Assignment and Disintegration Scheme of the 6.75-Hour Molybdenum

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Contrary to previous reports, deuteron bombardment of molybdenum does not produce any measurable amount of the 6.75-hour Mo activity. The assignment of this activity has, therefore, been reinvestigated and correctly found to be still Mo^{93}. Decay does not take place by positron emission or K-capture. A disintegration scheme has been worked out using absorption, beta-ray spectrograph, and coincidence counter measurements. By successive emission of three gamma-rays of energies 0.30, 0.70 and 1.7 Mev of which the first is about 90 percent internally converted, the 6.75-hour Mo^{93} passes into a long-lived isomeric state. Mo K_α x-rays resulting from the conversion have been found.

INTRODUCTION

BY private communication,[1] a 7-hour Mo activity had been reported from a proton bombardment of columbium and tentatively assigned to Mo^{93}. The characteristic radiations were not given. Later workers[2] changed the value of the half-life to 6.7 hours and assigned the activity to Mo^{93} by producing it from columbium by protons and deuterons and from zirconium by alpha-particles. They reported beta-particles of energies 0.3 and 0.7 Mev and one gamma-ray of energy 1.6 Mev. Absence of x-rays was indicated and the method of decay by positron emission was confirmed by others[3] on the basis of Cb+d and Mo+d but no check on the energies of the emitted radiations was available. Recently, however, it has been pointed out[4] that though the value of the half-life was 6.75 hours and the maximum energies of the charged particles were 0.35 and 0.65 in fair agreement with those previously reported, these were not positrons but negative electrons from internally converted gamma-rays. Neither positrons nor x-rays were observed. The presence of a long-lived isomer, Mo^{93}, was also indicated.

The above observations, however, had left the disintegration scheme of the 6.75-hour activity in a very unsatisfactory state. Supposing, even, that the decay took place by internal conversion to the long-lived Mo^{93} isomeric state, the absence of x-rays, resulting from the conversion of the gamma-rays, was not consistent with this mode of disintegration. A study of the decay scheme was, therefore, deemed necessary. This paper reports the results obtained from absorption measurements, beta-ray spectrograph and coincidence methods which were brought together to work out the detailed mode of disintegration.

Another peculiarity connected with the 6.75-hour Mo activity was noticed in connection with our studies of nuclei in the Cb−Mo region. Contrary to the reports of others,[3] that this activity was produced by a deuteron bombardment of Mo, repeated efforts to produce it from metallic Mo with 10-Mev deuterons failed. The ease with which (d, p) reactions proceed, makes this phenomenon difficult to understand. To make certain that other Mo activities were not masking out the 6.75-hour activity, enriched molybdenum** with a concentration of 92.07 percent of Mo^{92} was bombarded by deuterons in order to produce this activity more strongly, but still without success. The correctness of the assignment of the activity to Mo^{93} is thus thrown into considerable doubt. It is, therefore, necessary to check the assignment by using enriched isotopes of neighboring elements.

ASSIGNMENT OF THE 6.75-HOUR ACTIVITY

The Mo fraction obtained from a bombardment of Cb with 10-Mev deuterons was followed with Geiger counters and with a Wulf unifilar electrometer provided with an ionization chamber. The value of the half-life from an interval of over ten half-lives is found to be 6.75±0.05 hours. It was also observed by the deflection of the charged particles from the sample in a magnetic field, that contrary to the previous reports,[2,3] these particles were negatively charged. No positrons of this half-life were detected. These observations open the question whether the 6.75-hour activity is to be assigned to the Mo^{93} resulting from $Cb^{93}(d, 2n)$ or to some other Mo isotope.

Bombardments with 5-Mev protons on Cb also produced the 6.75-hour activity quite readily. The proton bombardment does not lead to any definite assignment because the possible reactions may be $Cb^{93}(p, n)Mo^{93}$ or $Cb^{93}(p, \gamma)Mo^{94}$. It is, however, shown[5] that (p, n) reactions have larger cross sections than (p, γ) reactions in this region of atomic nuclei. It is, thus, plausible that an activity which is produced so readily would be caused by a (p, n) reaction rather than a (p, γ) reaction and, therefore, be assigned to Mo^{93}.

The assignment can be made more definite by the use of separated isotopes of Zr, since as reported

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[1] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).
[2] D. N. Kundu and M. L. Pool, Phys. Rev. **70**, 111 (1946).
[3] M. L. Wiedenbeck, Phys. Rev. **70**, 435 (1946).
[4] D. N. Kundu and M. L. Pool, Phys. Rev. **76**, 183 (1949).

** Supplied by Y-12 Plant, Carbide and Carbon Chemicals Corporation, through the Isotopes Division, A.E.C., Oak Ridge, Tennessee.

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

earlier,[2] the 6.75-hour activity is produced from Zr with 20-Mev alpha-particles. A rotating target arrangement[6] was employed to bombard two samples of Zr enriched respectively in isotopes Zr^{90} and Zr^{91}. It was found that the 6.75-hour activity was produced from Zr^{90} in quantities three times as much as from Zr^{91}. It has been observed that (α, n) reactions proceed more readily, at the alpha-energy used, than $(\alpha, 2n)$ reactions. The ratio $(\alpha, n)/(\alpha, 2n)$ is 45:1 in case[7] of Ru and 72:1 in case[8] of Y. The assignment is, therefore, in favor of Mo^{93} resulting from the reaction $Zr(\alpha, n)$.

The possibility of the assignment to Mo^{94} was further excluded by observing that a fast neutron bombardment on enriched Mo^{95} did not produce the 6.75-hour Mo activity.

The final confirmation was made by a simultaneous bombardment of Mo enriched separately in Mo^{92} and Mo^{94} isotopes, with fast neutrons. In this case, the 6.75-hour activity was produced from Mo^{94} but not from Mo^{92}. The 6.75-hour activity is, therefore, definitely assigned to Mo^{93}.

ABSENCE OF (d, p), (n, γ) AND (γ, n) REACTIONS

It was mentioned previously that the 6.75-hour Mo activity was produced from Mo^{94} by fast neutrons. The yield from the reaction Mo^{94} $(n, 2n)$ was, however, very low.

With regard to the failure of the 10-Mev deuterons to produce any appreciable amount of the 6.75-hour activity from Mo^{92}, it was suggested that the growth of the 5.9-hour Tc^{99} isomer from the 2.79-day Mo^{99} might possibly mask the decay of the 6.75-hour Mo activity. Such a possibility had to be discarded, however, subsequent to deuteron bombardments of Mo of ordinary isotopic composition, enriched Mo^{92}, and enriched Mo^{98}. The percent of Mo^{92} contained in the three samples was 15.84, 92.07, and 0.65 respectively and of Mo^{98} content was 23.78, 1.65, and 95.00 respectively. The percents are so widely different that if in one sample the 6.75-hour decay were compensated exactly by the 5.9-hour growth, then in the other two samples the decay-growth ratio ought to be distinctly seen.

The above results are consistent with the findings recently reported by others[9] that the 6.75-hour Mo

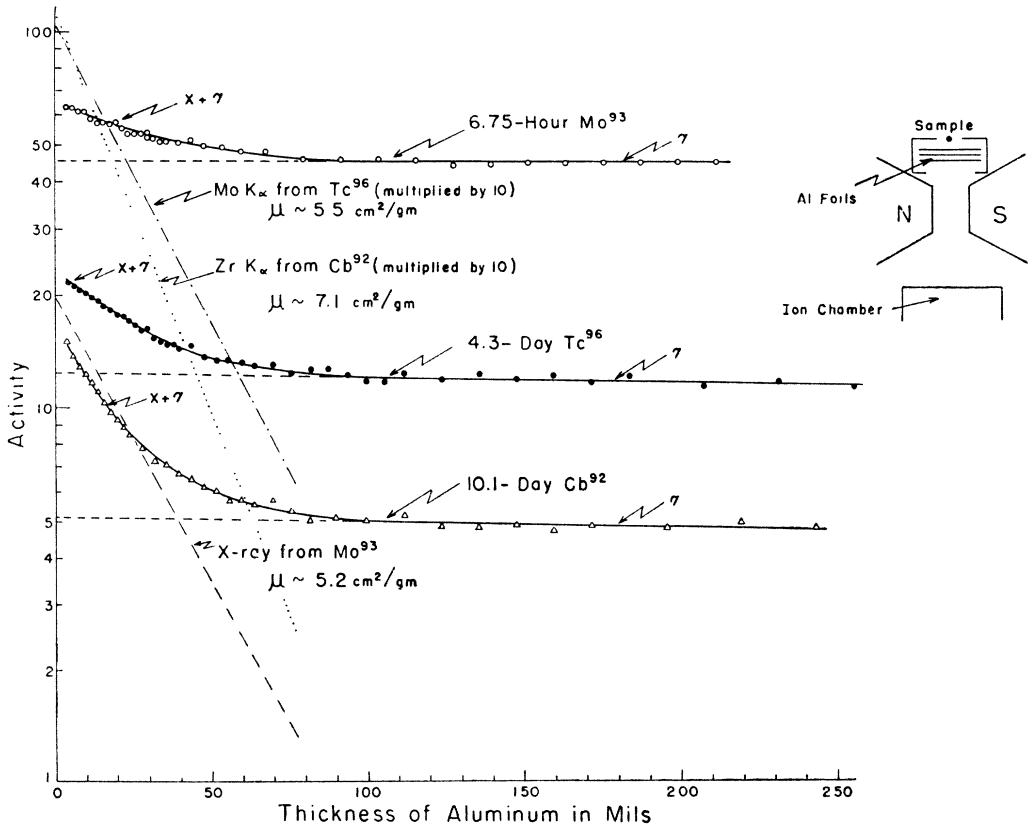

FIG. 1. Comparison of the absorption coefficient in aluminum of the x-rays from the 6.75-hour Mo^{93} with those of the K_α x-rays of Mo and Zr.

[6] D. N. Kundu and M. L. Pool, Phys. Rev. **74**, 1775 (1948).
[7] D. T. Eggen and M. L. Pool, Phys. Rev. **75**, 1464 (1949).
[8] Scott, Robertson, and Pool, Phys. Rev. **76**, 183 (1949).
[9] R. B. Duffield and J. D. Knight, Phys. Rev. **76**, 573 (1949).

activity was not produced by the (n, γ) reaction with slow neutrons. The additional fact that $Mo^{94}(\gamma, n)$ did not produce this activity either, may be understood in the light of the present data that the yield from $Mo^{94}(n, 2n)$ reaction was very low.

The observation that the $Mo^{92}(d, p)$ and $Mo^{92}(n, \gamma)$ reactions were not detected is probably related to the fact that Mo^{92} happens to be a nucleus with the magic number 50 and, therefore, has a very low neutron capture cross section.

COMPONENTS IN THE PRODUCTS OF DISINTEGRATION

Absorption measurements with thin aluminum foils show two end points with values for the maximum energies at 0.30±0.05 and 0.65±0.05 Mev. As mentioned previously these particles were verified to be negatively charged. Since Tc^{93} is not a stable isotope, these electrons are to be suspected as resulting from the internal conversion of γ-rays. In this connection, it may be noted that the ratio of the ionization for the γ-rays and for charged particles indicated that more than one gamma-ray must be involved in each disintegration process.

An absorption measurement with lead showed that apart from the hard γ-ray of 1.7±0.1 Mev, there is a second component of energy 0.70±0.05 Mev. The relative ionization at zero thickness caused by the 0.70-Mev component is 0.9 of that produced by the 1.7 Mev-component. This indicates that there are as many gamma-photons of higher energy as of the lower energy, and also that the small number of 0.65-Mev electrons may possibly result from the internal conversion, to a small extent, of the 0.70-Mev gamma-rays.

The conclusive proof that the 0.30-Mev group was caused by the internal conversion of another gamma-ray was obtained by the use of a beta-ray spectrograph. Two lines were photographed which corresponded approximately to the K and L conversion lines of molybdenum.

To find out if any of the 0.30-Mev group of gamma-rays remained unconverted, the sample was covered with 29 mils of Mo foil which would stop both the groups of charged particles and a thin aluminum absorption made. It was found that charged particles of these energies could still be detected. This shows that the conversion is not complete and also supports the view that these particles are not genuine beta-rays but they are conversion electrons.

The reported absence of x-rays was next examined. Absorption measurements were made in the usual way by placing the sample in a magnetic field and inserting thin aluminum foils immediately in front of the window of the ionization chamber or Geiger counter. In agreement with earlier reports, no definite indication of x-rays could be found. But it was suspected that the diminution of the ionization by x-rays might, perhaps, be just compensated by the increase in the ionization due to the secondary electrons which the gamma-rays

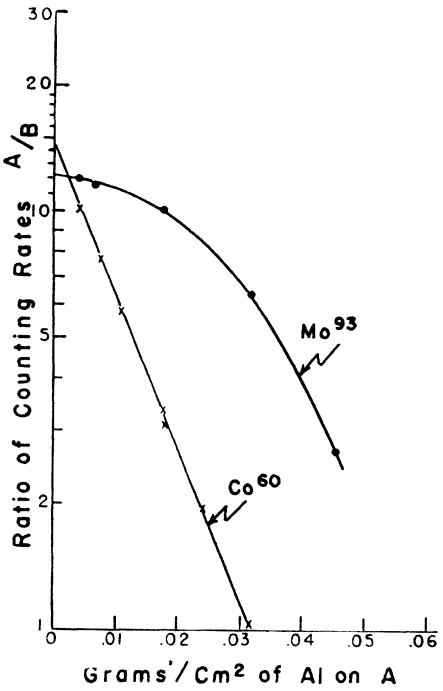

FIG. 2. Comparison of the beta-gamma-counting ratio in the 6.75-hour Mo^{93} with that for Co^{60}.

knocked out of the added aluminum foils. An arrangement was, therefore, devised to place the source and also the absorbing foils both in a strong magnetic field. No secondary electrons resulting from the aluminum foils could then reach the ionization chamber or Geiger counter. The schematic arrangement and the results of measurements are shown in Fig. 1. Distinct evidence of x-rays is indicated. The wave-length of the x-rays is calculated as 0.70A from the half-value thickness equivalent to an absorption coefficient of 5.52 cm^2/g. These x-rays are thus suspected to be Mo K_α. This was proved conclusively by comparing the observed absorption coefficient of x-rays in the 6.75-hour activity with the coefficients of known Mo and Zr x-rays under the same geometry and with intensities of the same order of magnitude. Mo K_α x-rays were obtained from the 4.3-day Tc^{96} and the Zr K_α x-rays from the 10.1-day Cb^{92}. The absorption coefficients were found to be 5.55 and 7.1 cm^2/g respectively. The absorption coefficients thus arrived at are different enough to easily distinguish between Mo x-rays and those from neighboring elements.

During the decay of the 6.75-hour Mo^{93}, the ratio of the ionization from the γ-rays to that from the x-rays is 2.6 for the ionization chamber used. For this chamber, each γ-quantum produces about the same ionization as each 0.70A X-quantum. Each x-ray quantum must, therefore, be associated with at least two γ-quanta. Which of these radiations are in series was determined by coincidence counter measurements, presented in the following section.

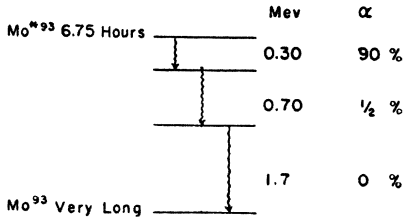

FIG. 3. Disintegration scheme of 6.75-hour Mo^{93}.

COINCIDENCE COUNTER DATA

The coincidence study of the radiations from Mo^{93} was made with thin end-window beta-ray Geiger tubes. When used for the detection of gamma-rays only, thick Lucite absorber-radiators were placed in front of the windows. The gamma-ray energy *versus* gamma-ray detection efficiency for this arrangement was found to be very nearly linear[10] over the calibrated range from 0.4 Mev to 2.8 Mev.

The beta-gamma-coincidence rate per beta-particle *versus* beta-ray absorber thickness was consistent with a simple beta-ray spectrum in series with a total of 2.5 ± 0.05 Mev of gamma-ray energy.

Lead absorption measurements of the gamma-gamma-coincidence rate per gamma-ray indicated that the 2.5 Mev total gamma-ray energy is split principally into two components of approximate energies 1.8 Mev and 0.7 Mev.

The beta-gamma-counting ratio extrapolated to zero beta-ray absorber thickness when compared with that for Co^{60} (0.31 maximum beta-ray energy, 2.50 total gamma-ray energy[11]) indicates that the 0.30-Mev transition in Mo^{93} is 0.9 internally converted. Figure 2 shows the ratio of the counting rate in the beta-ray tube to the counting rate in the calibrated gamma-ray tube with a thick Lucite radiator as a function of the grams/cm^2 of aluminum in front of the beta-tube. It may be shown that

$$\frac{(N_{\beta 1}/N_{\gamma 1})E_1}{(N_{\beta 2}/N_{\gamma 2})E_2}=1+\alpha,$$

when there is a primary beta-spectrum. If, however, there is no primary beta-spectrum, as in the present case, the right-hand side of the above equation reduces simply to α. In the above $N_{\beta 1}/N_{\gamma 1}$ = extrapolated e^--gamma-counting ratio in Mo^{93}, $N_{\beta 2}/N_{\gamma 2}$ = extrapolated beta-gamma-ratio in Co^{60}, E_1 and E_2 are the total gamma-ray energies in series with the beta-spectra of Mo^{93} and Co^{60}, respectively, and α = the total internal conversion coefficient.

The beta-beta-coincidence rate per beta-particle *versus* aluminum absorber thickness in front of both tubes indicates that no radiation of energy greater than 0.1 Mev, other than the 0.3-Mev transition, is internally converted more than one-half percent.

A delayed coincidence study of beta-gamma-, gamma-beta- and gamma-gamma-coincidence revealed that the half-lives of the intermediate states involving the 0.3-, 0.7-, and 1.7-Mev components are less than 1×10^{-7} seconds.

DISINTEGRATION SCHEME

Summarizing the observations of the two foregoing sections, it is found that in the decay of Mo^{93}, there are three gamma-rays of 0.30, 0.70, and 1.7 Mev. The first of these is almost completely internally converted, giving rise to Mo K_α x-rays. The other two gamma-rays are in cascade with the first gamma. The 0.70-Mev gamma is very slightly converted to the extent of only one-half percent at most. The mode of decay is not by K-capture, unless it be to the extent of a small percent. No positron emission was observed.

The absence of positrons and the presence of Mo K_α x-rays alone coupled with the fact that a high percent of the 0.30-Mev gamma-rays is internally converted indicate that there must be a Mo^{93} isomer, and that this isomer must be long-lived.

Two more facts, already in the literature, also substantiate the above remarks. The first is that the 17-minute Mo activity has been shown[4] to be Mo^{92} and not Mo^{93}, as previously reported by others.[1] The second fact is that the Tc^{93} isomers of half-lives 4.5 minutes and 2.75 hours decay into Mo^{93} primarily by positron emission and K-capture respectively.[6] No growth of the 6.75-hour Mo was observed from Tc^{93}. These observations also indicate that Mo^{93} must have a very long-lived isomer.

On the basis of the above findings, the decay scheme shown in Fig. 3 is, therefore, suggested. After three successive gamma-emissions, the 6.75-hour activity passes into the proposed long-lived isomer Mo^{93}.

ACKNOWLEDGMENT

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[10] Lind, Brown and DuMond, Phys. Rev. **76**, 591 (1949).
[11] C. E. Mandeville and M. V. Scherb, Nucleonics, **3**, 2 (1948).