Disintegration of Ca^{47}, Sc^{47}, Va^{48}, and Rh^{100}†

LUIS MARQUEZ*
Institute for Nuclear Studies, University of Chicago, Chicago, Illinois
(Received August 17, 1953)

The disintegration of four radioactive isotopes have been investigated, with the following results: Ca^{47} decays by the emission of two beta-ray groups with end-point energies and relative intensities of 685 kev, 81 percent; and 2060 kev, 19 percent. Sc^{47} decays by the emission of two beta-ray groups of end-point energies and relative intensities of 280 kev, 28 percent; and 490 kev, 72 percent. V^{48} decays by the emission of one single positron groups with end-point energy of 693 kev. Rh^{100} decays by the emission of five positron groups with end-point energies and relative intensities of 150 kev, 0.06 percent; 540 kev, 3.64 percent; 1260 kev, 12.8 percent; 2070 kev, 38.5 percent; and 2615 kev, 45.0 percent. Procedures for preparation and chemical purification of these nuclides are described; and also their other characteristics such as half-lives, conversion electrons of associated gamma rays, etc.

INTRODUCTION

THE characteristics of the yields obtained in spallation reactions when elements are irradiated with protons of a few hundred Mev show that nuclides removed from stability by several neutrons are still formed in sizeable yield.[1] This is true for both the neutron-deficient and the neutron-excess nuclides. The protons of this energy have a considerable range in matter. Since synchrocyclotrons can provide currents of protons of this energy of the order of one microampere, they are among the most suitable machines to prepare certain radioisotopes, particularly those which are far from stability.

We have used the 450-Mev synchrocyclotron at the University of Chicago to prepare several radioisotopes and to study their disintegration. The criterion in choosing the isotopes to be studied was to take those which from the available information seemed to have a large *ft* value and still could be made in sufficient yield in the synchrocyclotron.

BETA-RAY SPECTROMETER

The beta-ray spectrometer used for the experiments was the double-magnetic lens nuclear spectrometer designed by Anderson and Agnew.[2] It had also the additional helical baffle that was placed in the machine by Fan.[3] The samples were counted with an end-window Geiger counter having a mica window of 2.5-cm diameter and a thickness of 1.5 mg/cm². It had a plateau of 150 volts. All the samples were mounted on a Zapon film supported in a hard rubber frame. The thickness of the Zapon film was 0.1 mg/cm². The calibration of the spectrometer was done with the conversion line of Ba^{137m} in equilibrium with Cs^{137}. The energy of this gamma ray has been very accurately determined by Muller *et al.*[4] The line profile that this spectrometer gives has been shown by Anderson[2] and also by Bloom.[5] The resolution of the machine can be changed by varying the opening of the iris diaphragm near the detector. We carried out most of the measurements with resolutions of 5.1 percent and of 3.5 percent. We mean by resolution the full width at half-maximum of the line profile plotted as a function of current or momentum, divided by the value of the current or momentum at the maximum. In our machine the momentum of the electron being detected is proportional to the current in the coils corresponding to the maximum in the line profile.

We carried out the measurements for Ca^{47}, Sc^{47}, and V^{48} with 5.1 percent resolution and the measurements for Rh^{100} with 3.5 percent resolution.

RESULTS

The results for each isotope will be described separately. For most of the isotopes several runs were made. Some of the conditions were changed from one experiment to the other, like the length of irradiation, the weight of the target, etc. The figures quoted are then what we consider a typical run, or what is necessary to get enough activity.

The chemical procedures were derived from procedures described by Meinke[6] and from the well known books by Scott[7] and Treadwell.[8] In the description of the chemical procedures we will use terms commonly used by radiochemists, such as carriers, hold-back carriers, etc. Also, when we say 10 mg of Sc carrier is added, or some similar expression, it is understood that the Sc is dissolved and ionized and suitable precautions are taken to ensure complete exchange. In comparing our results with those of other workers, we shall refer

† This work was supported by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.
* Now at Centro Brasileiro de Pesquisas Fisicas, Rio de Janeiro, Brazil.
[1] For a summary of spallation results see the review article by D. H. Templeton, Ann. Rev. Nuc. Sci. **2**, 93 (1953).
[2] H. L. Anderson and H. M. Agnew, Rev. Sci. Instr. **20**, 869 (1949).
[3] C. Y. Fan, Phys. Rev. **87**, 252 (1952).
[4] D. E. Muller *et al.*, Phys. Rev. **88**, 775 (1952).
[5] S. D. Bloom, Phys. Rev. **88**, 312 (1952).
[6] W. W. Meinke, University of California Radiation Laboratory Report UCRL-432, 1949 (unpublished).
[7] W. W. Scott, *Standard Methods of Chemical Analysis* (D. Van Nostrand Company, Inc., New York, 1939).
[8] F. P. Treadwell, *Analytical Chemistry* (John Wiley and Sons, New York, 1949).

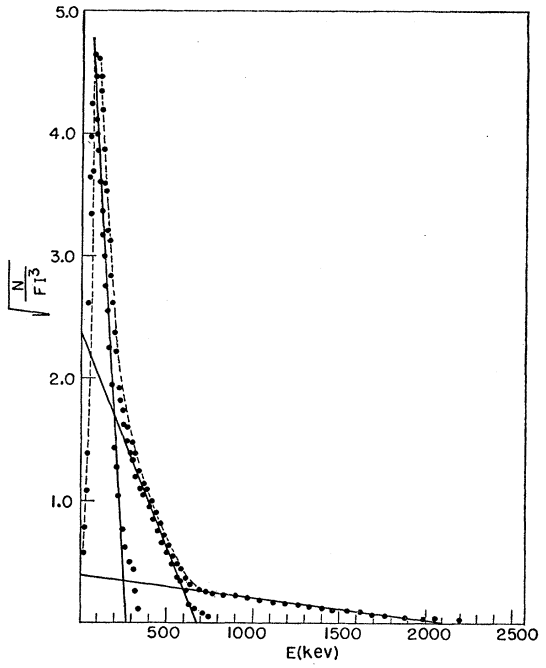

FIG. 1. The Fermi plot of the beta spectra of Ca^{45} and Ca^{47}.

only to those which appeared after the publication of the tables of Way *et al.*[9] and the table of Hollander, Perlman, and Seaborg.[10] For previous work the readers are referred to these tables.

Ca^{47}

About 10 g of $K_2Cr_2O_7$ were irradiated for two hours in the circulating beam of the synchrocyclotron at 74-inch radius, which corresponds to about 420 Mev. The $K_2Cr_2O_7$ was wrapped in an aluminum foil or placed in a thin-walled copper vial specially made for it. The $K_2Cr_2O_7$ was dissolved in hot water, one mg of Ca carrier was added, and the solution was boiled. The Ca was precipitated as $CaCO_3$ by adding NaOH and Na_2CO_3 solutions until the mixture was strongly basic. The $CaCO_3$ was centrifuged, washed with a dilute solution of Na_2CO_3, dissolved in one ml of concentrated HCl, and diluted with 20 ml of water. Hold-back carriers of the elements Ti, V, Cr, Mn, Fe, Co, Ni, and Cu were added. H_2S was passed through the solution and the sulfides were centrifuged. The excess H_2S was boiled off, the solution was oxidized with HNO_3 and the insoluble hydroxides were precipitated by adding NH_4OH until there was a slight excess and the hydroxides were centrifuged. H_2S was passed through the ammonia solution and the sulfides were centrifuged. The Ca was precipitated as $CaCO_3$ by adding an excess of NaOH and Na_2CO_3 solution and boiling; the $CaCO_3$ precipitate was washed with water, redissolved, reprecipitated, and washed with water again. Finally, a fraction of the finely divided $CaCO_3$ was transferred to a Zapon film, dried in a heat lamp, placed in the spectrometer, and measured.

The Fermi plot of the beta-spectrum obtained right after the precipitation of the $CaCO_3$ is shown in Fig. 1. As expected, the Ca^{45} activity was formed also and shows in the spectrum. The Fermi plot could be resolved into the three allowed components shown in the figure. One has an end-point energy of 685 ± 6 kev, a relative intensity of 81 percent, and corresponds to Ca^{47}. Another has an end-point energy of 2060 ± 20 kev, a relative intensity of 19 percent, and also corresponds to Ca^{47}. The third group is the well known beta of Ca^{45} and we find for it an end-point energy 261 ± 4 kev, in agreement with other measurements.[9,10]

The two beta groups of Ca^{47} show allowed spectra within the limits of the experimental errors and to the extent that their overlapping will not mask it. For instance, the Fermi plot of the 2060-kev group is a straight line from about 800 kev to its end point. After subtracting the hard group, the Fermi plot of the 685-kev group shows a straight line from 320 kev to its end point. The 2060-kev group was observed to decay for several half-lives and the value obtained for its half-life was 4.3 ± 0.2 days, which is compatible with the best value of the half-life assigned to Ca^{47} in the literature.[9,10] The group of 685-kev end point decayed and deviated from a straight line, becoming concave upward, and it was possible to resolve it into the group of 685-kev end-point energy, decaying with the half-life of Ca^{47}, and another group which was growing and corresponded to Sc^{47} and had an end-point energy of about 485 ± 15 kev. Sc^{47} could not be studied very well in this way because of the interference of Ca^{45} and Ca^{47}, and it was investigated separately and is described in the next section.

We should mention that the conversion electrons of the 1375-kev gamma ray were not observed, probably because this gamma ray has a very small conversion coefficient and because the resolution was not good enough to detect them above the beta-spectrum. We carried out the measurement, for Ca^{47}, with a resolution of 5.1 percent.

Sc^{47}

The preparation of Sc^{47} was made by milking it from Ca^{47} in order to have it completely free from other Sc activities. Consequently, the irradiation and chemistry were the same as those described under Ca^{47}, except that the amount of Ca carrier was increased to 10 mg. After the Ca^{47} was purified, the Sc^{47} daughter was allowed to grow for about one week, 1 mg of Sc carrier was added to the solution, which was boiled, and the Sc was precipitated as the hydroxide by adding NH_4Cl and NH_4OH. The Sc was washed and dissolved in HCl, 10 mg of Ca hold-back were added, and the

[9] K. Way *et al.*, *Nuclear Data*, National Bureau of Standards Circular No. 499 (U. S. Government Printing Office, Washington, D. C., 1950).

[10] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

Sc was reprecipitated the same way. The Sc was washed, dissolved in HNO_3, evaporated to dryness, dissolved in a small amount of water, and a fraction of it was transferred to a Zapon film, dried, and placed in the machine for measuring. The Fermi plot of the beta spectrum of Sc^{47} is shown in Fig. 2. It can be resolved into two components having allowed shape, with end-point energies and relative intensities 280±3 kev, 28 percent; and 490±5 kev, 72 percent. There was a faint line of electrons at an energy of 218±10 kev which is the conversion electron of the gamma ray whose energy is the difference of the end-point energies of the beta rays. The conversion coefficient could not be determined with any accuracy because it is obtained by subtracting two large numbers whose difference is of the same order as the statistical error. We estimate that it is of the order of 10^{-2}. We also counted a sample of Sc^{47} with an end-window Geiger counter for more than seven half-lives to test its purity and to determine its half-life. The decay was exponential within experimental error and the half-life obtained was 3.44±0.05 days. Our results on the disintegration of Sc^{47} are in qualitative agreement, but in quantitative disagreement, with the recently reported measurements of Cheng and Pool.[11]

V^{48}

This positron emitter is known to decay to the second excited state of Ti^{48} with an energy of 690 kev. Its spectrum was investigated to see if the decay to the first excited state could be detected and also because there has been another positron group reported[12] associated with the low-intensity 2.31-Mev gamma ray.

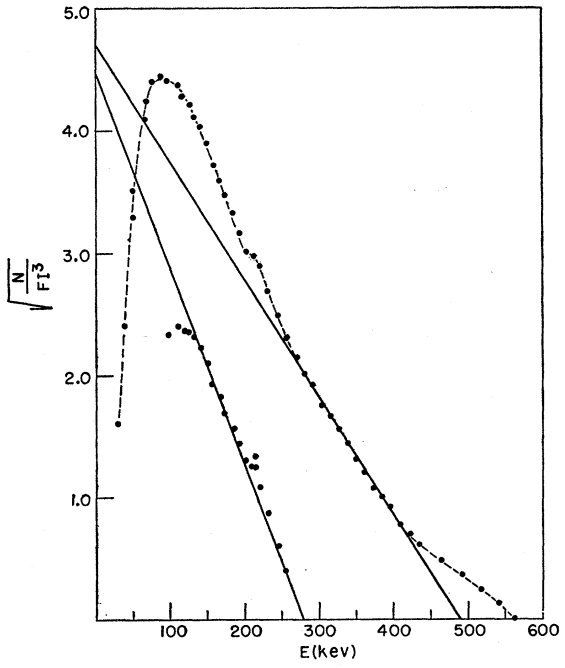

FIG. 2. The Fermi plot of the beta spectrum of Sc^{47}.

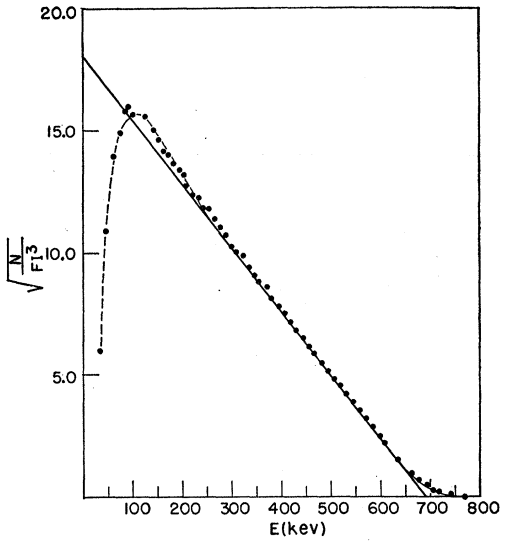

FIG. 3. The Fermi plot of the beta spectrum of V^{48}.

The activity was prepared by spallation of Cr as $K_2Cr_2O_7$ as described under Ca^{47}. The $K_2Cr_2O_7$ was dissolved in hot water, 1 mg of vanadic acid was added, the solution was boiled, 30 mg of Fe were added, and the Fe hydroxide was precipitated with ammonia and centrifuged. The hydroxide carried the vanadium; it was thoroughly washed with water, dissolved in 8*N* HCl, hold-back carriers of the elements P, S, Ca, Sc, Ti, Cr, Mn, Fe, Co, Ni, and Cu were added, and the iron was extracted in isopropyl ether. The solution was neutralized, made slightly acid with acetic acid, and the lead vanadate was precipitated by adding lead acetate. The precipitate was centrifuged, washed, and dissolved in hot HCl, diluted with water, and the lead was removed by passing H_2S. The whole procedure was repeated to get greater purity and finally the solution containing the vanadic acid was evaporated to near dryness, diluted with a few drops of water, and a fraction of that was placed in a Zapon film, dried, and placed in the machine for measurement.

The Fermi plot of the positron spectrum obtained is shown in Fig. 3. The end-point energy is 692±5 kev and the Fermi plot is straight within experimental error, and therefore the spectrum is allowed. We could not detect any other positron group; the few experimental points shown beyond the end point are due to the finite resolution of the instrument, scattering in the machine, etc. This happens in every beta spectrum. If there were a positron group of end-point energy greater than 692 kev, we could place an upper limit of its relative intensity as a function of its end-point energy under the assumption that the shape of its spectrum would be the same as the shape of the 692-kev positron.

[11] L. S. Cheng and M. L. Pool, Phys. Rev. **90**, 886 (1953).

[12] P. L. Roggenkamp *et al.*, Phys. Rev. **88**, 1262 (1952).

TABLE I. The relation between the upper limit of the relative intensity of the second positron group in V^{48}, if it exists; and its end-point energy.

End-point energy of assumed positron group (kev)	Upper limit to its relative intensity (percent)
760	1.0
780	0.6
800	0.3
820	0.2
•	•
•	•
•	•
870 or greater	0.1

The result of this calculation is shown on Table I. In the same way it was found that the transitions to the first excited state have a relative intensity of less than 0.1 percent. Our results are in excellent agreement with the results of Casson *et al.*[13] on the gamma rays following the decay of V^{48}.

Rh^{100}

This isotope was prepared by milking it from its parent Pd^{100}. Eight grams of Ag were irradiated in the circulating beam of the synchrocyclotron. The target was allowed to decay for 4 days in order to get rid of the Pd^{101}. After that the target with 10 mg of Pd was dissolved in boiling HNO_3. The solution was diluted to 500 ml and all the Ag was precipitated with HCl as AgCl. The AgCl was filtered and the Pd was precipitated as the palladium dimethylglyoxime by adding 20 ml of 1 percent alcohol solution of dimethylglyoxime and neutralizing in part the excess of acid. The precipitate was filtered, washed, and dissolved in boiling HNO_3, carriers of the elements Nb, Mo, Ru, Rh, and Cd were added, and the solution was diluted with H_2O and the Pd was precipitated again with dimethylglyoxime. The precipitate was centrifuged, washec, and dissolved again with HNO_3. The Pd was repurified by reprecipitating it with dimethylglyoxime. After dissolving it in HNO_3, 1 mg of Rh carrier was added and the solution was diluted and allowed to stand for three days. After that the Pd was precipitated with dimethylglyoxime and centrifuged, the supernatant was evaporated to dryness; the excess of dimethylglyoxime was destroyed with HNO_3. The HNO_3 was evaporated by adding 1 ml of H_2SO_4 and boiling to fumes. The solution was cooled; 20 ml of H_2O were added and the Rh was precipitated as the metal with $TiCl_3$. The Rh metal was washed with a dilute solution of H_2SO_4, then with H_2O, and then was transferred to a Zapon backing, dried in a heat lamp, and placed in the machine for measurements.

TABLE II. The end-point energy and relative intensity of the positrons from Rh^{100}.

Group	End-point energy (kev)	Relative intensity (percent)
β_1	150±30	0.06
β_2	540±25	3.64
β_3	1260±10	12.8
β_4	2070±20	38.5
β_5	2615±20	45.0

The Fermi plot of the position spectrum is shown in Fig. 4. A glance at its shape shows that it is extremely complex. We could resolve it into the five positron groups which are shown in the figure and whose properties are summarized in Table II. The relative intensity of the positrons given in the table does not take into account electron capture either to the same or to different levels.

When we started to measure the spectrum of Rh^{100} we had hoped to be able to decide whether or not the hardest beta has allowed shape; but as it turned out it could be examined only for a very narrow region, from about 2200 kev to its end-point, and probably this is not enough to make a categoric statement. If its Fermi plot is not straight, then the line shown in the figure is

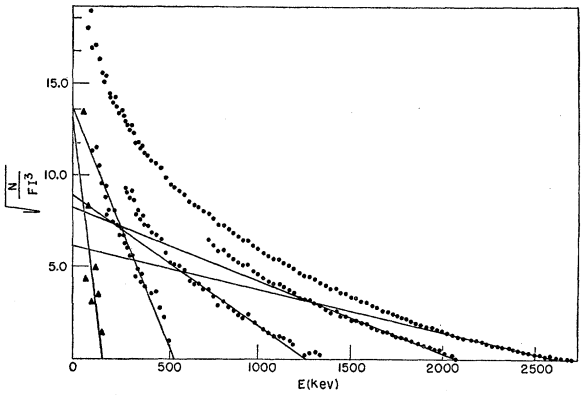

FIG. 4. The Fermi plot of the beta spectrum of Rh^{100}.

probably an average position for it. At any rate, we resolved the spectrum under the assumption that all the beta groups have allowed shape, and the three hardest beta components seem to fit the experimental spectrum quite well. The two softest betas are present in small relative intensity, and after the subtracting of the hardest groups their points scatter considerably. This is the reason for the large error in their end-point energies. We estimate that the error in their relative intensity could be as large as a factor of one-half of its value for β_1 and one-fourth of its value for β_2.

We realize that to resolve a complex beta spectrum into five components is somewhat uncertain, and perhaps one could even find another completely different set of five components fitting the data just as well.

In order to check the result of the beta-spectrum analysis, we decided to determine the energy and intensity of the conversion electrons from the gamma rays following the decay of Rh^{100}. For this prupose we prepared a very strong sample of Rh^{100} by the same technique described, but making a longer irradiation, and

[13] H. Casson *et al.*, this issue [Phys. Rev. **92**, 1517 (1953)].

we looked for the conversion electrons of the gamma rays. The spectrum obtained from the conversion electron is shown in Fig. 5. Eleven conversion lines were identified as belonging to the decay of Rh^{100} because they were observed to decay with the half-life of Rh^{100}. The only impurities observed in the electron spectrum were the K- and L-conversion lines shown at the low current of the spectrum; they decayed with 4-day half-life and probably correspond to some traces of Pd^{100} which was carried with the Rh. We found that the energy of the gamma ray originating from the decay of Pd^{100} and giving rise to these K and L electrons is 80.7 ± 0.4 kev.

The properties of the eleven conversion electrons found associated with the decay of Rh^{100} are summarized in Table III. For this run, the momentum and the current were related by the equation $\eta=5.695\times10^{-2}I$; where η is the momentum in units of mc; and I the current in the coils in amperes.

The combination of eleven gamma rays and five positron groups could be fitted into a self-consistent

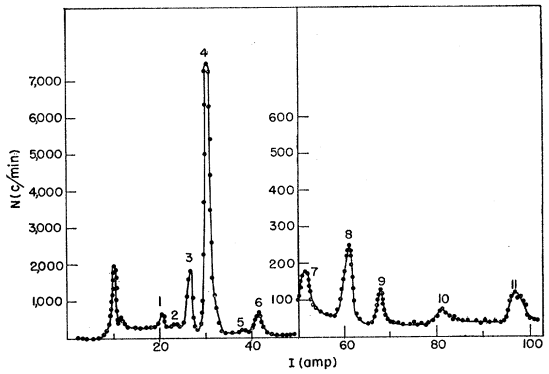

FIG. 5. The spectrum of the conversion electrons following the decay of Rh^{100}.

decay scheme on which the position of all the gamma rays and positron groups could be checked by one or more additions or subtractions applying the conservation of energy; the small residues left in these operations indicate the magnitude of the errors in the energy of the gamma rays. The only gamma ray which could not be checked this way was γ_1 which, for this reason, we placed in the top of the decay scheme, since this seemed to be the most likely place for a gamma ray with such a property.

A sample of Rh^{100} was counted with an end-window Geiger counter for ten half-lives, and we found for its half-life the value 20.8 ± 0.1 hr. In one of the runs in the spectrometer, we measured both the positron spectrum and the conversion line of the 535.3-kev gamma ray, and from the area in the plots of N/I against I we obtained the ratio of the number of electrons belonging to that gamma ray to the total number of positrons, and this was 0.062. Since the relative intensity of all the conversion electrons are given in relation to the intensity of the conversion electrons of the 535.3 gamma ray, we can relate the conversion electrons to the positron groups through the above ratio.

TABLE III. The energy and intensity of the conversion electrons of the gamma rays of Rh^{100} arising from the decay of Rh^{100}.

Current (amp)	Momentum of conversion electron (in units of mc)	Energy of conversion electron (kev)	Energy of gamma ray (kev)	Relative intensity of conversion electron ($\gamma_4=100$)	Name of gamma ray
20.70	1.1788	279.0	301.2	5.3	γ_1
23.80	1.3553	349.6	371.8	0.9	γ_2
26.75	1.5233	420.2	442.4	21.3	γ_3
30.50	1.7369	513.1	535.3	100.0	γ_4
38.50	2.1924	720.2	742.4	0.6	γ_5
41.50	2.3633	800.3	822.5	9.7	γ_6
52.0	2.9612	1085.9	1108.1	1.5	γ_7
61.0	3.4737	1336.0	1358.2	2.8	γ_8
68.1	3.8780	1535.2	1557.4	1.1	γ_9
81.4	4.6354	1911.9	1934.1	0.4	γ_{10}
97.0	5.5238	2357.1	2379.3	1.0	γ_{11}

CONCLUSION

The disintegration characteristics of Ca^{47}, Sc^{47}, V^{48}, and Rh^{100} have been established through this work. The decay schemes of Ca^{47}, Sc^{47}, and Rh^{100} are shown in

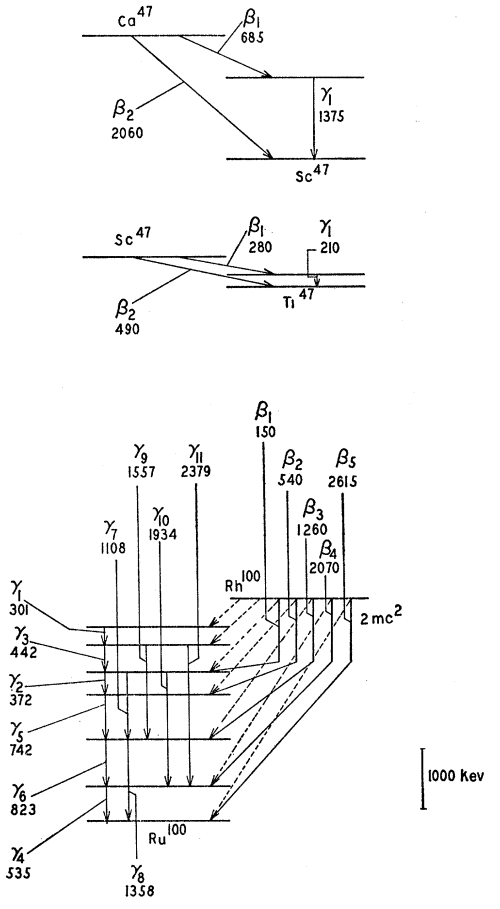

FIG. 6. The decay schemes of Ca^{47}, Sc^{47}, and Rh^{100}.

TABLE IV. The characteristics of the beta groups investigated in this work.

Isotope	Half-life	Group	End-point energy (kev)	Relative intensity (percent)	logft
Ca^{47}	4.3 days	β_1	685	81	6.0
		β_2	2060	19	8.5
Sc^{47}	3.44 days	β_1	280	28	5.0
		β_2	490	72	5.4
V^{48}	16.0 days	β_1	692	48	6.1
		β_2	2012	0.05	11.1
Rh^{100}	20.8 hr	β_1	150	0.003	5.7
		β_2	540	0.18	6.6
		β_3	1260	0.62	7.7
		β_4	2070	1.88	8.2
		β_4	2615	2.19	8.6

Fig. 6. The relevant information for the beta decay is shown for each beta group of each isotope in Table IV. The relative intensities given in this table for positron emitters are corrected for electron capture. In order to make this correction for Rh^{100}, we assumed that γ_4 is an electric quadrupole transition (this is very likely for γ_4 according to the work of Scharff-Goldhaber[14]), and that γ_8 is either electric quadrupole or magnetic dipole. This seems to be correct from consideration of the logft values and the competition among the gamma-ray decays; the ratio calculated from this assumption for the total number of positron emissions to the total number of electron captures is 4.9 percent and 5.0 percent, respectively; which are almost the same and in agreement with Lindner and Perlman.[15] To calculate this ratio we used the conversion coefficients of Rose *et al.*[16] As explained under the paragraph headed Rh^{100}, the spectrum of β_5 could not be explored over a region wide enough to decide definitely whether or not its Fermi plot is a straight line, although the results were compatible with a straight Fermi plot for it. The large value of logft of 8.5 for the beta decay of β_2 in Ca^{47} led us to re-examine its beta spectrum very closely and carefully. We concluded that a straight line was the best fit for the Fermi plot of its beta spectrum from 800 kev to its end-point energy; and in particular it was a much better fit than we obtained when applying the correction corresponding to the "unique" first forbidden transition.

[14] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).

ACKNOWLEDGMENT

I am indebted to H. L. Anderson, who by making available to me the beta-ray spectrometer and the synchrocyclotron, made this work possible. I am also indebted to L. Kornblith, Jr., and C. A. Bordeaux and the crew of the synchrocyclotron for their help in performing the irradiations; and to H. Casson *et al.* for letting me know the results of their experiments before publication.

[15] M. Lindner and I. Perlman, Phys. Rev. **73**, 1124 (1948).

[16] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023 (unpublished).