Radioactivity of Ti^{44}†

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Ti^{44} has been produced by the $Sc^{45}(d,3n)Ti^{44}$ reaction. The radiations of Ti^{44} were studied with gamma-scintillation spectrometers alone and in coincidence. A decay scheme is suggested in which Ti^{44} electron captures predominantly to a 144-kev level in Sc^{44} which in turn de-excites by two gamma rays in cascade. A Ti^{44} half-life of approximately 10^3 years is deduced.

SHARP and Diamond[1,2] produced 10^2 counts/minute of a long-lived titanium activity by bombarding scandium oxide with 30–45 Mev protons and assigned the activity to Ti^{44}. They reported that Ti^{44} decays by electron capture with a half-life of $\geqslant 23$ years and a gamma ray of 160 ± 60 kev.

We have produced approximately 10^5 counts/minute of Ti^{44} by the $Sc^{45}(d,3n)Ti^{44}$ reaction. The scandium[3] target material was chemically purified prior to bombardment with tributyl phosphate extractions (to remove thorium) and thenoyltrifluoroacetone extractions (to remove rare earths and zirconium), both under proper acid conditions. Scandium oxide, prepared from 110 mg of purified scandium, was pressed into the grooves of an aluminum target holder and bombarded with 20-Mev deuterons for 1000 microampere hours in the Argonne cyclotron. The threshold[4] for the $Sc^{45}(d,3n)Ti^{44}$ reaction is $(14.3+X)$ Mev, where X is the electron capture disintegration energy of Ti^{44}. The other titanium isotopes produced by the (d,n) and $(d,2n)$ reactions are Ti^{46} (stable) and Ti^{45} (3.1 hr).

Approximately three weeks after the bombardment, the scandium oxide was removed from the aluminum target and dissolved in hot HCl. The solution was cooled, titanium carrier and hydrogen peroxide were added, and the excess peroxide destroyed by boiling. The titanium fraction was chemically separated from the scandium by several cupferron precipitations from chilled $6N$ HCl followed by the extraction of the cupferron complex into chloroform.[1] The cupferron complex was destroyed by hot sulfuric acid prior to repeating the above chemical procedure.

The chemically separated titanium fraction containing the Ti^{44} was examined with a twenty-channel gamma scintillation spectrometer equipped with NaI crystals of $\frac{1}{8}$-in. and $2\frac{1}{2}$-in. thicknesses. The gamma spectrum of Ti^{44} measured with the $\frac{1}{8}$-in. crystal is shown in Fig. 1. An intense gamma-ray peak was observed at about 72 kev, and a peak of about one-tenth its intensity at approximately 144 kev. No gamma rays of higher energy were observed which could be assigned to the Ti^{44} decay. Gamma rays of 511 and 1160 kev were observed to grow into a chemically separated titanium sample with a half-life of 4 hours. This is evidence for the presence of an electron capturing Ti^{44} isotope in that Sc^{44} decays mainly by positron emission with a 3.9-hour half-life to a 1160-kev excited state[5] of Ca^{44}.

The full width of the 72-kev gamma-ray peak at half-maximum intensity was measured in our experimental arrangement to be 32.7% (see Fig. 1) compared to 21.5% and 19.7% for the same quantities for the 59.6-kev gamma ray of Am^{241} and the 88-kev gamma ray of Cd^{109}, respectively. These results indicate that the 72-kev peak is composite. The 72-kev peak can be decomposed into two peaks of equal intensities with energies of 68 and 76-kev and full widths at half-maximum of each peak of approximately 20%, a value which agrees well with experimental results discussed above for single gammas of these energies.

Coincidence experiments also showed that the 72-kev gamma-ray peak is composite. Furthermore, by accepting in the single-channel analyzer only the high-energy

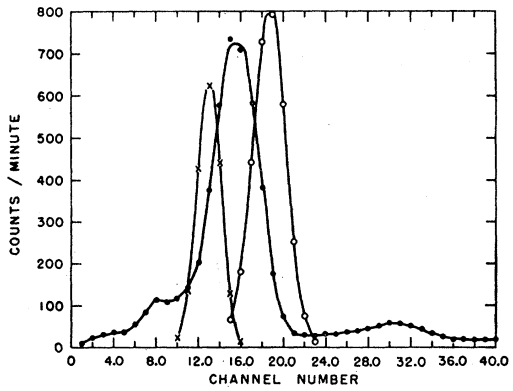

FIG. 1. Gamma spectrum of Ti^{44}. (●) Gamma spectrum of Ti^{44} with $\frac{1}{8}$-in. NaI crystal; (×) 59.6-kev gamma ray of Am^{241}, measured with one g/cm² of copper absorber; (○) 88-kev gamma ray of Cd^{109}.

† Based on work performed under the auspices of the U. S. Atomic Energy Commission.

[1] R. A. Sharp and R. M. Diamond, Phys. Rev. **93**, 358 (1954).

[2] R. A. Sharp and R. M. Diamond, Phys. Rev. **96**, 1713 (1954).

[3] We are indebted to D. F. Peppard for supplying the scandium and for his suggestions on the chemical separation of scandium from various trace impurities. For chemical separation of Sc, Th, and Zr see Peppard, Mason, and Maier, J. Inorg. Nuclear Chem. **3**, 215 (1956).

[4] Summaries of the necessary reaction energies for this calculation are given by D. M. Van Patter and W. Whaling, Revs. Modern Phys. **26**, 402 (1954); R. W. King, Revs. Modern Phys. **26**, 327 (1954).

[5] J. W. Blue and E. Bleuler, Phys. Rev. **100**, 1324 (1955).

side of the 72-kev peak, the coincidence spectrum (Fig. 2) was moved to lower energy (by 4 or 5 kev) and the full width at half-maximum intensity compared more favorably with values obtained for single gamma rays.

Further evidence for two gamma rays of approximately 72 kev was obtained from collimation and absorption experiments which showed the 144-kev peak to be mainly a sum peak. Figure 3 shows the results obtained with copper absorbers of 0.54, 0.99, 2.03, and 5.09 g/cm². The half-thickness for a gamma ray of 144 kev is about 2.8 g/cm² of copper. From Fig. 3 one can see that 0.99 g/cm² of copper reduces the 144-kev peak to almost the background resulting from the 511-kev annihilation radiation of the daughter Sc^{44}. In addition, 0.54 g/cm² of copper absorber reduces the peak at 144 kev about a factor of 2 more than it reduces the 72-kev peak in agreement with the interpretation that the 144-kev peak is a sum peak of two gamma rays of approximately 72 kev. From the absorption data one concludes that the upper limit for the intensity of a 144-kev gamma ray is one percent of the 72-kev gamma-ray intensity.

An attempt was made to measure the number of Ti^{44} "72-kev" gammas per Sc^{44} positron in an equilibrium sample. Making use of a photopeak counting efficiency ratio[6] of 3.1 for the "72-kev" gammas to the 511-kev gammas with a $2\frac{1}{2}$-in. NaI crystal, we calculate 2.2±0.4 "72-kev" gammas per positron. After correcting for the measured positron emission branching ratio[5] of 93% for Sc^{44}, we compute 2.0±0.4 "72-kev" gamma rays per disintegration of Sc^{44}.

This suggests that the electron capture decay of Ti^{44} leads predominantly to a 144-kev excited state in Sc^{44} which in turn is de-excited by a cascade of two approximately "72-kev" gamma rays.[7] The experimental result

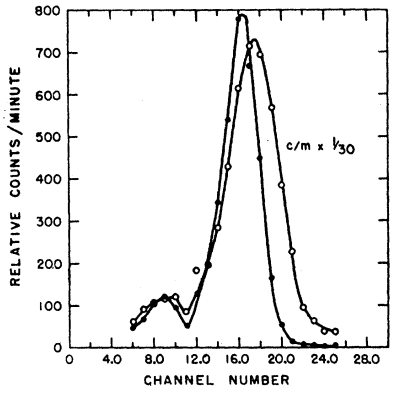

FIG. 2. Coincidence spectrum of Ti^{44}. (o) Gamma spectrum of Ti^{44} in the 72-kev energy region (singles). (●) Gamma spectrum in coincidence with single-channel analyzer gated to accept gammas with energies corresponding to channels 18 to 22.

[6] D. W. Engelkemeir (private communication).

[7] In a private communication with J. C. Roy, J. R. Simanton, and T. P. Kohman, we learned that they have independently reached this same conclusion.

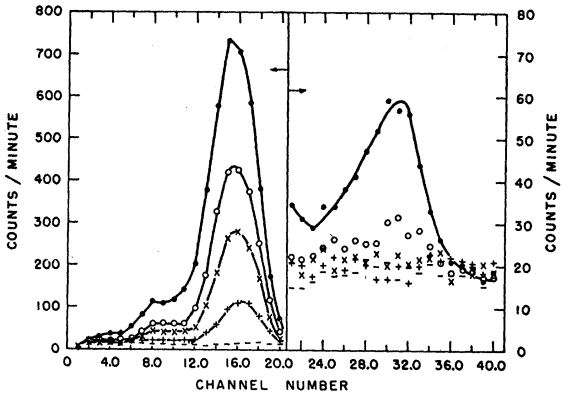

FIG. 3. Ti^{44} gamma spectrum in energy region 0–185-kev with various thickness copper absorbers. (●) No absorber. (o) 0.54 g/cm². (×) 0.99 g/cm². (+) 2.03 g/cm². (—) 5.09 g/cm².

of 2.0±0.4 "72-kev" Ti^{44} gammas per Sc^{44} disintegration in an equilibrium sample indicates that the internal conversion coefficients of the "72-kev" gammas (68 and 76-kev) are small[8] (<20%). For nuclei with $Z=21$, transitions of approximately 70 kev with internal conversion coefficients of <20% are either of electric or magnetic dipole character.[9] If the internal conversion coefficients of both the "72-kev" gamma rays were approximately 0.10,[9] an equilibrium sample of $Ti^{44} \xrightarrow{e.c.} Sc^{44} \rightarrow$ would have 2.2±0.4 "72-kev" transitions per Sc^{44} disintegration, a result in agreement with the decay scheme proposed in Fig. 4.

In addition to a closed shell of 20 protons and a closed shell of 20 neutrons, Sc^{44} has one proton and three neutrons. The shell model[10] predicts that the levels of lowest energy following the closed shells at 20 are the $1f_{7/2}$, $2p_{3/2}$, $1f_{5/2}$, and the $2p_{1/2}$ levels, respectively. Since the low-energy excited states of Sc^{44} are expected to be made up of an even number of negative-

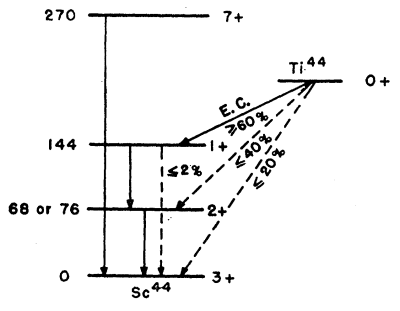

FIG. 4. Proposed decay scheme of Ti^{44}.

[8] This is not true if the Ti^{44} decay leads to an excited state of Sc^{44} which de-excites by 3 or more "72-kev" transitions in coincidence. With the $2\frac{1}{2}$-in. NaI crystal, however, no sum peaks were observed with energies greater than that of the sum peak at 144 kev.

[9] M. E. Rose, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Appendix IV, pp. 907, 908.

[10] M. G. Mayer and J. H. D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley and Sons, Inc., New York, 1955).

parity particles, one predicts these states to have positive parity. The ground state of Sc^{44} has been assigned a spin and parity[5] of 2+ or 3+. The above information favors $M1$ assignments over $E1$ for both of the low-energy gamma rays in the Ti^{44} decay.

The experimental observations of (1) approximately two "72-kev" gammas per Sc^{44} disintegration and (2) the low intensity of $\leqslant 2\%$ for the 144-kev crossover transition are consistent with spins and parities of 3+, 2+, and 1+ for the ground, first excited, and second excited states of Sc^{44} as shown in Fig. 4. Assuming a ground state of 0+ for Ti^{44}, electron capture to the 1+, 144-kev level, is allowed and approximately two "72-kev" transitions per disintegration are expected, in that electron capture to the first-excited state and to the ground state are at least second forbidden. The above spin sequence (Fig. 4) implies a multipolarity of $E2$ for the 144-kev crossover transition. This assignment for the 144-kev gamma is consistent with the experimental limit placed on the intensity of the crossover transition since the calculated theoretical lifetime[11] of a 144-kev $E2$ transition is approximately 10^4 times as long as a 72-kev $M1$ transition.

A ground state spin of 2+ for Sc^{44} with first and second excited states of 2+ and 1+, respectively, is not likely since this scheme predicts a strong 144-kev $M1$ transition. The same argument can be used against the spin sequence 3+, 3+, and 2+. First and second excited state spins of 1+ and 0+, respectively, with a 2+ ground state are consistent with a low-intensity crossover gamma but from this spin sequence one predicts too small a number of "72-kev" gammas per Sc^{44} disintegration. Electron capture to both the 1+ and 0+ excited states would be allowed with the transition to the first excited state favored by energy considerations. Assuming no electron capture to the ground state (2+), a second forbidden transition, and the electron capture probability to the first excited state equal to or greater than the electron capture probability to the second excited state, one calculates ≤ 1.5 "72-kev" transitions per Sc^{44} disintegration. This value is smaller than the experimental value.

The half-life of Ti^{44} has been estimated to be about 10^3 years by the following method. The disintegration rate of Sc^{46} from the $Sc^{45}(d,p)Sc^{46}$ reaction was measured in addition to the disintegration rate of Ti^{44} from the $Sc^{45}(d,3n)Ti^{44}$ reaction. Employing a half-life of 84 days[12] for Sc^{46} and a $\sigma(d,p)/\sigma(d,3n)$ cross-section ratio of 2 for 20-Mev deuterons on Sc^{45}, we calculate a half-life of 10^3 years for Ti^{44}. The above cross-section ratio was estimated from fragmentary cross-section data in the region of scandium and is subject to considerable error. An increase in the above cross-section ratio would decrease the Ti^{44} half-life.

If the electron capture transition to the 144-kev level is allowed, a half-life of 10^3 years implies that the energy available for electron capture to the 144-kev level is only a few kev. The energy available for electron capture to the approximately 72-kev level from the above argument would be approximately 80 kev. Assuming a logft value of $\geqslant 10$ for this as a second forbidden transition, one deduces the half-life for electron capture to the 72-kev state to be $\geqslant 10^5$ years. A branching of 1% to the 72-kev level is well within the limit set in this experiment.

The sequence of spins of 4+, 3+, and 2+ for the ground, first excited, and second excited states of Sc^{44} would allow a sizeable amount of energy available for electron capture to the 144-kev level to be consistent with a half-life of 10^3 years. A measurable competition for electron capture to the 270-kev state would be prohibited because of its very large spin. A spin of 4+ for the ground state of Sc^{44}, however, is unlikely in that Sc^{44} decays by an allowed transition[5] to a probable (2+) state in Ca^{44}.

If the assumption of positive parity for the low-lying levels of Sc^{44} is in error, spin sequences involving negative parity levels can be suggested which are consistent with the experimental data. For example from the spin sequence 3+, 2+, 1−, one predicts the 144-kev level (1−) to decay principally by a "72-kev" $E1$ transition since the $M2$ 144-kev crossover transition has a much longer theoretical lifetime.[11] A half-life of 10^3 years for the first forbidden electron-capture transition to the 144-kev level (1−) would be consistent with a larger disintegration energy than that deduced for the decay scheme discussed earlier with the 144-kev level populated by an allowed transition. Electron capture branching to the 2+ first excited state and to the 3+ ground state would be small since these are $\geqslant$ second forbidden transitions.

In summary from our experiments the preferred spins and parities of the Sc^{44} levels are 3+, 2+, and 1+ for the ground state, 72-kev, and 144-kev levels, respectively. Ti^{44} should prove to be a useful titanium tracer owing to its favorable radiation and long half-life (all other known radioactive isotopes of titanium have half-lives $\leqslant 3.1$ hr).

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[11] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 627.

[12] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 490 (1953).