Study of Several Neutron-Deficient Hafnium Nuclides*

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The half-life of ^{169}Hf was found to be 3.26 ± 0.05 min. Several γ rays of ^{169}Hf were determined, and a partial decay scheme of ^{169}Hf proposed. The previously reported 1.5 h ^{169}Hf was found to be in error. The half-lives of ^{168}Hf, ^{170}Hf, and ^{171}Hf were redetermined to be 25.95 ± 0.20 min, 15.92 ± 0.15 h, and 12.09 ± 0.35 h, respectively. Several new γ rays of ^{168}Hf were observed. γ spectra of ^{170}Hf and ^{171}Hf were redetermined, and comparisons were made with previous investigations.

The neutron-deficient hafnium isotopes between mass 170 and mass 173 were first produced and identified in the bombardments of ytterbium with 20- and 38-MeV α particles and of lutecium with protons of energies from 15 to 75 MeV.[1] ^{168}Hf, ^{169}Hf, and ^{170}Hf were later studied from the bombardments of lutecium oxide with 300- to 400-MeV protons.[2] In these studies, the genetic relationships and mass assignments of these nuclides were established by measuring the well characterized radiations in the different lutecium nuclides from a series of timed chemical separations of lutecium from the original hafnium fraction without mass separations. In the first experiment, the identifications were further tested by observing the change of relative amounts of different nuclides produced by varying the bombarding energy. More studies[3,4] of these hafnium isotopes were carried out subsequent to the earlier investigations and discrepancies began to emerge. In addition to the large difference of the half-life of ^{170}Hf found in the first two investigations, the earlier reported 1.5 h ^{169}Hf[2] was not observed and the half-life of ^{169}Hf was found to have an upper limit of 8 min.[4] The half-lives of other nuclides also disagreed substantially from the earlier values. Recent experiments[5,6] have used isotopically pure samples of ^{170}Hf and ^{171}Hf and their half-lives were found to be still different from the previous results. Another effort to study ^{169}Hf was made recently[7] by using rapid high-purity chemical separations and otherwise the same principles as the earliest investigations.[1,2] The ^{169}Hf was produced in the spallation of tantalum by the bombardment of 660-MeV protons and ^{169}Lu was separated periodically from the chemically pure hafnium fraction. The half-life of ^{169}Hf was found to be 5.0 ± 0.5 min. The first seven columns of half-lives in Table I represent the experimental results concerning these nuclides prior to this investigation.

The purpose of this work was to study these nuclides again (from ^{168}Hf to ^{171}Hf), with special emphasis on measuring ^{169}Hf directly. Efforts were made again to search for the 1.5 h ^{169}Hf reported earlier[2] by preparing isotopically pure ^{169}Hf samples and direct observation of the short-lived ^{169}Hf[4,7] was attempted. For the latter purpose, the neutron-deficient nuclides were produced by irradiating an ytterbium target with ^{3}He ions. The irradiated targets could be assayed directly with a high-resolution Ge(Li) x-ray detector (full width at half maximum of 650 eV at 50 keV) immediately after the bombardment. The resolution of the detector was sufficient to separate the $K\alpha_1$ x rays of Lu from $K\alpha_2$ x rays of Lu and $K\alpha_1$ x rays of Yb. Even without chemical separation, the measured Lu $K\alpha_1$ x rays will include only contributions from K electron capture of neutron-deficient hafnium nuclides and from isomeric transitions of Lu isomers with transition energies exceeding the K-shell binding energy of lutecium. The half-lives produced were sufficiently different and the number of components small enough for the decay curve of the Lu $K\alpha_1$ x rays to be resolved by the least-squares computer program CLSQ.[8] By varying the energy of the bombarding particles to optimize the production of certain nuclides, the mass assignments could be assured. For the longer-lived nuclides, mass separation of the irradiated target was performed. High-resolution Ge(Li) detectors were used to measure the γ radiations of the samples.

SEARCH FOR 1.5-h ^{169}Hf

Isotopically pure ^{169}Hf was prepared in the following manner. An ytterbium oxide target was irradiated with 40-MeV ^{3}He ions in the Brookhaven 60-in. cyclotron. After 1 h of bombardment, the irradiated ytterbium oxide was introduced into the ion source of the Brookhaven electromagnetic isotope separator. The carrier-free lutecium and hafnium nuclides formed during the irradiation and the target material were converted to volatile chlorid-

TABLE I. Comparison of half-lives of neutron-deficient hafnium nuclides obtained by different investigators.

Nuclide	Half-life							
	Ref. 1	Ref. 2	Ref. 3	Ref. 4	Ref. 5	Ref. 6	Ref. 7	Present work
^{168}Hf	···	22 min	···	25 min	···	···	···	25.95±0.20 min
^{169}Hf	···	1.5 h	···	<8 min	···	···	5.0±0.5 min	3.26±0.05 min
^{170}Hf	112±2 min	9 h	12.2±0.5 h	12 h	···	16.25±0.25 h	···	15.92±0.15 h
^{171}Hf	16.0±0.5 h	···	10.7±0.3 h	11 h	12.4±0.3 h	···	···	12.09±0.35 h

es by a controlled stream of carbon tetrachloride vapor over the heated sample in the ion source. They were subsequently ionized, accelerated, and mass analyzed. The collected mass 169 sample was used for x-ray and γ-ray measurements. The K x rays were measured on a thin-window high-resolution 100-mm^2×5-mm Ge(Li) detector. The γ rays were measured on a calibrated 30-cm^3 Ge(Li) detector. The time elapsed between the end of bombardment and the first count was about 3 h. No trace of Lu K x rays (from ^{169}Hf) was observed, and neither were any γ rays decaying with the half-life of 1.5 h. The K x rays and the prominent γ rays corresponding to both ^{169}Lu and ^{169}Yb were observed. The 170 and 171 mass samples showed characteristic radiations of ^{170}Hf and ^{171}Hf; the amounts of ^{170}Lu and ^{171}Lu present in these samples at the time of the first count were smaller than the corresponding amounts of ^{170}Hf and ^{171}Hf. By reason of analogy, the amount of ^{169}Hf produced in the target should exceed the amount of ^{169}Lu produced during the irradiation. Therefore, the half-life of ^{169}Hf has to be considerably shorter than 1.5 h to escape observation. This result is consistent with previous reports which gave an upper limit of 8 min[4] and a value of 5.0 min,[7] respectively, for ^{169}Hf.

3.3-min ^{169}Hf

An enriched ^{170}Yb sample[9] was irradiated with 40-MeV ^{3}He ions in the Brookhaven 60-in. cyclotron for one minute. Immediately after the irradiation, the target was removed from the cyclotron and the K x rays of the sample were assayed with a thin-window high-resolution Ge(Li) x-ray detector. The measurements started about five minutes after the end of bombardment and were made for short intervals almost continuously in the first half hour and continued at suitable intervals for about one day. The Lu $K\alpha_1$ x ray was found to decay with three major half-life components: 3.3 min, 26 min, and 16 h. The relative intensities of these components after saturation corrections were 1:0.3:1. When the energy of the ^{3}He beam was lowered to 32 MeV in another experiment, these ratios changed to 1:0.012:3.3. When the energy of the ^{3}He beam was raised to 49 MeV, the ratio of the 3.3-min component to the 26-min component became 1:4.5. Duplicate runs at these energies gave reproducible results.

Since the 16-h half-life has been assigned to ^{170}Hf by mass separation (see below), these data show that the 3.3- and 26-min half-lives must be assigned to ^{169}Hf and ^{168}Hf, respectively, the three isotopes being produced from ^{170}Yb by the (^{3}He, 3n), (^{3}He, 4n), and (^{3}He, 5n) reactions, which have calculated thresholds of 14 , 23, and 31 MeV, respectively.[10] The 3.3-min activity cannot be identical with the 2.7-min ^{169m}Lu[11] because the energy of the isomeric transition in ^{169m}Lu is only 29 keV, too low for conversion in the K shell, and thus cannot be accompanied by Lu K x rays.

In addition to the Lu K x rays, several γ radiations were found to have half-life of 3.3 min. The decay of the annihilation radiations of the same samples also exhibited a 3.3-min component. The energies and the relative intensities of these γ rays are given in Table II. The weighted average of the half-life of ^{169}Hf from the measurements of these γ rays and the Lu $K\alpha_1$ x ray is 3.26±0.05 min.

Even though the 191-keV γ ray in ^{169}Lu was assayed from these samples, yet the abundances of the γ rays of ^{169}Hf could not be evaluated from these measurements. Due to the large difference in their half-lives and consequently in their levels of radioactivity, it was not possible to measure the growth of the 191-keV γ ray in ^{169}Lu in the presence of ^{169}Hf accurately enough to determine the relative initial amounts of ^{169}Lu and ^{169}Hf. However, one can assume that the ^{169}Hf-^{169}Lu pair has the same production cross-section ratio as the ^{170}Hf-^{170}Lu pair (which is 1.5); the abundance

TABLE II. γ rays of the 3.3-min ^{169}Hf.

Energy (keV)	Relative intensity
123	5.6±0.6
368	11.9±1.2
491	100
511	82±20

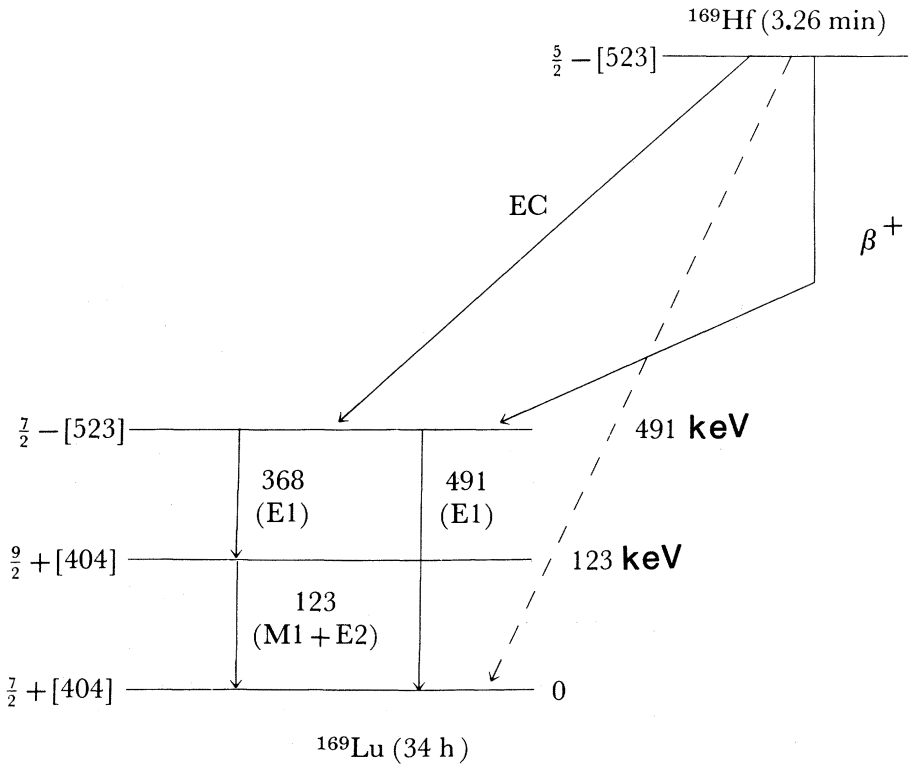

FIG. 1. Partial decay scheme of ^{169}Hf.

of the 491-keV γ ray in ^{169}Hf calculated based on this assumption is 83%.

According to the Nilsson formulation,[12] the ground states of ^{169}Hf and ^{169}Lu may be described by $\frac{5}{2}$−[523] and $\frac{7}{2}$+[404] orbitals, respectively. Using the analogy of the levels of the neighboring odd-mass lutecium isotope ^{171}Lu, a partial decay scheme of ^{169}Hf can be proposed as shown in Fig. 1. The conversion coefficients of these transitions for several multipole orders[13] are given in Table III. It is consistent that the 368- and the 123-keV γ rays are in cascade and form an alternate route for the depopulation of the 491-keV level and the multipole orders of the 368-, 123-, and 491-keV transitions should be $E1$, $M1$ or $E2$, and $E1$, respectively. Using the proper conversion coefficients, one can conclude that ^{169}Hf decays predominantly (>90%) through the 491-keV level in ^{169}Lu. The comparison of the intensity of the 123-keV transition with the intensity of the 368-keV transition requires either that the multipole order of the 123-keV transition be predominantly $E2$ or that another electron capture branch decays from ^{169}Hf to the 123-keV level. Since this latter transition would be accompanied by a change of two units of angular momentum and change of parity, its branching ratio must be extremely small. Therefore, the 123-keV transition should be mostly $E2$. A small branch of the ^{169}Hf decaying directly to the ground state in ^{169}Lu cannot be ruled out but would be very difficult to observe.

From the relative intensities in Table II, one can determine the K to positron ratio obtained in ^{169}Hf decay to be 2.7. The estimated β^+ end-point energy is 2.2 MeV.[14] Since the depopulation is primarily to the 491-keV level in ^{169}Lu, this corresponds to a mass difference of 3.7 MeV between the ground states of ^{169}Hf and ^{169}Lu, in good agreement with the calculated values of 4.0 MeV[10] and 3.6 MeV.[15]

^{168}Hf

The half-life and prominent γ radiations of ^{168}Hf were studied in the same experiments in which the 3.3-min ^{169}Hf was investigated. No chemical or mass separations were carried out. By varying the bombarding energy of the ^{3}He beam, the mass

TABLE III. Conversion coefficients of $Z=71$ (see Ref. 13).

Energy (keV)	Total conversion coefficient $E1$	$E2$	$M1$	$M2$
123	0.205	1.57	2.11	16.27
368	0.0126	0.043	0.101	0.364
491	0.006 29	0.0186	0.0469	0.142

TABLE IV. γ rays of the 26-min ^{168}Hf.

Energy (keV)	Relative intensity
157	100
184	147 ± 15

assignment was ascertained. The energies and relative intensities of the γ rays observed are given in Table IV. However, none of the previously reported γ rays[2] were observed in this investigation. The weighted average of the half-life of ^{168}Hf obtained in this work is 25.95 ± 0.20 min.

Note added in proof: We became aware of an article by Arlt, Malek, Musiol, Pfreffer, and Strusny[16] after this paper was submitted for publication. These authors found the half-life of ^{169}Hf to be 3.2 ± 0.1 min. They also observed the same γ rays as we did for ^{168}Hf and for ^{169}Hf.

^{170}Hf

An ytterbium oxide target was bombarded with a 40-MeV ^{3}He beam for about one hour. The irradiated ytterbium oxide was then used for mass separation in the Brookhaven isotope separator. The mass 170 sample was assayed on a calibrated 30-cm^3 Ge(Li) detector for γ radiations. A computer program for resolving the γ spectra was used.[17] The decay of these individual γ rays was analyzed with the CLSQ program.[8] The half-life of ^{170}Hf obtained by averaging the various measurements in this work is 15.92 ± 0.15 h, in good agreement with the most recent literature value.[6] The relative intensities of the γ rays of ^{170}Hf are given in Table V. Also listed for comparison in Table V are the relative intensities of these γ rays from earlier investigations.[4,6]

^{171}Hf

An isotopically pure ^{171}Hf sample was prepared in the same manner as ^{170}Hf. The mass 171 sample was also assayed on a calibrated 30-cm^3 Ge(Li) detector. Using the same methods of analyzing, the half-life of ^{171}Hf was found to be 12.09 ± 0.35 h, again in good agreement with the most recent literature value.[5] The relative intensities of γ rays of ^{171}Hf obtained both in this work and in another recent investigation[5] are listed in Table VI.

TABLE V. γ rays of the 16-h ^{170}Hf.

Energy (keV)	Relative intensity		
	This work[a]	Ref. 4	Ref. 6
99	17.5	14.3	20.1
116	2.9		2.4
119		2.4	
120	54.2	54.3	57.3
146	4.7	3.6	4.4
165	100	100	100
208	15.7	20.7	12.1
225	5.9	5.8	3.3
309	10.4	20.8	7.9
482	16.4	...	14.0
501	11.4	3.8	14.1
573	51.3	20.2	55.2
621	72.7	24.6	68.4

[a]The uncertainty of the relative intensity obtained in this work is about 10%.

TABLE VI. γ rays of the 12-h ^{171}Hf.

Energy (keV)	Relative intensity	
	This work[a]	Ref. 5
86	3.7	7.0
99	9.0	14.0
107	6.3	...
113	8.2	9.0
122	100	100
137	52.4	45.0
147	22.5	18.0
176	13.5	...
188	11.5	5.5
193	14.0	10.7
269	18.1	17.5
295	49.0	64.0
347	56.7	81.0
469	47.1	85.0
853	35.6	...

[a]The uncertainty of the relative intensity obtained in this work is about 10%.

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Evidence for the Missing 2− State in Ag^{110}†

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In spite of ingenious investigations of the isomeric decay of 253-day Ag^{110m} by several groups of authors, the missing low-energy first excited (2−) state in Ag^{110} has so far escaped detection. High-precision studies of the γ rays from the $Ag^{109}(n,\gamma)Ag^{110}$ reaction, as well as coincidence studies, now provide conclusive evidence for this level at an energy of 1.28 ± 0.10 keV.

The 253-day isomer of Ag^{110}, although it has been the subject of many investigations, still poses an unresolved problem. Namely, while the spins and parities of the isomeric state and the ground state of Ag^{110} are known to be 6+ and 1+, respectively, the only transition observed in the decay of Ag^{110m} is an $M4$ transition. It is thus necessary to postulate the existence of a "hidden" $E1$ transition from an intermediate 2− level, for which no conclusive evidence has yet been obtained.

The existing experimental evidence is as follows: A spin 6 for the 253-day isomer has been determined in an atomic-beam experiment.[1] Even parity for this level is inferred from its allowed β decay to an even-parity level in Cd^{110}.[2] The 1+ spin and parity of the ground state of Ag^{110} are inferred from its allowed β decay to 0+ and 2+ levels in Cd^{110}.[2] These assignments are in accordance with the shell-model configurations $[(g_{9/2}^{-3})_{7/2,9}(d_{5/2}^{-1})_n]_{1,6}$ expected for this nucleus. The nuclear magnetic moments for both states, computed on the j-j coupling model, also support this assumption.[3] The $M4$ multipole order of the 116.41-keV isomeric transition has been determined unambiguously by Geiger[4] in measurements of its K/L and $L_I/L_{II}/L_{III}$ internal-conversion coefficient ratios. Searches for the dipole transition have been carried out by several groups. Katoh and Yoshizawa[2] were able to deduce an upper limit of 25 keV from the absence of conversion-electron lines in the singles spectrum. Pasternak and Nardi[5] searched in vain for γ rays coincident with Ag K x rays from the internal conversion of the isomeric transition and thus reduced the upper limit to 5 keV, unless $\tau_{1/2} > 2$ μsec. Thus a low-energy $E1$ transition with a typical hindrance factor might have escaped detection. Hamilton, Jansen, Goudsmit, and Sattler[6] searched for low-energy conversion lines from Ag^{110m} with an iron-free double-focusing spectrometer. They did not find any conversion lines of a transition with $E \geq 4$ keV. Recently, positive evidence for a low-energy transition in the decay of Ag^{110m} has been reported by Rivier and Gizon[7]: Since a 3.18 ± 0.15-keV level had been deduced