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Contract No. W-7405-eng-48

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October 21, 1949

Berkeley, California

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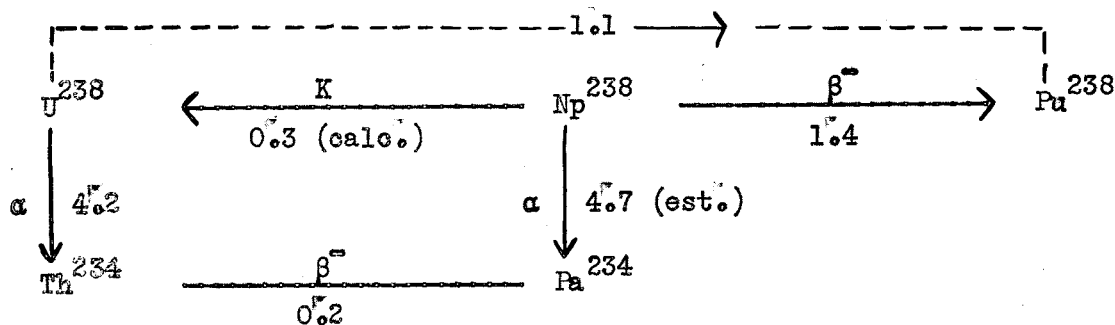
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HALF-LIFE FOR DOUBLE BETA-DECAY

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Fireman<sup>(1)</sup> has reported the results of a rather difficult beta-particle coincidence counting experiment in which the decay of  $\text{Sn}^{124}$  by the simultaneous emission of two negative beta-particles, with a half-life between  $0.4 \times 10^{16}$  years and  $0.9 \times 10^{16}$  years, seems to have been observed. This note reports the results obtained from a different and somewhat simpler method of looking for the phenomenon of simultaneous emission of two beta-particles. These results are negative so far and show that this process is considerably less probable in the case chosen by us than in that reported by Fireman.

Our method consists of looking in uranium samples for 90-year  $\text{Pu}^{238}$  which would come from  $\text{U}^{238}$  by the double beta-particle mechanism since  $\text{Np}^{238}$  is heavier than  $\text{U}^{238}$ , which in turn is substantially heavier than  $\text{Pu}^{238}$ , in the isobaric triplet  ${}_{92}^{238}\text{U}$  -  ${}_{93}^{238}\text{Np}$  -  ${}_{94}^{238}\text{Pu}$ . This chemical method of investigation is particularly applicable to this isobaric triplet because there appears to be no other mechanism to account for the  $\text{Pu}^{238}$  should it be found. The energetics of the situation are summarized in the following diagram, where the disintegration energies are derived from sources which may be traced through a recent compilation.<sup>(2)</sup>



The alpha-disintegration energy of  $\text{Np}^{238}$  is estimated from alpha-decay systematics.<sup>(3)</sup>

Our experiment consisted of taking 14 kg. of very pure, 6-year old,  $\text{UO}_2$  and extracting and separating the plutonium fraction by chemical means. The method

consisted essentially of dissolving the oxide in nitric acid and precipitating Pu(IV) with lanthanum fluoride, followed by solution of the lanthanum fluoride and oxidation of the plutonium to Pu(VI), which was extracted into diethyl ether and then re-extracted into water. Similar cycles were repeated five times in order to separate completely from  $UX_1$  and to reduce the amount of lanthanum carrier, after which the final sample was plated out on flat platinum with total carrier weight probably less than 50 micrograms. The use of tracer  $Pu^{239}$  in this separation established that the chemical yield amounted to 10%.

This final sample was measured for the presence of the 5.51 Mev alpha-particles of  $Pu^{238}$  on the alpha-pulse analyzer apparatus in this laboratory<sup>(4)</sup>. This analysis showed that the counting rate of the  $Pu^{238}$   $\alpha$ -particles at essentially 50% counting yield amounted to  $0.00 \pm 0.01$  counts per minute above background. This indicates that the "half-life" of  $U^{238}$  for simultaneous emission of two beta-particles, for which a total energy of 1.1 Mev is available, is greater than  $6 \times 10^{18}$  years.

This experiment could be extended to reach longer half-lives through the use of larger and older sources of uranium such as pitchblende ore. In this case, of course, the plutonium fraction so isolated will contain a certain amount of  $Pu^{239}$  as has already been demonstrated<sup>(5)</sup>. The extraction of plutonium from a ton of pitchblende (50% uranium) with 10% yield could detect a half-life as long as some  $10^{22}$  years for the same limits of counting accuracy.

This result appears to disagree with that of Fireman although it may not be possible to be positive about this in view of the difference in energies and atomic numbers and possible difference in degree of forbiddenness involved. The theory for the double beta-decay process sets widely differing ranges of half-life depending upon whether the process can take place without neutrino emission<sup>(6)</sup>. Fireman's results are in the range predicted for double beta-decay without neutrino emission while our half-life limit seems to be above this predicted range and perhaps points

toward the emission of two neutrinos in this process.

It is a pleasure to acknowledge the assistance of Dr. L. B. Magnusson in the chemical procedure. This work was performed under the auspices of the U. S. Atomic Energy Commission.

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