

Decay of Molybdenum-91

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A study has been made of the radioactivity of Mo^{91} and Mo^{91m} , produced by x-irradiation of natural molybdenum, in order to clarify the decay scheme of this isotope. The gamma-ray spectrum was measured using scintillation spectrometers and coincidence circuits. Three gamma rays which had a half-life of 64 seconds were found at 1.50, 1.20, and 0.65 Mev. The first two were in coincidence with positrons but not with each other, while the 0.65-Mev gamma ray was not in coincidence with positrons or with the two other gamma rays. The intensities of the gamma rays were in the ratios 1.5 Mev:1.2 Mev:0.65 Mev = 39:39:100. By using a plastic scintillator, positrons from the ground state of Mo^{91} (15 minute half-life) were found to have an end-point energy of 3.3 Mev while two positron groups from the isomeric state were found with energies differing by 0.2 ± 0.1 Mev.

On the available evidence a decay scheme is suggested.

THE decay scheme of Mo^{91} has been studied by a number of authors. The ground state, which has a half-life of 15 minutes, emits positrons, and an isomeric state which has a half-life of about 70 seconds decays either by gamma-ray de-excitation to the ground state or by positron emission. At the University of Illinois, Axel, Fox, and Parker¹ measured the branching ratio of the decay of the isomeric state, gamma/positron, and found it to be 2.4:1. This group found three gamma rays, of energies 0.67, 1.22, and 1.55 Mev, which were associated, by decay period, with the isomeric-state decay, and suggested that higher energy ones of low intensity might be present.

Katz, Baker, and Montalbetti² measured the maximum energy of the ground-state positrons and found it to be 3.3 ± 0.1 Mev.

A complete list of these and other earlier references has been compiled by Way *et al.*³

More recently, and since the present work was commenced, an account has been published of a more detailed investigation at the University of Illinois by Smith, Gove, Henry, and Becker,⁴ and a complete decay scheme has been drawn up by these authors. The results of the present work are in almost complete agreement with those of the Illinois group, but as the methods employed were in some ways different, and as the results provide an independent confirmation of the decay scheme suggested by the Illinois group, it is felt that a short account of the Glasgow work and results is of value.

In the present work we have had two objectives: (1) to study the spectrum of the gamma rays from this isotope in order to find their relative intensities and to discover, if possible, the higher energy gamma rays suggested by Axel *et al.*¹; (2) to clarify the decay

scheme by an investigation of gamma-gamma and positron-gamma coincidences, and by a study of the positrons emitted by Mo^{91} and Mo^{91m} .

APPARATUS AND PROCEDURE

Mo^{91} was produced by the reaction $\text{Mo}^{92}(\gamma, n)\text{Mo}^{91}$ by the irradiation of natural molybdenum in the x-ray beam of a 23-Mev synchrotron.

For examining the β and γ spectra of the Mo^{91} , plastic and 2-in. thick NaI scintillators were used in conjunction with DuMont 6292 photomultipliers, conventional circuits, and a 100-channel pulse height analyzer.

In the γ - γ and γ - β^+ coincidence work, there was in addition to the above apparatus a 2-in. thick NaI crystal spectrometer used with a single-channel pulse-height analyzer. The output from this analyzer could operate a gate in the input of the 100-channel pulse-height analyzer. By using this equipment, the beta and gamma spectra in coincidence with pulses from the total energy peak ("photopeak") from a particular gamma line could be determined. Corrections were made, if necessary, for the fact that Compton recoils due to higher energy gamma rays could operate the gate as well as photoelectrons from the desired gamma line. In addition, corrections were made for the effects of longer lived activities induced in the other molybdenum isotopes.

GAMMA-RAY MEASUREMENTS

A typical gamma-ray spectrum of Mo^{91} and Mo^{91m} obtained in this work is shown in Fig. 1. The spectrum shows the presence of lines at 0.51, 0.65 ± 0.02 , 1.20 ± 0.02 , and 1.50 ± 0.02 Mev. The 0.51-Mev gamma rays were identified as positron annihilation quanta. In some of the runs weak peaks were also present at 1.7 Mev and 2.0 Mev, but these were thought to be due to the addition of 0.51- with 1.20-Mev gamma rays and 0.51- with 1.50-Mev gamma rays. This was confirmed by measurements taken with the source at varying

¹ Axel, Fox, and Parker, Phys. Rev. **97**, 975 (1955).

² Katz, Baker, and Montalbetti, Can. J. Phys. **31**, 250 (1953).

³ *Nuclear Level Schemes, A=40→A=92*, compiled by Way, King, McGinnis, and van Lieshout, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, 1955).

⁴ Smith, Gove, Henry, and Becker, Phys. Rev. **104**, 706 (1956).

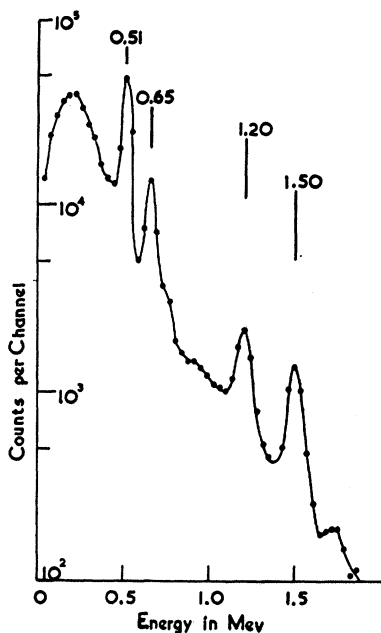


FIG. 1. The gamma spectrum of Mo^{91} on a semilogarithmic plot. The energy scale was calibrated by using Na^{22} and Cs^{137} .

distances from the crystal. No other evidence was found for lines other than the four first given above.

By using the gamma spectra obtained in a number of runs, and knowing the relative efficiency of the crystal for the different gamma-ray energies, the relative strengths of the 0.65-, 1.20- and 1.50-Mev lines were computed. It was found that the intensities of the rays were in the ratios

$$0.65 \text{ Mev} : 1.20 \text{ Mev} : 1.50 \text{ Mev} = 100 : 39 : 39.$$

It will be seen that the two higher energy lines are of approximately equal intensity.

By using the single-channel pulse-height analyzer, set to accept pulses from the photopeaks of the three lines, measurements were made of the half-lives of the three gamma rays. All three had the same half-life to within the experimental accuracy, the average value being 64 ± 1 seconds.

γ - γ COINCIDENCE MEASUREMENTS

With the single-channel pulse-height analyzer set on the photopeaks of the 0.51-, 1.20- and 1.50-Mev lines, respectively, determinations were made of the spectra of gamma rays in coincidence with these three lines. These gamma-gamma coincidence results are presented graphically in Figs. 2(a), (b), (c), and are summarized below. The range of energy investigated in each case is given in parentheses.

(a) In coincidence with 0.51-Mev gamma rays ($0.2 < E < 1.8$ Mev)—0.51 Mev, 1.20- and 1.50-Mev gamma rays.

(b) With 1.20- and 1.5-Mev gamma rays ($0.3 < E < 2.0$ Mev)—0.51-Mev gamma rays.

(c) With 1.20-Mev gamma rays ($0.1 < E < 0.7$ Mev)—0.51-Mev gamma rays.

Thus while the 1.20- and 1.50-Mev lines are associated with a positron decay of the isomeric state, the 0.65-Mev line is not. Moreover, the 1.2-Mev and 1.5-Mev rays are not in coincidence, even though this is suggested by their having equal intensities. A special search was made for a 0.3-Mev gamma ray which might be expected to be in coincidence with the 1.20-Mev line (1.5-Mev minus 1.2 Mev) but no such line was found [Fig. 2(c)]. The large number of counts between 0.1 and 0.3 Mev is due to scattering of the 0.5-Mev annihilation quanta, and is relatively high because of the thickness of the source being used. This explanation was confirmed by using a thick Cu^{62} positron source.

POSITRON ENERGY MEASUREMENTS

1. 15-Minute Decay

A 0.01-inch-thick Mo sheet was irradiated in the x-ray beam for 30 minutes, and then placed before a

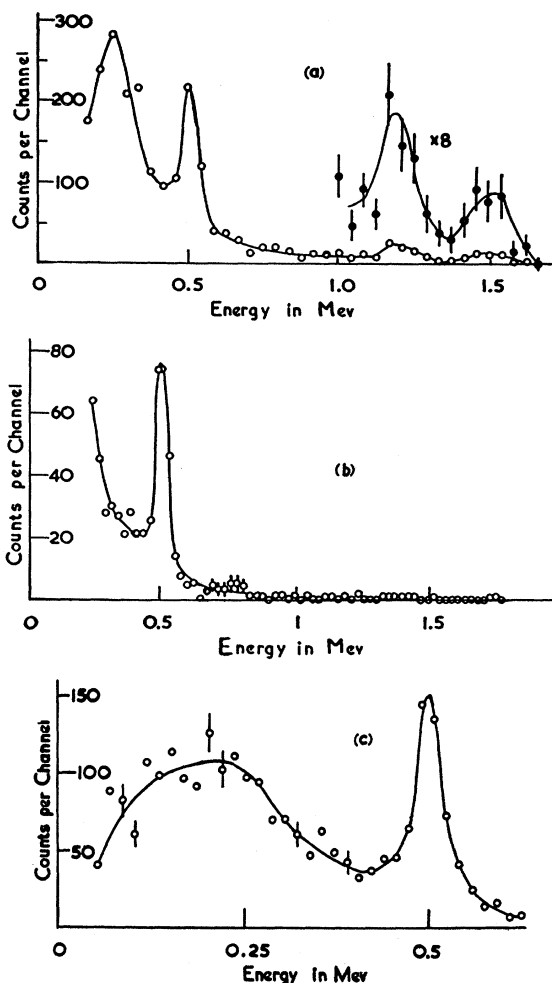


FIG. 2. The spectra of gamma rays in coincidence with (a) 0.51-, (b) 1.2- and 1.5-, and (c) 1.2-Mev gamma rays. The energy scale was calibrated by using Na^{22} .

1-inch-thick plastic scintillator, the spectrum from which was displayed on a 100-channel pulse-height analyzer. After allowing time for the isomeric-state activity to die away, the positron spectrum of the 15-minute activity was determined. The average of several runs gave an end-point energy of 3.3 ± 0.2 Mev, in agreement with previous measurements of this energy by Katz, Baker, and Montalbetti.²

2. 64-Second Decay

Immediately after x-ray irradiation a sheet of Mo was placed between a plastic scintillator and a 2-in. thick NaI crystal. As described in a previous paragraph, the output from the plastic scintillator went to the 100-channel pulse-height analyzer, while pulses from the NaI crystal spectrometer were fed to a single-channel pulse-height analyzer which in turn operated the coincidence gate in the 100-channel pulse-height analyzer circuit. By a proper setting of the single-channel analyzer, the spectra of positrons in coincidence with either 1.5- or 1.2-Mev gamma rays could thus be obtained.

Although low counting rates made an absolute measurement of the end points of the positron energies untrustworthy, a comparison of the curves obtained in coincidence with the 1.20-Mev and 1.50-Mev lines, respectively, showed that the former had an end-point energy 0.2 ± 0.1 Mev higher than the latter. It is therefore concluded that positron decay of the isomeric state takes place by two branches to two excited states of Nb^{91} , one of which de-excites by means of a 1.20-Mev gamma ray, and the other by a 1.50-Mev gamma ray.

DISCUSSION

A decay scheme for molybdenum can now be drawn up using the results obtained in this work and previous results obtained by other authors.³

The ground state (15 minutes) and isomeric state (64 seconds) of Mo^{91} have been assigned spins and parities of $9/2^+$ and $1/2^-$, respectively, and the same assignment has been made to the ground state and to the 0.1-Mev isomeric state of Nb^{91} .

Relative to the ground state of Mo^{91} , the energy of the ground state of Nb^{91} is fixed by the measurement of the 3.3-Mev 15-minute positron decay of molybdenum. The Mo^{91} isomeric state is shown to be 0.65 Mev above its ground state by the energy of the gamma ray which is not associated with positron decay of the isomeric state.

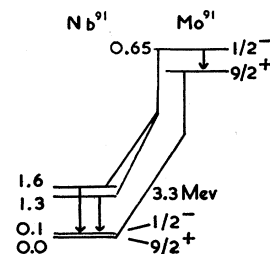


FIG. 3. The proposed decay scheme of Mo^{91} .

The positrons from the $1/2^-$ isomeric state have allowed ft values, and it is therefore most probable that the gamma rays emitted after the positron decay go to the $1/2^-$ isomeric state of Nb rather than to the $9/2^+$ ground state. It is therefore possible to fix the positions of the two states from which these two gamma rays come at $0.1 + 1.2$ Mev and $0.1 + 1.5$ Mev above the Nb^{91} ground state. The possibility of the two gamma rays starting from the same level and de-exciting to two lower levels is excluded by the lack of 0.3-Mev gamma rays and by the fact that the coincidence positrons are of different energies.

Knowing the positions of these two excited levels and that of the isomeric state of Mo^{91} , one may calculate the energies of the positrons in coincidence with the 1.2- and 1.5-Mev gamma rays, and these energies are found to be 2.65 Mev and 2.35 Mev, respectively. The values are consistent with the shapes of the positron spectra found experimentally, and the energy difference, 0.3 Mev, is in agreement with that found experimentally.

The proposed decay scheme is illustrated in Fig. 3.

It will be seen that even if there is no direct positron decay to the 0.1-Mev level of Nb^{91} , the relative probabilities of decay of the isomeric state of Mo^{91} via gamma de-excitation as against positron emission is $100 : (2 \times 39)$, or 1.28:1. This is not in agreement with the value of 2.4:1 for this ratio found by Axel, Fox, and Parker,¹ but agrees with the later Illinois value.⁴

To summarize: (1) No gamma rays of energy higher than 1.50 Mev have been found in the decay of Mo^{91} , and the relative probabilities of decay of the isomeric state via gamma de-excitation as against positron emission is at the most 1.28:1. (2) By coincidence and energy measurements, a decay scheme for Mo^{91} has been established. This decay scheme, as stated before, is almost identical with that independently proposed by Smith, Gove, Henry, and Becker⁴ and the agreement of the results from the two laboratories is strong evidence for the truth of this picture.