The Isolation of ^{227}Ac from ^{231}Pa and the Gamma-ray Spectra of ^{231}Pa, ^{227}Th, and ^{223}Fr

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A method for the isolation of ^{227}Ac and its daughter nuclides from ^{231}Pa was established. The γ-ray spectra of ^{231}Pa, ^{227}Th, and ^{223}Fr were investigated, and the γ-ray tables of these nuclides were presented. A decay scheme of ^{223}Fr was corrected so as to explain these γ-ray data.

Several handicaps for the radiochemical study of the natural actinium series are known. Some of them are as follows:

1. It is difficult to keep enough ^{231}Pa or ^{227}Ac.
2. No confidential method for the separation of ^{231}Pa and ^{227}Ac has been established.
3. The exact information about the complex γ-rays of the nuclides of the actinium series is not enough for the γ-ray spectrometries of these nuclides.

From the standpoint of radiochemical common sense, ^{231}Pa is a precursor of ^{227}Ac. However, it is widely believed that the radiochemical purity of ^{227}Ac isolated from ^{231}Pa is not enough, because the complete removal of the colloidal or polymeric species of ^{231}Pa is very difficult. ^{227}Ac is almost non-γ-radioactive. The decay chain is complicated. Also ^{231}Pa, ^{227}Th, and ^{223}Fr have many γ-rays of approximately equal energies. Therefore, it is difficult to certify the radiochemical purity by γ-ray spectrometry in spite of the recent development of solid-state detectors. On the other hand, ^{227}Ac is produced as the daughter of ^{227}Ra, which is produced by means of the ^{226}Ra(n, γ) ^{227}Ra reaction. This method is recommended today. However, we do not think that the production of ^{227}Ac from ^{226}Ra is a popular technique or a conventional method, considering that the geochemical abundance of ^{231}Pa is of the same order as that of ^{226}Ra. Therefore, we tried to isolate ^{227}Ac from ^{231}Pa. Recently, we presented a method for the mutual separation of ^{227}Ac, ^{227}Th, ^{223}Ra, and ^{223}Fr.[1] The combination of these two methods will be an answer to the second difficulty with the radiochemistry of the actinium series mentioned above.

Then we determined the γ-ray spectra of ^{231}Pa, ^{227}Th, ^{223}Ra, and ^{223}Fr by means of a high-resolution Ge(Li) detector or an intrinsic germanium detector with a higher resolution. Moreover, we will present the data on the γ-rays of these nuclides for the sake of the γ-ray spectrometric determination of these nuclides.

Experimental

Radioisotopes. ^{231}Pa was supplied by The Radiochemical-Centre (Amersham, England). A 10-mg portion of the ^{231}Pa was purified by the method reported by Suzuki and Inoue.[2] The purified ^{231}Pa was left to stand for seven years to grow enough ^{227}Ac and then used for the experiment.

Reagents. All the chemicals used were of an analytical grade. HBEHP (bis(2-ethylhexyl) phosphate) was purified by the method reported previously.[3]

Apparatus. The apparatuses used were as follows. Separatory funnel shaker: IWAKI KM type(200—400 min^{-1}); coaxial Ge(Li) detector: ORTEC model 8101—1820 (effective volume, *ca.* 60 cm^3); planar intrinsic germanium detector: ORTEC model 16300-1000 (Be window 25 μm thick; effective volume, *ca.* 1.4 cm^3); silicon surface-barrier α-ray detector: ORTEC model BE-16-160-100 (active area, 160 mm^2; depletion depth, 100 μm); multichannel pulse-height analyzer: TOSHIBA USC-1 model 10 4096 ch. (controlled by a minicomputer TOSBAC-40A); linear amplifier: ORTEC model 472 spectroscopy amplifier (the shaping time constant was fixed to 2 μs throughout the spectrum accumulation).

The dead-time ratio was monitored by USC-1 and was found to be less than 10%.

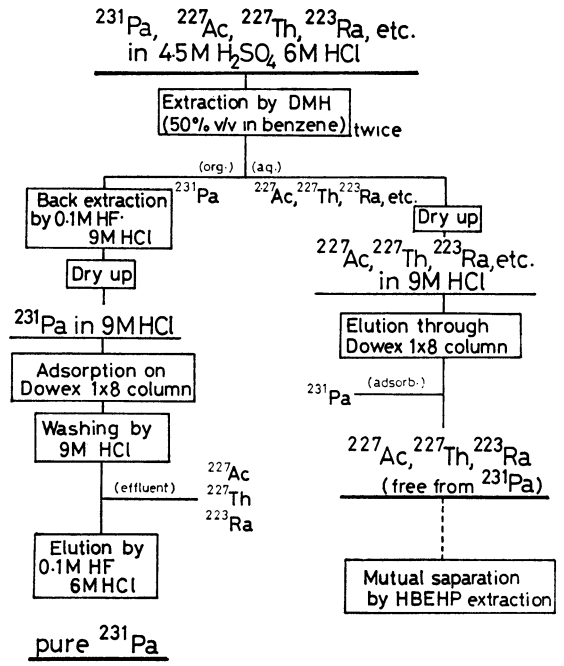

Fig. 1. The separation method for ^{231}Pa and ^{227}Ac.
In this figure 1 M = 1 mol/dm^3.

Procedures. Separation of ^{231}Pa and Its Daughter Nuclides: A flow chart for the chemical separation of ^{231}Pa and ^{227}Ac is shown in Fig. 1. The meaning and the performance of the separation method will be described in the next section. From the isolated ^{227}Ac fraction, ^{227}Ac, ^{227}Th, and ^{223}Ra were purified by the method reported in Ref. 1. ^{223}Fr was milked from the purified ^{227}Ac by the method reported in Ref. 1.

Alpha-ray Spectrometry: In order to examine the radiochemical purity of the purified ^{231}Pa and ^{227}Ac, the α-ray spectra were compared. Thin sources for good resolution in the α-ray spectrum were prepared by the evaporation of aliquots of solutions of these nuclides onto glass plates. The apparent resolution of an α-ray peak was less than 20 keV in FWHM.

Gamma-ray Spectrometry: The γ-ray spectra of ^{231}Pa, ^{223}Fr,

and ^{223}Ra were determined by means of the Ge(Li) detector. The γ-ray spectrum of ^{227}Th was determined by the intrinsic germanium detector, because the spectrum is too complex to resolve each γ-ray by the Ge(Li) detector. For the calibration of energy and the efficiency of the acquisition system, the standard sources of ^{241}Am, ^{109}Cd, ^{144}Ce, ^{133}Ba, ^{22}Na, ^{106}Ru, ^{137}Cs, ^{54}Mn, and ^{60}Co were used. In addition, KX-rays of Ac, Ra, Rn, Po, Bi, and Pb were applied to the inner calibration. The highest resolution observed for the Ge(Li) detector was 1.2 keV in FWHM for 300 keV γ-ray of ^{231}Pa. The resolution of the intrinsic germanium detector was 300 eV at 20 keV, 400 eV at 50 keV, 500 eV at 100 keV, and 800 eV at 300 keV.

Plate sources of ^{231}Pa, ^{227}Th, and ^{223}Ra were prepared by the evaporation of aliquots of the purified stock solutions onto stainless steel plates. Liquid sources of ^{223}Fr were prepared by pipetting aliquots of the purified solution into polyethylene test tubes. The calibration factor for the self-absorption by the liquid sources was calculated by comparing the relative intensities determined with respect to the two kinds of sources of ^{227}Th, which has many γ-rays identical with ^{223}Fr.

All the spectra were analyzed by means of a nonlinear least-mean-squares fitting method to evaluate the peak position and the peak area. The least-mean-squares fitting method is based upon peak response functions rather than analytical functions in order to account for the histogram nature of the γ-ray spectrum accumulated by a multichannel pulse-height analyzer. Moreover, the peak response functions were determined from the γ-ray peak shapes of the γ-rays of the standard sources.

The relative intensities of the γ-rays of ^{227}Th, ^{223}Fr, and ^{223}Ra were determined by the decay analysis of the calculated peak areas. The abundances of the γ-rays of ^{223}Fr were calculated by normalizing the relative intensities to the α-disintegration rate of ^{227}Ac, which was in radioequilibrium with ^{223}Fr.

Results and Discussion

Radiochemical Purity of the Purified ^{231}Pa and ^{227}Ac. From the point of view of the decontamination of ^{227}Ac for ^{231}Pa, the anion-exchange chromatographic separation method reported by Suzuki and Inoue[2] is the best method if we can suppress the irreversible formation of colloidal or polymeric species of Pa(V), which is not adsorbed by the anion-exchange resin column. Therefore, we applied this method to the final stage of the separation of ^{227}Ac and ^{231}Pa, as is shown in Fig. 1. However, it was often observed that a small fraction of ^{231}Pa was eluted out with a loading solution when more than 1 mg of ^{231}Pa was introduced into the separation column. The fast, irreversible polymerization of Pa(V) in a hydrochloric acid solution has been reported.[4] Therefore, this eluted fraction of ^{231}Pa can be considered to be the polymeric species.

To get enough ^{227}Ac from ^{231}Pa, we must chemically process a weighable amount of ^{231}Pa. Quantitative information has not yet been obtained about the chemical behavior of polymeric Pa(V) in an aqueous solution. However, it is known that the polymerization of Pa(V) in an aqueous solution is negligible unless the solution is left to stand too long after the preparation and unless the concentration of Pa(V) is more than 10^{-7} mol/dm^3.[4-6] These facts show that, in order to suppress the formation of polymeric Pa(V) in a hydrochloric acid solution, the concentration of Pa(V) should be lower than 10^{-7} mol/dm^3.

On the basis of these considerations, we applied a preliminary separation based on the DMH(2,6-dimethyl-4-heptanol)-extraction of ^{231}Pa from a mixed acid solution of a sulfuric acid and a hydrochloric acid in order to lower the concentration of ^{231}Pa in the ^{227}Ac fraction. Pa(V) forms stable anionic complexes in a concentrated sulfuric acid solution and does not show the tendency to polymerize. Moreover, the extraction of macro amount of Pa(V) from the mixed acid gives very reproducible results. The extraction ratio of Pa(V) exceeds 99.95% by the use of DMH-extraction process shown in Fig. 1, while Ac is not extracted. The isolated ^{227}Ac fraction was processed by the HBEHP-extraction method to get the purified ^{227}Ac, ^{227}Th, ^{223}Ra, and ^{223}Fr.[1]

An α-ray spectrum of the purified ^{231}Pa is compared with a spectrum of ^{231}Pa before the separation in Fig. 2.

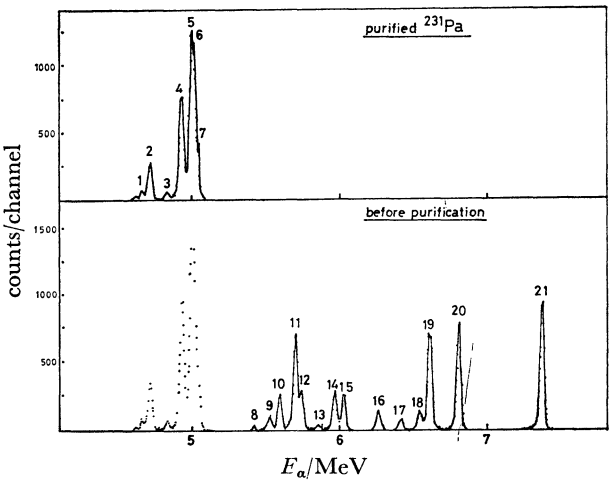

Fig. 2. Alpha-ray spectra of ^{231}Pa.
1, 4.671 MeV ^{231}Pa; 2, 4.727 MeV ^{231}Pa; 3, 4.844 MeV ^{231}Pa; 4, 4.943 MeV ^{231}Pa; 5, 5.007 MeV ^{231}Pa; 6, 5.023 MeV ^{231}Pa; 7, 5.052 MeV ^{231}Pa; 8, 5.432 MeV ^{227}Th; 9, 5.538 MeV ^{227}Th; 10, 5.605 MeV ^{223}Ra; 11, 5.71 MeV (complex) ^{227}Th and ^{223}Ra; 12, 5.755 MeV ^{227}Th; 13, 5.864 MeV ^{227}Th; 14, 5.976 MeV ^{227}Th; 15, 6.037 MeV ^{227}Th; 16, 6.278 MeV ^{211}Bi; 17, 6.423 MeV ^{219}Rn; 18, 6.551 MeV ^{219}Rn; 19, 6.622 MeV ^{211}Bi; 20, 6.817 MeV ^{219}Rn; 21, 7.384 MeV ^{215}Po.

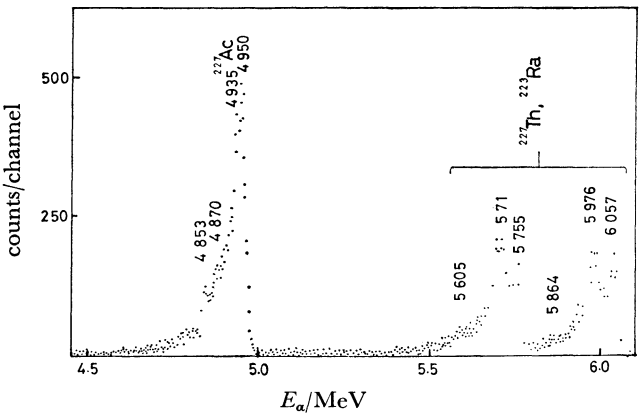

Fig. 3. Alpha-ray spectrum of the purified ^{227}Ac.

An α-ray spectrum of ^{227}Ac is also shown in Fig. 3. In Fig. 3, no α-rays of ^{231}Pa are observed, while α-rays of ^{227}Th and ^{223}Ra are observed. These daughter nuclides grow fast during the sample preparation and the spectrum acquisition. The branching ratio of the α-decay of ^{227}Ac is only 1.38%.[7] Therefore by comparing these three α-ray spectra, it can be concluded that the purified ^{227}Ac is completely free from the contamination of ^{231}Pa. On the other hand, no α-rays of ^{227}Th, ^{223}Ra, and their daughter nuclides appeared in the purified ^{231}Pa sample. This is evidence that the purified ^{231}Pa is free from any contamination of the daughter nuclides.

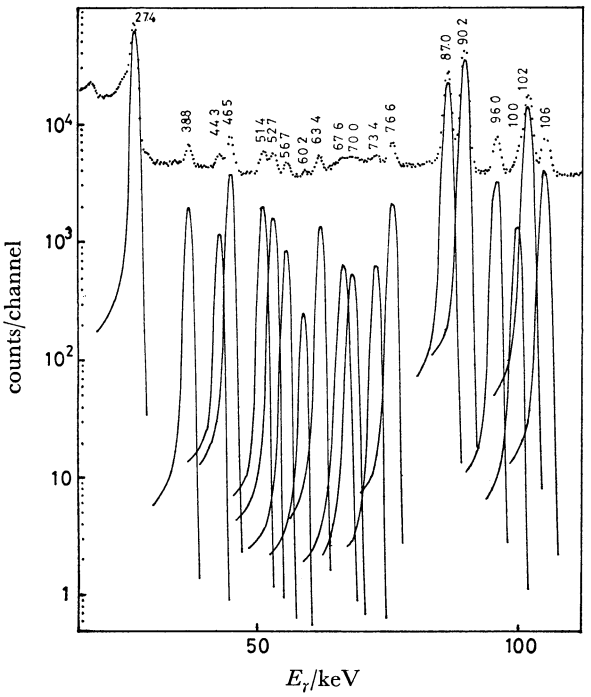

Fig. 4. Gamma-ray spectrum of ^{231}Pa, from 20 keV to 110 keV.

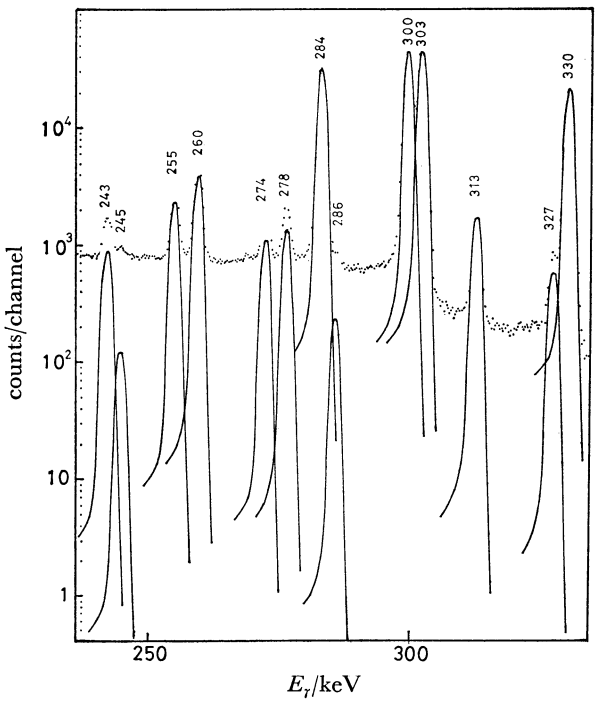

Fig. 5. Gamma-ray spectrum of ^{231}Pa, from 240 keV to 335 keV.

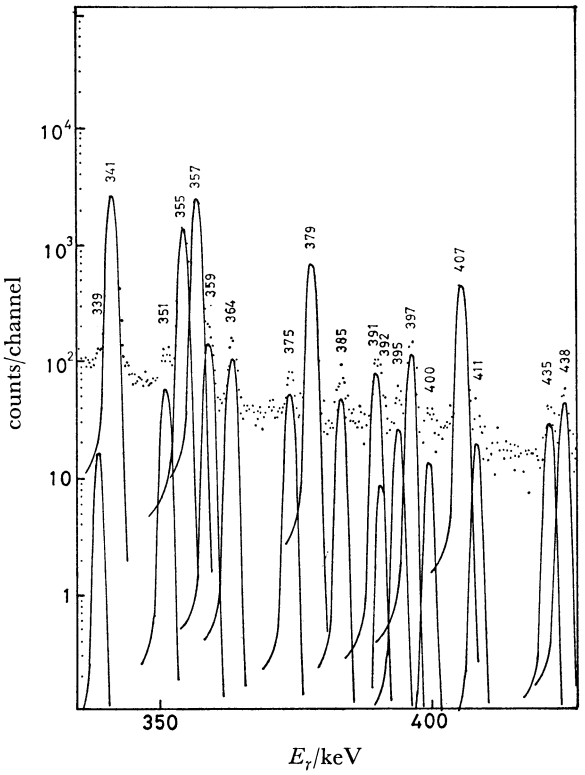

Fig. 6. Gamma-ray spectrum of ^{231}Pa, from 335 keV to 440 keV.

Gamma-ray Spectrum of ^{231}Pa. The γ-ray spectra of ^{231}Pa are shown in Figs. 4—6. In these figures, the peak components are shown as the peak shapes to illustrate the analysis of the γ-ray spectrum described in the Experimental section. The relative intensities of some γ-rays are shown in Table 1.

It is worthwhile to point out the following facts from Table 1:

1. Our data show the best correlation with the data of de Pinho *et al.*[8] The correlation is almost complete for the γ-rays whose intensities relative to that of 303

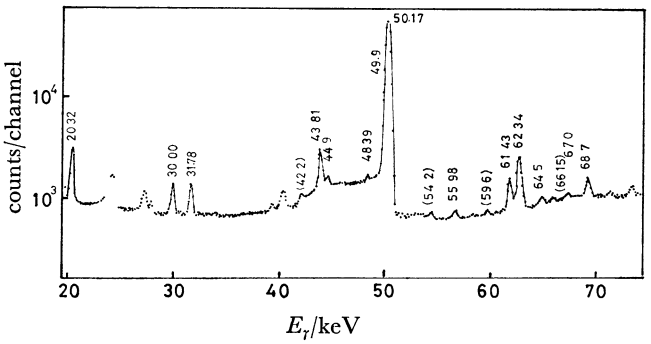

Fig. 7. Gamma-ray spectrum of ^{227}Th, from 20 keV to 73 keV.
The γ-rays in parentheses are decayed also but the intensities are not certain.

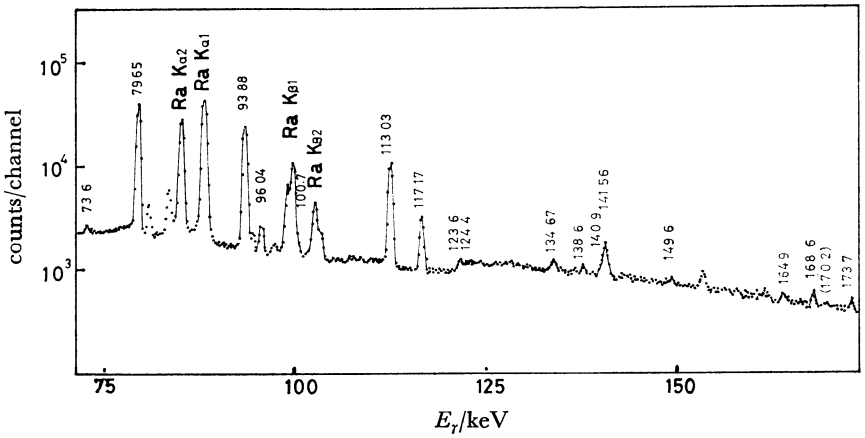

Fig. 8. Gamma-ray spectrum of ^{227}Th, from 72 keV to 175 keV. The γ-rays in parentheses are decayed also but the intensities are not certain.

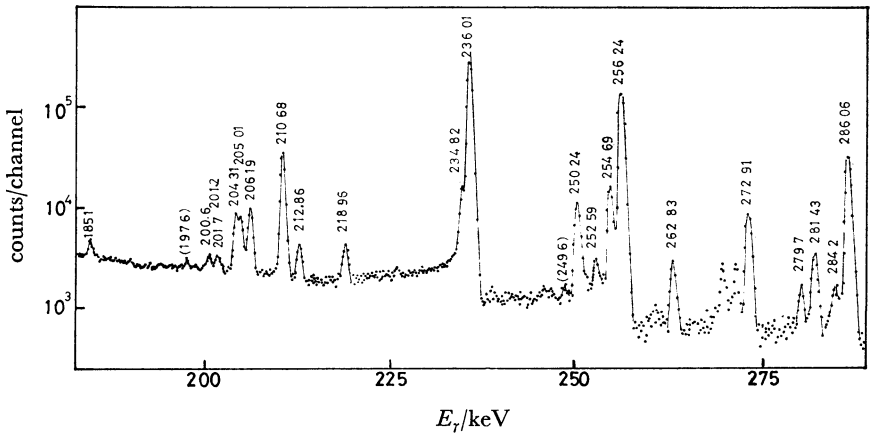

Fig. 9. Gamma-ray spectrum of ^{227}Th, from 180 keV to 288 keV. The γ-rays in parentheses are decayed also but the intensities are not certain.

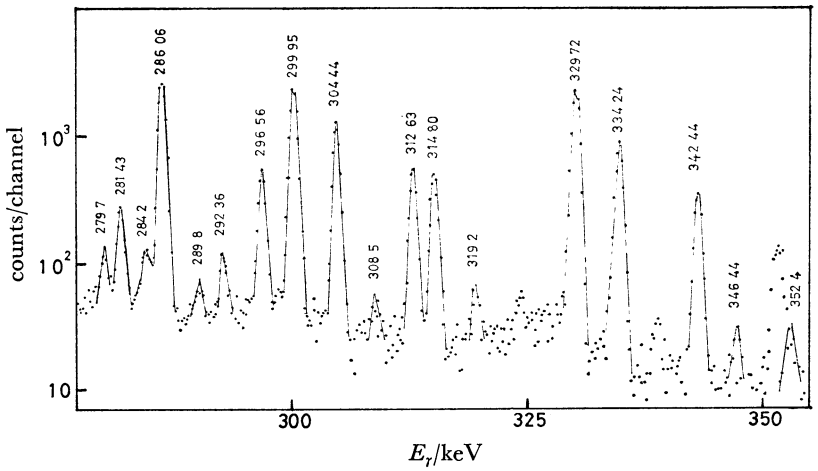

Fig. 10. Gamma-ray spectrum of ^{227}Th, from 278 keV to 355 keV.

keV γ-ray are greater than 0.01.

2. The data of Lange and Hagee[9] are quite erroneous, though they were employed in the γ-ray catalogue of Wakat.[10]

3. The data of Zaddach[11] are the latest. His data about abundances are identical with the data of Leang and since Zaddach did not describe his experimental technique, we considered the data of Leang. As will be described below, Leang's data are not very accurate and the intensities of the 255 keV, 284 keV, 300 keV,

TABLE 1. TABLE OF γ-RAYS OF ^{231}Pa

E_γ/keV	Relative intensity			
	This work	References		
		9, 10[a]	8	11, 12
27.4	4.1	(obsd)[b]	3.97	2.98
38.8	0.044	(obsd)	0.063	0.043
44.3	0.027	(obsd)	0.026	
46.5	0.077	(obsd)	0.089	0.077
52.7	0.035	(obsd)	0.036	
56.7	0.018		0.016	
63.4	0.016		0.021	
73.4	0.015		0.011	
76.7	0.036	(obsd)	0.029	(obsd)
96.0	0.039		0.038	0.030
100	0.017		0.013	
243	0.017	0.025	0.015	(obsd)
255	0.043	0.027	0.043	0.057
260	0.076	0.082	0.074	0.078
274	0.021	(obsd)	0.025	0.03
278	0.025	(obsd)	0.029	0.03
284	0.67	0.43	0.67	0.70
300	0.97	1.00	0.97	1.00
303	1.00		1.00	1.00
313	0.039	0.018	0.040	0.047
327	0.016		0.013	
330	0.55	0.15	0.56	0.57
341	0.070	0.016	0.070	(obsd)
355	0.038		0.040	0.043
357	0.066	0.020	0.075	0.065
379	0.019	0.003	0.021	0.017
407	0.018	(obsd)	0.015	0.009

The γ-rays which are important for the γ-ray spectrometry of the actinium series are tabulated in this table.
a) Intensities relative to the doublet of 300 keV and 303 keV. b) Observed.

and 313 keV γ-rays are obviously overcounted. These deviations, except that of the γ-ray of 255 keV, can be explained as resulting from the contamination of ^{233}Pa, which was spiked as a tracer in his experiment. Indeed, this contamination was mentioned by Leang in his raw γ-ray spectrum.[12]

As has been described above, we can conclude that our data and de Pinho's data about the relative intensity have a higher reliability than the other data. The abundances of the γ-rays of ^{231}Pa were determined by Leang; the abundance of 303 keV γ-ray is 2.3%.[12] As will be described below, the abundance of the γ-rays of ^{231}Pa are much smaller than those of ^{227}Th. Also, the measurement of the intensities of the main γ-rays of ^{231}Pa, *i.e.*, 284 keV, 300 keV, 303 keV, 341 keV, and 357 keV, is almost impossible if ^{227}Ac, ^{227}Th, or ^{223}Ra is in the sample of ^{231}Pa. Especially, it should be remenbered that 300 keV γ-rays of ^{231}Pa and ^{227}Th appear as a singlet peak even when a high-resolution Ge(Li) detector is used. Therefore, it is necessary to separate completely ^{227}Ac and its daughter nuclides from ^{231}Pa for the γ-ray spectrometric determination of ^{231}Pa. It is also necessary to certify the correlation between the relative intensities of the observed γ-rays and our data.

Gamma-ray Spectrum of ^{227}Th. The γ-ray spectra of ^{227}Th, as obtained by the intrinsic germanium detector, are shown in Figs. 7—10. Even in an initially pure ^{227}Th sample, ^{223}Ra and its daughter nuclides grow rather fast. Therefore, as has been described in the Experimental section, the decay in the γ-rays was examined until the transient equilibrium was attained between ^{227}Th, ^{223}Ra, and their daughter nuclides. Some examples of the decay curves are shown in Fig. 11. As is shown in Fig. 11, γ-rays of ^{227}Th can be easily determined to be monotonously decaying γ-rays. In Table 2, the relative intensities are shown along with the literature values.[13,14]

The data of Hesselink *et al.*[14] are the latest and were obtained by α,γ coincidence employing a Ge(Li) detector with a resolution of 2.3 keV at 120 keV and a silicon surface-barrier detector with a resolution of 30 keV. It is clear from Figs. 2 and 7—10 that the Ge(Li) detector and the silicon surface-barrier detector do not have efficient enough resolutions for the γ-ray spectroscopy of ^{227}Th. On the other hand, the works of Briancon *et al.*[13] consist of γ,γ coincidence and conversion electron spectroscopy. They also employed two Ge(Li) detectors with resolutions of 0.9 keV and 2.5 keV for 100 keV γ-ray. The resolutions of the Ge(Li) detectors employed by Briancon *et al.* were not good enough to resolve the complex structure of the γ-ray spectrum of ^{227}Th at the energy regions of 40 keV to 50 keV, 80 keV to 100 keV, 200 keV to 213 keV, 234 keV to 236 keV, 250 keV to 257 keV, 269 keV to 273 keV, 279 keV to 305 keV, 312 keV to 315 keV, and 350 keV to 353 keV. In spite of these difficulties, the two works agree well about existence of the main γ-rays. However, about the intensities of almost all the γ-rays and the existence of some weak γ-rays, the agreement between the two works is far from satisfactory. For the γ-ray spectrometry of ^{227}Th, the

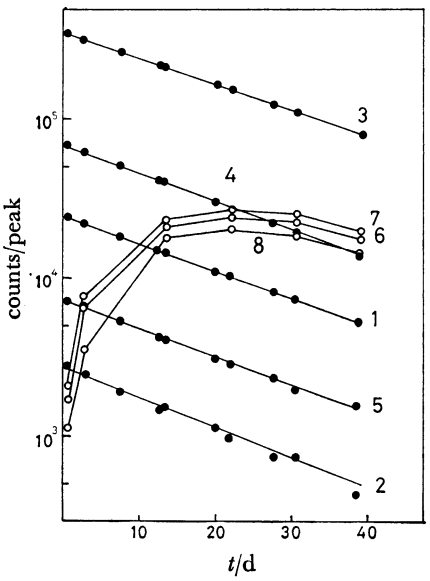

Fig. 11. Examples of decay or growth curves of some γ-rays found in initially pure ^{227}Th. 1, 20.32 keV; 2, 30.00 keV; 3, 50.17 keV; 4, 236.01 keV; 5, 299.95 keV; 6, 72.86 keV; 7, 269.46 keV; 8, 351.1 keV.

Table 2. Table of γ-rays of ^{227}Th

E_γ/keV	Relative intensity[a] This work	Ref. 14	Ref. 13	Transition
20.32(5)	0.030(2)[b]		0.037	50.2—29.9
30.00(5)	0.005(1)		0.007	29.9—0
31.78(8)	0.005(1)		0.006	61.5—29.9
43.81(9)	0.017(3)	0.002	0.017	329.8—286.1
44.5(1)	0.002(1)	0.001		174.6—130.0
48.39(8)	0.003(1)	0.004	0.0007	334.2—286.1
49.9(2)	0.062(9)	0.002	0.04	79.8—29.9
50.17(6)	0.59(2)	0.72	0.59	50.2—0
55.98(8)	0.0006(1)	0.0002	0.0004	432.4—376.4
61.43(8)	0.007(1)		0.006	61.5—0
62.34(7)	0.017(1)	0.021	0.015	123.9—61.5
64.5(1)	0.0011(6)	0.002	0.001	350.5—286.1
67.0(1)	0.0016(5)			
68.7(1)	0.0061(3)	0.002	0.004	130.3—61.5
73.6(1)	0.0012(2)	0.0014	0.0015	123.9—50.2
79.65(2)	0.162(4)	0.15	0.15	79.8—0
93.88(9)	0.127(4)	0.11	0.10	123.9—29.9
96.04(8)	0.007(1)	0.004	0.005	376.4—280.5
100.7(1)	0.0028(5)	0.007	0.006	130.0—29.9
113.03(1)	0.067(1)	0.045	0.053	174.6—61.5
117.17(1)	0.016(1)	0.013	0.013	247.5—130.3
123.6(2)	0.0011(6)	0.0007	0.0007	247.3—123.9
124.4(4)	0.0009(1)	0.0002	0.0004	174.6—50.2
134.67(9)	0.0022(4)	0.002	0.002	369.4—234.8
138.6(6)	0.001			
140.9(2)	0.0023(3)	0.003		314.8—174.6
141.56(6)	0.0086(6)	0.005	0.01	376.4—234.8
149.6(1)	0.0007(4)	0.002	0.0007	280.5—130.5
164.9(7)	0.0009(1)		0.001	445.1—280.5
168.6(2)	0.0010(3)	0.001	0.001	342.9—174.6
173.7(2)	0.0014(3)	0.0009	0.001	234.8—61.5
185.1(4)	0.0029(5)	0.0022	0.003	234.8—50.2
200.6(2)	0.0017(6)	0.0004	0.003	280.5—79.8
201.2(2)	0.0017(6)			
201.7(2)	0.0017(6)	0.0018	0.003	376.4—174.6
204.31(3)	0.015(1)	0.019	0.016	334.2—130.2
205.01(5)	0.011(1)	0.014	0.011	234.8—29.9
206.19(3)	0.018(1)	0.022	0.016	329.9—123.9 or 286.1—79.8
210.68(2)	0.089(2)	0.10	0.082	334.2—123.9
212.86(4)	0.0062(5)	0.007	0.006	342.9—130.3
218.96(5)	0.0069(8)	0.004	0.005	342.9—123.9
234.82(6)	0.040(2)	0.05	0.04	234.8—0
236.01(1)	0.96(1)	0.95	0.97	286.1—50.2
250.24(2)	0.037(2)	0.023	0.031	329.8—79.8
252.59(6)	0.007(1)	0.009	0.007	376.4—123.9
254.69(2)	0.058(2)	0.075	0.052	334.2—79.8
256.24(2)	0.522(9)	0.52	0.54	286.1—29.9
262.83(6)	0.0054(3)	0.008	0.007	342.4—79.8
272.91(3)	0.038(1)	0.041	0.037	334.2—61.5
279.7(1)	0.0043(8)	0.008	0.005	329.8—50.2
281.43(6)	0.014(1)	0.012	0.012	342.9—61.5
284.2(3)	0.0045(4)	0.002	0.004	334.2—50.2
286.06(2)	0.157(2)	0.14	0.11	286.1—0
289.8(3)	0.0018(6)	0.0003	0.0005	369.4—79.8
292.36(5)	0.0045(6)	0.005	0.005	342.4—50.2
296.56(3)	0.037(5)	0.032	0.036	376.4—79.8
299.95(3)	0.172(4)	0.16	0.16	329.8—29.9
304.44(3)	0.085(1)	0.11	0.075	334.2—29.9
308.5(1)	0.0016(4)	0.001	0.001	432.4—123.9
312.63(2)	0.041(2)	0.043	0.037	342.4—29.9
314.80(3)	0.038(1)	0.047	0.035	376.4—61.5
319.2(3)	0.0025(5)	0.002	0.003	369.4—50.2
329.72(3)	0.212(6)	0.24	0.21	329.8—0
334.24(2)	0.083(3)	0.095	0.082	334.2—0
342.44(5)	0.036(4)	0.016	0.031	342.4—0
346.44(5)	0.0018(3)	0.0007	0.0007	376.4—29.9
352.4(1)	0.0008(3)	0.0007	0.001	432.4—79.8
384.5(1)	0.0038(9)	0.0002	0.004	424.0—61.5

a) Intensities relative to the doublet of 234.82 keV and 236.01 keV. b) Numbers in parentheses are estimated standard deviations in units of the last significant digit.

relative intensities of the γ-rays of 50 keV, 236 keV, 286 keV, and 300 keV are very important parameters as measures of the radiochemical purity of the sample, because the contamination by ^{231}Pa or ^{227}Ac influences the relative intensities of these γ-rays. These γ-rays are observed as multiplet γ-ray peaks, and the multiplicity depends on the resolution of the detector employed. Therefore, it is necessary to get data about the intensities of these γ-rays based upon more accurate basis than the two works in the literature in order to improve the accuracy of the γ-ray spectrometry of ^{227}Th.

On the other hand, the excellency of our measurement depends on the highest resolution of the detector employed. As is shown in Figs. 7—10, we obtained well-resolved γ-ray spectra, and so we can now consider the reason for the disagreement between the literature data.

Obviously, there is no doubt about the existence of

Table 3. Table of γ-rays of ^{223}Ra

E_γ/keV	Relative intensity This work	Ref. 15	Nuclide
123	0.105	0.082	^{223}Ra
132	0.017	0.009	^{219}Rn
144	0.267	0.221	^{223}Ra
154	0.459	0.386	^{223}Ra
159	0.058	0.050	^{223}Ra
180	0.015	0.011	^{223}Ra
269	1.00	1.00	^{223}Ra
271	0.818	0.786	^{219}Rn
324	0.265	0.264	^{223}Ra
328	0.013	0.014	^{223}Ra
338	0.186	0.193	^{223}Ra
343	0.011	0.014	^{223}Ra
351	0.951	0.950	^{211}Bi
372	0.037	0.039	^{223}Ra
402	0.457	0.479	^{219}Rn
405	0.271	0.293	^{211}Pb
427	0.122	0.136	^{211}Pb

Only the γ-rays, which are important for γ-ray spectrometry of the actinium series, are tabulated.

the three γ-rays of 20.32 keV, 30.00 keV, and 31.78 keV reported by Briancon *et al.*[13] However, we could not find any γ-rays except the KX escape peaks of 50.17 keV γ-ray at 39.22 keV and 40.32 keV. It was found the intensity ratio of these two peaks in identical to that of the K_α and K_β rays of Ge. Thus, the intensity of the γ-ray of 40.1 keV reported in Ref. 13 seems to be doubtful. Similarly, we did not observe the 224.7 keV and 246.4 keV γ-rays except the KX escape peaks of the 236.01 keV and 256.24 keV γ-rays respectively, while these two γ-rays were reported in Ref. 14. For the sake of further consideration, a γ-ray table of ^{223}Ra and its daughter nuclides is shown in Table 3. In the γ-ray spectrum of ^{223}Ra, KX-rays of the daughter elements are also observed. In ^{227}Th, we observed rapidly growing KX-rays and γ-rays of 69.8 keV, 72.9 keV, 75.3 keV, 89.9 keV, 95.0 keV, 269.5 keV, 271.2 keV, 324.2 keV, 338.3 keV, and 351.1 keV. Many γ-rays with energies approximately equal to these KX-rays and γ-rays were reported in Refs. 13 and 14. For the γ-ray spectrometry of ^{227}Th, these γ-rays are not important because the many other intense γ-rays can be utilized. However for ^{223}Ra, 269 keV, 324 keV, and 338 keV γ-rays are important for the γ-ray spectrometry. According to Refs. 13 and 14, it seems necessary to correct the contribution by the interfering γ-rays in order to evaluate the intensities of these γ-rays accurately when the sample is contaminated with ^{227}Ac or ^{227}Th. However as is shown about the 269.46 keV γ-ray in Fig. 11, the correction is negligibly small.

According to Refs. 13 and 14, the abundances of the 50 keV, 236 keV, 256 keV, 286 keV, 300 keV, 304 keV, and 330 keV γ-rays of ^{227}Th are large enough to apply these γ-rays to the γ-ray spectrometry of ^{227}Th. We measured the abundance of the overlapped γ-rays of 234.82 keV and 236.01 keV and got a value of $(12.1\pm 0.6)\%$.[16] However, it should be mentioned that all these seven γ-rays are observed as complex overlapping peaks even when the spectrum is determined by a detector with the highest resolution. Therefore, it is very important to take into account the complex nature of the γ-ray spectrum of ^{227}Th in order to apply the γ-ray spectrometric technique to the determination of ^{227}Th. Today, ^{227}Th is often used as a radiochemical tracer for ^{231}Pa or ^{227}Ac in geochemical application. It should be mentioned that measurement by a few γ-rays, such as 50 keV, 236 keV, or 256 keV, can not be correct because of the complex nature of the decay chain and the γ-ray spectrum of ^{227}Th. The relative intensities of the observed γ-rays should be carefully examined to account for the radiochemical purity of the sample.

Gamma-ray Spectrum and Decay Scheme of ^{223}Fr. A γ-ray spectrum of ^{223}Fr is shown in Fig. 12. We observed 16 γ-rays which decayed with a half-life of ^{223}Fr in the energy region from 50 keV to 876 keV. Our observation is an additional proof of the characteristics of the γ-ray spectrum of ^{223}Fr, *i.e.*, the existence of two grounps of γ-rays, one in an energy region below 369 keV and the other in a region higher than 700 keV.[17,18] In Table 4, the relative intensities, abundances, and characteristics of the γ-rays are shown.

Among the works done previously, the work of Maria *et al.*[18] is the most comprehensive. However, we found some disagreements between the results. We found the relative intensity of the 50 keV γ-ray and the 235 keV γ-ray to be 13.9 whereas their value is only 10.[18] Another difference is in the point that they assigned the very weak γ-rays to ^{223}Fr. They used a large amount of ^{227}Ac precursor (740 MBq). Their experimental conditions are more suitable for the detection of weak γ-rays than our experimental conditions. Even so, an ambiguity remains with regard to the γ-rays of 256 keV and 286 keV, which were reported in Ref. 18. As has already been shown in Table 2, it is well known that the three γ-rays of 236 keV, 256 keV, and 286 keV are emitted from the same excited state of ^{223}Ra. Therefore, their result is erroneous in the point that they missed the 236 keV γ-ray, which is the most intense γ-ray among the three γ-rays. We did not observe any γ-rays between 240 keV and 289 keV which decayed with the half-life of ^{223}Fr. A possible explanation for these disagreements

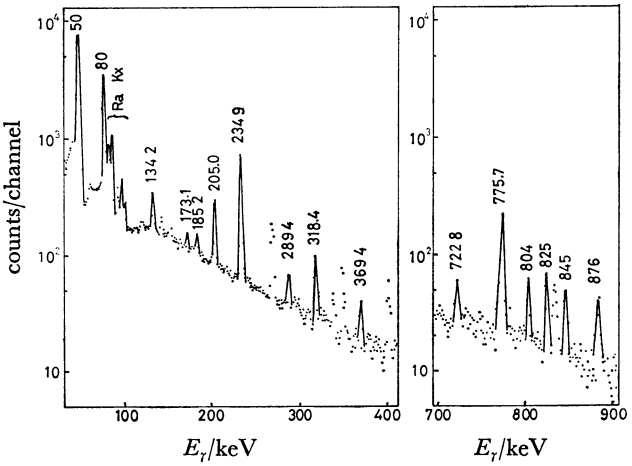

Fig. 12. Gamma-ray spectrum of ^{223}Fr.

TABLE 4. TABLE OF γ-RAYS OF ^{223}Fr

E_γ/keV	Transition	Relative intensity		Abundance (%)
		This work	Ref. 18	
50.0(2)[a]	50.2—0; El	13.9(5)	10	47(1)
80.0(3)	79.8—0; El	3.3(2)	2.4	10.9(5)
134.2(6)	369—235; El	0.17(3)	0.16	0.6(1)
173.1(6)	235—61.5; Ml, El	0.044(3)	0.04	0.15(2)
185.2(5)	235—50.2; El	0.075(6)	0.09	0.25(2)
205.0(5)	235—29.9; Ml, El	0.33(2)	0.34	1.1(1)
234.9(4)	235—0; Ml	1.00	1.00	3.4(1)
289.4(6)	369—79.8; Ml	0.08(1)	0.072	0.27(1)
318.4(5)	369—50.2; Ml	0.14(1)	0.162	0.47(2)
369.4(4)	369—0; El	0.041(4)	0.032	0.14(1)
722.8(6)	804—79.8	0.022(5)	0.015	0.07
775.7(7)	824—50.2	0.14(1)	0.123	0.46(3)
804(1)	804—0	0.025(5)	0.017	0.08(2)
825(1)	824—0	0.027(5)	0.014	0.09(2)
845(1)	908—61.5	0.018(7)	0.014	0.06(2)
876(1)	908—29.9	0.014(6)	0.013	0.05(2)

a) Numbers in parentheses are estimated standard deviations in units of the last significant digit.

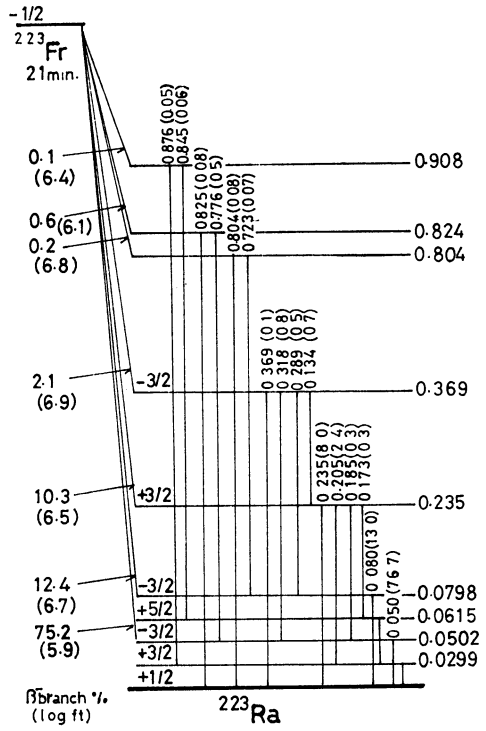

Fig. 13. Decay scheme of ^{223}Fr. Energies are given in MeV unit. Probabilities of γ-transitions per 100 β^- disintegrations are given in parentheses.

is as follows. As has been described before, the four γ-rays of 236 keV, 256 keV, 286 keV, and 50 keV are the main γ-rays of ^{227}Th. Moreover, the intensity of the overlapped γ-rays at 50 keV relative to the overlapped γ-rays at 236 keV is 0.65 for ^{227}Th. Therefore, it is reasonable to assume a trace contamination of ^{227}Th or ^{227}Ac in ^{223}Fr as an explanation of the low value of the relative intensity and of the exitence of the 256 keV and 286 keV γ-rays in the γ-ray spectrum of ^{223}Fr.

By investigations of the α-decay of ^{227}Th, we can utilize much information about the level structure of ^{223}Ra.[19] On this basis, we can conclude that all the γ-rays can be explained by the seven β^--transitions to the 50 keV (−3/2), 79.8 keV (+3/2), 235 keV (+3/2), 368 keV (+1/2), 804 keV, 824 keV, and 908 keV levels of ^{223}Ra. The decay scheme thus constructed is shown in Fig. 13. In this figure, the 29.9 keV (+3/2) and 61.5 keV (+5/2) levels are assumed to be the rotational excited states.[18,19] The classfications of the six γ-transitions from the 908 keV, 824 keV, and 804 keV levels are not known. However, the contribution of the conversion process to these γ-transitions can be neglected because of the high transition energies. For the other ten γ-transitions, the conversion coefficients were calculated by the use of the table of Hager and Seltzer.[20] Maria *et al.* also proposed a decay scheme.[18] Our modification involves the branching ratio of the most intense β^--transition to the 50 keV level.

As a conventional rapid method for the determination of non-γ-radioactive ^{227}Ac, γ-ray spectrometry of ^{223}Fr which is in radioequilibrium with ^{227}Ac is considered to be promising. Practically the methods can be classified into two types. One type is based on the measurement of the purified ^{223}Fr. The other type is based on the measurement of ^{223}Fr which is in the partial radioequilibrium with ^{227}Ac, when the other daughter nuclides has been removed from the sample. And then, the γ-ray spectrum of almost pure ^{223}Fr will be determined. In order to apply the former method, ^{223}Fr should be isolated from ^{227}Ac by a method which has a definite mean retention time for ^{223}Fr. Also, the mean retention time should be less than 1 min in the cow phase of ^{227}Ac to get more than 95% of ^{223}Fr. Therefore, the latter method seems to be more reliable. Of course, it is necessary to correct the contribution by ^{227}Th when the intensity of the sample is not large enough. However, the correction can be done with a high precision by referring to the relative intensities of the γ-rays of ^{227}Th tabulated in Table 2. Moreover, whether or not there is a necessity to correct can be easily desided when an abnormally low value of the relative intensity of the 50 keV γ-ray is measured.

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