energies around 200 kev. When unshielded so as to respond to low-energy betas, coincidence counts were observed with gamma energies greater than 250 kev.

It is thus possible to arrange the observed transitions in a rather simple decay scheme which essentially satisfies every observation, as shown in Fig. 4.

The final titanium 47 nucleus with 22 protons and 25 neutrons, from shell theory, would have five particles in the $f_{7/2}$ level and an $f_{7/2}$ ground state. It is, however, possible that the five $f_{7/2}$ particles couple to form a 5/2 odd ground state, similar to the coupling observed in Mn^{55}. In this event the first excited level is $f_{7/2}$ and the beta transition goes to the excited state with no change in spin or parity, since the scandium 47 is likely to have an $f_{7/2}$ ground state as does the similar Sc^{45}. Selection rules do not forbid a decay to the 5/2 odd ground state. Such a transition, however, would not be a simple one-particle process, thus it would probably have a much longer lifetime than the simple $f_{7/2}$ to $f_{7/2}$ transition. The 159.5-kev radiation is then an $M1$ transition. The Ca^{47} nucleus is given an assignment of $f_{7/2}$ from the shell theory. It then seems difficult to explain the lack of a beta transition to the ground state of Sc^{47}.

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A Long-Lived Activity in Neutron-Irradiated Niobium

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An activity has been found in samples of niobium metal irradiated for long periods in the Chalk River reactor. Extensive chemical tests showed that the activity is niobium. The activity is most likely the long-lived ground state of Nb^{94}. If so, its half-life is estimated as $(2.2\pm0.5)\times10^4$ years from its yield. It has a 0.50 ± 0.05-Mev beta and three gammas, 0.70 ± 0.01 Mev (92 percent), 0.87 ± 0.01 Mev (92 percent), and 1.57 ± 0.02 Mev (8 percent). The capture cross section of Nb^{94} is 15 ± 4 barns.

I. INTRODUCTION

A NUMBER of investigators have reported the presence of a 6.6-min activity in neutron-irradiated niobium.[1,2] The same activity has also been produced by a (d,p) reaction on niobium.[3] This activity has been assigned as class A (element and mass number certain) to Nb^{94}.[4] The activity has been found to decay almost entirely by isomeric transition to a long-lived ground state.[5,6] The energy of the transition is 41.5 kev as determined from the energies of the K, L, and M conversion electrons.[7] In addition, about 0.1 percent of the activity has been found to decay by emitting a 1.3-Mev β[8] and a 0.9-Mev γ.[9]

Goldhaber and Muehlhause[6] set a lower limit of $\gg$100 years for the half-life of the ground state. Hein, Fowler, and McFarland,[10] who studied niobium metal that had been irradiated four months in the Argonne reactor, set a minimum half-life of 5×10^4 years.

This laboratory has had some samples of niobium irradiated for a long period in the Chalk River reactor in order to look for a long-lived activity which might be the ground state of Nb^{94}. An activity has been found which the authors feel is the ground state of Nb^{94}.

II. CHEMICAL SEPARATIONS

Two samples of niobium metal were irradiated in the Chalk River reactor at an average flux of $4-5\times10^{13}$ n/cm^2 sec, one sample for a total $nvt=1.33\times10^{21}$ n/cm^2, the other for a total $nvt=2.5\times10^{21}$ n/cm^2. The total nvt received was checked by cobalt flux monitors. The niobium metal was spectrographically standardized niobium powder. A spectrographic analysis, made by the supplier, showed very faint lines of Mg, Cu, Fe, Si, and Ca, and no lines of Al, Ag, B, Co, Cr, Mn, Mo, Ni, Pb, Sn, Ta, Th, Ti, V, Zr, or Zn.

The samples were returned to the laboratory about two months after the end of irradiation. Decay curves and aluminum absorption curves on the irradiated metal showed that the major beta activity was Ta^{182}. Scintillation spectrometer scans on the metal showed the 0.76-Mev gamma of Nb^{95} in addition to the characteristic spectrum of Ta^{182}. Aluminum absorption curves made with a low-absorption counter also showed the presence of the weak beta of Nb^{95}.

An initial chemical purification of the niobium by

* Operated by the General Electric Company for the U. S. Atomic Energy Commission.

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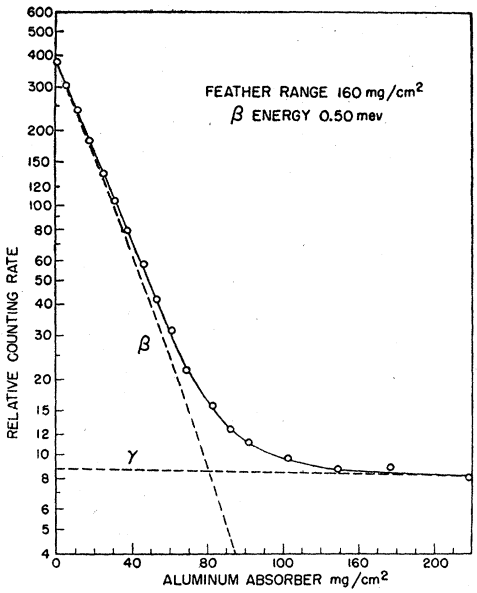

FIG. 1. Aluminum absorption curve of Nb^{94}.

the procedure used in analyzing niobium in fission products[11] showed that the bulk of the activity was niobium or tantalum which follows Nb in the procedure. Two methods have been used to purify the niobium from radioactive tantalum. The first method is based on the extraction of the niobium from 8*N* HCl into a solution of methyldioctlyamine in xylene.[12] A sample of

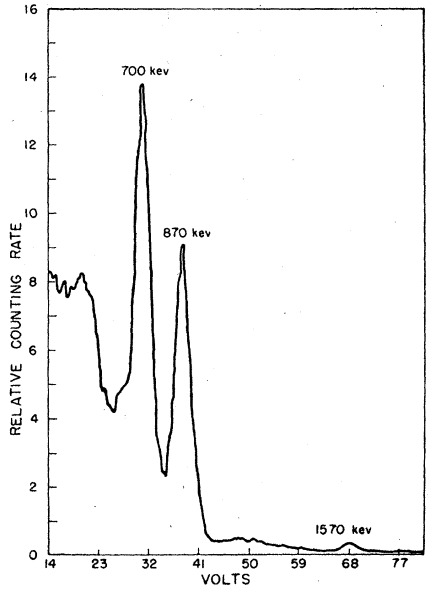

FIG. 2. Scintillation spectrogram of Nb^{94}.

[11] L. E. Glendenin, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), paper 253, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 1523.

[12] G. W. Leddicotte and F. L. Moore, J. Am. Chem. Soc. **74**, 1618 (1952).

niobium was purified by four methyldioctlyamine extractions plus three cupferron scavenging extractions. A small yield of niobium was obtained which was fairly free from Ta^{182}. The purified niobium contained mainly Nb^{95}, and, when followed for decay, showed in addition, Ta^{182} plus a still longer-lived activity.

The other method that has been used to separate the niobium from the tantalum impurity is based on partition chromatography.[13] In this method, the niobium is complexed with HF, a little Ta carrier is added, and the sample added to a cellulose packed column. The column is then treated with three solutions; first with water saturated methyl ethyl ketone to remove the Ta, then methyl ethyl ketone containing about 1 percent HF to fix Ti, Sn, and Zr on the column, then finally methyl ethyl ketone containing $12\frac{1}{2}$ percent HF to elute the niobium. The niobium containing solution is then evaporated, the residue ignited, and the resulting Nb_2O_5 purified further by the standard niobium fission product procedure.[11] Scintillation spectrograms showed that the above procedure gave a very clean separation of Ta and Nb. The purified niobium from a sample that had decayed about fourteen months showed only the long-lived activity; the niobium from a sample that had decayed five months showed mainly Nb^{95} plus a little of the same long-lived activity.

TABLE I. Specific beta and gamma activities of long-lived niobium.

Radiation	Energy, Mev	Counts/min mg
β^-	0.50	530 000
γ	0.70	455 000
γ	0.87	470 000
γ	1.57	41 000

In order to prove that the long-lived activity is actually niobium, further chemical tests were run. Neither an AgCl or an Sb_2S_3 scavenging precipitation carried the activity. Some of each sample of the cellulose column purified niobium was then run through an anion exchange column. The column, packed with Dowex-1, is similar to the one used by Kraus and Moore[14] to separate Nb and Zr. The long-lived activity and the Nb^{95} in the five-month decay sample showed no fractionation in going through the column as shown by scintillation spectrograms. The long-lived activity in the 14-month decay sample gave the same elution pattern as the Nb^{95}.

Since the long-lived activity follows niobium chemistry and is not fractionated from Nb^{95} in the anion exchange column, it can be assigned to an isotope of niobium. Also, the niobium from the same irradiation and purified by the two different methods has the same specific activity of the long-lived isotope. The specific

[13] A. F. Williams, J. Chem. Soc. **1952**, 3155.

[14] K. A. Kraus and G. E. Moore, J. Am. Chem. Soc. **73**, 9 (1951).

activity of the long-lived isotope in the niobium from the two irradiations is proportional to the *nvt* received.

III. DETERMINATION OF ENERGY OF RADIATIONS AND SPECIFIC ACTIVITY

The long-lived activity is most likely the ground state of Nb^{94}. The beta energy of the activity has been determined by Feather analysis of aluminum absorption curves taken on samples known to be free of Ta^{182} (see Fig. 1). An energy of 0.50 ± 0.05 Mev has been obtained for the beta. The energies of the gamma rays have been determined with a scintillation spectrometer by comparison with the known energies of the Cs^{137}-Ba^{137} gamma and the Co^{60} gammas (see Fig. 2). The energies found are 0.70 ± 0.01, 0.87 ± 0.01, and 1.57 ± 0.02 Mev.

The specific beta activity of the niobium has been obtained by counting with an end-window proportional counter. Approximate specific gamma activities have also been obtained by comparing the scintillation spectrograms of the niobium with those of $Cs^{137}-Ba^{137}$, Nb^{95}, and Co^{60} of known disintegration rates. Table I gives the specific β and γ activities found on a sample

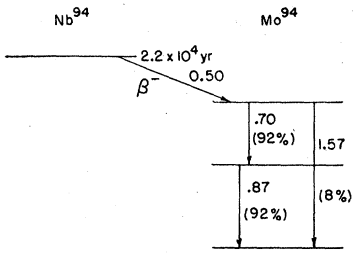

FIG. 3. Decay scheme of Nb^{94}.

in which all the Nb^{95} had decayed and which was free of Ta^{182}.

The specific activity of Nb^{95} in the purified niobium from the two irradiations has also been obtained by 4π counting, by gamma counting, and by beta counting with a low absorption counter. A specific activity corrected back to the end of irradiation of 2.2×10^8 β/min mg was obtained for the shorter irradiation, and 6.5×10^8 β/min mg for the longer irradiation.

The activity of the highly purified niobium in which all the Nb^{95} has decayed has been followed for several months and has shown no decay. From the decay

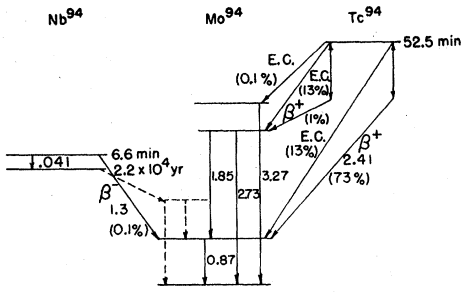

FIG. 4. Decay schemes of Nb^{94m} and Tc^{94} (after Goldhaber and Hill see reference 8). The broken lines indicate our proposed decay scheme of Nb^{94}.

curve, a half-life of >3 years can be set. A proportional counter x-ray spectrometer did not show the presence of an appreciable amount of x-ray activity; consequently the activity decays largely, if not entirely, by β emission.

IV. DISCUSSION OF RESULTS

A half-life of the Nb^{94} has been calculated from the specific activity, the total *nvt*, and the capture cross section of Nb^{93}. Based upon a capture cross section of Nb^{93} of 1.1 ± 0.1 barns,[15] a half-life of $(2.2\pm0.5)\times10^4$ years is obtained for the Nb^{94}. An approximate capture cross section of 15 ± 4 barns for Nb^{94} has been calculated from the specific activity of Nb^{95}, the average flux during the irradiation, and a capture cross section of 1.1 ± 0.1 barns for Nb^{93}.

A possible decay scheme for the Nb^{94} has been proposed based upon the results of this investigation and upon the decay schemes given for Nb^{94m} and Tc^{94}. The proposed decay scheme is given in Fig. 3, while the decay schemes given by Goldhaber and Hill[8] for Nb^{94m} and Tc^{94} are given in Fig. 4. On the basis of the Tc^{94} and Nb^{94m} decay schemes, it is expected that the 0.70-Mev gamma precedes the 0.87-Mev gamma in the gamma cascade.

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[15] D. J. Hughes *et al.*, *Atomic Energy Levels*, U. S. Atomic Energy Commission Report AECU-2040, May, 1952 (unpublished).