Decay of Ho^{163}†

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A sample of Ho^{163} was prepared by neutron irradiation of Er^{162} followed by chemical purification and isotope separation. This nuclide Ho^{163} appears to decay by electron capture from the M and higher shells. A half-life of 33 ± 23 yr has been estimated by counting the M Auger electrons and M x rays from a sample over a period of two years. A search for L x rays has been made. The M-to-L capture ratio is greater than 1040. From this datum the Q value for electron capture can be deduced to be 9.1 ± 1.5 keV.

HOLMIUM-163 is one of six unstable neutron deficient nuclei (Ti^{44}, Ho^{163}, Pt^{193}, Hg^{194}, Pb^{202}, and Pb^{205}) for which K electron capture appears to be energetically impossible. However, Ho^{163} is the only nucleus in this group which also cannot capture from the L shell. This nucleus must have a very small Q value for electron capture and a very long half-life.

A sample of erbium enriched in the mass-162 isotope was irradiated for six months at the Materials Testing Reactor to produce the material used in these studies. After chemical fractionation of the products using ion-exchange chromatography, a mass spectrum of the holmium fraction revealed the presence of a long-lived isotope with mass number 163. In order to study the radiations accompanying the electron-capture decay of this isotope to stable Dy^{163}, a sample free from radioactive Ho^{166} was obtained by dispersing a portion of this holmium fraction using the isotope-separation facility at the Niels Bohr Institute in Copenhagen. The Ho^{163} was collected on 10-mil aluminum foil at the full 50 kV separator accelerating voltage. When this sample was examined using a windowless proportional counter, only very soft radiations of approximately 1.3 keV could be detected. These radiations have been assigned as Auger electrons and x radiations following M electron capture.[1,2]

A method for investigating possible decay of this sample has been devised.[3] The shape of the Fe^{55} K x-ray spectrum observed in a windowless flow proportional counter of the Sugarman type has been investigated as a function of the applied voltage, using a counting gas with composition 90% argon, 10% methane. A 200-V interval (2100 to 2300 V) has been found over which ideal proportional operation was closely approximated. In this region the shape of the observed pulse-height spectrum remained constant (superposable) when plotted using logarithmic axes for both the number of events and the corresponding pulse amplitude. Since the relative shape of the spectrum remained invariant in this voltage range, a reproducible measure of the counting rate could be found by recording a spectrum while the counter is operated within the 200-V interval and then summing the events contained in all channels between predetermined upper and lower limits. These limits were located as the channels where the events accumulated equaled some specified fraction of the events recorded in the peak of the spectrum.

Spectra due to similarly mounted sources of Fe^{55}, Zn^{65}, and Ho^{163} were observed under these conditions for 24 months. Typical spectra obtained are shown in Fig. 1 using linear axes. The shaded portion shown in

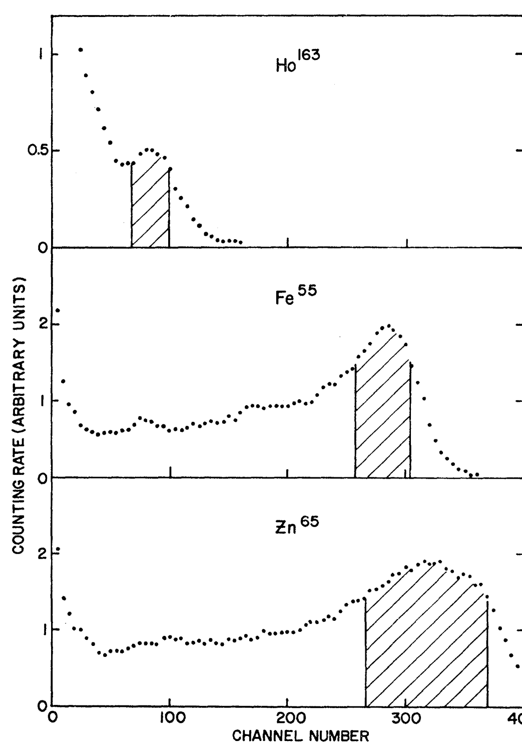

FIG. 1. X-ray spectra of Ho^{163}, Fe^{55}, and Zn^{65} taken with a windowless proportional counter. The Zn^{65} spectrum was recorded with one-half the amplifier gain used for the other spectra.

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[1] R. A. Naumann, M. C. Michel, and J. C. Power, J. Inorg. Nucl. Chem. **15**, 195 (1960).
[2] R. A. Naumann, M. C. Michel, and J. C. Power, Bull. Am. Phys. Soc. **6**, 73 (1961).
[3] J. S. Evans, thesis, Princeton University, 1965, PPAD-572F (unpublished).

each spectrum represents the spectral region summed to investigate the decay of these samples. For Fe^{55} and Zn^{65}, those channels where the counting rate was greater than 75% of the peak height were included in the summation. For Ho^{163}, channels with a counting rate greater than 80% of the peak height have been included in the summation.

The results from approximately 24 months of observation are shown in Fig. 2. The points have been fitted to exponential decay curves using a least-squares method in which all points have been given equal weight. The values of the half-lives have been computed from the slopes of the lines, and errors have been computed from the scatter of experimental points with respect to the least-squares fitted curve. These values are given in Table I together with the accepted half-lives for Fe^{55} and Zn^{65}. A comparison of these values indicates the reliability of our counting method for low-energy radiations.

TABLE I. Half-life determinations.

Nuclide	Half-life	Reported half-life
Zn^{65}	264+9 days	245.0 ±0.8 days[a] 243.5 ±0.8 days[b] 246.4 ±2.2 days[c] 245.7 ±1.1 days[d] 249.7 ±1.4 days[e]
Fe^{55}	2.41±0.12 yr	2.94±0.03 yr[f] 2.60±0.02 yr[g]
Ho^{163}	33±23 yr	

[a] J. Tabailem, J. Phys. Radium **14**, 553 (1953).
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[g] R. P. Schuman, M. E. Jones, and A. C. Mewherter, J. Inorg. Nucl. Chem. **3**, 160 (1956).

Possible changes in the Ho^{163} source during the observation period must be considered with respect to the decay we observe in this sample. Two possible alterations are (a) surface loss of active material, and (b) diffusion of the Ho^{163} atoms into the interior of the aluminum backing plate. Using the Aarhus University computer program based on Linhard's theory of the ranges of heavy ions in matter,[4] one calculates that 50-keV Ho^{163} ions penetrate aluminum to a mean depth of 6 $\mu g/cm^2$ with a rms deviation of 1.5 $\mu g/cm^2$.[5] This computation implies that the bulk of the Ho^{163} is located several hundred atom diameters below the surface, so that surface loss appears unlikely. The diffusion of rare earths through aluminum at room temperature is negligible.

Accordingly, we believe that the decrease of Ho^{163} activity that we have observed is due to radioactive decay of the sample in accord with a half-life of 33±23 yr. In any event this figure must represent a lower limit of the half-life for the decay of this species.

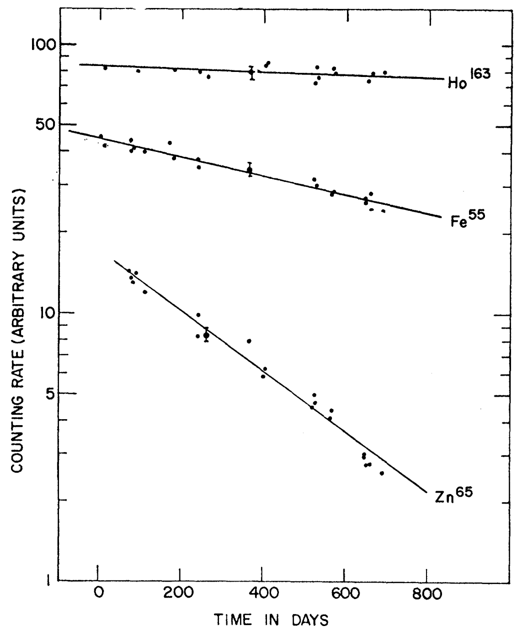

FIG. 2. Decay curves for Ho^{163}, Fe^{55}, and Zn^{65}. The lines are the least-squares fit to the data.

In order to set limits for the Ho^{163} electron-capture Q value, a search has been made for x rays indicative of L electron capture. A cleaved cylindrical NaI(Tl) detector 3.1 mm diam by 6 mm in thickness with a 0.127-mm-thick beryllium window contained in a lead shield has been used. Spectra covering the range 6 to 20 keV were recorded with the source placed directly on the counter window for 142 h. A background was taken for a similar period. An upper limit of 550 counts/h above background was observed in the region of 7 keV in this period. The integrated counting rate of the Ho^{163} source above background osberved in the proportional counter was approximately 44 000 counts/h in the M Auger-x-ray region. By assuming an L fluorescence yield $\omega_L=0.200$, an M fluorescence yield $\omega_M=0.015$,[6] and the same geometrical efficiencies for the x-ray and gas proportional counters, one determines a lower limit for the M-to-L capture ratio of 1040. Since the attenuation of the Ho^{163} M Auger and x radiation due to the 6-$\mu g/cm^2$ penetration depth in aluminum has been neglected, this limit is conservative.

An upper limit for Q, the electron-capture energy for Ho^{163}, may now be estimated using the allowed electron-capture result.

$$\lambda_{M_1}/\lambda_{L_1}=[(Q-W_{M_1})/(Q-W_{L_1})]^2|g_{M_1}/g_{L_1}|^2,$$

where $\lambda_{M_1}/\lambda_{L_1}$ is the M-to-L electron-capture ratio. One

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TABLE II. Logft values for allowed unhindered β transitions $\frac{5}{2}-$ [523] $\rightarrow \frac{7}{2}-$ [523].

Parent			Daughter				
Nucleus	Spin	Process	Nucleus	Spin	State populated (keV)	Q (keV)	logft
$_{64}Gd_{97}^{161}$	$(\frac{5}{2}-)$	$\beta-$	$_{65}Tb_{96}^{161}$	$(\frac{7}{2}-)$	418	2020	4.9
$_{67}Ho_{94}^{161}$	$(\frac{7}{2}-)$	EC	$_{66}Dy_{95}^{161}$	$\frac{5}{2}-$	26	1100[a]	6.0[a]
$_{68}Er_{95}^{163}$	$(\frac{5}{2}-)$	EC, $\beta+$	$_{67}Ho_{96}^{163}$	$(\frac{7}{2}-)$	0	1210±6	4.8
$_{68}Er_{97}^{165}$	$\frac{5}{2}-$	EC	$_{67}Ho_{98}^{165}$	$\frac{7}{2}-$	0	371±5	4.62+0.02
$_{70}Yb_{97}^{167}$	$(\frac{5}{2}-)$	EC, $\beta+$	$_{69}Tm_{98}^{167}$	$(\frac{7}{2}-)$	293	1970±30	4.5
$_{67}Ho_{96}^{163}$	$(\frac{7}{2}-)$	EC	$_{66}Dy_{97}^{163}$	$\frac{5}{2}-$	0		

[a] The decay energy has been estimated in the *Nuclear Data Sheets* from systematics. The resulting logft value seems to be too high.

estimates the ratio of the densities of the large components of the Dirac wave functions for the M_1 and L_1 electron shells $|g_{M_1}/g_{L_1}|^2=0.220$ by linear interpolation of the values given in the tables of Robinson.[7] Using the electron binding energies $W_{M_1}=2.05$ keV and $W_{L_1}=9.05$ keV,[8] one finds $Q\leq 9.1\pm 1.5$ keV.

The asymptotic quantum numbers [523] $\frac{7}{2}-$ have been assigned to the last odd proton in the ground state of Ho^{163}.[9] The measured nuclear spins of Ho^{161} and Ho^{165} of $\frac{7}{2}$ support this assignment.[10] Similarly the asymptotic quantum numbers [523] $\frac{5}{2}-$ have been assigned to the last odd neutron in the ground state of Dy^{163}, corresponding to a rotational sequence based on the ground state.[11] The electron capture then involves the transition [523] $\frac{7}{2}-$ to [523] $\frac{5}{2}-$ or $\Delta N=\Delta N_3=\Delta\Lambda=0$, that is, an allowed unhindered transition in the classification of Alaga.[12] The known logft values for β processes involving these proton and neutron Nilsson states are collected in Table II. An average logft value of 4.7 is found.

A theoretical estimate of the half-life of Ho^{163} is possible using the equation for the electron-capture rate for an allowed (Gamow-Teller) transition given in Table I of Robinson.[7]

$$\lambda=\tfrac{4}{3}g_A{}^2\langle\sigma\rangle^2(n_{-1}g_{-1}{}^2q_{-1}{}^2+n_1f_1{}^2q_1{}^2),$$

where we have used Konopinski's[13] notation. The value for the coupling constant is $g_A=g_A{}^C=1.63\times 10^{-49}$ erg cm^3.[14] We have made the approximation that the β moment is given by its value in mirror nuclei,

$$\langle\sigma\rangle=(-1)^{I+\frac{1}{2}-L}[(2l'+1)/(L+\tfrac{1}{2})]^{1/2},$$
$$\langle\sigma\rangle^2=12/7,$$

where $q_{-1}{}^2$ and $q_1{}^2$ are the squares of the neutrino energies for capture from the M_1 and M_{11} shells, respectively. We have used the wave functions of Brewer, Harmer, and Hay[15] for $Z=65$. The error incurred by this approximation should be small compared to the error in the β moment. The wave functions are $g_{-1}{}^2=8.78\times 10^{-5}$ and $f_1{}^2=3.57\times 10^{-7}$. A value of the half-life $T_{1/2}=59$ yr was calculated. This value is comparable to the value of 33 ± 23 yr found by direct observation.

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