Radiations from the Active Isotopes of Ytterbium*

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By using Yb of high purity (99.8%) irradiated in the maximum flux of the Argonne pile and studied by scintillation and magnetic photographic spectrometers, a reevaluation of the energies of the radiations has been made. Several previously unreported gamma rays are found and nuclear level schemes for Tm^{169}, Lu^{175}, and Lu^{177} are proposed. Several of the levels appear to be rotational states in the unified nuclear model. Yb^{169} decays with a half-life of 30.6 days by K capture, followed by eleven gamma rays in Tm^{169}. Rotational levels lie at 8.4, 118.3, and 139.1 kev. The gamma energies are 8.4, 20.6, 63.2, 93.6, 109.9, 118.3, 130.7, 177.7, 198.6, 261.0, and 308.3 kev. Yb^{175} decays with a half-life of 4.2 days by β emission (471 kev max) followed by five gamma rays in Lu^{175}. Rotational levels exist at 114.1, and 251.9 kev. The gamma energies are 114.1, 137.8, 145.0, 282.9, and 397.0 kev. Yb^{177} decays with a half-life of 1.88 hour by β emission followed by gamma transitions in Lu^{177}. In addition to any lower energy gamma rays, two high-energy transitions are found at 1.080 and 1.228 Mev. The latter is a cross over for the 1.080- and 0.148-Mev gammas which are in coincidence. The expected well-known daughter product Lu^{177}, if present at all, is too weak to be observed by the magnetic spectrometers.

YTTERBIUM exists in nature as seven stable isotopes, with masses ranging from 168 up to 176, except for 169 and 175. Neutron capture might be expected to produce radioactive Yb^{169}, Yb^{175}, and Yb^{177}. Previous studies have been made[1–3] on these activities with some disagreement in reported results. Since Yb samples of high purity (99.8%) are now available and the neutron flux density in the pile is much greater than was used in the earlier irradiations, a reinvestigation of the radioactivities seemed worthwhile.

Specimens were irradiated in the maximum flux region of the Argonne pile and were studied in both magnetic photographic and scintillation spectrometers. The beta spectrum of Yb^{175} was observed in the double-focusing magnetic spectrometer. Studies of the short-lived (1.88 hour) activity were made on spectrometers located adjacent to the pile.

TABLE I. Conversion electron energies due to transitions in Tm (Auger lines omitted) in kev.

Electron energy	Interpretation	Energy sum	Electron energy	Interpretation	Energy sum
6.1	M	8.4	101.1	L_3	109.8
7.9	N	8.4	107.5	M	109.8
10.5	L_1	20.6	108.7	L_2	118.3
18.3	M	20.6	109.5	N	110.0
20.3	N	20.8	118.3	K	177.7
34.2	K	93.6	120.4	L_1	130.5
50.5	K	109.9	120.9	L_2	130.5
53.0	L_1	63.1	121.9	L_3	130.6
53.5	L_2	63.1	128.6	M	130.9
54.5	L_3	63.2	130.4	N	130.9
58.9	K	118.3	139.4	K	198.8
60.9	M	63.2	167.6	L_1	177.7
62.7	N	63.2	175.2	M	177.5
71.3	K	130.7	176.9	N	177.4
83.6	L_1	93.7	188.4	L	198.5
83.9	L_2	93.5	196.0	M	198.3
84.7	L_3	93.4	198.0	N	198.5
91.3	M	93.6	201.6	K	261.0
93.1	N	93.6	248.7	K	308.1
99.9	L_1	110.0	298.2	L_1	308.3
100.2	L_2	109.8	306.4	M	308.7

Due to the very great source strength, spectrograms could be obtained with exposures of a few hours, with excellent geometry. Some sixty electron conversion lines were observed and measured. The electron lines appeared to belong to two distinct groups as judged by their half-lives and their K–L–M energy fits. One group satisfying the work functions of Tm ($Z=69$) decayed with a half-life of 30.6 days. These transitions are believed to occur in Tm^{169} following K capture in Yb^{169}. Since Yb^{168} is not abundant (0.14%) in normal Yb, it must have a very large capture cross section to produce the high yield of Yb^{169}. The energies of the electron lines that decay with the 30.6-day half-life, exclusive of those of Auger origin, are presented in Table I. The interpretation of these lines confirms the existence of eleven gamma rays. Certain of these (8.4 and 261 kev) had not been observed before. Some gamma rays previously reported[3] could not be found, particularly those of energy 142.6 and 160 kev. Conversion lines attributed to these gamma energies are otherwise interpreted.

Table II shows the energies of the gamma rays together with their K/L ratios and the relative intensities of the L lines, where observable. The no-screen photographic emulsion appears able to record energies as low as 5 kev. The lowest energy gamma ray (8.4 kev), which is of considerable theoretical interest, can energetically yield on conversion only M and N electrons. These observed electron lines at 6.1 and 7.9 kev are believed to be not of Auger origin because of their sharpness. The relative intensities of many of the electron lines were determined from microphotometer traces of the photographic plates. Corrections were made for varying radius and emulsion sensitivity with energy. In some cases only a visual estimate could be made of the relative intensities.

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1 Cork, Keller, Rutledge, and Stoddard, Phys. Rev. **78**, 95 (1950).

2 A. Sunyar and J. Mihelich, Phys. Rev. **81**, 300 (1951).

3 Martin, Jensen, Hughes, and Nichols, Phys. Rev. **82**, 579 (1951).

Since most of the gamma rays enter into coincidence with others, and with Tm x-rays, the lifetimes of all unstable levels must be short. A comparison of the observed line intensities with the calculated internal conversion coefficients of Rose *et al.*[4] for the K, L_1, and L_2 shells and with the relative conversion coefficients for the L_3 subshell given[5] by Church and Monahan, makes possible an assignment of multipolarities for most of the gamma rays. Where the ratio of intensities for the L lines was not supported by the K/L ratio, in making an assignment of multipolarity, less weight was given to the latter because of the large and somewhat uncertain variation in emulsion sensitivity with energy. The preferred assignments of multipolarity are shown in column 6 of Table II. It appears in several cases that the observed intensities can best be satisfied by a mixture of $M1$ and $E2$ radiations, although for the 63.2-, 93.6-, and 109.9-kev transitions the possibility of an $E1$ assignment cannot be eliminated on the basis of the relative line intensities alone.

TABLE II. Gamma energies in Tm^{169} with intensities and multipolarities. (The symbol x indicates that only one L line was observed.)

Gamma energy kev	K/L	Rel. intensity L_1	L_2	L_3	Multipolarity
8.4					
20.6		x			$M1$
63.2		10	4.3	5.5	$M1$, $E2$
93.6	1.6±0.2	10	1.8	1.0	$M1$, $E2$
109.9	2.9±0.3	10	1.9	0.6	$M1$, $E2$
118.3	0.9±0.3		x		$E2$
130.7	0.8±0.2	[10	]	9	$E2$
177.7	5.6±0.2	x			$M1$, $E2$
198.6	6.6±0.2	x			$M1$, $E2$
261.0					
308.3	3.5±0.2	[x	]		$E2$

Although the resolution of the scintillation spectrometer is not comparable with that of the magnetic instruments, important information on coincidences and summations was obtained with it. A single crystal yields the distribution shown in Curve A of Fig. 1, indicating six peaks, some of which are composite. A coincidence curve with thulium x-rays (50 kev) is identical in shape with the singles curve. A similar result is obtained with the fixed channel set on 20 kev. Since there exists the possibility of back-scattered iodine x-rays (29 kev) from the crystal and the subsequent escape peaks of the thulium x-rays, the above similarity in results is not unexpected. The high-energy side of the 115-kev peak (130.7) was found to be in coincidence with the low-energy side of the 190-kev peak (177.7). The 261-kev peak was in coincidence with the 110 or 118 or both but not with the peaks at 130, 190, 261, or 308 kev. With good geometry the 308-kev peak is found in coincidence only with the 50-kev peak. On the other hand, with 2π solid angle a coincidence peak appears at 115 kev which is attributed to summations between thulium x-rays and the 63-kev gamma. When the source was placed in a narrow well in the NaI crystal so as to give summation effects, the 20-kev peak disappeared and the 50- and 190-kev peaks were lessened in height while those at 115, 261, and 308 kev were augmented as shown in Curve B, Fig. 1.

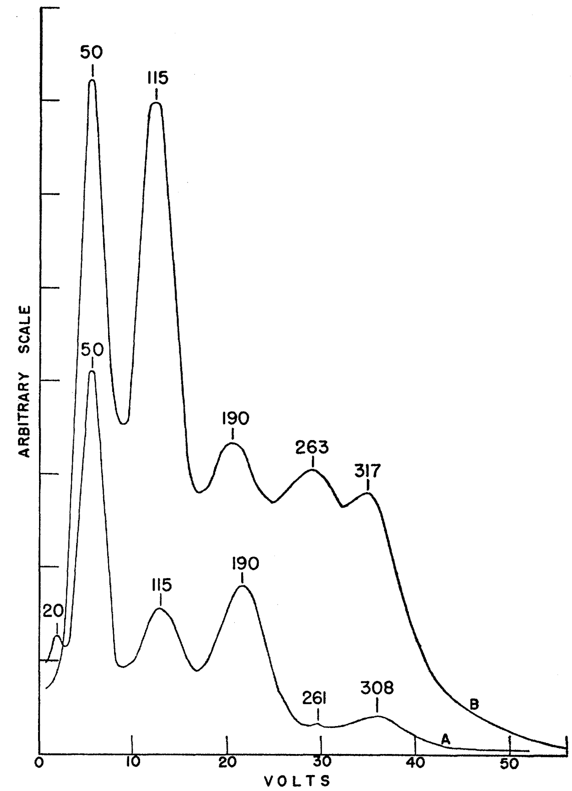

FIG. 1. Scintillation spectrometer peaks; A—singles, B—source in crystal well. (Energies in kev.)

A nuclear level scheme for Tm^{169} is presented in Fig. 2. The spin of the ground state for the odd-A ($Z=69$, $N=100$) nucleus has been measured to be 1/2. This is interpreted from the shell model as an $s_{\frac{1}{2}}$ state. Some calculations of the low-energy gamma rays to be expected in Tm^{169}, assuming them to be transitions between rotational states in the unified nuclear model, have been made by Mottelson and Nilsson.[6] Their expression for the energy E_I in terms of the spin I is

$$E_I=(\hbar^2/2J)\{I(I+1)+a(-1)^{I+\frac{1}{2}}(I+\tfrac{1}{2})\},$$

where J is the moment of inertia and a is a constant as I takes values 1/2, 3/2, 5/2, 7/2, 9/2, etc. Using the values for the low-energy gammas reported[3] by Jensen *et al.*, which are somewhat in error, they derived a value

[4] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report No. ORNL-1023, 1951 (unpublished); and subsequent letters.

[5] E. L. Church and J. E. Monahan, Phys. Rev. **98**, 718 (1955).

[6] B. R. Mottelson and S. G. Nilsson, Z. Physik **141**, 217 (1955).

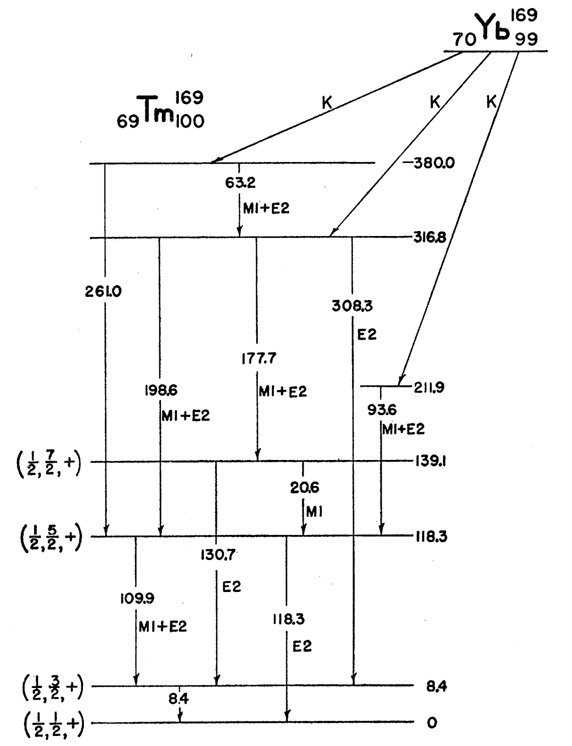

FIG. 2. Nuclear level scheme for Tm^{169} following K capture in Yb^{169}. (Energies in kev.) Spin along axis of symmetry, total spin, and parity are shown in parentheses.

of a equal to -0.74. From our values for the energies of the first three levels (8.4, 118.3, and 139.1 kev), a is obtained by comparing each of the upper energies with the lowest. Its value is found to be -0.775 and the excellent agreement between calculated and observed energies for the first three levels is shown in Table III. The lack of agreement for the two higher spins indicates that these possible rotational levels are not observed. Since the rotational levels are of the same parity, short-lived transitions between them can be only $M1$ or $E2$ in nature, in agreement with the multipolarity assignments noted in Table II.

From the results with the source in the crystal well, support is obtained for the arrangement of levels as shown in Fig. 2. A metastable level of half-life 6×10^{-7} second had been observed[2] in Tm^{169} and it was believed to be derived from K capture directly. It now appears

TABLE III. Energies in kev of rotational levels, together with observed energies.

I	Calculated	Observed
3/2	...	8.4
5/2	118.8	118.3
7/2	138.6	139.1
		316.8
9/2	337	
11/2	368	
		380.0

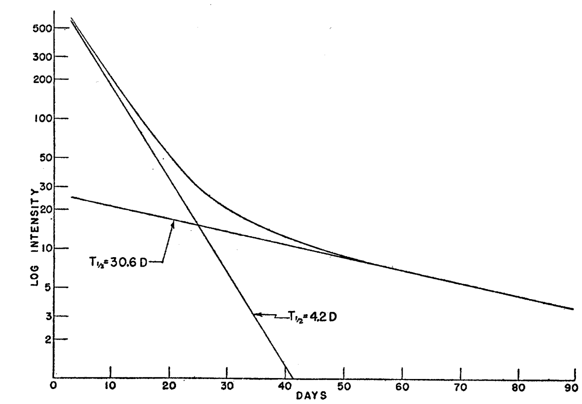

FIG. 3. Analysis of the decay curve for Yb.

from the summation evidence that the 316.8-kev level is most likely the delayed state. The presence of an unsummed x-ray peak strongly suggests a K-capture branch terminating at this level. The highest summation level appears to be 317 kev, which is reasonable since the transitions above it could not be included because of the delay. The increased peak height at 115 kev could be due to the sum of 63 kev and Tm x-rays (50 kev). The placement of the strongly converted 93-kev gamma transition is based on the augmented peak at 263 kev which could be the sum of 118, 93, and 50 kev.

The level at 211.9 kev is expected to be a single particle state, which may be either $d_{5/2}$ or $g_{7/2}$ in agreement with the shell model and the $M1$ character of the 93.6-kev transition. The 316.8-kev level should also be a single-particle level. It is observed that if this level is assigned a spin of 3/2 and the moment of inertia is assumed to be the same as in the ground state, the first rotational excitation will come 62.2 kev higher, as compared with the observed transition of 63.2 kev.

An analysis of the rate of decay shown in Fig. 3 indicates half-lives somewhat different than previously found.[1] This must be due to the higher purity of the Yb specimen. The long half-life of Yb^{169} appears to be 30.6 ± 0.2 days. The previously reported intermediate

TABLE IV. Conversion electron energies in kev in Lu^{175} following β emission from Yb^{175}.

Electron energy	Interpretation	Energy sum
50.5	K	113.8
74.5	K	137.8
81.7	K	145.0
103.1	L_1	114.0
103.6	L_2	114.0
104.9	L_3	114.1
111.7	M	114.2
113.5	N	114.0
127.3	L_1	138.2
219.6	K	282.9
272.6	L_2	283.0
333.5	K	396.8
386.5	L	397.4
394.9	M	397.4

half-life of 6.7 days attributed to Lu^{177} could not be observed. For Yb^{175} the half-life is 4.2±0.1 days. The absence or extreme weakness of the 6.7 day Lu^{177} activity as noted both in the decay curves and in the lack of conversion lines for the well known gamma transitions in Hf raises a serious question. The previous observation of the rather strong activity must have been due to Lu being present in the Yb as an impurity. Since Yb^{176} is abundant (12.7%) in all Yb, it is difficult to understand why Yb^{177} with its half-life of 1.88 hours would not build up the Lu^{177} daughter activity. Either the capture cross section in Yb^{176} is very small, which does not appear to be the case, or some assignment of mass may be in error.

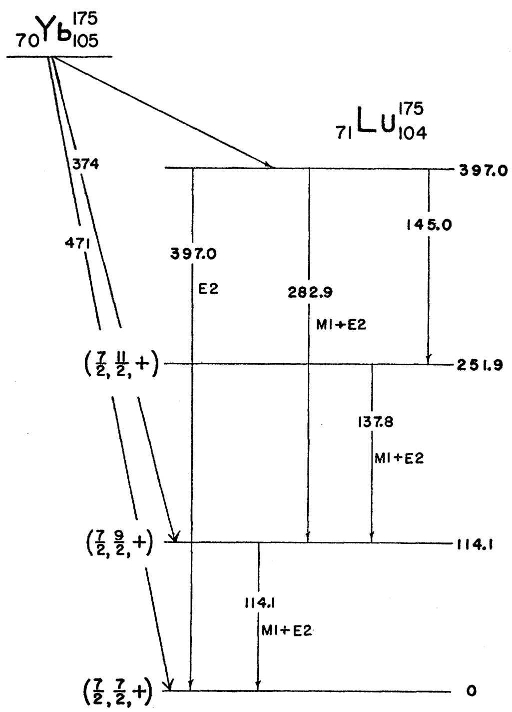

FIG. 4. Nuclear level scheme for Lu^{175} following β emission from Yb^{175}. (Energies in kev.)

The electron conversion lines that decay with the 4.2-day half-life are shown in Table IV together with their interpretations. It appears without doubt that there are five gamma rays, whose energies and relative conversion intensities are presented in Table V. This is in agreement with the recent results[7] of Mize, Bunker, and Starner on Yb^{175}. Only one of these gamma rays (114 kev) is observed[8] in the decay of Hf^{175}. In the Coulomb excitation of Lu^{175} both of the levels 114 and 252 kev are found to exist.[9] The K/L ratios for the 114- and 397-kev gammas were determined from microphotometer traces of the photographic plates. The values for the 138- and 283-kev transitions are based upon visual estimates of the blackness of the lines. The choice of $M1$, $E2$ for the 114-kev gamma is based upon the agreement with calculations[4] for the L_1/L_2 ratio, and is supported by agreement with the empirically expected[10] K/L ratio. To satisfy the observed K/L ratio and the presence of a strong L_2 line for the 283-kev gamma a mixture of $E2$ and $M1$ transitions also appears likely. The other selections are based upon the K/L ratios together with the assumption of very short lifetimes. A nuclear level scheme for Lu^{175} based upon excellent energy fits is presented in Fig. 4.

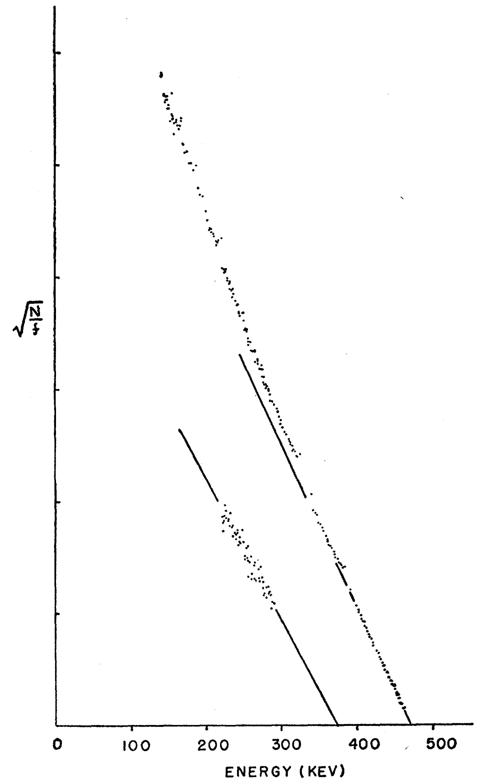

FIG. 5. Kurie plot of the β spectrum of Yb^{175}.

The spin of the ground state in Lu^{175} has been measured to be 7/2, which from shell theory is interpreted as $g_{7/2}$. If the lower energy levels are regarded as ro-

TABLE V. Gamma energies in kev in Lu^{175} and relative electron line intensities. (The symbol x indicates that only one L line was observed.)

Gamma energy	K/L	L_1	L_2	L_3	Multi-polarity
114.1	2.9±0.4	10	4.1	2.7	$M1$, $E2$
137.8	~2				$E2$
145.0					
282.9	~6		x		$M1$, $E2$
397.0	5.4±0.3				$E2$

[7] Mize, Bunker, and Starner, Phys. Rev. **99**, 671 (1955).
[8] Burford, Perkins, and Haynes, Phys. Rev. **99**, 3 (1955).
[9] G. Temmer and N. Heydenburg, Phys. Rev. **94**, 1399 (1954).
[10] M. Goldhaber and A. Sunyar, Phys. Rev. **83**, 906 (1951).

TABLE VI. Relative intensities of the K–L–L auger lines.

	$K-L_I-L_I$	$K-L_I-L_{II}$	$K-L_I-L_{III}$	$K-L_{II}-L_{II}$	$K-L_{II}-L_{III}$	$K-L_{III}-L_{III}$
Observed	1.0	1.6	1.5	...	2.0	0.9
Calculated[a]	1.0	1.02	1.65	0.24	1.46	0.81

[a] See reference 13.

tational in nature, then their energies may be calculated from that of the gamma ray of lowest energy. As the spins are given successive values of 9/2, 11/2, etc. and the 114-kev gamma regarded as basic, then the following levels should have energies of 253.6 and 418.5 kev. The 253-kev value is in good agreement with an experimental level, as shown in Fig. 4. The level at 397 kev is sufficiently divergent from the expected 418-kev value, to assume that it is not rotational. The transitions between the rotational levels, with no change in parity, should be $M1$ or $E2$ or a combination of both, agreeing with the choices indicated in Table V. The 397-kev level is expected to be a single-particle state. The multipolarities of the various gamma rays, together with the fact that the energy available in beta decay will permit at most an ordinary first forbidden transition, suggest that this level has spin 9/2 and even parity, with an $f_{7/2}$ ground state for Yb^{175}. The spin of 9/2 for the 397-kev level is in agreement with the angular correlation work of Akerlind *et al.*[11]

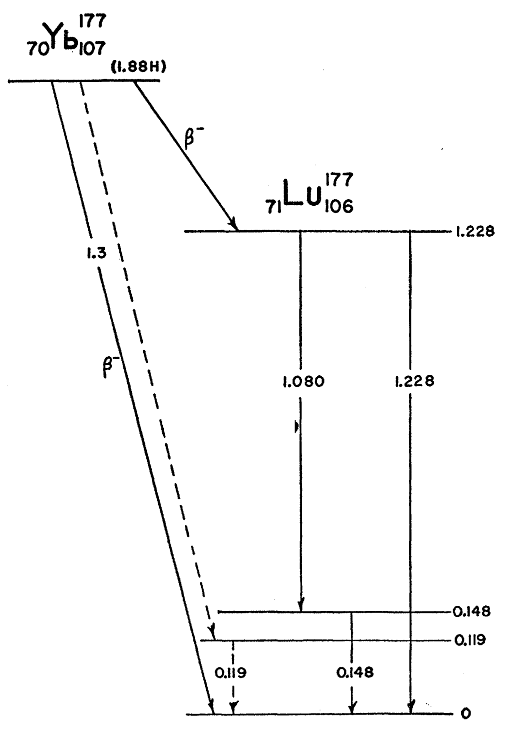

FIG. 6. Nuclear levels in Lu^{177} following β emission in Yb^{177}. (Energies in Mev.)

The beta spectrum of Yb^{175} was observed with the double-focusing magnetic spectrometer, using a Scotch tape source and Zapon ($\sim$15 micrograms per square centimeter) counter window, with a resolution of 0.5%. The Kurie plot is shown in Fig. 5. The spectrum appears to be complex, with an upper energy limit of 471 ± 3 kev. After subtraction of this high-energy component, there is found to be a rather large scatter in the points of the residual Kurie plot. A least squares fit to these points in regions where no interference from internal conversion lines is expected gives a component of maximum energy 374 ± 30 kev, whose intensity is about 25% that of the high-energy component. The level scheme requires an additional low-energy component at about 80 kev, which would be difficult to observe due to the many internal conversion lines in this region.

The activity in Yb^{177} was induced by short irradiation in the maximum flux region of the pile and was studied in a ten channel scintillation spectrometer and magnetic photographic spectrometer near the pile. A recent paper reports[12] the existence of two gamma rays of energy 119 and 146 kev and three beta rays with a maximum energy of 1.3 Mev. In the present investigation two additional high-energy gamma rays are found with energies of $1.228+0.005$ and 1.080 ± 0.005 Mev. The former is undoubtedly a cross-over transition for the 1.080- and 0.148-Mev gammas which are found to be in coincidence. All gamma rays appear to decay with a half-life of 1.88 ± 0.1 hr. The gamma ray reported at 119 kev has not been identified but is included as a dotted line in the provisional level scheme shown in Fig. 6. It has previously been assumed that the ground state of Lu^{177} has a half-life of 6.7 days followed by the emission of certain well-known highly converted gamma rays in Hf^{177}. It is quite certain that this activity, if present at all, is extremely weak in the high-purity Yb source.

Auger electrons following K-capture to Tm^{169} were also observed. Their relative intensities, determined from microphotometer traces of the photographic plates, are presented in Table VI, together with the calculated intensities of Hill[13] for $Z=80$.

[11] Akerlind, Hartmann, and Wiedling, Phil. Mag. **46**, 448 (1955).
[12] H. deWaard Phil. Mag. **46**, 445 (1955).
[13] R. D. Hill, Phys. Rev. **91**, 770 (1953).