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Collected Papers on Disintegration Studies of Short-lived Nuclides

Abstract

Accurate values for the half-life of Ca-49, Sc-49, Ti-51, Nb-92m, 97m, 97g, Rh-103m 105, 106, Ba-137m and Pa-234m were determined after the above nuclides were purified radiochemically.

Results are shown below together with literature values:

Nuclide	Half-life	
	Present value	Literature value
Ca-49	8.715±0.016 min	8.75±0.20 min
Sc-49	57.26 ±0.39 min	57.5 ±0.1 min
Ti-51	5.752±0.007 min	5.79±0.03 min
Y-89m	15.65 ±0.021 sec	16.2 ±0.1 sec
Nb-92m	10.09 ±0.02 day	10.15±0.03 day
Nb-97m	57.95 ±0.52 sec	60 ±8 sec
Nb-97g	73.74 ±0.04 min	74.0 ±0.2 min
Rh-103m	56.62 ±0.64 min	57.5 ±0.5 min
Rh-105	35.47 ±0.08 hr	35.88±0.02 hr
Rh-106	29.80 ±0.08 sec	30 sec
Ba-137m	2.527±0.003 min	2.60 ±0.05 min
Pa-234m	1.183±0.037 min	1.21 ±0.03 min

April, 1969

Ed. Tomitaro ISHIMORI
Kaoru UENO

短寿命核種の壊変に関する報文集

要　旨

放射化学的に精製した，Ca-49；Sc-49；Ti-51；Nb-92m, 97m, 97g；Rh-103m, 105, 106；Ba-137m および Pa-234m の半減期の精密測定をおこなった．

得られた結果は表のとおり．

１９６９年４月

編者　石　森　富　太　郎
上　野　　馨

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Introduction

Recent developments in radiochemical separation make possible to get better knowledges on radioactive disintegration processes. Especially rapid separation technique are sometimes indispensable for studying short-lived nuclides.

The radiochmistry laboratory, JAERI, is interrested in the exact measurements of half-lives for short-lived nuclides and studied them with assistance of several resident students sent from Tohoku University, Kanazawa University or Ibaraki General Vocational Training Center. Eight short papers along this line are compiled in this issue.

In the present studies, entirely new or somewhat modified methods were established for the purification of Ti from Sc, Ca from Sc, Y from Zr, Nb from Zr, Rh from Ru, Ba from Cs and Pa from Th. By using these methods, short half-lives of Ca-49, Sc-49, Ti-51, Y-89m, Nb-92m, 97m, 97g, Rh-103m, 105, 106, Ba-137m and Pa-234m were determined. The decay curves for simple A→B type disintegration were analyzed on an IBM-7044 computer, using Rogers' FRANTIC program [NYO-2303(1962)] in order to find values of half-life very accurately. For A→B→C type disintegration a careful graphical method was applied to analyze. Beta activity was measured with a proportional gas-flow counter while γ-activity with a scintillation counter. For short-lived nuclides, a 400 channel multiscaler was also used. Gamma-ray spectra were read by 256 channel pulse height analyzer and $3'' \times 3''$ NaI(Tl) scintillation crystal covered with plastic absorber.

English expressions in the present article were corrected by Dr. Hiroshi ONISHI, JAERI. The authors thank him sincerely.

1. Half-lives of Calcium-49 and Scandium-49

by M. SAEKI, K. UENO and T. ISHIMORI

There are many studies[1] in the decay scheme of Ca-49 but few in its half-life. Specially, there is no report in which radiochemical purification adopted before measurement.

In the present work, the decay of radioactivity due to radiocalcium purified with precipitation was carefully measured with a scintillation counter in order to find accurate values of half-life for Ca-49 and its daughter, Sc-49.

1. 1 Preparation and Identification of Ca-49 and Sc-49

About 50 mg of specpure calcium carbonate of natural isotopic composition obtained from Johnson Matthey & Co., Limited, was irradiated at the pneumatic tube ($\sim 3 \times 10^{11}$ n/cm^2/sec) of JRR-1 reactor for 10 min. The irradiated target was dissolved in conc. HCl and boiled. Then, 5 mg of iron (III) carrier was added, and the radioactive impurities were scavenged with ferric hydroxide. This step was repeated. Ammonium oxalate solution was added to the supernatant. Calcium oxalate formed was centrifuged and washed with dil. ammonium oxalate solution. The final precipitate was dissolved in conc. HCl and transferred into a 2m*l* test tube equipped with a stopper.

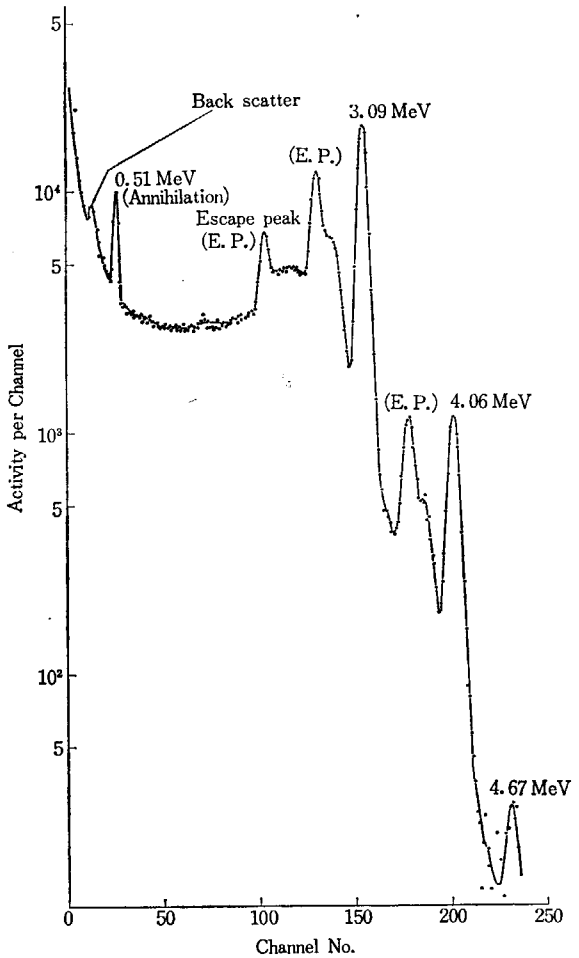

Fig. 1 γ ray spectrum of Ca-49

Fig. 1 shows the γ-ray spectrum obtained. There, 3 photo-peaks are seen at 4.67, 4.06 and 3.09 MeV. Any other peak is not found except for a 0.51 MeV annihilation peak. These values for peak energy are in excellent agreement with those given in the literatures[2)3)] as shown in TABLE 1. On these facts, it is shown that the activity studied should be assigned to those of Ca-49 and Sc-49, the daughter nuclide.

TABLE 1 Peak energy in MeV

Peak \ Reporter	ISHIMORI T. *et al.*	MARTIN D. W. *et al.*	O'KELLEY G. D. *et al.*
P_1	4.67	4.7	4.68
P_2	4.06	4.04	4.05
P_3	3.09	3.07	3.10
Standard	^{24}Na, ^{60}Co, ^{22}Na, ^{137}Cs	^{24}Na, Po-Be	^{24}Na, ^{88}Y, TnC"

1.2 Half-lives of Ca-49 and Sc-49

The decay of the activity was traced by counting with a NaI (Tl) scintillator (7/4"× 2") for about 1000 min. Fig. 2 shows one of the decay curves observed. It consisted of the three components, due to Ca-49, its daughter Sc-49 and the longer lived impurities.

Half-life of Sc-49 was analyzed by the conventional peeling method and least square method. Results obtained are summarized in TABLE 2 with the literature values[3)4)]. The averaged value presently obtained, 57.26 ± 0.39 min, agrees with the literature values.

TABLE 2 Half-life of Sc-49

Run No.	Half-life (min)	Stand. Dev. (min)
1	56.81	——
2	57.76	——
3	57.20	——
Average	57.26	0.39
Ref. 3	57.2	0.7
Ref. 4	57.5	0.1

Half-life of Ca-49 was obtained graphically. Further, using the same data, the calculation with the computer was carried out for fixed half-life of Sc-49, 57.26 min. The results obtained in both ways are summarized in TABLE 3 where the literature values[2)4)5)] are given for comparison. Values graphically obtained show a good agreement with those obtained with the computer but their mean value has a greater standard deviation. This is due to the adoption of weighted mean for the computer values. It is thought that the present value is more accurate than the old values.

The authors wish to express their sincere thanks for the experimental assistance given by Mrs. R. OKITSU.

TABLE 3 Half-life of Ca-49

	Run No.	Half-life (min)	Stand. Dev. (min)
Computer	1	8.68	0.037
	2	8.72	0.019
	3	8.74	0.046
	Average	8.71_5	0.016
Graphically	1	8.73	—
	2	8.65	—
	3	8.66	—
	Average	8.68_0	0.036
	Ref. 2)	8.9	0.2
	Ref. 3)	8.75	0.20
	Ref. 5)	8.5	—

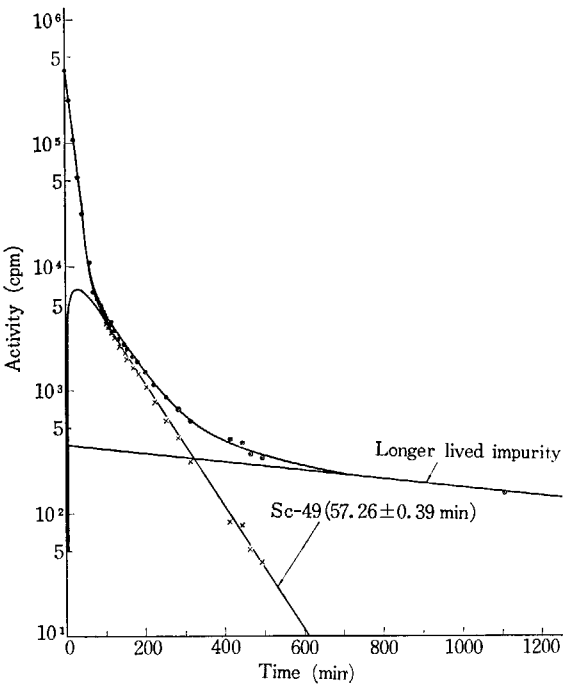

Fig. 2 Decay curve of Ca-49

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2. Half-life of Titanium-51

by T. ISHIMORI, K. UENO, M. HOSHI, M. SAEKI and K. AWA*

The half-life of Ti-51 was investigated by many workers[5]. Their data were obtained without any chemical separation. The purpose of the present study was to determine accurate half-life of Ti-51 after purification with simple solvent extraction techniques.

2. 1 Neutron Irradiation of Titanium Metal

Titanium metal, in which known impurity is manganese, was irradiated for 1 min with a neutron flux of 1×10^{13} n/cm^2/sec in JRR-2. In addition to Ti-51 as the main product, following radioactive nuclides were expected to be formed when irradiation condition, isotopic composition of titanium, impurities of the target and activation cross section were considered:

Sc-46 by (n, p) reaction of Ti-46
Sc-47 by (n, p) reaction of Ti-47
Sc-48 by (n, p) reaction of Ti-48
Sc-49 by (n, p) reaction of Ti-49
Sc-50 by (n, p) reaction of Ti-50
Mn-56 by (n, γ) reaction of Mn-55

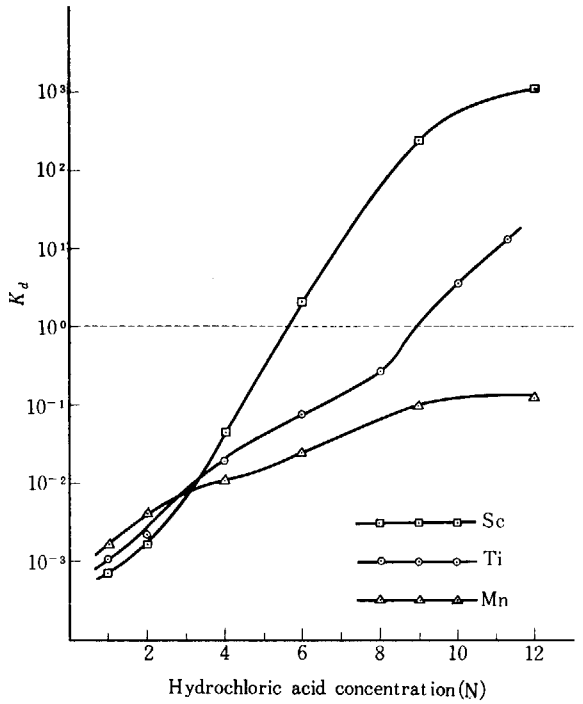

Fig. 1 Variation of K_d values for Ti, Sc and Mn with hydrochloric acid concentration[6]

2. 2 Purification of Ti-51

The solvent extraction behavior of Ti, Mn and Sc[6] in the system of 100% TBP vs. HCl is shown in

* Resident student for 1966 sent from Ibaraki General Vocational Training Center.

TABLE 1 γ-ray energy and its relative intensity of Ti-51.

Authors	Energy (MeV)	Relative intensity
ISHIMORI T. *et al.*	0.322	100
	0.62	1.3
	0.94	7.0
JORDAN W. C. *et al.*[7]	0.32	100
	0.61	1
	0.92	5
ROBINSON R. L. *et al.*[9]	0.319	100
	0.613	1.6
	0.934	5.8
BURKER M. E. *et al.*[8]	0.323	100
	0.605	1.46
	0.928	4.4

Fig. 1 as functions of the HCl concentration. Basing on these data, the following purification process of Ti-51 was adopted.

The irradiated target was dissolved quickly in warm conc. HCl. Then it was adjusted to 8N HCl solution by the addition of a few drops of water. From the resultant solution, scandium was extracted $4 \sim 5$ times with equal volumes of 50% TBP toluene solution. After that, the aqueous solution containing Ti-51 was made up to 10N HCl by the addition of a few m*l* of conc. HCl. Then the radioactive titanium was extracted with 100% TBP from the aqueous solution in order to minimize the contamination with radioactive manganese. The organic phase obtained was transferred into a test tube for γ-ray measurements.

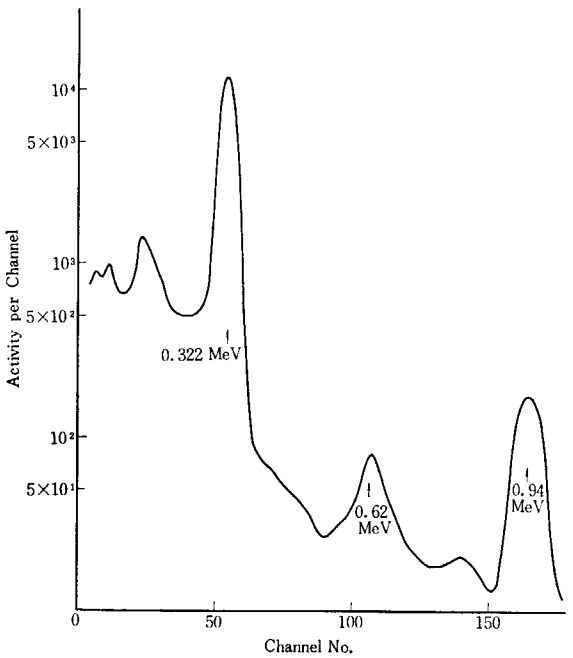

Fig. 2 γ-ray spectrum of Ti-51

2. 3 Identification of Ti-51

Gamma-ray spectrum of the sample purified in 2.2 was measured with a pulse-height analyzer for 3 min. From the spectrum obtained, the activities of background and long-lived components were subtracted. The γ-ray spectrum given in Fig. 2 shows that separate peaks at 0.322, 0.62 and 0.94 MeV. In view of fact that separation was complete, it is obvious that most of the activities of the sample was due to titanium and the present irradiation condition eliminated the possibility of production of other radioactive titanium nuclides except Ti-51.

Furthermore, the calculated relative intensities compared to the 0.3 MeV peak of the γ-ray spectrum are summarized in TABLE 1. All peak energies and relative intensities are in good agreement with results for Ti-51 reported by other investigators[7)~10)]. Therefore, it is concluded that the radionuclide studied is Ti-51.

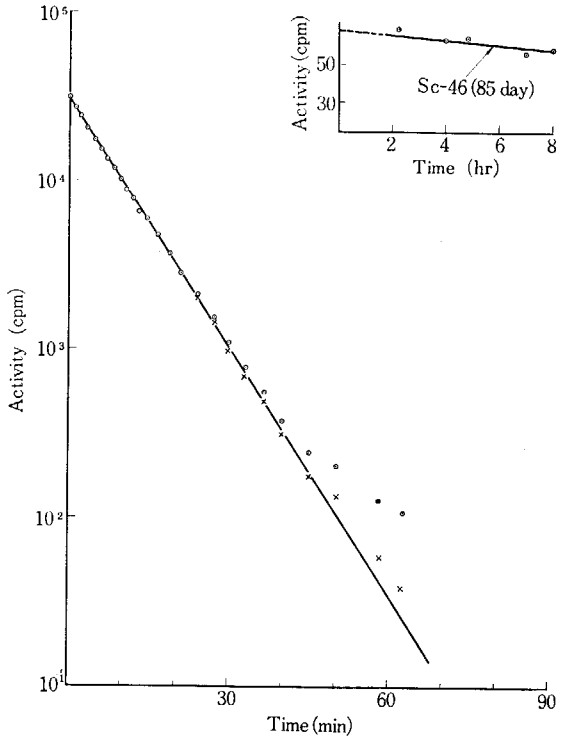

Fig. 3 Decay curve of Ti-51

TABLE 2 Half-life of Ti-51

Reported by		Half-life (min)	Formation
Present study	1	5.732 ± 0.009	
	2	5.820 ± 0.019	
	3	5.756 ± 0.021	
	4	5.800 ± 0.053	^{50}Ti (n, γ)
	5	5.821 ± 0.038	
	6	5.815 ± 0.081	
	Weighted mean	5.752 ± 0.007	
HAMMOND *et al.*[3)] (1953)		5.89 ± 0.2	^{48}Ca (α, n); ^{54}Cr (n, α)
YAFFE *et al.*[2)] (1953)		5.79 ± 0.03	^{50}Ti (n, γ)
BURKER *et al.*[5)] (1955)		5.80 ± 0.03	^{50}Ti (n, γ)

2. 4 Half-life Determination

The decay of the activity was traced with a scintillation counter. The counting rates were corrected for the dead time of 1.65×10^{-5} sec, and the activity of the background was subtracted. Decay data thus corrected were given in circles in Fig. 3. Here, the disintegration rate was followed from about 4×10^4 cpm down to about 60 cpm resulting from long-lived contaminants. The long-lived component was assigned to Sc-46 according to the decay curve given in the inset of Fig. 3.

The decay data after the correction for the contribution of long-lived Sc-46 were plotted in crosses in Fig. 3 and analyzed as a single component disintegration by the method of least squares using an IBM computer.

Half-life determinations were made on purified samples from different irradiations. The half-life values obtained are summarized in TABLE 2 together with those from the literature[1)-5)]. The weighted mean value, 5.752 ± 0.007 min, is lower than any other value reported previuosly.

The authors express their deepest gratitude to Mrs. R. OKITSU for her assistance given for the present study.

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3. The Disintegration of Yttrium-89m

by K. UENO, M. HOSHI, M. SAEKI,
T. KON* and T. ISHIMORI

Yttrium-89m is the daughter nuclide of Zr-89. As Zr-89 has a relatively long half-life, Y-89m can be milked from the Zr-89 cow, which is prepared by the ^{90}Zr (γ, n) reaction:

$$^{90}Zr(\gamma, n)^{89m}Zr \quad (4.4\,m \downarrow IT)$$

$$^{90}Zr(\gamma, n)^{89}Zr \xrightarrow[79hr]{\beta^-} {}^{89m}Y \xrightarrow{IT} {}^{89}Y \quad (\text{stable})$$

In the present study, Y-89m was stripped rapidly from tri-*n*-butyl phosphate solution of ^{89}Zr-^{89m}Y with 10N HCl, and served for the measurement of its half-life.

3. 1 Preparation of Zr-89 Cow Solution

Zirconium-89 was prepared by irradiating 60 mg of zirconium metal, reactor grade, natural isotopic composition, with 20 MeV Bremsstrahlung from the JAERI linear accelerator for 2 hrs. This length of irradiation is short enough to depress the formation of Zr-93 and -95 as the products of ^{94}Zr (γ, n) and ^{96}Zr (γ, n) reactions.

About 2 hrs later, the target was dissolved in a dilute HF. The solution was converted to 10N HCl solution. From the resultant solution radiozirconium was extracted with 2 m*l* of 100% TBP. The organic phase was then washed with equal volumes of 10N HCl. Thus, the cow solution free from radioyttrium which was formed as the product of Zr (γ, p) reaction, was prepared in a TBP solution.

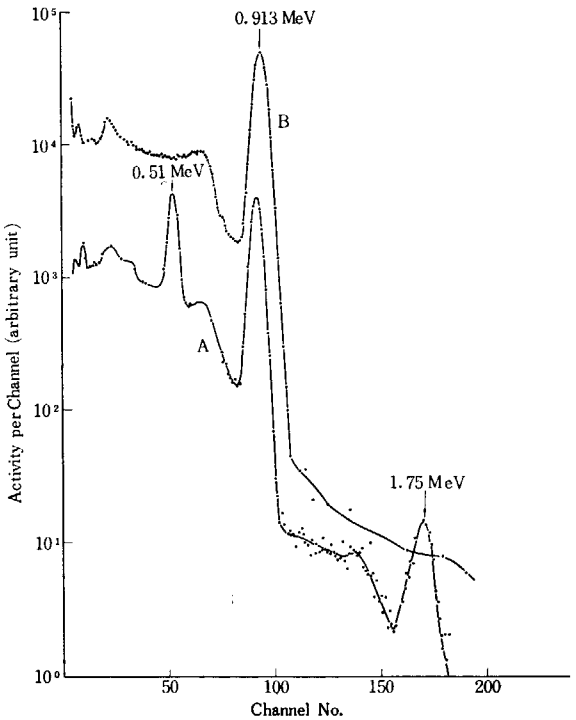

Fig. 1 γ-ray spectrum of Y-89m

* Resident student for Summer 1966, sent from Tohoku Univ.

The activity of the cow solution was measured for about 2 months. According to the decay curve obtained, which showed 78.6 hr component [2)3)] besides very small amounts of longer-lived impurities, the initial activity consisted of more than 99% of Zr-89 and about 1% of long-lived components.

As is seen in Fig. 1 (curve A), the γ-ray spectrum of the young cow showed a rather simple shape. Two peaks found at 0.51 and 1.75 MeV should be assigned to Zr-89. The other peak at 0.913 MeV should be assigned to Y-89m. Besides those, no peak was found.

Finally daughters of long-lived Zr-95 and -93 have much longer half-lives than that of Y-89m.

From these facts, it is clear that the cow gave very pure Y-89m according to the well scheduled procedure given below.

3. 2 Milking of Y-89m

The cow solution was set aside for 1 min in order to equilibrate the daughter nuclide, Y-89m, with Zr-89.

Yttrium-89m was milked with 2 m*l* of 10N HCl from the organic cow. The aqueous phase containing radioyttrium was rapidly collected in a test tube for γ-ray measurements.

3. 3 Identification of Y-89m

The γ-ray spectrum of Y-89m was measured in the following way: Yttrium-89m was separated according to the procedure given in 3.2. Its γ-ray was measured with a pulse-height analyzer for 4 min. From the spectrum obtained, background and activities of long-lived contaminants were subtracted for 4 min. This procedure was repeated 10 times with freshly prepared Y-89m and each spectrum was accumulated. Curve B of Fig. 1 shows the γ-ray spectrum in which one peak is observed at 0.913 MeV. This value agrees excellently with the results of other authors[1)3)4)].

TABLE 1 Values for half-life of Y-89m

Half-life (sec)	Reported by
15.62 ± 0.097	Present authors
15.61 ± 0.043	〃
15.68 ± 0.028	〃
15.63 ± 0.060	〃
15.65 ± 0.021 (Weighted mean)	〃
16 ± 3	GOLDHABER M. *et al.* (1953)
16.1 ± 0.2	SWANN C. P. *et al.* (1955)
16.3 ± 1.3	BRAMLITT E. *et al.* (1964)
16.2 ± 0.1	ARINO H. *et al.* (1964)

3. 4 Half-life of Y-89m

The decay of the Y-89m sample was measured with a 400 channel multiscaler. At the beginning the activity was measured every 1 sec. After Y-89m decayed off, the counting was continued in order to identify the long-lived components.

As is seen in the inset of Fig. 2, the long-lived component showed half-life of about 80 hrs. Further, it gave 1.75 MeV peak in its γ-ray spectrum. Therefore, the long-lived component was assigned to Zr-89.

Counted data were corrected for dead time and length of counting[5]. From there values shown as circles, in Fig. 2 the contribution of long-lived component together with the background were subtracted. The final values, shown as crosses in Fig. 2, were fed to the IBM computer and treated as values of a simple A→B type disintegration.

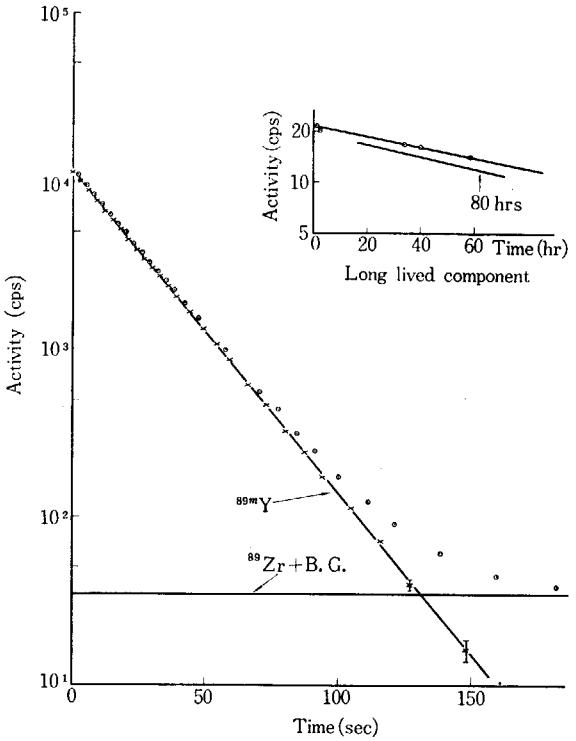

Fig. 2 Decay curve of Y-89m

The resulted values for the half-life are shown in Table 1 together with their weighted mean. Further, values taken from the literature[6)-9)] are also given in the same table in order to compare to the present value.

It is found that the present value for the half-life is a few percent shorter than those of other authors. Besides that the present value for the standard deviation is also smaller than those of the former workers.

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4. Half-life of Niobium-92m

by H. SUGIURA*, M. SAEKI** and T. ISHIMORI

Niobium-92m was prepared by ^{93}Nb (γ, n) reaction and identified with its γ-spectrum after being purified according to the method given by KIMURA K. *et el.*[1] The decay of the activity was followed very carefully for about 120 days. Basing on the decay curves obtained, the half-life of Nb-92m was determined. Short-lived isomers were not found.

4. 1 Preparation of Nb-92m

Niobium pentaoxide of "Specpure" grade obtained from Johnson Matthey & Co. Limited, was taken as the target material. This lot contained following impurities:

Ta	K	Fe, Si	Na, Ca	Cu, Mg, Al
1000	400	5	2	1 ppm

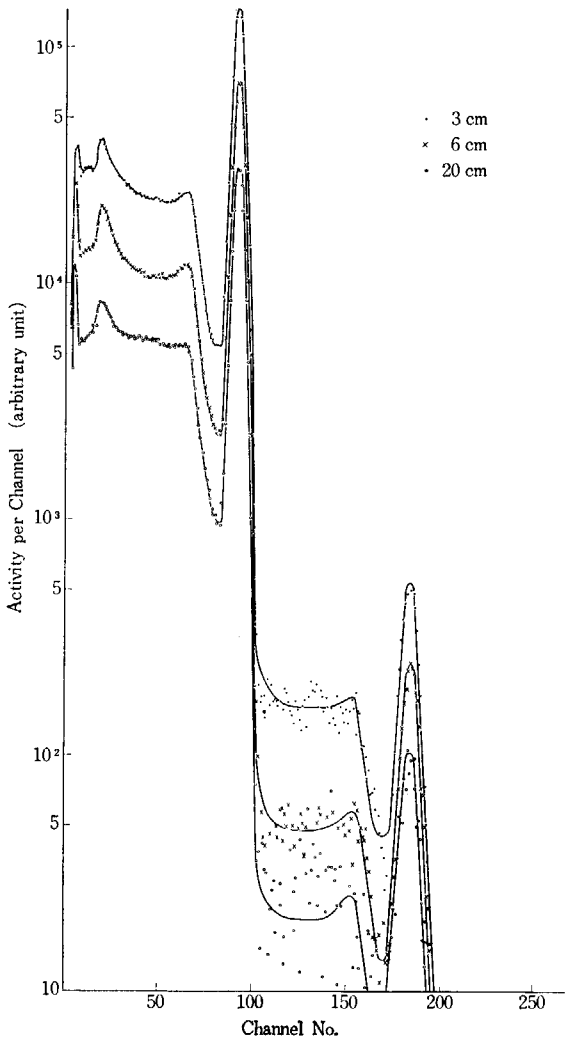

Fig. 1 γ-ray spectra of Nb-92m distance from the scintillator to the sample

* Resident student for Summer 1967, sent from Kanazawa University. Responsible only for the experimental part.

** Responsible only for the evaluation of results obtained.

The target material of 97.30 mg was wrapped with an aluminum foil and irradiated with 20 MeV Bremsstrahlung of the JAERI LINIAC for 2 hours. The irradiated target was treated with a mixture of hydrofloric and hydrochloric acids on a water bath. The solution was evaporated to dryness the residue was taken up with a mixture of the acids. Finally the solution was adjusted to 3N HF + 6N HCl. The resultant solution was treated according to the procedure given in 5.1 in order to obtain pure niobium in an organic solution.

4. 2 Identification of Nb-92m

The γ-spectra of the activity obtained are shown in Fig. 1. They were measured by putting the samples at different distances from the scintillator. The shape of the curves do not change among these, showing that peaks observed are photopeaks. As shown in TABLE 1, these peaks correspond well[2)3)], to those of 10 days "Nb-92", both in the peak-energy and the relative intensity. In the present work, the peak at 0.904 MeV found by BUNKER *et al.* was difficult to be idntified because of its low intensity.

Thus, it is proved that the nuclide in the present study work should be assigned to that studied by BUNKER *et al*[2)]. or WEST *et al.*[3)]. Although they called the nuclide Nb-92, recent studies[4)5)] show that this nuclide should be assigned to Nb-92m basing on its probable spin.

4. 3 Half-lite of Nb-92m

The decay of the activity prepared according to the procedure given at 4.1 was traced for about 120 days. Special attention was paid for finding short-lived isomers. However, as shown in Fig. 2, measured values fall on a straight line in the log (activity) vs. time plane. Therefore, it is concluded that the 13 hr-

TABLE 1 Nuclear proporty of Nb-92 isomers

Authors	Formation reaction	γ-ray energy (MeV)	Half-life (day)	Remarks
Present authors	^{93}Nb (γ, n)^{92m}Nb	0.935 1.79	10.09±0.02	no 13hr isomer <1hr or>350y ?
BUNKER *et al.*	^{89}Y (α, n)^{92}Nb	1.83 ±0.10 (0.90) 0.932±0.008(100) 0.904±0.008(1.92)	10.16±0.03	no 13hr isomer <1hr or>350y ?
WEST *at el.*	^{93}Nb (d, p2n)^{92}Nb	0.934(97.4) 1.82 (2.6)	10.15±0.03	β^+; (5.6±0.6) ×10^{-2} %
MOCK *et al*	^{93}Nb (γ, n)^{92}Nb	—	9.8±0.7	—
JAMES	^{93}Nb(p, pn)^{92}Nb	0.933 1.84	—	13±2hr isomer (E.C.; 2.35MeVγ)
WIEDENBECK	^{93}Nb(α, ^{3}H)^{92}Nb	—	—	21.6 hr
SHELINE *et al.*	^{93}Nb(d, t)^{92}Nb	—	—	10.1d(2^+)meta stable ground state~10^5y
SWEET *et al.*	^{93}Nb(p, d)^{92}Nb	—	—	10.1d(2^+)meta stable

isomer of JAMES[6], was not found in the present study. On the other hand, as it took several hours from the end of the irradiation to the beginning of the measurement, the possible isomer of less than 1 hour is not denied in the present experiments.

Further, the long-lived isomer which should be assigned to the ground state is not seen in Fig. 2, as the decay curve goes down linearly to very low activity. For studying this isomer, much longer irradiation is desired.

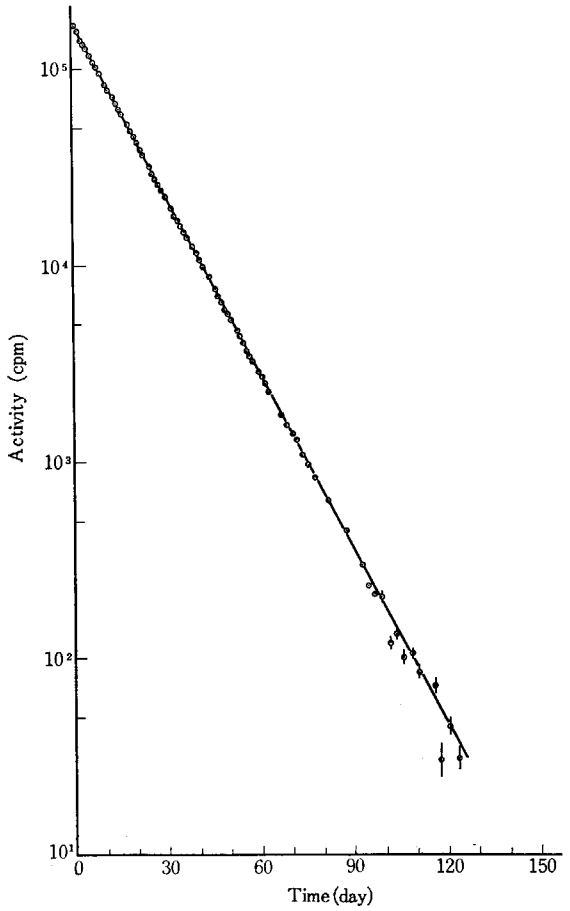

Fig. 2 Decay curue of Nb-92m

TABLE 2 Half-life of Nb-92m

Authors	Half-life	Stand. Dev.
Present authors 1	10.09 (day)	—— (day)
〃 2	10.09	——
〃 3	10.13	——
〃 4	10.06	——
〃 mean	10.09	±0.02
BUNKER *et al.*	10.16	±0.03
WEST *et al.*	10.15	±0.03
MOCK *et al.*	9.8	±0.7

Four runs of the measurement were carried out. The decay curves obtained always showed that of a typical one-component disintegration as is shown in Fig. 2. From the slope, the half-life was calculated gragphically. Results are shown in TABLE 2 together with those reported by former investigators[2)3)7)].

References

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5. The Disintegration of Niobium -97m and 97g

by F. HAGIYA*, K. KIMURA and T. ISHIMORI

5. 1 Preparation of Nb-97m, 97g

In this study, carrier-free niobium saparation was achieved with tributyl-phosphate as an extractant from HCl-HF mixture solution.

Results obtained in preliminary experiment indicate that, although *Kd* values both for niobium and zirconium decrease by the addition of HF to HCl solution of the elements, those only for niobium shows increasing *Kd* values with further increase of HF concentration. As seen from Fig. 1, carrier-free separation of niobium would be possible by extraction with tributyl phosphate from 6M solution containing more than 3M HF.

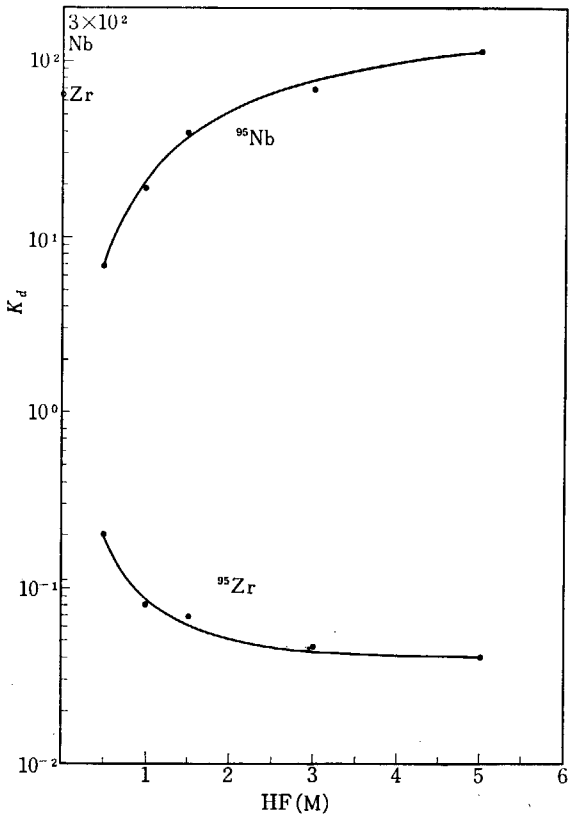

Fig. 1 Distribution ratios of Zr-95 and Nb-95 between 100% tri-butyl phospahte and 6M HCl—xM HF.

In fact, the separation of niobium from zirconium was successfully demonstrated in case of the Zr-Nb-95 pair under the above conditions, and further applied to the case of Zr-Nb-97 (m and g) mixtures. Fig. 2 shows a recommended procedure. It takes less than 3 min for single extraction step followed by single scrubbing.

Niobium was also separated from the fission products by a combination of the separation method of zirconium and niobium by ISHIMORI *et al.*[1] and this method.

* Resident student for 1965 sent from Ibaraki General Vocational Training Center.

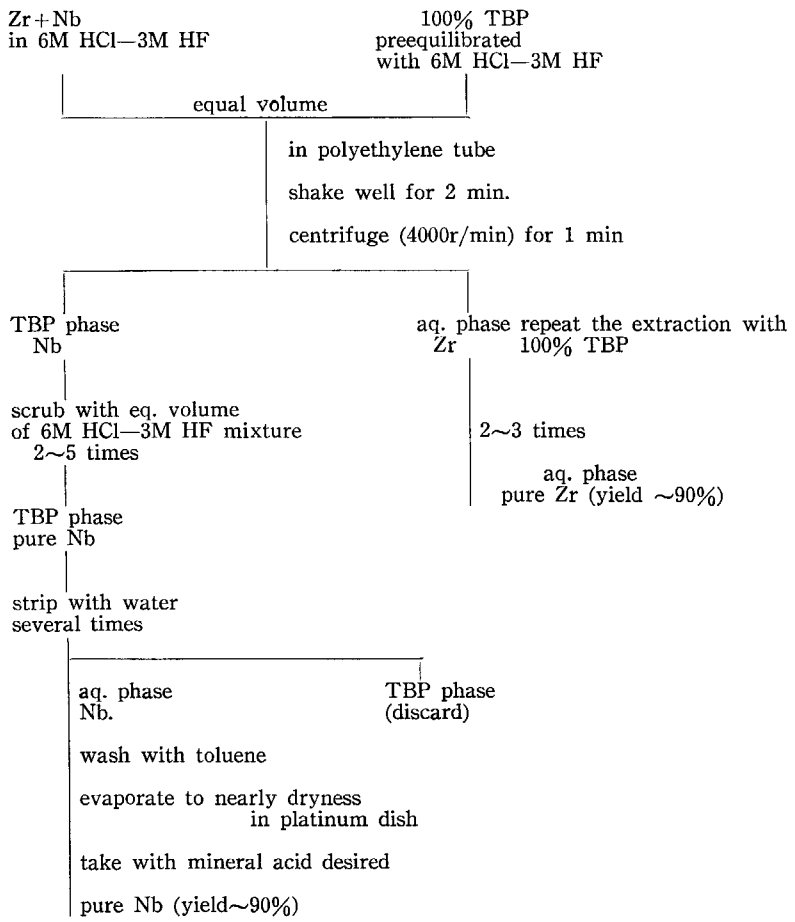

Fig. 2 Separation scheme of niobium from zirconium

5. 2 Experimental Notes

All chemical reagents and target used were of analytical grade.

Zr-Nb-97 samples were obtained by an irradiation of 100mg of $ZrO(NO_3)_2 \cdot 2H_2O$ in JRR-2 with neutron flux of 1×10^{13} n/cm^2/sec for 20 min. After dissolution of the zirconyl salt, it was made up to 3M HF-6M HCl solution. Up to 20 mg/m*l* of zirconium concentration did not affect the procedure.

A scintillation counter with well-type NaI crystal was used for γ-ray measurements. For the half-life measurements of Nb-97m, a 400 channel multiscaler unit was also used with an external timing pulse unit. Gamma-ray spectra were read by the use of a 256 channel pulse height analyser made by JAERI Electronics Shop with 3″×3″ NaI (Tl) crystal attached with plastic β-ray absorber.

Decay curves measured were analyzed by an electronic computer IBM-7044 with FRANTIC-7044 cord after ROGERS[2], and a very accurate half-life for each nuclide was obtained.

5. 3 Identification of Nb-97g

Niobium was milked from the irradiated zirconium sample by the above-mentioned procedure. The γ-ray spectra of niobium sample prepared showed only one photopeak at 0.665 MeV by 10 min accumulation after very short-lived nuclide decayed off. This result agrees very well with the results of other authors[3)~7)] for Nb-97g. Therefore γ-activity of the sample was assigned to that of Nb-97.

5. 4 Half-life of Nb-97g

Fig. 3 shows one of the decay curves obtained. Very low counting rate of long-lived component was found to be due to 35 day Nb-95 by the analysis of decay curve and the γ-ray spectrum obtained by 60

min accumulation. **Fig. 4** shows the decay curve of shorter half-lived component, i. e. Nb-97g, after subtraction of the long-lived component. Dead time of the counting system was estimated to be 2.75×10^{-8} min by two-halves source method and by the least square analysis of counting loss at higher counting

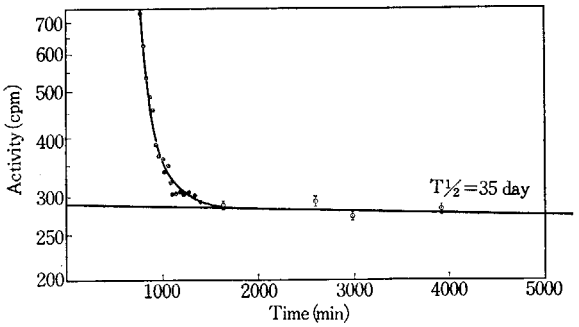

Fig. 3 A part of decay curve of niobium fraction

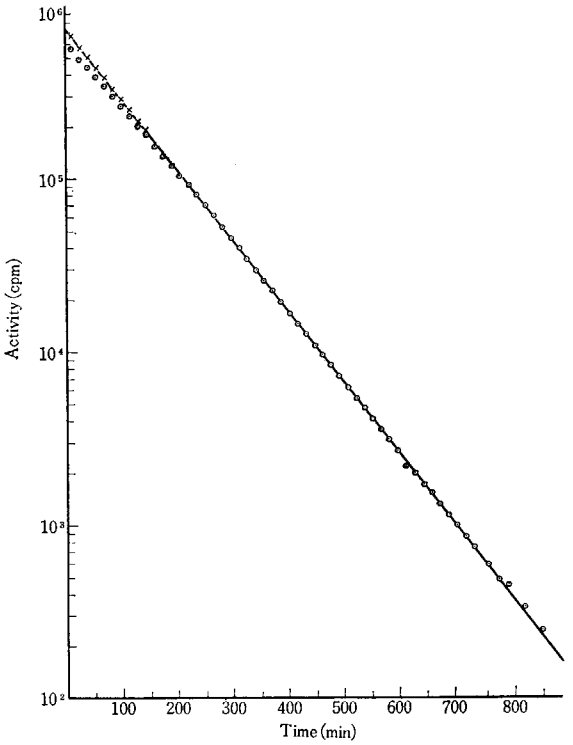

Fig. 4 Decay curve of Nb-97g

rate by IBM computer, and the correction for dead time was made.

Analysis of three experiments gave the values in TABLE 1 for the half-life of Nb-97g.

TABLE 1 Half-life of Nb-97g

Exp. No.	Half-life of Nb-97g
I	73.7047 ± 0.0908 min
II	73.8882 ± 0.0585
III	73.4373 ± 0.0903
Weighted mean	73.74 ± 0.04

The mean value is to be compared with previously reported values of 72.1[3], 74[4], 75[5], and 76 min[6].

5. 5 Identification of Nb-97m

The procedure adopted for the milking of Nb-97m was the same as the recommended procedure except for one washing of the organic phase. Only one photopeak was observed at 0.75 MeV on γ-ray spectrum, which decayed very quickly into 0.665 MeV peak of the first excited level of Mo-97. The 0.665 MeV peak first grew slightly and then decayed with the half-life of Nb-97g. All these facts gave positive evidences of that the short-lived nuclide in the sample was Nb-97m, and all these facts were also in good agreement with observations on Nb-97m by BURGUS *et al.*[4] Therefore the γ-activity with a short half-life in the sample was assigned to the activity due to Nb-97m.

5. 6 Half-life of Nb-97m

Fig. 5 illustrates the experimental steps, together with the time relations. By the first milking, Nb-95, -97m, and -97g that had been produced were removed. After a few minutes of equilibration of Nb-97m, the second milking was carried out at time d. The organic phase washed once was transferred into a sample tube and the γ-ray counting was immediately followed.

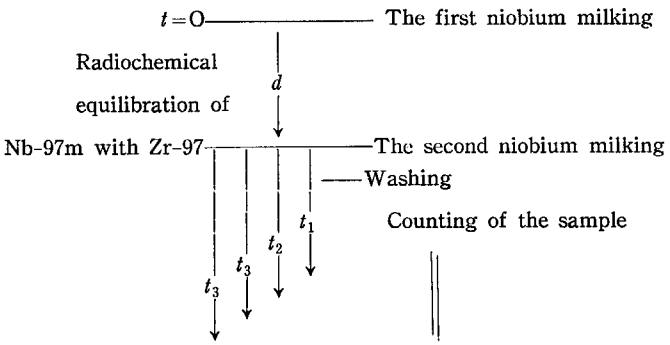

Fig. 5 The experimental steps with time relations.

A decay curve thus obtained was shown semi-logarithmically in **Fig. 6** after small corrections for the long-lived components and background. As seen in the figure, the counted values fell on the straight line corresponding to the half-life of Nb-97g after 12 min from the first milking.

Analysis of the curve was done graphically as follows: Total radioactivities measured, A, consisted of Nb-97m, A_2, and of Nb-97g, A_3;

$$A=A_2+A_3$$

Further A_3 was composed of the growth from Nb-97m after the second milking, A_{3G}, and the decay of Nb-97, A_{3D}, which had grown during the time between the first and second milkings, d, i. e.,

$$A_3=A_{3D}+A_{3G}$$

The terms in the right side, at time t from the second milking, can be written in the well-known forms,

$$A_{3D}=A_{3,d}e^{-\lambda_3 t}$$

and $$A_{3G}=\frac{\lambda_3}{\lambda_3-\lambda_2}A_{2,d}(e^{-\lambda_3 t}-e^{-\lambda_2 t}),$$

in which

$$A_{2d}=\frac{\lambda_2}{\lambda_2-\lambda_1}A_{1,0}(e^{-\lambda_1 d}-e^{-\lambda_2 d})$$

and $$A_{3,d}=\lambda_2\lambda_3 A_{1,0}\left(\frac{e^{-\lambda_1 d}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)}+\frac{e^{-\lambda_2 d}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)}+\frac{e^{-\lambda_3 d}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right),$$

where the subscripts 1, 2, and 3 denote Zr-97, Nb-97m, and Nb-97, respectively; and 0 and d represent the time of the first and second milking respectively. The radioactivity of Nb-97, A_3, is then shown as

$$A_3=\frac{A_{1,0}\lambda_2\lambda_3}{(\lambda_1-\lambda_2)(\lambda_2-\lambda_3)(\lambda_3-\lambda_1)}[(\lambda_3-\lambda_1)e^{-\lambda_2(t+d)}(e^{-(\lambda_1-\lambda_3)d}-1)-(\lambda_1-\lambda_2)e^{-\lambda_3(t+d)}(1-e^{(\lambda_3-\lambda_1)d})]$$

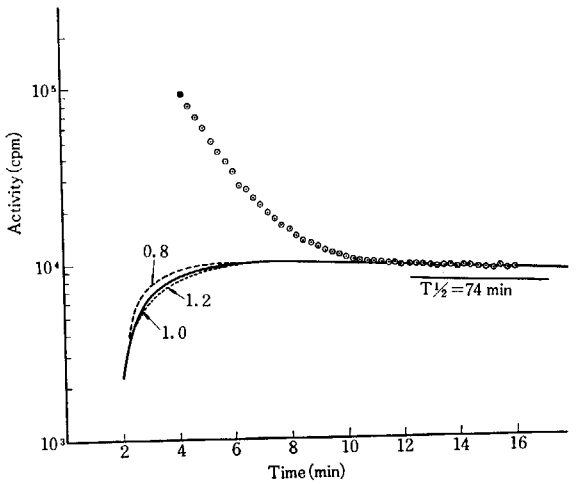

Fig. 6 Decay curve of Nb-97m+97g sample and calculated growth curves of Nb-97

The multiplier of the right side out of the brackets is only a factor which does not affect the shape of the decay-growth curve determined by the multiplicand in the brackets, A'_3. The ratio of the counting efficiencies of Nb-97 to Zr-97 is also not a shape-determining factor. Therefore

$$A_3=K(A'_3).$$

In this case, λ_1 and λ_3 are known (for λ_3, the value found in the previous section of the part is chosen) and d is fixed experimentally, and hence, a set of values for λ_2, a set of the corresponding curves of A_3 are obtainable by varying t.

Since the observed curves show the straight line corresponded to the decay of Nb-97 in the region of $t<(T)_2$, that in the half-life of the second nuclide, Nb-97m, one can easily fit the set of curves of A'_3 to the experimentally observed curve at the range of $t>(T)_2$, as shown in Fig. 6. A'_3 curves of Nb-97g seen in Fig. 6 were calculated by assuming $(T)_2=0.8$, 1.0, or 1.2 min, and substituting the experimental value of d, 2.5 min.

The decay curve of Nb-97m is then obtained by subtracting the Nb-97 component from the observed values at each time of counting, t. These curves of Nb-97m are almost straight lines, giving slightly different values of "apparent" half-life.

At the "true" value of half-life, the ratio of "apparent" half-life to the assumed half-life must be unity. The ratios obtained were plotted against the values of the assumed half-life, and the true value of the half-life was easily read out as about 58 sec.

In this manner, the following values are obtained for the half-life of Nb-97m:

$(T)_2=58.25$ sec
57.39
58.20
58.50
57.40 Av. 57.95 ± 0.52 sec.

Only one value, 60 ± 8 sec[4], has been found in the literature.

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6. Half-lives of Rhodium-106, -105 and -103m

by Y. KOBAYASHI

According to the nuclear data, Ru-105 ($t_{1/2}$=45hr), Ru-103 ($t_{1/2}$=40d) and Ru-97 ($t_{1/2}$=2.9d) are produced by the (n, γ) reactions of natural ruthenium. They disintegrate into Rh-105, Rh-103m and Tc-97, respectively. At a given cooling time of the irradiated ruthenium, predominant Rh-105 or -103m is resulted by the radiochemical separation of ruthenium and rhodium whereas Tc-99 is practically negligible for its long half-life (10^6y). Rhodium-106 exists in fission products as the disintegration product of Ru-106 ($t_{1/2}$=1y).

6. 1 Preparation

Commercial ruthenium metal is often contaminated with iridium. Therefore it was purified in advance according to a similar procedure to that given below. In the present study, Rh-105 or -103m was separated from the purified ruthenium target which was irradiated for 20 min with a neutron flux of 1×10^{13} n/cm^2/sec in JRR-2 and cooled for one day or one month. Rhodium-106 was prepared from a commercial Ru-106.

The separation of radiorhodium from the irradiated ruthenium target or Ru-106 source was carried out according to the procedure previously reported[1]:

Irradiated target or Ru-106 source with the carrier was dissolved in a sodium hypochlorite solution. The solution was acidified with dil. H_2SO_4 and ruthenium tetroxide produced was removed completely from the aqueous solution by extracting it several times with an equal portions of CCl_4. Radiorhodium left in the aqueous solution was precipitated with ferric hydroxide. Finally ferric hydroxide containing radiorhodium was dissolved in 9M HCl and ferric chloride was extracted with isopropyl ether. Radiorhodium was left in the aqueous solution, which was then evaporated to dryness on a platinum disc.

For Rh-106 the step of coprecipitation with ferric hydroxide was omitted because of the short half-life.

6. 2 Measurements of Radioactivities

Rhodium-106, -105 or -103m was identified by the measurement of γ-ray spectrum of the nuclide or its parents. Rhodium-105 was counted with a gas-flow proportional counter, whereas Rh-103m with a 0.1″ ×1″ NaI(Tl) scintillation counter. For Rh-106, a well-type NaI(Tl) scintillation counter with a 400 channel multiscaler was used. The scaler was set one channel per second.

6. 3 Results

The analyses of the decay curves were carried out with an IBM computer using Rogers' FRANTIC programme. In the calculation of the half-lives, values for the half-lives of Ru-106 and Ru-103 were assumed to be 1.00y and 40.0d, respectively. The values of the half-lives obtained are given in TABLE 1 with the literature values.

TABLE 1 Half-lives of Rn-106, -105 and -103m

Nuclide	Half-life		Unit
	New value	Literature	
Rh-106	29.80 ± 0.08	30.36±0.15[2], 30[3], 40[4]	sec
Rh-105	35.47 ± 0.08	35.30±0.05[5], 35±1[6]. 35.88±0.02[7], 35.4±0.1[8]	hr
Rh-103m	56.6 ± 0.6	57.5±0.5[9], 56[10] 52 ±2[1], 45[2]	min

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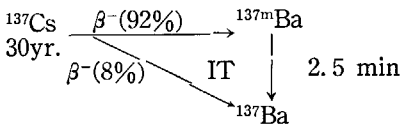
7. The Disintegration of Barium-137m

by K. KIMURA

7. 1 Cesium-137 Cow Solution

Cesium-137 source was imported from the Oak Ridge National Laboratory, Tenn., USA, and used without further purification.

Cesium-137 decays in the following way:



Accordingly about 20min standing of the cow solution was enough to attain the radiochemical equilibrium between Cs-137 and Ba-137m.

7. 2 Milking of Ba-137m

Carrier-free separation of Ba-137m from Cs-137 was carried out by using zirconium phosphate* as inorganic exchanger. Cesium was adsorbed on the phosphate column from 2N HNO_3 solution, whereas barium passed through.

Cs(Ba)-137 solution was fed onto a small Ionite C column (0.2 cm^2 × 3 cm; 150~200 mesh). The column

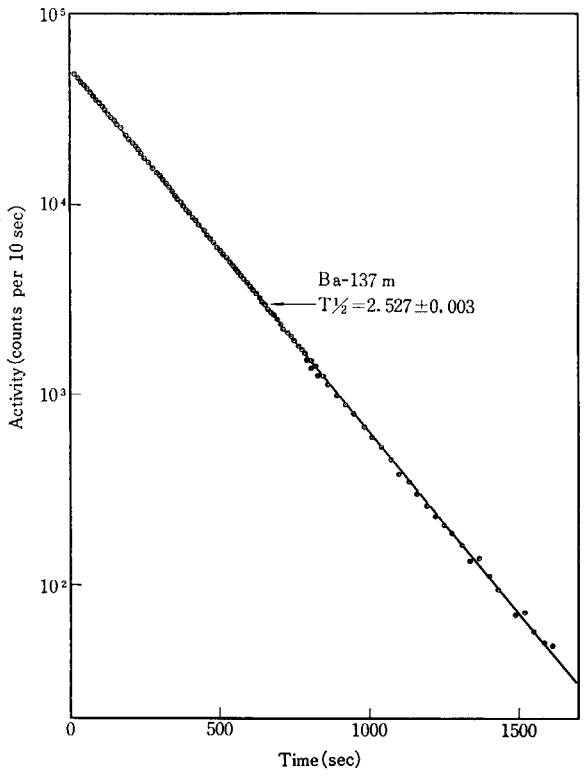


Fig. 1 Decay curve of Ba-137m

* Trade name: Ionite C, manufactured by Shin Nippon Kinzoku Kagaku Co, Ltd.

was washed with a small portion of 2N HNO_3 and allowed to set aside for about 10 min to equilibrate the daughter nuclide. Then a milking of Ba-137m was achieved by washing the column with 2N HNO_3. Suitable drops of Ba-137m were collected for γ-ray measurement. The time required for the elution was less than 10 min

7. 3 Half-life of Ba-137m

γ-spectrum of the sample showed only one photo peak at 0.66 MeV, that is well-known as an exited level of Ba-137. No other peak was detected even a long accumlation after Ba-137m decayed out. Therefore, the Ba-137m sample obtained was pure enough for measurement of its half-life, except a very small amount of Cs-137 contaminated.

Decay curve of the sample was measured by a 400 channel multiscaler unit with an external timing unit. Fig. 1 shows one of the decay curves measured after small corrections for the contamination of Cs-137, dead time of the counter used, and background count. Then data were analyzed by an IBM computer.

Experimental values of half-life are given in TABLE 1. The mean value is slightly shorter than value, 2.554±0.003 min, by MERRITT and TAYLOR.

TABLE 1 Half-life of Ba-137m

Exp. No.	Half-life
I	151.50 ± 0.320 sec
II	151.67 ± 0.229 sec
Weighted mean	151.613± 0.186 sec 2.527± 0.003 min

The author is very grateful to Mrs. R. OKITSU for her invaluable assistance during this work.

8. Half-life of Protactinium-234m

by M. SAEKI*, K. KIMURA and T. ISHIMORI

8. 1 Preparation of Th-234 Cow Solution

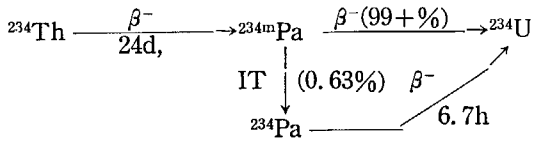
During a trial operation of the pilot plant for fuel-reprocessing, JAERI, by using about 200 kg of natu ral uranium, most of Th-234, the decay product of U-238, was separated from the bulk of uranium and concentrated into the aqueous waste liquor of the first extraction column of the plant. There, the reprocessing was going to be carried out by a modified Purex method.

Thorium-234 in the aqueous waste was purified both by coprecipitation with lanthanum hydroxide and by the solvent extraction with undiluted tri-*n*-butyl phosphate. Finally Th-234 was made up to 4 m*l* of 7N HCl solution and stocked as the cow solution for the milking of Pa-234m.

The cow solution thus prepared gave the γ-ray spectrum where 0.03, 0.06 and 0.09 MeV peaks of Th-234 and 0.77 and 1.02 MeV peaks of Pa-234m were found.

8. 2 Milking of Pa-234m

Thorium-234 disintegrates in the following way:



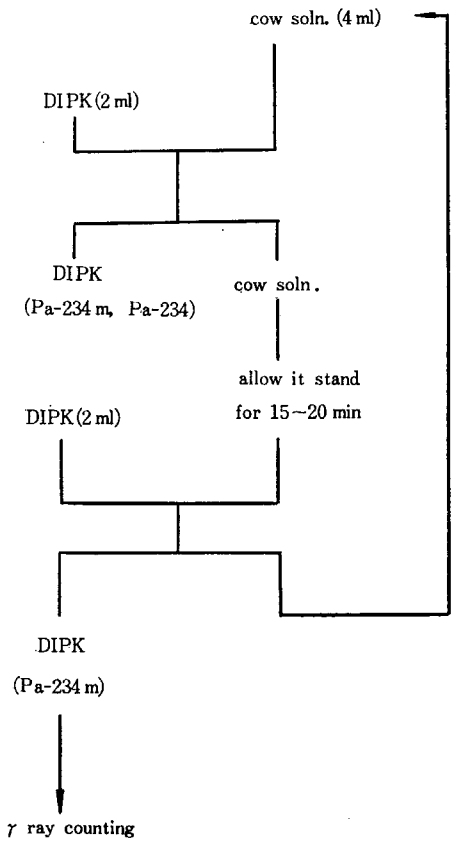


Fig. 1 Milking of Pa-234m

* Resident student for summer, 1965, sent from Kanazawa Univ.

Accordingly about 15~20 min standing of the cow solution was enough to make the radiochemical equilibrium between Th-234 and Pa-234m established. In order to minimize the contamination with Pa-234, it was necessary to separate protactinium from the cow in advance, and then, after standing the cow for 15 ~20 min, Pa-234m formed in the cow solution was isolated by the di-iso-propyl ketone (DIPK) extraction.[1)]

Two m*l* of DIPK was added to the cow solution and shaken for 30 sec vigorously. The organic phase was separated from the aqueous phase with the aid of a centrifuge at 3000 rpm for 10 sec. The organic phase was transferred to a small test tube for the measurement of radioactivity. The time required for this milking procedure is only 1.5~2 min including the 10 sec centrifugation step. The procedure is shown schematically in Fig. 1

8. 3. Identification of Pa-234m

The gamma-ray spectrum of Pa-234m prepared was measured by putting the sample for counting 3cm from the scintillator. According to the conventional peeling-off method, the gamma-ray spectrum obtained was analyzed. The results are shown in Fig. 2. TABLE 1 shows both energies and relative intensities of

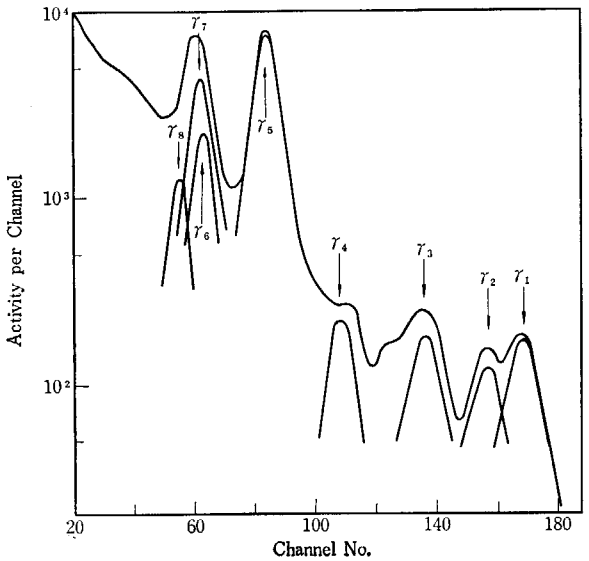


Fig. 2 γ-ray spectrum of Pa-234m

TABLE 1 Photo peaks of Pa-234m

Peak	E (Mev)	Relative Int.
γ_1	1.87	5.9
γ_2	1.76	3.2*
γ_3	1.55	3.4**
γ_4	1.26	4.1
γ_5	1.02	100.0
γ_6	0.78	18.9
γ_7	0.77	34.9
γ_8	0.69	8.7

* Corrected for Chance Coincidence (γ_7 and γ_5)
** Corrected for Chance Coincidence (γ_6 and γ_7)

the photopeaks observed in Fig. 2 after the corrections both for the chance coincidence and for the energy

dependence of the counting efficiency. Here, the relative intensities were calculated taking the intensity of the 1.02 MeV photopeak as unity.

These results are in good agreement with those of HOK *et al.*[2] and Cross[3]. It was, therefore, ascertained that the gamma-activity of the sample was assigned to that of Pa-234m.

8. 4 Half-life of Pa-234m

According to the prescription given in 8. 2 the sample solution of Pa-234m was prepared. The half-life of Pa-234m was measured with a NaI(Tl) scintillation counter for 6 sec at every 12~20 sec.

After the activity of Pa-234m decayed off, the counting was lasted pretty long in order to find the long-lived impurity.

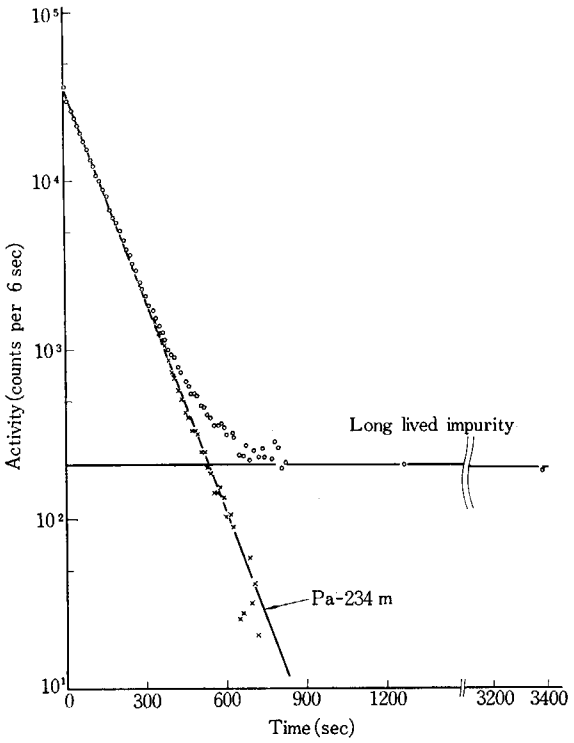


Fig. 3 Decay curve of milked solution

TABLE 2 Half-ilfe of Pa-234m

Measured by	Half life (min)	Standard deviation (min)
The present authors 1	1.176	±0.048
〃 2	1.180	±0.098
〃 3	1.240	±0.185
〃 4	1.193	±0.073
〃 (mean)	1.183	±0.037
BARENDREGT F. *et al.*	1.175	—
CURIE M. *et al.*	1.14	—
BJØRNHOLM S. *et al.*	1.14	±0.01
BRAUNSTEIN J. *et al.*	1.21	±0.03
HOK ONG PING *et al.*	1.25	±0.10

Counted values were corrected for dead time of the counting apparatus, background and length of counting. These corrected values are shown with circles in **Fig. 3.** The short-lived component found by the correction for the long-lived component is shown with lines and crosses.

Four measurements of the half-life were carried out according to the above procedure. Data obtained were analyzed with IBM-7044 and FRANTIC-7044 code. The values obtained are summarized in **TABLE 2** together with those from the literature[2)4)~7)]. The present values agree well each other, except for the third run which gave a little higher value with a large standard deviation than those of the other runs.

References

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