Decay of ^{50}V

D. E. Alburger, E. K. Warburton, and J. B. Cumming
Brookhaven National Laboratory, Upton, New York 11973
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A 4.25-kg sample of vanadium metal was counted with a high purity Ge detector for 135.5 d. Ninety-six peaks were observed in the γ-ray spectrum between 220 and 2615 keV, all but one of which could be assigned to known contaminant radioactivities mostly contained in the V sample itself. After making a 4.0% calculated correction for the intensity of a ^{234}Pa^{m} γ ray of 1554.1(5) keV, the unassigned peak had an energy of 1553.77(6) keV and contained 1020(70) counts. This energy agrees with a recent precision determination of 1553.768(8) keV for the γ ray from the first excited state of ^{50}Ti following the β^- decay of ^{50}Sc. The 1554-keV γ ray from the V sample is assigned to the electron capture decay of ^{50}V to ^{50}Ti. Based on an absolute γ-ray efficiency calibration of the system, a partial half-life $T_{1/2}=1.5(3)\times10^{17}$ yr and $\log f_0 t=23.4(1)$ were derived for the ^{50}V e.c. branch. The expected 783.3-keV γ ray, which would follow the β^- decay of ^{50}V to ^{50}Cr, was in an unresolved region of contaminant peaks allowing only an upper limit of <560 counts to be placed on its intensity. This corresponds to $T_{1/2}>4.3\times10^{17}$ yr and $\log f_0 t>24.0$ for the β^- branch. The total half-life derived for ^{50}V is $T_{1/2}=1.5^{+0.3}_{-0.7}\times10^{17}$ yr.

I. INTRODUCTION

There are two naturally occurring isotopes of vanadium, ^{51}V (99.750%) and ^{50}V (0.250%). It is known[1] that ^{50}V has a spin-parity $J^\pi=6^+$ and that it is unstable against electron capture decay to ^{50}Ti ($Q_{\text{e.c.}}=2206.7$ keV) as well as β^- decay to ^{50}Cr ($Q_{\beta-}=1038.1$ keV), both of which have $J^\pi=0^+$ in their ground states. However, in both cases there is enough energy for nonunique fourth forbidden decays to take place to $J^\pi=2^+$ first-excited states, 1554 keV in ^{50}Ti and 783 keV in ^{50}Cr, followed by γ-ray emission. No other excited states can be reached.

Several searches[2-8] have beem made for the γ rays which would reveal the e.c. or β^- decay branches of ^{50}V, but neither has been detected. In the most recent study, Pape *et al.*[2] counted a 4 kg sample of V for 1173 h (49 d) with a Ge(Li) detector of 70 cm^3 volume. They placed upper limits on the intensities of 1554- and 783-keV γ rays corresponding to $T_{1/2}>8.8\times10^{17}$ yr for e.c. decay and $T_{1/2}>7.0\times10^{17}$ yr for β^- decay. For both branches $\log f_0 t>24.2$ was deduced. In their experiment the vanadium was in the form of metal chips and the overall resolution (FWHM) in the final spectrum was 5 keV for 1333-keV γ rays.

Several improvements in methods and techniques are now in more general use and it was anticipated that considerably greater sensitivity in the search for ^{50}V decay could be achieved. In brief, these included higher efficiency for detecting the γ rays, procedures for obtaining better energy resolution, computer fitting programs, and more complete and precise listings of the energies and intensities of contaminant γ rays.

II. SAMPLE PREPARATION AND COUNTING GEOMETRY

Pure vanadium metal is available commercially in the form of chips, as in the sample used by Pape *et al.*[2] However, vanadium metal chips occupy three to four times the volume of the V itself. For a high-efficiency counting experiment using a Ge detector, the ideal sample would be a solid close-fitting cylindrical cap. By taking into account the self-absorption factors for γ rays of 783 and 1554 keV, and the decreasing solid angle for the material at the outer edges, as the size is increased, it was concluded that not much would be gained by increasing the cap thickness beyond 3 cm. On the basis of these considerations, a 4560-g quantity of V chips was purchased.[9]

With the cooperation of the Brookhaven National Laboratory (BNL) Metallurgical Services Group, Department of Energy and Environment, the V chips were processed by dividing the material into ten equal batches and melting each batch in a vacuum furnace to form a flat disc having a diameter of ~8.1 cm and a thickness of ~1.7 cm. A single cap-shaped casting was not possible to fabricate with the equipment available. Attempts to bend one of the castings did not succeed because of the extreme hardness and brittleness of the material. Thus, to approximate the desired geometry, eight of the castings were cut on opposite sides into forms having a trapezoidal cross section so as to fit snugly around the standard 7.0-cm diam Ge detector housing (vertical mounting) as shown in Fig. 1. The ninth casting was cut in an octagonal shape so that it rested inside the assembly of the eight pieces, and against the top of the Ge detector housing. The tenth piece was placed on the very top. Material left over from the above cuttings was then divided into eight equal lots and each of these was remelted into a disk form somewhat smaller in diameter than the original melts, and about 1.2 cm thick. These were again cut in trapezoidal shapes so as to fit closely around the outside of the first set of eight pieces as shown in Fig. 1. The net effect was to approximate the geometry of a single cylindrical cast cap with top and side thicknesses of ~3 cm. The various pieces were held in place with tape and the total weight of V metal was 4245 g. After placing the V sample over the

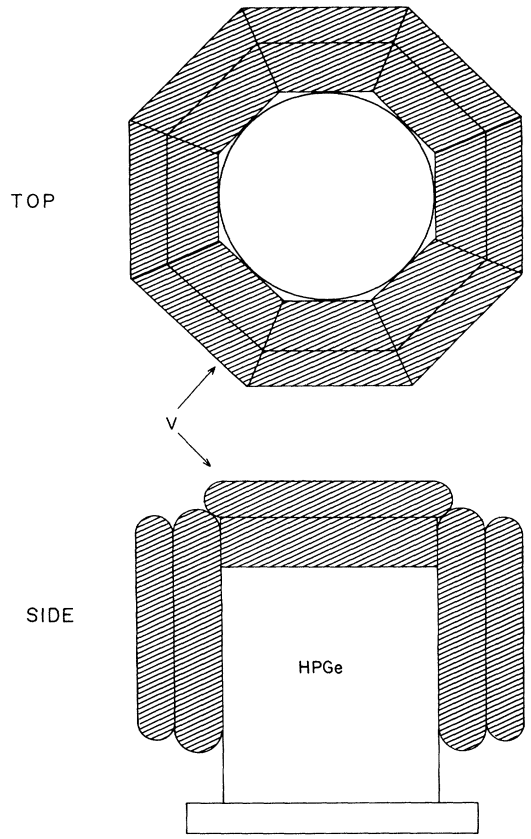

FIG. 1. Geometry of the vanadium castings cut to fit around the HPGe detector housing.

detector the entire assembly was shielded on all sides with a thickness of 7.5 cm or more of "prewar" iron blocks known to be free of ^{60}Co contamination. The experiments were carried out in one of the low-background counting rooms in the chemistry building at BNL.

III. γ-RAY EFFICIENCY CALIBRATION

The HPGe (high-purity germanium) detector used in this work was a well-type ORTEC unit that had been specially procured for experimental searches for the decay of ^{180}Ta. These studies[10] both preceded and followed the tests on the V. The active volume of this detector was $\gtrsim 100$ cm^3 and its efficiency, expressed as a percentage of the efficiency of a 7.6×7.6 cm NaI(Tl) detector for ^{60}Co 1333-keV γ rays at $d=25$ cm, was quoted as 25.5% by ORTEC. The energy resolution for 1333-keV γ rays was 1.92 keV (FWHM), a value that remained constant during the V measurements. The relative photopeak efficiency versus γ-ray energy was determined with a point source of ^{56}Co on the detector axis at a distance of ~ 5 cm with no absorbers.

In order to establish the effective absolute efficiencies for possible γ rays of 783 and 1554 keV from the ^{50}V, a cylindrical liquid container was fabricated from 0.025 cm thick galvanized sheet steel with dimensions such that, when placed over the detector and filled with 750 cm^3 of water, the liquid approximated the configuration of the V sample with respect to the Ge detector. Two 750-cm^3 samples were prepared from the same source of distilled water, one plain and the other containing 100.0 g of dissolved KCl. An identical sample of KCl salt was set aside for later checks on the γ-ray emission rate, if needed. The rate of the 1460-keV photopeak was 0.004/sec for the plain water and 1.177/sec for the one containing the KCl (5-h run). The difference, 1.173/sec, was due to the KCl alone. Based on the summary information[11] for the decay of ^{40}K and the isotopic abundance, it was calculated that the rate of emission of 1460-keV γ rays is 1.72 γ's per g KCl per sec, or 172/sec for the entire sample. Thus the effective absolute photopeak efficiency at 1460 keV for the V geometry was 0.68% per γ ray emitted. In order to derive the appropriate photopeak efficiencies for the 783- and 1554-keV γ rays, it was first necessary to unfold the relatively small self-absorption effect for the 1460-keV γ rays in water (calculated to be a factor of 0.93) and then fold in the respective self-absorption factors for the 783- and 1554-keV γ rays emitted in the V sample. Tables[12] of γ-ray absorption coefficients were used for this purpose along with simple numerical integrations. As an aid in these calculations a two-dimensional map of relative γ-ray peak efficiency was measured by recording spectra with a point source of ^{60}Co at a number of positions out to a distance of 3 cm from the detector container in a plane passing through the detector axis. Self-absorption reduced the efficiency by a factor of 0.67 for the 1554-keV γ ray and a factor of 0.58 for the 783-keV γ ray. Finally, the relative γ-ray efficiency function measured with the ^{56}Co source was used, and the following absolute photopeak efficiencies were derived for the V sample.

$$\epsilon_{1554}=0.47(9)\% ,$$

$$\epsilon_{783}=0.74(18)\% .$$

The uncertainties lie mostly in the differences in the geometries of the V and water samples and the inexact calculations of the self-absorption effects.

As a partial check on the self-absorption calculations an effective γ-ray efficiency curve for the V sample was derived from the contaminant peaks in the spectrum. As shown later, 95% of the intensities of these lines were from the V itself, and it was assumed that the contaminants were uniformly dispersed in the sample. Over the energy range 583—2615 keV, the measured intensities, I(peak), of the strongest lines from the decays of ^{212}Bi and ^{208}Tl (see later discussion) were compared with published γ-ray intensities, $I(\gamma)$.[11] Whereas the ^{56}Co efficiency curve referred to above was nearly linear on a $\log[I(\text{peak})/I(\gamma)]$ vs $\log E(\gamma)$ plot, as is usually the case for Ge detectors, the corresponding efficiency curve derived from the ^{212}Bi-^{208}Tl lines showed a pronounced curvature, clearly indicating the effect of increasing self-absorption with decreasing γ-ray energy. Within accuracies of $\sim 10\%$ the effective relative γ-ray efficiencies found in this way for the V agreed with the self-absorption calculations discussed above.

IV. MEASUREMENTS AND PEAK ASSIGNMENTS

All of the measurements made use of standard electronics including a Northern Scientific Co. 700 series 4096-

channel pulse-height analyzer. This was operated in an automatic sequential mode such that the spectrum was recorded for 90 000 sec (25 h) after which the data were written on magnetic tape along with a tag number, also automatically advanced, the memory cleared, and a new run started. The 25-h counting time was selected so that the strongest of the background peaks would have sufficient intensities for analysis of their shapes and positions, yet if a run had to be rejected there would not be a significant loss of data.

The analysis was performed on the Σ-7 computer at the BNL Tandem Van de Graaff facility. Ten runs were rejected because of unacceptable resolution resulting from gain shifts. The following peaks were selected as references: 583.19, 727.3, 911.1, 1460.8, and 1592.5 keV (2614.5 two escape), and their pulse-height positions in one of the early runs were chosen as standards. By shifting both the gain and base line, all of the accepted spectra were in turn normalized so as to closely match the positions of the five standard peaks. In most cases all five lines could be matched to within 0.1—0.2 channels relative to the reference peaks (dispersion 0.681 keV/channel). Thus, the final spectrum, which included 130 runs for a total of 135.5 d, nearly preserved the energy resolution of individual runs. Based on the computer fits discussed below, the resolution at 1333 keV for the grand sum was determined to be 2.18 keV (FWHM) compared with the 1.92 keV for the checks before and after the V runs, thus indicating an acceptably small increase in linewidth as a result of the shifting and summing procedure. It may be noted that the gain-shift factors required for normalization varied over the range 0.9973—1.0017, corresponding to a peak shift of 6 keV at 1333 keV. Also, the base-line shift varied as much as 2.2 channels or 1.5 keV. Although the gain and base-line shifts were generally very slow in terms of the individual 25-h runs, the analytical procedure described above was obviously essential for preserving the quality of the final spectrum.

The properly shifted V spectra were summed first in four groups of 30—40 runs and then a grand sum of all runs. In the grand sum there were 96 discernible peaks between 220 and 2615 keV. Gamma rays were identified by comparing their energies and relative intensities with γ-ray compilations in Ref. 11 as well as with the catalogs of γ rays prepared by Reus and Westmeier.[13] The majority of the peaks could be assigned to members of the ^{232}Th-^{208}Pb and ^{238}U-^{206}Pb decay chains since their energies agreed with those quoted to within the stated uncertainties. Several lines were unexpected and showed significant variations in their relative intensities in the separate 30- or 40-d sums. These included[14] the 834.843(6)-keV line of ^{54}Mn, the 1115.546(4)-keV line of ^{65}Zn, the 1157.002(3)-keV line of ^{44}Ti-^{44}Sc, the two ^{60}Co lines at 1173.238(4) and 1332.502(5) keV, and the two principal lines of ^{56}Co at 846.764(6) and 1238.287(6) keV. Some of these were also used in the energy calibration described below. It was apparent that test sources were being used from time to time on other equipment in the general area and that their γ radiations were detected in spite of the iron shielding. However, all of these lines were weak and their background contributions were negligible.

An initial inspection of the grand sum spectrum showed that a line of about 1554 keV was present. The first question to address was whether or not this line could be assigned to a contaminant radioactivity. As an aid in understanding the background lines a 4-d run was made with the V sample removed. Rates for a few selected peaks with and without the V are given in Table I. Except for the ^{40}K γ ray of 1460.8 keV, the lines are due to members of the ^{232}Th-^{208}Pb decay chain. Their intensities with the V sample in place were higher than the room background by a factor of ~20 and thus originated ~95% from the V itself. In the region of the expected 1554-keV line the Compton background level with the V in place was found to be only 2.6 times higher than with it removed, while the ratio was 2.3 for the region of the expected 783.3-keV peak. Thus, the amplitude of the continuum in the 783- and 1554-keV regions without the V sample appeared to be due mostly to cosmic rays, while the corresponding amplitude with the V resulted from Compton yields of contaminant γ rays coming from the sample.

The only known long-lived contaminant radioactivity that could have been a candidate for explaining the observed 1554-keV peak was ^{234}Pa^m, a member of the ^{238}U-^{206}Pb decay chain, which emits[11] a γ ray of 1554.1(5) keV. ^{234}Pa^m was shown to be present by the appearance of an isolated peak of 1001.1 keV which had a net intensity of 5561 counts (also present was the ^{234}Pa^m 766.6-keV line—see Fig. 3). However, there are two γ rays in ^{228}Ac decay of 1001.1(8) and 1002.0(8) keV at this same position. Their intensities were calculated based on the strong ^{228}Ac line at 968.8 keV, which contained 64 470 counts. The combined intensity ratio[11] of the 1001.1-and 1002.0-keV lines is 0.63% with respect to the 968.8-keV peak. The ^{228}Ac contribution to the 1001.1-keV peak was therefore 406 counts, leaving 5155 counts for the ^{234}Pa^m γ ray. By using the known[11] intensity ratio

TABLE I. Comparison of background radiations with and without the V sample.

E_γ (keV)[a]	Peak rate in counts/h With V	Without V	Ratio of rates
238 + 241	776	54.6	14.2
583.2	262	11.3	23.2
727.3	55.9	2.8	20.0
860.5	32.9	1.6	20.6
1460.8[b]	11.1	12.2	0.91
2614.5	119.9	6.1	19.7
783[c]	1.57[d,e]	0.69[d]	2.28
1554[c]	0.467[d,f]	0.177[d]	2.64

[a]From members of the ^{232}Th-^{208}Pb decay chain unless otherwise noted.
[b]^{40}K.
[c]Energy region.
[d]Continuum background level in counts per channel (0.681 keV/channel).
[e]Region includes other peaks (see Fig. 3).
[f]Refers to background level under the observed 1554-keV peak.

$$I(1554.1)/I(1001.2)=0.0107\ ,$$

and correcting for detector efficiency (including self-absorption), the intensity of the ^{234}Pa^{m} 1554.1-keV peak was calculated to be 42 counts.

The region of the grand sum spectrum containing the 1554-keV peak is shown in Fig. 2 and includes the known lines at 1556.972(72) and 1580.522(23) keV, both occurring in the decay of the ^{228}Ac member of the ^{232}Th-^{208}Pb decay chain. Gamma-ray peak positions and intensities were extracted using the program SAMPO.[15] The energy calibration was obtained by a quadratic fit to 15 contaminant peaks which had accurately known energies. Accurate values for γ rays from the ^{232}Th chain have been reported by Helmer,[16] while the ^{40}K value was from Helmer, Gehrke, and Greenwood.[17] With the position of the 1556.972(72)-keV peak fixed, and allowing for the presence of a ^{234}Pa^{m} 1554.1(5) γ ray of 4.0% relative intensity, it was determined that the unassigned peak had an energy of 1553.77(6) keV and a net intensity of 1020(70) counts. The four separate 30- to 40-d sums all showed evidence for the 1554-keV peak with relative yields in statistical agreement.

The region of the final spectrum that would contain the

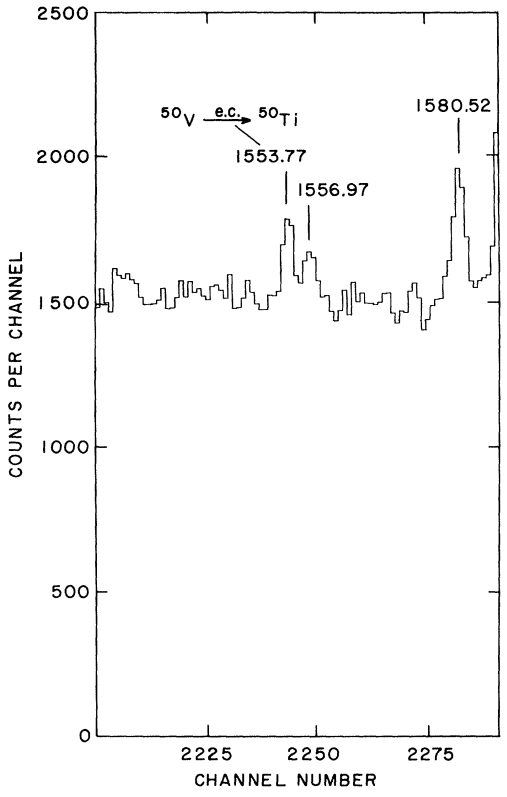

FIG. 2. Region of the spectrum from the V sample accumulated in 135.5 d showing evidence for a γ ray of 1553.77(6) keV assigned to the e.c. decay of ^{50}V to ^{50}Ti. A SAMPO fit gave an area of 1020(70) counts for the 1554-keV peak [corrected for a 4.0% contribution due to a 1554.1(5)-keV γ ray of ^{234}Pa^{m}]. The dispersion is 0.681 keV/channel. The 1556.97- and 1580.52-keV peaks are due to the ^{228}Ac member of the ^{232}Th-^{208}Pb decay chain.

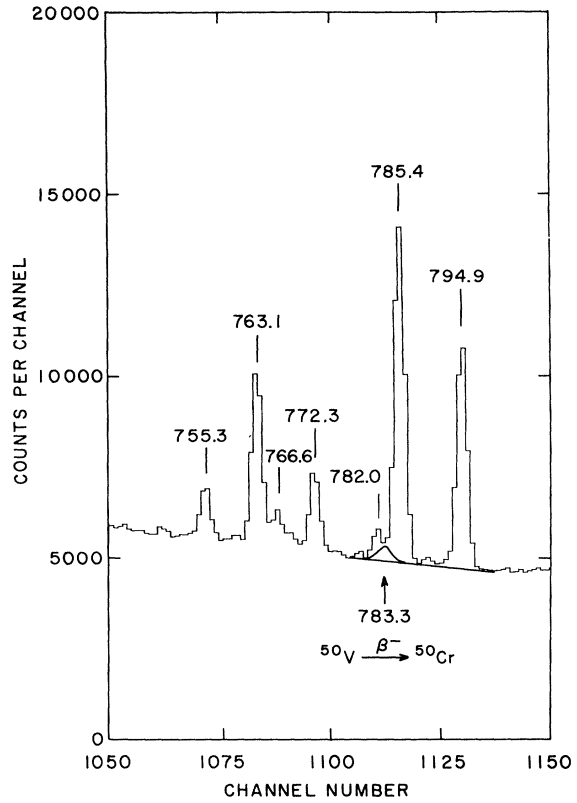

FIG. 3. Region of the spectrum showing the expected position (arrow) of a γ ray of 783.3(1) keV which would follow the β^- decay of ^{50}V to ^{50}Cr. According to a SAMPO fit, the area of the 783-keV peak is <560 counts (drawn in). The 766.6-keV peak is due to ^{234}Pa^{m}, while all others are from members of the ^{232}Th-^{208}Pb decay chain.

783.3(1)-keV γ ray following ^{50}V β^- decay to ^{50}Cr is shown in Fig. 3. This peak would appear at channel 1112.5(2), just at the minimum between the known peaks of 782.0 and 785.4 keV. Based on a SAMPO fit to the lines in this vicinity, the amplitude of the 783.3-keV peak can be no greater than indicated in Fig. 3, with a corresponding area of <560 counts.

V. ANALYSIS AND DISCUSSION

The positive indication of an e.c. branch of ^{50}V as given in Fig. 2 provided an incentive to make precision energy measurements on the γ rays occurring in the β^- decay of 1.7-min ^{50}Sc. For the γ ray from the ^{50}Ti first-excited state the result[18] was 1553.768(8) keV, in agreement with the earlier value[19] of 1553.7(2) keV, but 25 times as accurate. Thus it is seen that the γ ray of 1553.77(6) keV from the V sample agrees well within the uncertainty with the expected energy. As discussed in the preceding section this γ ray cannot be ascribed to a long-lived background activity, and, as far as can be determined, it cannot be assigned to a summing effect or as an escape peak. The fact that it has an intensity greater than some of the known contaminant peaks, such as the nearby one at 1556.97 keV, makes it unlikely that it is a previously unobserved contaminant line. A run[10] of 110 d using the

same detector and similar analysis procedures in a search for ^{180}Ta decay gave a good measure of the yield in the 1554-keV region without a V sample. There was no evidence whatsoever for a 1554-keV peak in that experiment, even though the Compton background rate was 2.6 times lower than the rate in the V runs. We have not been able to explain the 1554-keV line other than to assign it to the e.c. decay of ^{50}V.

By using the effective photopeak efficiency at 1554 keV cited above, together with the weight of the V sample, the ^{50}V abundance of 0.250%, the area of 1020(70) counts under the 1554-keV peak, and the total counting time of 135.5 d, the decay constant for the e.c. branch was calculated to be $1.47(32)\times10^{-25}$ sec^{-1}. Thus, for the electron-capture branch

$$T_{1/2}=1.5(3)\times10^{17}\text{ yr},$$

$$\log f_0 t=23.4(1).$$

The uncertainty in the $T_{1/2}$ value is almost entirely due to the uncertainty in the γ-ray detection efficiency.

Similar calculations were made for the β^- branch using the limit of <560 counts for the 783.3-keV peak from the analysis of Fig. 3. For β^- decay we find

$$T_{1/2}>4.3\times10^{17}\text{ yr},$$

$$\log f_0 t>24.0.$$

In both cases it was assumed that the relative e.c. or β^- branching intensity to the ground state is negligible.

Our results on ^{50}V decay are summarized in Fig. 4. Even though the β^- decay branch was not observed, a total half-life

$$T_{1/2}=(1.5^{+0.3}_{-0.7})\times10^{17}\text{ yr}$$

can be derived from the present work. The present partial half-life for e.c. decay is a factor of 6 shorter than the lower limit reported by Pape *et al.*,[2] although it is consistent with all of the previous[3–8] lower limits. The reason for the discrepancy with Ref. 2 is not clear. There

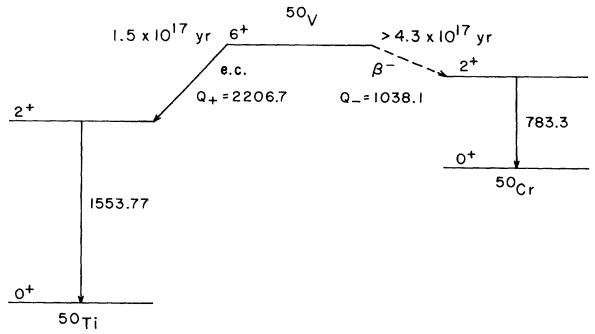

FIG. 4. Decay scheme proposed for ^{50}V. A total half-life $T_{1/2}=(1.5^{+0.3}_{-0.7})\times10^{17}$ yr was derived for ^{50}V.

is very limited information on the systematics of fourth forbidden nonunique transitions, but the value $\log f_0 t=23.4$ for e.c. obtained in the present work seems to be consistent with the three previous cases[20] (all β^- decays) including ^{96}Zr ($\log f_0 t>21.5$), ^{115}In ($\log f_0 t=22.6$), and ^{113}Cd ($\log f_0 t=23.2$).

Nishimura[21] has carried out shell-model calculations on ^{50}V decay following the formulation of Behrens and Bühring,[22] Stech and Schülke,[23] and Schülke.[24] For e.c. decay he predicted $T_{1/2}=1.51\times10^{16}$ yr and for β^- decay $T_{1/2}=9.04\times10^{17}$ yr. Our experimental results are consistent with the longer half-life calculated for β^- decay, although the predicted value for e.c. decay is a factor of 10 shorter than our result.

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