even-even nuclei. Then although Xe^{134} is only two neutrons removed from a closed shell, its low-lying levels, like the surprising case of Xe^{136}, appear to possess collective properties. The asymmetric rotor model of Davydov and Filippov[20] has had moderate success in predicting some of the low-lying energy states, and gamma-ray transition probabilities between them, for this class of nuclei. It gives the energy states of an asymmetric top as a function of γ, where γ determines the deviation of the nuclear shape from axial symmetry. The ratio of the energy of the second 2+ state (1.62 Mev) to that of the first 2+ state (0.85 Mev) observed in the present case corresponds closely to $\gamma=30°$. The Davydov-Filippov model predicts the first 3+ level in Xe^{134} to be at 2.54 Mev, the sum of the first and second 2+ levels. As shown in Fig. 15, there is a level close to this energy at 2.48 Mev which is indeed compatible with a 3+ assignment; in further agreement with the predictions of the model for $\gamma=30°$, gamma emission occurs from this level to the second 2+ and not to the first 2+ state. In the case considered, the ratio $B(E2, 2'\rightarrow 2)/B(E2, 2'\rightarrow 0)$ is expected to approach infinity, whereas we observe a much smaller value of 50 corresponding to $\gamma\approx 27°$. The model appears unsuccessful in predicting energies for any of the higher spin states of Xe^{134}.

The first few levels of the even xenon nuclides are shown schematically in Fig. 16. The continuous increase in first excited state energies as N approaches 82 is, of course, to be expected. However, it is surprising that the collective behavior as evidenced by the ratio of second to first excited state energies of ~ 2 does persist up to the closed shell. The graph suggests that second excited state doublets should exist in Xe^{126}, Xe^{128}, and Xe^{130}.

[20] A. S. Davydov and G. F. Filippov, Nuclear Phys. 8, 237 (1958).

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Neutron-Deficient Nuclides of Hafnium and Lutetium*

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New neutron-deficient nuclides of lutetium and hafnium were produced by bombarding lutetium oxide with 300- to 400-Mev protons. The genetic relationships and mass assignments were established by means of high-purity chemical separations and a series of chemical isolation experiments in which the daughter activity was determined as a function of time.

The positron spectra of the different nuclides were measured with an anthracene crystal detector and a 256-channel pulse height analyzer. Gamma radiation was also observed for Lu^{168}, Lu^{169}, Lu^{170}, Hf^{168}, and Hf^{169} by means of a NaI crystal detector and the pulse height analyzer. The half-lives and maximum positron energies observed are: Lu^{168}, $T_{\frac{1}{2}}=7.0$ min, $E_{\beta^+}=(1.20\pm 0.05)$ Mev; Lu^{169}, $T_{\frac{1}{2}}=1.5$ days; Lu^{170}, $T_{\frac{1}{2}}=1.9$ days, $E_{\beta^+}=(1.8\pm 0.1)$ Mev; Hf^{168}, $T_{\frac{1}{2}}=22$ min, $E_{\beta^+}=(1.7\pm 0.1)$ Mev; Hf^{169}, $T_{\frac{1}{2}}=1.5$ hr; Hf^{170}, $T_{\frac{1}{2}}=9$ hr.

INTRODUCTION

IN the course of a study of the spallation reactions of tantalum and tungsten with 300–400-Mev protons using the Carnegie Institute of Technology synchrocyclotron, several new neutron-deficient nuclides of hafnium and lutetium were observed. After performing a few preliminary experiments, a detailed study was undertaken to establish the half-lives, genetic relations, and radiation characteristics of these new nuclides. The investigation included the hafnium nuclides of Hf^{168}, Hf^{169}, and Hf^{170} and those of lutetium of the same mass numbers.

In these more detailed studies Lu_2O_3 was bombarded with 300–400 Mev protons producing only the neutron-deficient nuclides of hafnium and lutetium by (p,xn) and (p,pxn) reactions, and eliminating interfering neutron excess nuclides of these elements. In order to avoid separating the lutetium activities from the many other rare-earth activities produced in the bombardment, the lutetium decay products were chemically separated from the purified hafnium fraction. All lutetium nuclides produced, heavier than Lu^{168}, decay to stable ytterbium nuclides with the exception of Lu^{169}, which decays to 32-day Yb^{169}. The well-known spectrum of Yb^{169} was used as evidence for the mass assignment of Hf^{169} and Lu^{169}. The half-lives observed for these nuclides are listed in Table I. All the nuclides decay either completely or in part by electron-capture. Positron emission was observed in Hf^{168}, Hf^{169}, Lu^{168}, and Lu^{170}. In addition to these six nuclides, some evidence was found for Hf^{167} and Lu^{167} with half-lives

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TABLE I. Observed half-lives.

Hf^{168}	22 min	Lu^{168}	7.0 min
Hf^{169}	1.5 hr	Lu^{169}	1.5 days
Hf^{170}	9 hr	Lu^{170}	1.9 days

of ~10 min and ~4 min, respectively. No evidence was observed for a 55-min activity associated with Lu^{167} as reported by Aaron *et al.*[1] and Harmatz *et al.*[2]

EXPERIMENTAL

The targets used in the experiments consisted of tantalum and tungsten metal foils, and Lu_2O_3 wrapped in aluminum foil in quantities of 30–200 mg. The targets were irradiated in the internal beam of the 140-in. synchrocyclotron with 300–400-Mev protons for periods ranging from 5–15 min with a beam of about 10^{12} protons per cm^2-sec. Immediately after irradiation, the targets were chemically processed to separate hafnium and lutetium.

The gamma rays emitted by the different hafnium and lutetium isotopes were measured by using a 2 in. ×2 in. NaI crystal connected to a 256-channel pulse-height analyzer. The positron radiation from these nuclides was detected with a simple magnetic spectrometer and an anthracene crystal in connection with the 256-channel pulse-height analyzer.

The genetic relationships between the different nuclides was determined by choosing adequate milking-experiments and measuring techniques. Fast hafnium and lutetium chemical separation procedures were developed which provided very high decontamination factors from disturbing radioactivities.

The bombarded target (Lu_2O_3) was dissolved in 5*N* HNO_3 and evaporated to dryness. The residue was dissolved in 10 ml 2*N* HNO_3 containing 20 mg Zr-carrier (for hafnium), holdback carrier of Yb (200 mg), and 5 mg of each of the following elements: Cu, Ni, Ca, and Na. The zirconium (Hf) was extracted for 5 min with 10 ml of a 0.5 molar TTA-solution in xylene. The organic layer was washed twice with 10 ml 1*N* HNO_3 (Yb carrier was added to the wash-solution). The zirconium (Hf) was then back extracted with 10 ml of a mixture of 0.3*N* HNO_3+0.3*N* H_2F_2. To this solution 8 ml concentrated HNO_3 and 7 ml concentrated H_2F_2 were added together with 20 mg tantalum carrier. After cooling the solution to about 20°C, the zirconium (Hf) was precipitated with an excess of $Ba(NO_3)_2$ solution.

The $BaHfF_6$ was dissolved in saturated H_3BO_3 and HNO_3. Hafnium was then precipitated as $Hf(OH)_4$ by adding NH_4OH. For the experiments where hafnium was measured, the $Hf(OH)_4$ was dissolved in 6*N* HCl and after the addition of about 0.5 g La holdback carrier, the hafnium was precipitated with phenylarsonic acid.

In cases where the lutetium was isolated from the hafnium fraction, the $Hf(OH)_4$ was dissolved in 15 ml 2*N* HNO_3 and the zirconium (Hf) again extracted with 10 ml 0.5*M* TTA (in xylene). The organic layer was washed with 10 ml 6*N* HCl.

The lutetium was extracted by mixing the organic layer for 4 min with 10 ml 6*N* HCl, containing 10 mg Yb carrier. To the aqueous solution, 5 mg Zr carrier were added and the zirconium (Hf) precipitated with phenylarsonic acid. The scavenging with zirconium was repeated once or twice and finally 5 ml H_2F_2 and 0.5 g zirconium (Hf) holdback carrier added. The YbF_3 (LuF_3) produced was centrifuged and then transferred with a pipette to a filter disk.

When using Ta or W targets, the metal foils were dissolved in a HNO_3-H_2F_2 mixture and the hafnium and zirconium, respectively, precipitated as $BaHfF_6$ with an excess of $Ba(NO_3)_2$ solution. After converting to $Hf(OH)_4$ and dissolving in 2*N* HNO_3 it was treated as described for Lu_2O_3 targets.

Half-Life Measurements

All the following measurements were based on Lu_2O_3 targets unless otherwise stated.

Beta decay measurements on a hafnium sample, separated from the Lu_2O_3 target 40 min from the end of irradiation, first showed an increase in activity with time for the first 10–15 min. Following this, the decay curve was resolvable into periods with half-lives of 22 min, 1.5 hr, 13 hr, 44 hr, and long-lived activities of about 8 days and 70 days. This latter activity was probably due to Hf^{175}.

Beta decay measurements on the lutetium sample gave results which were dependent on growing time allowed for the lutetium decay products from the hafnium. In a fast separation of the lutetium from hafnium, a strong 7-min activity was observed, followed by a half-lives of 1.5 days, 2.0 days, and finally 8.5 days, probably associated with Lu^{171}.

A simple magnetic spectrometer was used to make crude separations of positrons, electrons, and electromagnetic radiations. The hafnium sample showed positron radiation which again increased in activity for the first 15–20 min and then decayed with half-lives of 22 min, 80 min, and 45 hr. No longer-lived activity was observed.

The magnetic spectrometer was also used to measure the decay of electromagnetic radiation alone, and negative electrons alone. Decay periods similar to those found with positrons alone were observed in all cases.

In one experiment, the lutetium was extracted from a freshly purified hafnium fraction after a growing time of 15 min. Use of the magnetic spectrometer to measure positron radiation indicated that 90% of the total lutetium activity decayed with a 7-min half-life.

[1] P. M. Aaron, A. V. Kalyamin, A. N. Murin, and V. A. Yakovlev, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **22**, 817 (1958).
[2] B. Harmatz, T. H. Handley, and J. M. Mihelich, Bull. Am. Phys. Soc. **3**, 358 (1958).

The remaining activity decayed with a 1.9 day half-life. These beta decay measurements indicate that the 7.0 min activity belongs to a lutetium nuclide which decays at least in part by positron emission. The results of all these measurements are listed in Table II.

Several lutetium chemical isolation experiments were performed varying the growth time for the lutetium, in order to determine the genetic relationships of the different hafnium and lutetium isotopes. Figure 1 shows the results of such an experiment in which the growth time was 30 min. The experiment proves that the 22-min hafnium isotope is the parent of the 7-min lutetium daughter. The most likely mass assignment for this chain is 168 since there was no evidence for a radioactive daughter of the 7-min isotope, and Yb^{168} is stable.

In another isolation experiment, the lutetium was extracted from the hafnium in intervals of 1.5 hr. The 1.5-day lutetium activity measured in these experiments at the time of chemical isolation from the hafnium parent, and plotted vs the isolation time, yielded a curve with a half life of 1.5 hours for the hafnium parent. The 1.5-day lutetium decayed into a 32-day nuclide which could be assigned to Yb^{169}. This was confirmed by detecting the well known gamma spectrum of Yb^{169} in all the isolated lutetium samples and noting that the intensity of this spectrum also decreased from sample to sample with a 1.5-hour half-life.

On still another experiment, a lutetium activity of 1.9-day half-life was successively separated from the hafnium sample. Extrapolation of the 1.9 day-lines to the separation time and plotting the total activity vs separation time yielded a curve indicating a 9-hour half-life for the hafnium parent. It was, of course, impossible to resolve the 1.9 day and 1.5 day periods in the lutetium gross decay curve; which instead indicated the presence of a nuclide with a 1.7-day half-life. However, choosing proper growth times for the lutetium daughters, it was possible to obtain samples in which one or the other of these two lutetium daughters were prominent. By use of the magnetic spectrometer and detection with an anthracene crystal, it was possible to identify positrons associated with the 1.9-day lutetium activity. This fact was also confirmed by counting annihilation radiation with a NaI crystal detector.

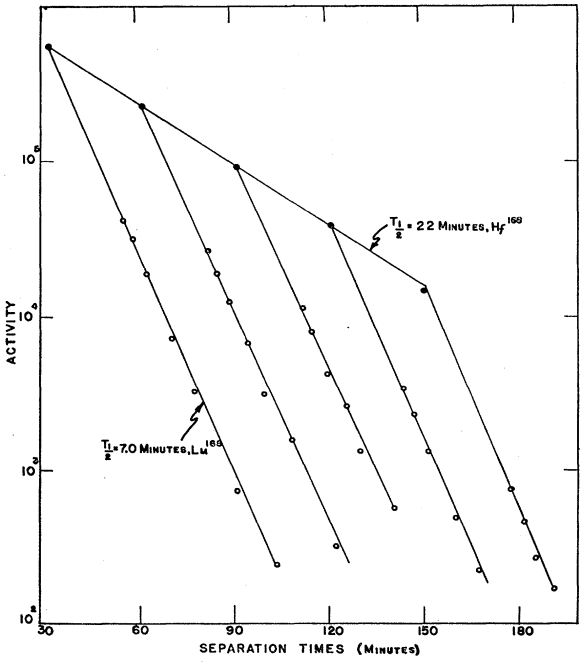

FIG. 1. Chemical isolation of Lu^{168} from Hf^{168} at successive times.

TABLE II. Decay periods associated with various radiations.

Radiation	Hafnium fraction	Half-lives observed		
Positrons	Initial growth	22 min	80 min	45 hr
Negatrons	Initial growth	24 min	11 hr	44 hr
Electromagnetic radiation	Initial growth	24 min	10 hr	44 hr

Radiation	Lutetium fraction Half-lives observed	
Positrons	7 min[a]	1.9 days
Negatrons	7 min	1.5 days
Electromagnetic radiation	7 min	1.7 days

[a] The 7-min positron activity accounted for 90% of the total positron activity associated with the sample.

A search was made for Lu^{167}, which was reported recently by two different groups to have a half-life of ~55 min.[1,2] To check this result, a tantalum target was used in order to get a higher yield of the very highly neutron deficient hafnium-lutetium isotopes. The hafnium fraction showed, besides the known half-lives, a component with a half-life of about 10 min. Lutetium was rapidly extracted from the hafnium and immediately beta counted. It initially decayed with a half-life of 4–7 min. No 55-min activity could be observed. In addition to the 4–7 min half-life, half lives of about 20 min and ~2 days were observed. The 20-min activity was produced from a parent activity with a half-life of about 10 min. From the experiments, one can conclude that Hf^{167} has a 10-min half-life, decays to an approximately 4-min Lu^{167} which in turn decays to Yb^{167} with a reported half-life of 19 min. It is possible however, that the measured 20-min activity is an unknown and unexplainable contamination, and that Hf^{167} has a half-life so short that no 55-min Lu^{167} could be extracted. The first lutetium milking from the hafnium fraction was made 50 min after the end of the irradiation, and that would mean the half-life of Hf^{167} has to be shorter than about 10 min. It is also possible, however, that the observed decay rates of 10 and 4 min may be due instead to the Hf^{166} and Lu^{166} decay chain.

Positron and Gamma Ray Energy Measurements

The energies of the positrons were measured with a 2 in.$\times\frac{1}{2}$ in. anthracene crystal scintillation spectrometer connected to a 256-channel pulse-height analyzer. The beta spectrometer was calibrated with standard beta sources such as C^{14}, Co^{60}, Tl^{204}, and Bi^{210}.

Two positron emitters were observed in the lutetium fraction, corresponding to the 7.0-min Lu^{168} and 1.9-day Lu^{170}. A series of Fermi plots, constructed from spectra at different times during the decay, yielded two positron groups (Fig. 2). Resolution of the Fermi plots yielded (1.20±0.05) Mev maximum positron energy for Lu^{168} and (1.8±0.1) Mev maximum positron energy for Lu^{170}.

The beta spectrum of the hafnium sample showed an increase in intensity for the first 10 min of measurement, due to the growth of the short-lived Lu^{168}. By analyzing the Fermi plots at different times (Fig. 3), it was possible to derive two positron groups from the hafnium samples. One had a maximum positron energy of (1.2±0.1) Mev and decreased with a half-life of ~25 min. This is probably the 1.2-Mev β^+ due to 7.0-min Lu^{168} growing to equilibrium with Hf^{168}. The second positron group had a maximum energy of (1.7±0.1) Mev, and decayed with a half-life of ~25 min. This positron group is probably associated with the decay of Hf^{168}. However, an exact assignment of these positron groups is difficult because of the number of isotopes and radiations present in the sample. At the same time that these measurements were made, Lu^{170} was growing in from Hf^{170}, giving rise to positrons with 1.8 Mev maximum energy. A positron group between 1.3–1.4 Mev was also observed. Resolution was very difficult, due to the presence of the other positron groups in greater intensity. However, this 1.3-Mev positron group appeared to be associated with the decay of Hf^{169}.

The gamma radiations from these nuclides was detected with a 2 in.×2 in. NaI crystal spectrometer, and analyzed with a 256-channel pulse-height analyzer. Table III summarizes the results.

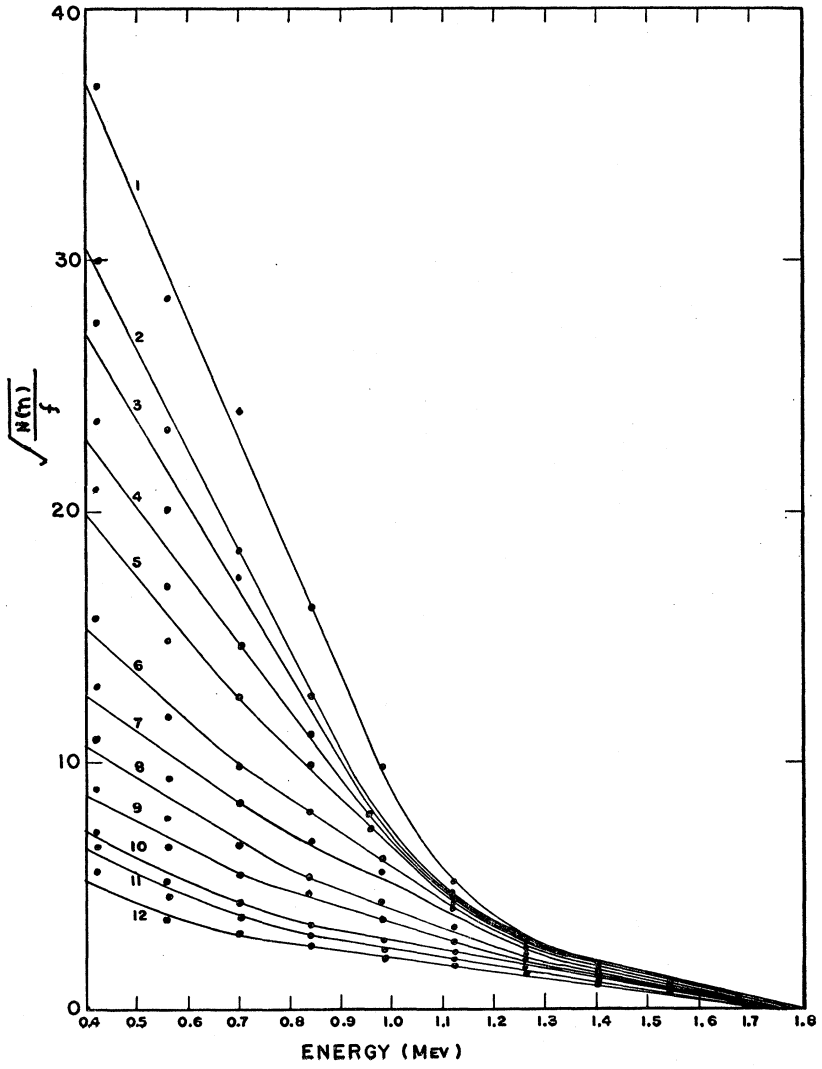

FIG. 2. Fermi plots of the lutetium fraction at different times. Curve 1, within 10 min of separation from hafnium. Curves 2–12, at 3 min intervals. Resolved maximum beta end points yielded: Lu^{168}, $E_{\beta^+}=(1.20\pm0.1)$ Mev; Lu^{170}, $E_{\beta^+}=(1.80\pm0.1)$ Mev.

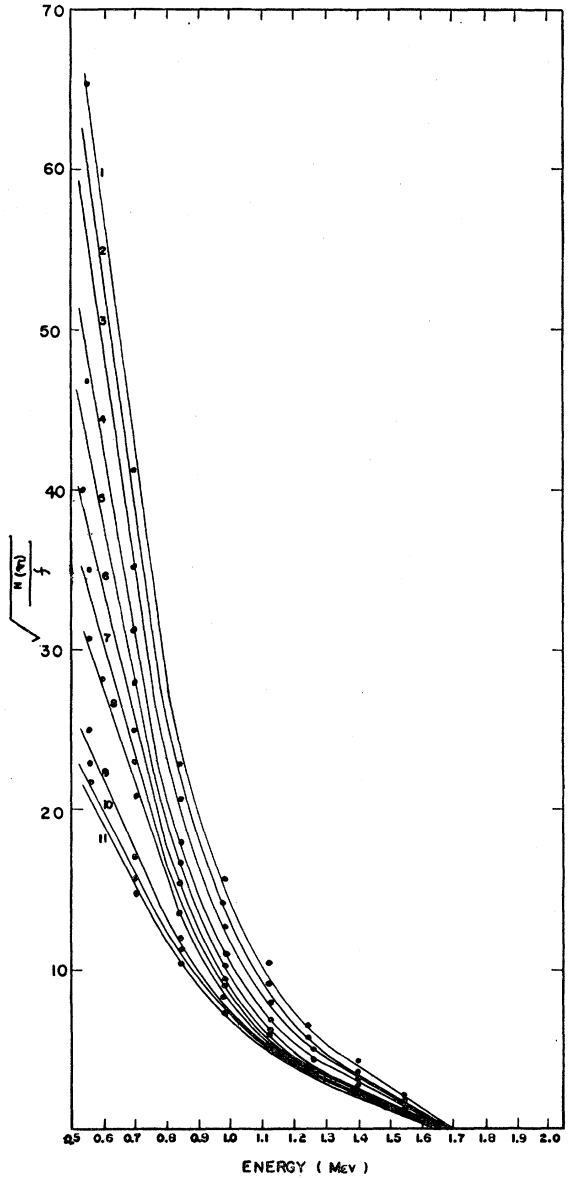

FIG. 3. Fermi plots of the hafnium fraction at different times. Curve 1, within 20 min of end of bombardment. Curves 2 and 3, at 10-min intervals; curves 4–8, at 20-min intervals; curves 9–12 at 1-hr intervals. Resolved maximum positron end points yielded: Lu^{168} (daughter of Hf^{168}), $E_{\beta^+}=(1.2\pm0.1)$ Mev; Hf^{168}, $E_{\beta^+}=(1.7\pm0.1)$ Mev.

It is very likely that in Hf^{168} and Hf^{169} γ rays with energies considerably higher than those listed are emitted, but as a consequence of their low intensity in comparison to the other gammas, an exact energy determination was not possible.

DISCUSSION

Five neutron-deficient hafnium isotopes are reported in the literature.[3] Hf^{175} is reported to have a half-life of 70 days, Hf^{172} about 5 years, and Hf^{170} 1.8 hr. The half-lives of Hf^{173} and Hf^{171} are uncertain, due to different values being reported by various investigators for these nuclides. Hf^{173} is reported with half-lives of 28 hr and 44 hr, while Hf^{171} is reported with 12 and 16 hr.

Hf^{170} was reported to decay by about 27% β^+ and 73% electron capture, having a maximum β^+ energy of 2.4 Mev and showing no γ emission. Beta-decay energy systematics give a Q^+ value of 2.0 Mev for Hf^{170}. This discrepancy makes it seem doubtful that this assignment could be correct. A tentative assignment for this positron decay to be associated with Hf^{169} was made in the Nuclear Data Sheets.[4] The Q^+ value for this isotope is $\sim$3.5 Mev. In the present investigation, a hafnium activity of 1.5-hr half-life was found in good agreement with the previously reported 1.8-hr value. It also was found that this activity belongs to Hf^{169} as a consequence of the observed decay chain Hf^{169} ($T_{\frac{1}{2}}=1.5$ hr) Lu^{169} ($T_{\frac{1}{2}}=1.5$ days) Yb^{169} ($T_{\frac{1}{2}}=32$ days). However, a disagreement exists in the observed positron energy for this isotope. The present measurements give a branching ratio f_E/f_{β^+} of about 2.5. However, the measured maximum beta energy is only about 1.3 Mev. A careful search for 2.4-Mev positrons was made, but no β^+ energy greater than 1.8 Mev could be detected at all in the hafnium sample.

The 1.8-hr hafnium activity found by Wilkinson[5] was produced by bombarding lutetium with 60-Mev protons. It is interesting that Nervik and Seaborg[6] report a 1.2-hr ytterbium activity which decays by positron emission with a maximum energy of 2.4 Mev. If it were possible that Wilkinson had a contamination of ytterbium in his hafnium fraction, it might explain his report of this β^+ energy.

A previously unreported activity with a half-life of (9 ± 2) hr was observed in the hafnium fraction, which cannot result from the reported 12- or 16-hr activities for Hf^{171}, because the half-life reported in this work was derived from milking a 1.9-day lutetium activity from the hafnium fraction. The 1.9-day lutetium activity is Lu^{170}. The Lu^{170} activity extracted from hafnium in intervals of 1.5 and 3.0 hr was detected by its positron-radiation.

As a result of this indirect determination, the error in half-life reported for Hf^{170} is relatively high, (9 ± 2) hr. No positron emission was found for this isotope, all decay presumably goes via electron capture to Lu^{170}. Also it was impossible to identify any γ radiation for this isotope because of the disturbing γ radiations from Hf^{171}, the half-life of which is very similar and also its γ spectrum is not well known.

As was mentioned, Nervik and Seaborg[6] found half-lives of 12 hr for Hf^{171} and 44 hr for Hf^{173}, whereas

[3] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 751 (1958).

[4] Nuclear Data Sheets, National Academy of Sciences, National Research Council No. NRC 59–2–80.

[5] G. Wilkinson and H. G. Hicks, Phys. Rev. **81**, 540 (1951).

[6] W. E. Nervik and G. T. Seaborg, Phys. Rev. **97**, 1092 (1955).

Wilkinson[5] reported 16 hr and 24 hr, respectively, for the same isotopes. Nervik and Seaborg produced these isotopes by the spallation of tantalum. However, Wilkinson produced them from a (p,xn) reaction on lutetium with protons varying between 30–60 Mev. A mixture of isotopes with half-lives of 9 and 16 hr may easily appear like a single half-life of 12 hr. Also the measured 44-hour half-life for Hf^{173} could be confused by the lutetium descendant activities: Lu^{169} ($T_{\frac{1}{2}}=1.5$ days) and Lu^{170} ($T_{\frac{1}{2}}=1.9$ days).

The third hafnium isotope reported, Hf^{168}, has a half-life of (22 ± 2) min, measured directly and indirectly by milking its 7.0-min lutetium daughter at intervals of 30 min. It is very difficult to ascertain if positrons are associated in this decay as a result of its descendant-activity, the β^+ emitter Lu^{168}. However, from the measured β-spectra of the hafnium and lutetium fractions, it seems likely that Hf^{168} decays by both electron capture and positrons. The contribution of the β^+ decay is definitely not very large, it may be in the order 1–3%. Wilkinson[5] was the first to report a 1.7-day activity in lutetium which he assigned to Lu^{170}. Later, Nervik and Seaborg,[6] confirmed by Gorodinskii,[7] reported the discovery of about a 2-day lutetium activity which they assigned to Lu^{169} because they could follow the decay into Yb^{169}, measured by its well-known γ spectrum. Mihelich *et al.*[8] reported the opinion that the 1.7-day lutetium activity of Wilkinson[5] might have been a mixture of Lu^{169} and Lu^{170}. They reported half-lives of 1.5 days and 1.9 days for Lu^{169} and Lu^{170}, respectively, in excellent agreement with the values reported here. The decay constant for Lu^{170} was measured, in the present work, by its positron radiation. With this half-life established, it was possible, by choosing adequate intervals for the isolation of lutetium from hafnium, to derive the 1.5-day half-life for Lu^{169}. Within the experimental limits, no positron radiation was observed for Lu^{169}. Lu^{170} decays, as already mentioned, by electron capture and a small branching of β^+ decay with a maximum energy of (1.8 ± 0.1) Mev.

TABLE III. Energies (in Mev) of gamma rays observed for the various nuclides.

Lu^{168}	Lu^{169}	Lu^{170}	Hf^{168}	Hf^{169}
0.088 ±0.002	0.024 ±0.002	0.084 ±0.002	0.129 ±0.005	0.049 ±0.005
0.223 ±0.005	0.062 ±0.003	0.15 ±0.01	0.17 ±0.01	0.115 ±0.005
0.71 ±0.02	0.111 ±0.003	0.19 ±0.01		
0.89 ±0.02	0.20 ±0.02	(0.51 ±0.01)[a]		
1.44 ±0.05	0.59 ±0.02			
1.81 ±0.05				
2.12 ±0.07				

[a] Annihilation radiation from β^+.

The Lu^{168}, which decays with a half-life of 7.0 min, has a f_E/f_{β^+} ratio of about 8. The maximum β^+ energy is found to be (1.20 ± 0.05) Mev. The γ spectrum could be measured very accurately for this isotope, due to the fact that this isotope could be isolated nearly free from other activities. No radioactive decay product of this radionuclide was observed, and therefore, the assignment must be Lu^{168}.

In recently published papers by Wilson and Pool,[9,10] it is reported that Lu^{168} and Lu^{170} were produced by separately bombarding enriched Yb^{168} in one case, and enriched Yb^{170} for the other, with ~5-Mev protons. Their reported γ energies are in good agreement with the ones found in the present investigation; however, they do not report any positron radiation associated with either Lu^{168} or Lu^{170}.

[7] G. M. Gorodinskii, A. N. Murin, V. N. Pokrovskii, B. K. Preobrazhenskii, and N. E. Tilov, Doklady Akad. Nauk S.S.S.R. **112**, 405 (1957).

[8] F. W. Mihelich, B. Harmatz, and T. H. Handley, Phys. Rev. **108**, 989 (1957).

[9] R. G. Wilson and M. L. Pool, Phys. Rev. **118**, 207 (1960).

[10] R. G. Wilson and M. L. Pool, Phys. Rev. **120**, 1843 (1960).