Radioactive Decay of Lutetium 173

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Ytterbium oxide enriched to 92.6% in the 173 mass number was irradiated with 6-Mev protons. An activity with a half-life of 625±50 days was produced and its assignment to Lu^{173} confirmed by the identification of the ytterbium K x-ray, gamma rays corresponding to transitions among the energy levels established in Yb^{173} by Coulomb excitation of the separated isotope, and the activities produced by the similar proton irradiations of the other enriched isotopes of ytterbium. The observed activity of Lu^{173} consists of the L and K x-rays of ytterbium and gamma rays with energies of 79±1, 101±1, 172±2, 273±2, 349±4, ~450, 556±4, and 630±4 kev and two other gamma rays not resolved in the gamma-ray spectrum but observed in gamma-gamma coincidence measurements. These two gamma rays have energies of approximately 179 and 282 kev. Because no positron radiation exists in the activity of Lu^{173}, the mode of decay is solely by electron capture to Yb^{173}. Gamma-gamma coincidence measurements have lead to the assignment of a 633-kev level with a probable spin of $\frac{5}{2}+$ and the confirmation of a 351-kev level in Yb^{173} in addition to the previously known 179.5- and 78.7-kev levels. The disintegrations of Lu^{173} to the 633-kev level are by L capture only. Branching ratios for the electron capture transitions of Lu^{173} to the levels of Yb^{173} and approximate relative probabilities for the transitions in Yb^{173} are given in a proposed energy level scheme for the decay of Lu^{173}. The choice of 9/2- for the ground-state spin of Lu^{173} is the most consistent with the proposed energy level scheme.

INTRODUCTION

An activity decaying by electron capture with a half-life of approximately 500 days has been assigned to Lu^{173}.[1] Energy levels of 78 and 181 kev were observed in Yb^{173} by Coulomb excitation of the separated isotope.[2] Spins of 7/2 and 9/2, respectively, were assigned to these levels. Earlier Coulomb excitation of natural ytterbium had resulted in the assignment of 78- and 180-kev levels to Yb^{173}.[3] The measured ground-state spin of Yb^{173} is 5/2.[4]

Recently a number of papers concerning the radioactivity of Lu^{173} produced by various reactions have appeared but significant discrepancies exist among the reported data. Conversion electron measurements on an approximately 450-day half-life activity produced by the proton irradiation of natural ytterbium oxide resulted in the assignment of 78.8-, 100.9-, 171.5-, and 272.7-kev transitions to the activity of Lu^{173}.[5]

Conversion electron measurements and gamma-ray studies have been made by four groups of workers on an activity in a lutetium fraction produced by the irradiation of tantalum with 660-Mev protons.[6–9] The half-life reported by these workers for this activity designated as Lu^{173} varies from 150 to 200 days. A total of six internal conversion transitions and seven gamma-rays were reported in the activity of Lu^{173} in these four references. Conflicting coincidence data, branching ratios, and energy level schemes for the decay of Lu^{173} were presented.

In a recent paper, gamma rays of energies 22, 79, 113, 145, 176, 274, 335, 440, 550, and 640 kev were associated with the activity of Lu^{173}.[10] The half-life of this observed activity produced by the irradiation of natural lutetium oxide with 24-Mev betatron bremsstrahlung was quoted as 1.4 years. Some gamma-gamma coincidence data were tabulated for these gamma rays.

Discrepancies on the following points are evident in the available information concerning the radioactive decay of Lu^{173}: (1) the value of the half-life, (2) the position of the 272-kev transition, (3) the existence of transitions of energy greater than 351 kev and a few of lower energy and the position of these in the energy level scheme, (4) the amount of electron capture transitions to the levels of Yb^{173}, especially to the ground state.

The enriched isotopes of ytterbium have recently become available. It is the purpose of the present investigation to resolve the discrepancies listed above by discussing the results of an examination of the activity of Lu^{173} produced by the proton irradiation of the enriched isotopes of ytterbium.

EXPERIMENTAL RESULTS

Ytterbium oxide enriched to 92.6% in the 173 mass number was irradiated with 6-Mev protons. The composition of the remaining portion is as follows in percent: 0.05 Yb^{168}, 0.05 Yb^{170}, 0.44 Yb^{171}, 2.33 Yb^{172}, 4.31 Yb^{174}, and 0.38 Yb^{176}. The atomic number of the resulting activity was determined by the identification of the ytterbium K x-ray which was compared with

[1] G. Wilkinson and H. G. Hicks, Phys. Rev. **81**, 540 (1951).
[2] Elbek, Nielsen, and Olesen, Phys. Rev. **108**, 406 (1957).
[3] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **100**, 150 (1955).
[4] A. H. Cooke and J. G. Park, Proc. Phys. Soc. (London) **A69**, 282 (1956).
[5] Mihelich, Harmatz, and Handley, Phys. Rev. **108**, 989 (1957).
[6] Bobrov, Gromov, Dzhelepov, and Preobrazhenskii, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **21**, 940 (1957) [Columbia Technical Translation (942)].
[7] Gorodinskii, Murin, Pokrovskii, and Preobrazhenskii, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **21**, 1004 (1957) [Columbia Technical Translation (1005)].
[8] Dzhelepov, Preobrazhenskii, and Sergiyenko, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **22**, 795 (1958) [Columbia Technical Translation (789)].
[9] Gorodinskii, Murin, Pokrovskii, and Preobrazhenskii, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **22**, 818 (1958) [Columbia Technical Translation (812)].
[10] Dillman, Henry, Gove, and Becker, Phys. Rev. **113**, 635 (1959).

the known K x-rays of europium, terbium, thullium, ytterbium, lutetium, and tantalum emitted from radioactive Gd^{153}, Dy^{159}, Yb^{169}, Tm^{170}, Hf^{175}, and W^{181}, respectively. Ion-exchange separation was deemed unnecessary.

In order to determine the mass number of the activity, similar proton irradiations were performed on each of the other enriched stable isotopes of ytterbium and the resulting activities intercompared. When Yb^{174} was irradiated, the well established 165-day activity of Lu^{174} was produced. When Yb^{173} was irradiated, the above activity was not observed but a longer half-life substance was found. This activity secured from Yb^{173} was not observed in any of the activities produced when the remaining enriched isotopes of ytterbium were irradiated with protons.

The half-life of the activity resulting from the proton irradiation of Yb^{173} is 625±50 days as determined by following the decay of the K x-ray and the 79- and 172-kev gamma rays for 200 days. It is certain that the half-life of this observed activity is not in the 170- to 200-day range and the original assignment of the 500-day activity to Lu^{173} is confirmed.

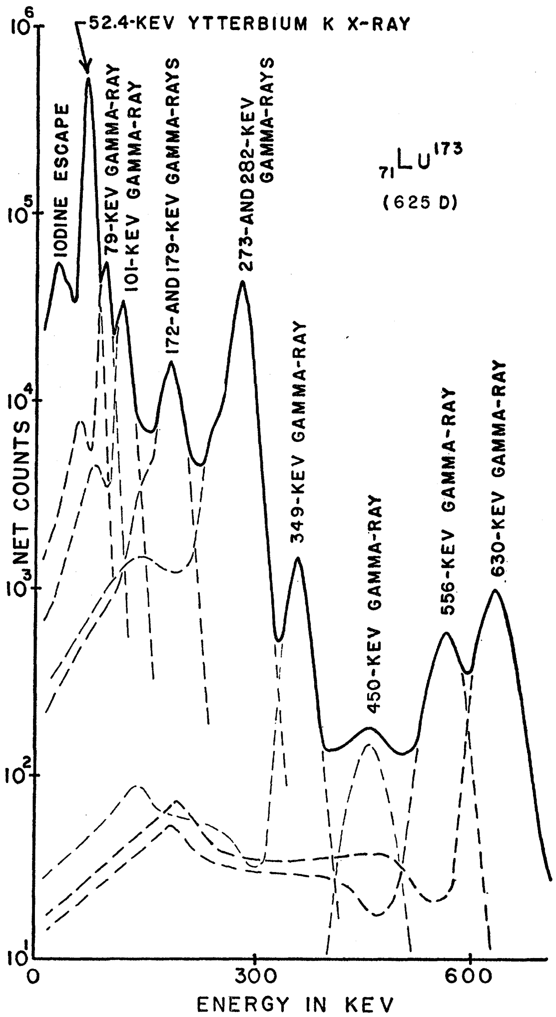

FIG. 1. Gamma-ray spectrum of the 625-day activity of Lu^{173} measured with a 3 in.×3 in. NaI scintillation crystal employing a geometry which minimized summation affects.

L and K x-rays were detected with a Geiger-tube used with aluminum and beryllium absorbers. Figure 1 shows the observed gamma-ray spectrum of Lu^{173} which includes 79±1-, 101±1-, 172±2-, 273±2-, 349±4-, ~450-, 556±4-, and 630±4-kev gamma-rays in addition to the previously mentioned ytterbium K x-ray. Two other gamma rays not resolved in the gamma-ray spectrum but observed in gamma-gamma coincidence measurements have energies of 179 and 282 kev. No evidence of positron activity was found in Lu^{173} by the method of plastic scintillation spectrometry, by the use of a Geiger-tube with aluminum and beryllium absorbers, nor by a search for annihilation radiation in the gamma-ray spectrum. Therefore, the mode of decay of Lu^{173} is solely by electron capture to Yb^{173}.

TABLE I. Gamma-gamma coincidence data for the activity of Lu^{173}. Energies are expressed in kev.

	630	556	349	273, 282	172, 179	101	79	K x-ray
K x-ray	no	yes	yes	yes	yes	yes	yes	yes
79	no	weak	no	yes	weak	yes	no	
101	no	no	no	no	weak	no		
172, 179	no	no	no	no	weak			
273, 282	no	no	?	weak				
349	no	no	no					
556	no	no						
630	no							

Table I displays the gamma-gamma coincidence information for the activity of Lu^{173} obtained by the use of a coincidence circuit of resolving time $2\tau=1.5$ μsec. In addition to the data shown in the table, the 273-kev gamma ray is in coincidence with two coincident K x-rays; and the 172, 179-kev peak is in coincidence with a weak 450-kev gamma-ray. One of the two coincident K x-rays in coincidence with the 273-kev gamma-ray originates from K capture preceding the 273-kev transition and the other, from K conversion of the 78.8-kev transition. 78.7- and 179.5-kev levels in Yb^{173} have been established by Coulomb excitation of the separated isotope.[2] A 351-kev level has been assigned to Yb^{173} by other workers from the observation of a 351-kev transition and from conversion electron coincidences between the 78.7- and 272.5-kev transitions.[8] This assignment is now confirmed by gamma-gamma coincidence measurements and the observation of a gamma ray of approximately 351 kev. Because the observed 630-kev gamma ray is in coincidence with no other gamma ray, a 556-kev gamma ray is in coincidence with the 78.7-kev gamma ray, a 450-kev gamma ray is in coincidence with the 172, 179-kev gamma peak, and a 282-kev gamma ray is in coincidence

with the 273-kev gamma ray, a 633-kev level is assigned to Yb^{173}. The 630-kev gamma ray and the K x-ray are not in coincidence. On the basis of the intensity of the coincidences between the 556-kev gamma ray and the K x-ray, it is concluded that if five percent or more of the electron capture transitions of Lu^{173} to the 633-kev level of Yb^{173} were by K capture, then coincidences between the 633-kev gamma-ray and the K x-ray could have been detected. The K x-ray in coincidence with the 556-kev gamma-ray is the x-ray resulting from K conversion of the coincident 78.7-kev transition.

The spins of the 351.1-, 179.5-, and 78.7-kev levels of Yb^{173} have been designated as $7/2+$,[7–9] $9/2-$, and $7/2-$, respectively, and the measured ground-state spin of Yb^{173} is $5/2-$. An examination of the relative probabilities of the transitions leaving the 351-kev level of Yb^{173} shows that the most probable transition involves no spin change but does involve a parity change, and the second most probable transition involves a spin change of one and a parity change. The choice of $5/2+$ for the spin of the 633-kev level of Yb^{173} leads to exactly the same set of conditions for the first and second most probable transitions from this level. Although this argument for a spin assignment to the 633-kev level is of questionable validity, it is the best which can be given with the available information. The choice of $9/2-$ for the ground-state spin of Lu^{173} is the most consistent with the proposed energy level scheme.

The fourth column of Table II gives the relative number of counts under the spectral distribution after correction for crystal efficiency for the observed radiations listed in the first column. The sixth column gives the relative number of the corresponding transitions calculated by using the ratios listed in column five which were derived from Rose's tables.[11] Conversion electron energies and multipole orders corresponding to the observed gamma rays are listed in the second column of Table II and the appropriate references in column three. Column eight shows the approximate relative number of K x-rays resulting from internal conversion of the transitions in Yb^{173}. The combined 172, 179-kev peak was divided by using the ratio of 172- to 179-kev transitions from reference 8. A rough division of the combined 273, 282-kev peak was obtained from analysis of gamma-gamma coincidence data.

Internal conversion of the transitions from the 633-kev level may be considered negligible for the following calculations. The approximate percentages of electron capture to the five energy level of Yb^{173} were obtained by accounting for the K x-rays observed in the activity of Lu^{173}. L capture to the other four levels was considered negligible with respect to K capture. One K x-ray was subtracted for every K capture required to balance the difference between the number of transitions from an excited level and the number of transitions to the same level. These differences were used as the relative number of electron capture transitions from Lu^{173} to the levels of Yb^{173}. K x-rays were then subtracted to account for K conversion of the transitions in Yb^{173}. The number of L capture transitions to the 633-kev level of Yb^{173} were determined from the number of transitions leaving this level. All the relative numbers for electron capture and transitions in Yb^{173} were then adjusted to yield a

TABLE II. Relative number of gamma rays, N_γ, corresponding transitions, N_{trans}, and K converted transitions, N_k, in the activity of Lu^{173} for energies, E_γ, expressed in kev. E_{ce} are internal conversion transition energies with references. N_{trans}/N_γ and N_{trans}/N_k are derived from Rose's tables.[a]

E_γ	E_{ce}[b]	Reference	N_γ	N_{trans}/N_γ	N_{trans}	N_{trans}/N_k	N_k
K x-ray	···	···	100	···	100	···	···
79±1	78.7, M_1	8,6,5	5.0	11.4	57.0	1.9	30
101±1	100.8, M_1	8,6,5	3.6	4.53	16.3	1.5	11
172±2	171.6, E_1	8,6,5	3.1	1.08	3.3	15	0.2
~179	179.5, E_2	8,6	1.0	1.40	1.4	6.4	0.2
273±2	272.4, E_1	8,6,5	17.1	1.02	17.5	51	0.3
~282	···	···	0.30	···	0.3	···	···
349±4	351.1, E_1	8	0.47	1.02	0.5	60	0
~450	···	···	0.09	···	0.1	···	···
556±4	···	···	0.60	···	0.6	···	···
630±4	···	···	1.1	···	1.1	···	···

[a] See reference 11.
[b] Note: Internal conversion transition energies corresponding to the four unreferenced gamma rays have recently been reported [see Harmatz, Handley, and Mihelich, Phys. Rev. 114, 1082 (1959)].

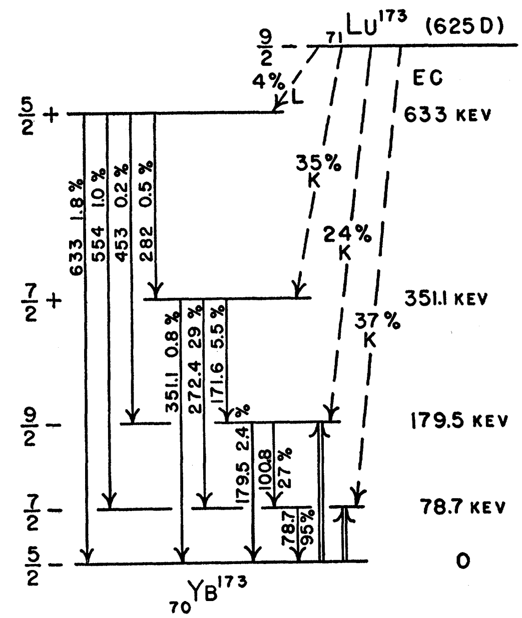

FIG. 2. Proposed energy level scheme for the decay of Lu^{173}. The percentage associated with each transition is the percentage of total disintegrations proceeding through that transition.

[11] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

total of 100% for the electron capture transitions to the five levels of Yb^{173}. The percentages of electron capture to the 633-, 351.1-, 179.5-, and 78.7-kev levels and the ground state of Yb^{173} are thus approximately 4, 35, 24, 37, and 0, respectively.

DISCUSSION

The discrepancies enumerated in the introduction are now considered. The 625-day half-life of Lu^{173} is closest to the originally assigned value of 500 days. The 273-kev transition is not to the ground state of Yb^{173} but originates from 351-kev level. Transitions of energy greater than 351-kev do exist in the activity of Lu^{173} and their positions in the energy level scheme are shown in Fig. 2. The 22-, 113-, 145-, and 335-kev gamma rays mentioned in the introduction are not observed by in the gamma-ray spectrum of Lu^{173} nor have any of them been observed by conversion electron measurements. A 22-kev gamma ray was shown not to exist in an amount greater than two percent of the K x-ray by absorption measurements with a scintillation spectrometer. Peaks at about 113 and 335 kev are observed in the gamma-ray spectrum of Lu^{173} when a geometry is used which allows coincidence summation to occur but these two peaks disappear rapidly when the K x-ray is absorbed. The conclusion is that the 113-kev peak is the coincidence sum peak of two K x-rays, and the 335-kev peak is that of a 273-kev gamma ray and a K x-ray. The results of this investigation show that few if any electron capture transitions of Lu^{173} occur to the ground state of Yb^{173}.

The gamma-gamma coincidence information obtained in this investigation agrees with that mentioned in the introduction[10] for the cases where the gamma rays of the two investigations correspond. In particular, the 172- and 273-kev peaks were each in coincidence with themselves and the 556-kev gamma ray was in coincidence with the 79-kev gamma ray only.

ACKNOWLEDGMENTS

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Isomeric States of Nd^{141} and Sm^{143}†

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Two previously unreported nuclear isomers which belong to the series having 81 neutrons and even numbers of protons have been found and studied. These are Nd^{141m} (half-life 63.7 seconds, gamma-ray energy 0.76 Mev) and Sm^{143m} (half-life 137 seconds, gamma-ray energy 0.68 Mev). The observed properties agree with the assignment of $h_{11/2}$ and $d_{3/2}$ to the upper and lower states, respectively, as predicted by the single particle model.

INTRODUCTION

SEVERAL isomers having even Z and 81 neutrons are known: Te^{133m}, Xe^{135m}, Ba^{137m}, and Ce^{139m}. All of these decay by $M4$ transitions involving the transition $(11/2,-)\rightarrow(\frac{3}{2},+)$. The energies of these transitions have a regular dependence on Z and their comparative lifetimes[1] ($T_{\frac{1}{2}\gamma}A^2E^9$) are very nearly equal. Predictions based on these facts indicated that Nd^{141m} and Sm^{143m} should also be easily observable. These predictions are verified by the work reported here.

EXPERIMENTAL

Targets of praseodymium metal, Pr_2O_3, La_2O_3, Sm_2O_3, and $Sm_2{}^{144}O_3$ were bombarded in the internal beam of the U.C.L.A. cyclotron. The powdered samples ranging in weight from 10 to 50 mg were wrapped in tantalum foil and bombarded for periods of 30 seconds to 5 minutes. Protons of 16 Mev were used for (p,n) reactions and of 20.6 Mev for the (p,pn) reaction.

The targets were removed from the cyclotron and placed in an aluminum container situated approximately 2 inches from a 2-in.×2-in. NaI(Tl) crystal coupled to a Du Mont 6292 photomultiplier tube and cathode follower preamplifier, followed by a linear amplifier (Baird-Atomic Model 215). The output from the amplifier was fed into a 100-channel pulse-height analyzer and, simultaneously, into a single-channel

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[1] M. Goldhaber and A. W. Sunyar, *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North Holland Publishing Company, Amsterdam, 1955), Chap. XVI.