ment with previously published experimental data. Beringer[2] found that the absorption in oxygen-nitrogen mixtures is proportional to the oxygen partial pressure. Lamont[3] measured directly the absorption in the atmosphere and found it to be about 15 db/km. On the basis of our experiments, we would predict the absorption resulting from oxygen in the atmosphere to be about 30 to 40 db/km.

A good deal of work remains to be done. Studies of the absorption in pure oxygen at low pressures should be carried out to determine the exact transition frequencies, any shift of frequency with pressure, any dependence of the line breadth upon rotational state, and any anomalous dependence of the line breadth on pressure. We intend to carry on further researches along these lines in this laboratory. The data given above from an incomplete study of the absorption phenomena in oxygen and are presented at this time so that the material already at hand might be made available without further delay to those interested in the problem.

ACKNOWLEDGMENT

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Radiations from Ge^{77} and Ge^{71} *

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The 12-hour Ge^{77} was found to emit beta-rays of energy 1.74 Mev and γ-rays of 0.5 Mev. Beta-gamma-coincidences were measured in the 12-hour period. The 40-hour positron emitter is not formed by slow neutrons on germanium. The 11-day Ge^{71} decays with the emission of the x-rays of gallium following orbital electron capture. Previously obtained absorption curves have given evidence of emission of charged particles by the 11-day period. This misinterpretation is discussed.
See note added in proof.

INTRODUCTION

THE measurements of this paper were accumulated during 1948 and the early part of 1949 and resulted from the exposure of four different quantities of GeO_2 in the Oak Ridge pile. Two activities, 12 hours and 11 days, were found in the germanium fraction, and the 40-hour daughter of the 12-hour period was found in the arsenic fraction. Owing to the distance of the laboratory from Oak Ridge, observations were never begun at a time less than twenty hours after removal of the irradiated material from the pile. The third sample of GeO_2 was irradiated for only twelve hours so as to enhance the intensity of the shorter germanium period. The first, second, and fourth samples were exposed for longer but unspecified periods as indicated by the fact that the 11-day activity was several times more intense in each case than in the third sample. Chemical separations were carried out for the removal of Ni, Fe, Cu, Ag, Na, Ca, and Ga as possible impurities.

Ge^{77}

The 12-hour Ge^{77} was found to emit beta-rays and gamma-rays. An absorption measurement of the beta-rays carried out within twenty-five hours after removal of the irradiated material from the pile is plotted in Fig. 1. The end point occurs at 0.78 g/cm², 1.74 Mev as calculated by Feather's equation.[1] This value is to be compared with a K-U extrapolation of cloud-chamber data giving 1.9 Mev,[2] and an aluminum absorption measurement giving 2.0 Mev.[3] The activity of the irradiated GeO_2 was followed in a single Geiger counter, shielded by 0.26 g/cm² of absorber, including wall thickness of the counter. This absorber thickness excluded from the counting rate the beta-rays of the forty-hour As^{77} which emits no gamma-rays. The decay curve is shown in Fig. 2 where it is clear

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[1] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).
[2] R. Sagane, G. Miyamoto, and M. Ikawa, Phys. Rev. **59**, 904 (1941).
[3] E. P. Steinberg and D. W. Engelkemeir, Plutonium Project Report, "*Nuclei formed in fission*," Rev. Mod. Phys. **18**, 513 (1946).

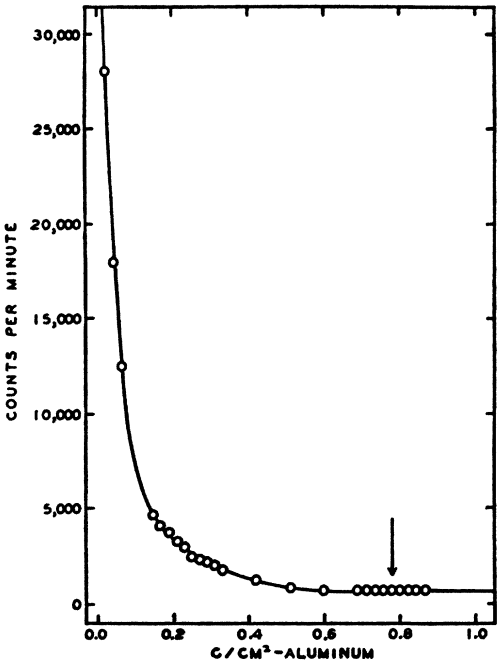

FIG. 1. Absorption in aluminum of the hard beta-rays of Ge^{77}. The end point corresponds to an energy of 1.74 Mev.

that only two periods, twelve hours and eleven days are present. No effects were observed from the 1.2-Mev positrons and accompanying annihilation radiation of the 40-hour Ge^{69}. This is consistent with the conclusion of McCown, Woodward, and Pool[4] that the 40-hour Ge^{69} is not produced by deuteron bombardment of germanium.

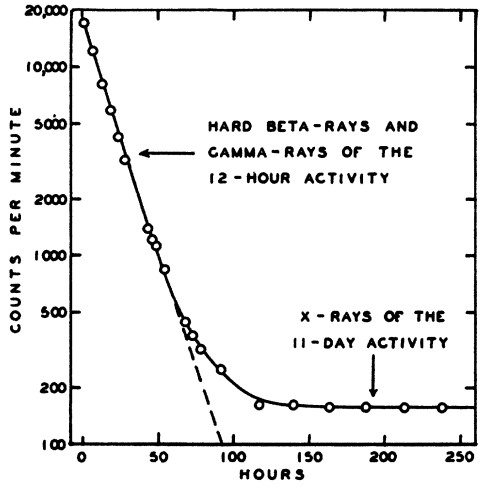

FIG. 2. Decay of the 12-hour Ge^{77}. The counter was shielded by 0.26 g/cm² of aluminum so as to suppress any effects from As^{77}. This decay curve shows that the 1.2-Mev positron emitter of 40-hour half-period is not formed by slow neutrons on germanium.

[4] D. A. McCown, L. L. Woodward, and M. L. Pool, Phys. Rev. **74**, 1311 (1948).

The beta-gamma-coincidence rate of the 12-hour Ge^{77} is given in Fig. 3 as a function of the surface density of aluminum placed before the beta-ray counter. It is seen to rise from a relatively low value at zero absorber thickness to 0.3×10^{-3} coincidence per beta-ray at an absorber thickness of about 100 mg/cm² and remains constant thereafter. The reduced coincidence rate below 100 mg/cm² is attributed to the presence of the x-rays of the 11-day Ge^{71} and the nuclear beta-rays of As^{77}. The beta-rays of As^{77} actually have a range of 0.192 g/cm² in aluminum, but at the time of the beta-gamma-coincidence measurements, the number of As^{77} beta-rays having a range in excess of 100 mg/cm² was small as compared with the number of hard beta-rays of Ge^{77} present. The coincidences per minute were observed to decay with the 12-hour half-period of Ge^{77} at all points along the curve, showing that the beta-rays of As^{77} are non-coincident with any gamma-radiation. The gamma-ray counter of the beta-gamma-coincidence counting arrangement was calibrated by the beta-gamma-coincidence rate of Sc^{46} (2 Mev of de-excitation energy). This calibration showed that the beta-gamma-coincidence rate of 0.3×10^{-3} coincidence per beta-ray beyond 100 mg/cm² corresponds to a de-excitation energy of 0.5 Mev; that is, the hard beta-rays of Ge^{77} lead to an excited state 0.5 Mev above the ground state of the As^{77} nucleus. When the gamma-rays of Ge^{77} were absorbed in lead, an energy of 0.5 Mev was obtained, in agreement with the value predicted by the coincidence rate.

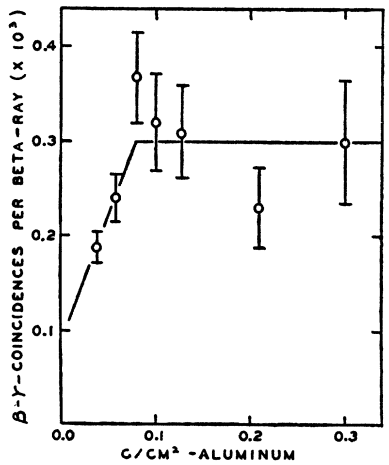

FIG. 3. The beta-gamma-coincidence rate of the 12-hour Ge^{77} as a function of the surface density of aluminum placed before the beta-ray counter. The coincidence rate below 100 mg/cm² is reduced owing to the presence of the beta-rays of As^{77} and the x-rays of Ge^{71}. Beyond 100 mg/cm², the beta-gamma-coincidence rate indicates that the hard beta-rays of Ge^{77} are followed by 0.5 Mev of gamma-ray energy.

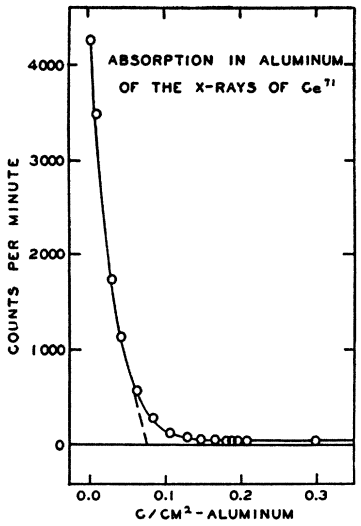

FIG. 4(a). Absorption in aluminum of the x-rays of Ge^{71}. The similarity of this curve to that of Fig. 4(b) led to an early misinterpretation of the data.

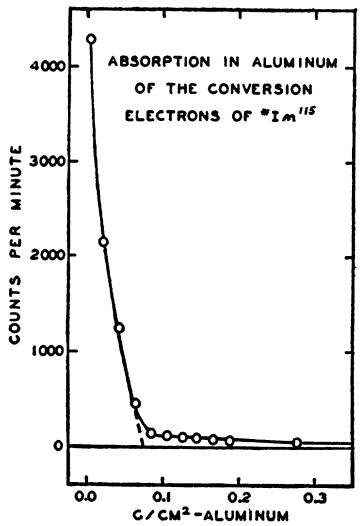

FIG. 4(b). Absorption in aluminum of the conversion electrons of the 4-hour isomer of indium (115).

Ge^{71}

The 11-day Ge^{71} was first reported by Seaborg, Livingood, and Friedlander,[5] who found the radiation to consist primarily of charged particles having an energy of 0.6 Mev. As stated by them, it seemed plausible that these were conversion electrons following K electron capture. Negatron emission by Ge^{71} was excluded by the fact that there exists no stable As^{71}. In the course of studying the radiations of the first three quantities of GeO_2, the absorption curve of Fig. 4(a) was obtained. On comparing its shape with those of some thirty-five nuclear beta-ray spectra having end points ranging from 0.15 to 2.22 Mev, it was concluded that the curve of 4(a) could not be that of a nuclear beta-ray spectrum. On the other hand, the curve shape was almost identical with that of the 0.338-Mev[6] conversion electrons of $*In^{115}$, shown in Fig. 4(b). The two curves strongly resemble each other in shape, half-value thickness, and extrapolated end point. It was concluded that the radiation of Ge^{71} was indeed conversion electrons following K electron capture. This view was strengthened when coincidences between the supposed conversion electrons and x-rays were noted. To ascertain whether the x-rays were from the gallium residual nucleus, 150 mg of aluminum were placed before the source to exclude the supposed conversion electrons, and critical absorption measurements were carried out on the residual x-ray activity. The radiation was much more heavily absorbed in copper than in zinc. This would be characteristic of the gallium $K\alpha$-line. The picture seemed fairly logical, and a preliminary abstract of these conclusions was given.[7]

Neither the writers nor Seaborg, Livingood, and Friedlander could dectect any gamma-radiation. This suggested that the supposed gamma-ray was very nearly totally converted. This feature could not be explained by a $0\rightarrow 0$ transition, because the ground state of the Ga^{71} residual nucleus has a spin of $\frac{3}{2}$. A search was made for an isomeric state of the Ga^{71} residual nucleus, but measurements carried out within 20 minutes after the germanium-gallium separation showed no activity in the gallium fraction.

Since no satisfactory explanation of the conversion coefficient of the supposed converted gamma-ray was reached, it was decided to prepare a fourth unit of Ge^{71} and make further observations. In the meantime, the paper by Pool *et al.*[4] appeared, reporting no charged particles whatever from Ge^{71}. This conclusion was reached from a study of decay curves and cloud-chamber observations.

After the fourth unit of Ge^{71} had been properly aged to avoid any 12-hour or 40-hour effects the critical absorption curves of Fig. 5 were obtained. The zinc and copper foils were placed before the source with no aluminum intervening. The rapid absorption in copper can be explained only if the bulk of the radiation is assumed to be the gallium $K\alpha$-line. This confirms the conclusion of the Ohio State University group that only K capture x-rays are emitted by the 11-day Ge^{71}. The previously reported "conversion electron-(x-ray)" coincidences[7] were

[5] G. T. Seaborg, J. J. Livingood, and G. Friedlander, Phys. Rev. **59**, 320 (1941).

[6] J. L. Lawson and J. M. Cork, Phys. Rev. **57**, 982 (1940).

[7] Woo, Mandeville, Scherb, and Keighton, Bull. Am. Phys. Soc. **24**, No. 1, 13 (1949).

actually (x-ray)—(x-ray) coincidences as might be expected.†

If the curve of 4(a) is treated as that of a nuclear beta-ray spectrum, the end point could reasonably occur near 0.2 g/cm² or about 0.6 Mev as it appeared to Seaborg, Livingood, and Friedlander.††

Note added in proof: Shortly after the writers submitted this paper for publication, they became aware of the study of the radiations of Ge^{71} made by L. Seren, H. N. Friedlander, and S. H. Turkel, Phys. Rev. **72**, 888 (1947). Those authors describe in the fine print of Table III of their paper critical absorption measurements identical with those of the present report, which show that only the x-rays of gallium are emitted by Ge^{71}. They also state specifically that no electrons or beta-rays are to be found in the radiations of Ge^{71} so that the only mode of decay is that of orbital electron capture. These data are apparently also contained in a restricted version of the work of Seren *et al.* which is referred to in connection with Ge^{71} in the recent isotopic table compiled by Seaborg and Perlman.

APPENDIX: CHEMICAL PROCEDURE FOR THE SEPARATION OF GERMANIUM AND ARSENIC AND THE PURIFICATION OF THE GERMANIUM FRACTION

Germanium oxide from the pile was fused with K_2CO_3 and Na_2CO_3 in a nickel crucible, and the melt was dissolved in cold H_2O. After filtering out insoluble material, 50 mg of As_2O_3, dissolved in K_2CO_3 solution, was added as a carrier. To the solution was added one-half volume of 48 percent hydrofluoric acid. The new solution was then saturated with H_2S to precipitate the arsenic. The As_2O_3 precipitate was washed with 16 percent hydrofluoric acid and with water. It was dissolved in ammonium hydroxide and several drops of H_2O_2, diluted with water, made 6 normal with HCl and again precipitated with H_2S. The precipitate was washed with water, hydrofluoric acid, water, alcohol, and dried.

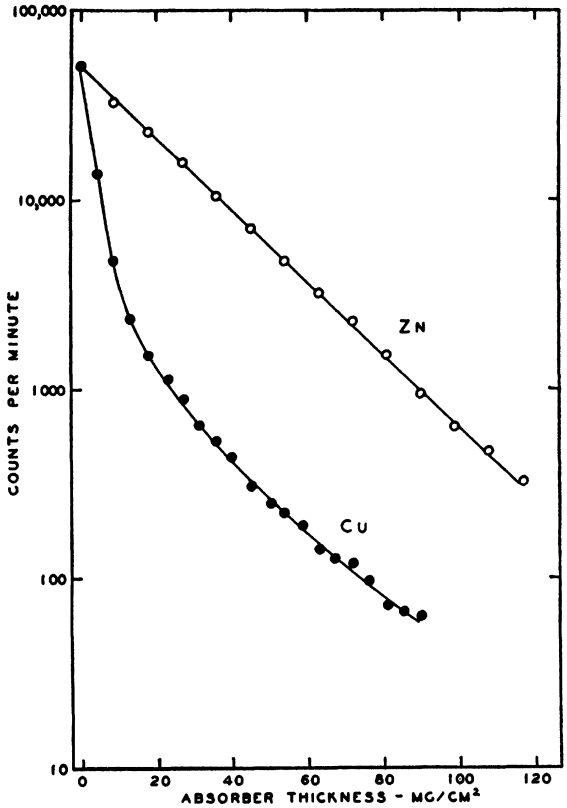

FIG. 5. Critical absorption of the x-rays of the 11-day Ge^{71}. The heavy absorption in copper indicates that most of the radiation is the gallium $K\alpha$-line.

The filtrate containing germanium was evaporated, after adding H_2SO_4 to expel the hydrofluoric acid. The residue was dissolved in water, and the acid concentration was adjusted to 25 percent free H_2SO_4. GeS_2 was precipitated with H_2S, washed with water, dissolved in hot $(NH_4)_2S$ and filtered. After adding H_2O_2 the solution was evaporated, the residue moistened with H_2SO_4, heated to expel $(NH_4)_2SO_4$ and free acid, and ignited to GeO_2. The procedure followed should have removed Ni, Fe, Cu, Ag, Ca, and Na from the GeO_2.

† Thought to be "K–L" coincidences.

†† L. C. Miller and L. F. Curtiss, Phys. Rev. **70**, 983 (1946), report 0.6 Mev beta-rays and 0.5 Mev gamma-rays as the radiations of the 11-day Ge^{71}. It seems probable that they have incorrectly assigned the beta-rays of As^{77} and the gamma-rays of Ge^{77} to Ge^{71}.