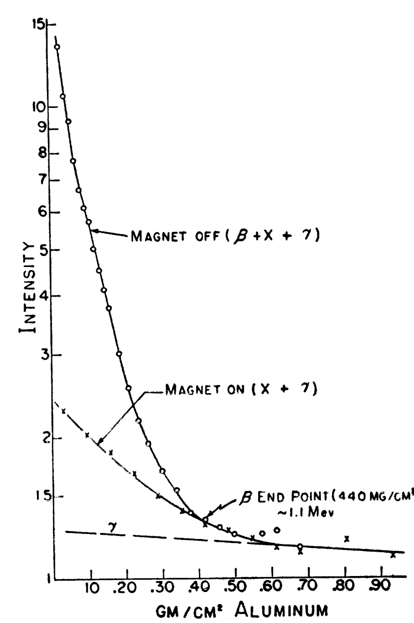

FIG. 2. Aluminum absorption curve showing the end-point energy of 1.1 Mev for the positrons in the 1.65-hour Ru^{95}. This curve also shows an x-ray of wave-length about 0.7A.

** Supplied by the Y-12 plant, Carbide and Carbon Chemical Corporation, through the Isotopes Division, United States Atomic Energy Commission, Oak Ridge, Tennessee.

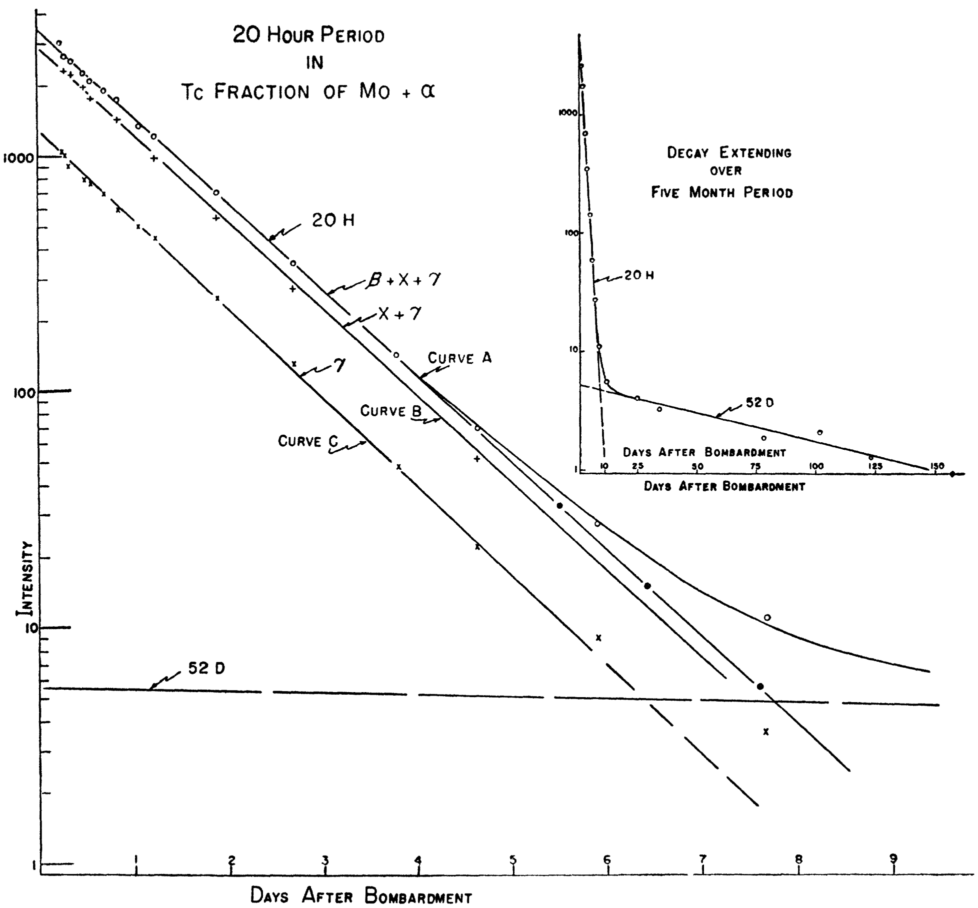

FIG. 3. Decay curves for the (*A*) total radiation, (*B*) total electromagnetic radiation, and (*C*) gamma-radiation of the Tc fraction of a Mo+α bombardment, showing the 20-hour Tc^{95} and 52-day Tc^{95} activities. The insert shows the intensity of the total radiation from the sample taken over a five month period.

TABLE I.

Isotope	Half-life	Characteristic	Radiations	Reaction
Ru^{95}	**1.65-h**	**1.1 Mev β^+** **K-capture**	**0.5 Mev γ** **0.95 Mev γ** **0.7 *A X***	**$Mo^{92}(\alpha,n)$** **$Ru^{96}(n,2n)$**
Ru^{97}	2.8-d	*K*-capture 0.2 Mev β^-	0.23 Mev γ 0.7 *AX*	$Ru^{96}(n,\gamma)$ $Ru^{96}(d,p)$ **$Ru^{98}(n,2n)$** **$Mo^{94}(\alpha,n)$**
Ru^{103}	45-d	0.2 Mev β^-	0.56 Mev γ	$Ru^{102}(n,\gamma)$ $Ru^{102}(d,p)$ **$Ru^{104}(n,2n)$** **$Mo^{100}(\alpha,n)$**
Tc^{95}	**20-h**	***K*-capture** **0.2 Mev β^-**	**0.78 Mev γ** **0.7 *A X***	**$Mo^{92}(\alpha,p)$** **$Mo^{95}(p,n)$**
Tc^{95}	52-d	*K*-capture	0.25 Mev γ 0.84 Mev γ	$Mo^{95}(p,n)$ $Mo^{95}(d,n)$ **$Mo^{92}(\alpha,n)$**

The decay of the activity from the bombarded $Mo^{92}O_3$ definitely shows the 1.65-hour and 20-hour activities and an indication of a long period presumed to be the 52-day Tc activity. The decay of the activity from the bombarded $Mo^{94}O_3$ is predominately the result of a 2.8-day activity. A small amount of 1.65-hour activity is present in the decay of the Mo^{94} isotope and is probably caused by a small percentage of Mo^{92} in the sample.

V. MASS ASSIGNMENTS

Since the 1.65-hour positron activity of Ru was produced by alpha-bombardment of molybdenum and fast neutron bombardment of ruthenium but was not produced by slow neutron bombardment of ruthenium, the 1.65-hour ac-

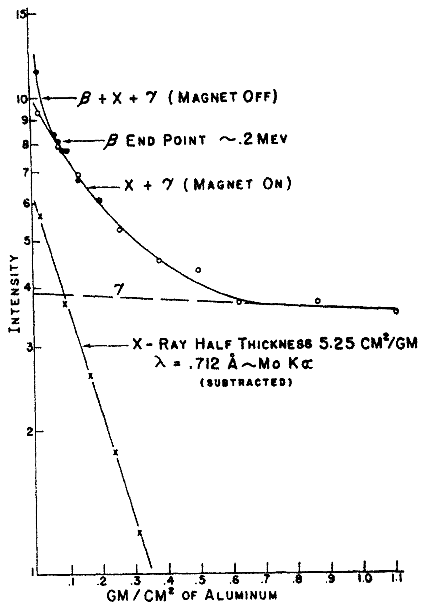

FIG. 4. Aluminum absorption curve taken during the 20-hour Tc^{95} period showing Mo $K\alpha$ x-rays.

tivity may be best assigned to Ru^{95}. The electromagnetically separated Mo^{92} bombarded with alpha-particles definitely confirms this assignment. Since the 20-hour Tc activity was shown to be produced by alpha-particle and by proton bombardments of molybdenum and also by the decay of the 1.65-hour Ru activity, the 20-hour activity is thus assigned to Tc^{95}. The alpha-particle bombardment of enriched Mo^{92} also confirms this assignment. The 2.8-day Ru activity which was produced by slow neutron and deuteron bombardments of ruthenium was assigned to Ru^{97}. The fast neutron bombardments of ruthenium and alpha-particle bombardments reported in this paper yield results which are consistent with this assignment. The alpha-particle bombardment of electromagnetically separated Mo^{94} confirms the assignment of the 2.8-day activity to Ru^{97}.

SUMMARY

The Tc region of a nuclear transmutation chart is shown in Fig. 6. In this chart, the isotopic positions of Tc^{95}, Ru^{95}, and Ru^{97} are indicated by heavy lines. New reactions are indicated by heavy arrows on which the nuclear reaction is noted.

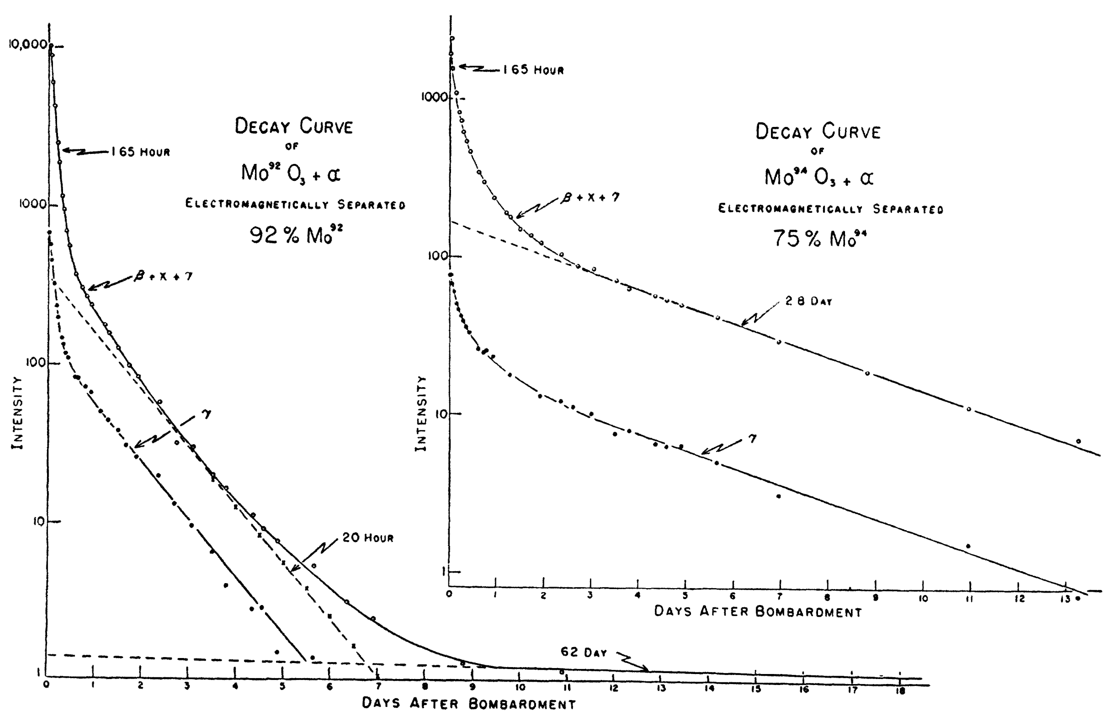

FIG. 5. Decay curves of the total radiation and gamma-radiation of electromagnetically separate Mo^{92} and Mo^{94} bombarded with alpha-particles. The 1.65-hour Ru activity and 20-hour Tc activity are found in the decay of the $Mo^{92}O_3+\alpha$ curves and the 2.8-day Ru activity is found in the $Mo^{94}O_3+\alpha$ curves.

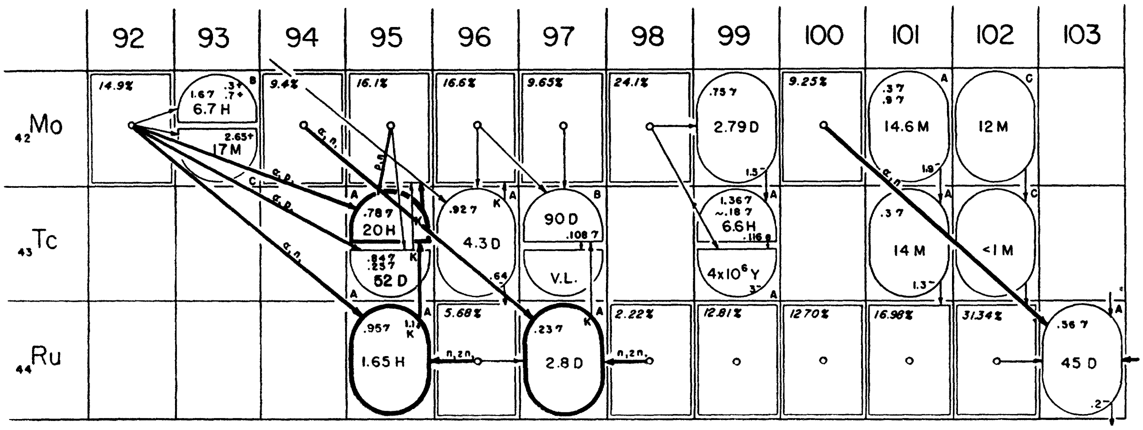

FIG. 6. The technetium region of the nuclear transmutation chart. Heavy lines and arrow indicate information reported in this paper.

Table I shows the characteristic radiations of the isotopes studied. The information reported in this paper is given in boldfaced type.

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On the Penetrating Showers of Cosmic Radiation

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Experiments have been performed on the penetrating showers of cosmic radiation accompanied and not accompanied by extensive air showers. In the accompanied showers the penetrating particles are very likely to pre-exist in the air; in the unaccompanied showers they are created in the absorbers surrounding the counters by a nucleonic component of the cosmic radiation.

1. INTRODUCTION

SEVERAL series of experiments have been performed in order to study penetrating showers associated and unassociated with the extensive cosmic-ray showers at sea level.

The experimental arrangement is shown in Fig. 1. The trays of unshielded counters *a*, *b*, *c*, each consisting of four counters in parallel (surface of each tray 400 cm²), were placed in a horizontal plane at the vertices of an equilateral triangle of 4-m sides, in boxes of light material, maintained at constant temperature, on the roof of the laboratory. The three-fold coincidences $a+b+c$ ($\sim 6h^{-1}$) were due to the extensive air showers.

The penetrating particles were recorded by the six trays, *A*, *B*, *C*, *D*, *E*, *F*, each consisting of four counters in parallel (surface of each tray 400 cm²) arranged in three telescopes, *AB*, *CD*, and *EF*, surrounded by lead. The thickness of the lead between *A* and *B* was 6 cm, between *C* and *D*, *E* and *F* it was 1 cm. The screens on the sides, fronts, and backs of all counters were 11 cm thick. Underneath each telescope 6 cm of lead were placed. Above the telescopes the thickness *T* was varied in different series of