Double Beta Decay of ^{238}U

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(Received 5 November 1990; revised manuscript received 26 August 1991)

The half-life for the decay of ^{238}U to ^{238}Pu has been measured to be $(2.0\pm0.6)\times10^{21}$ yr by chemically isolating and measuring, from the resultant alpha particles, the amount of plutonium that had accumulated in 33 yr from 8.47 kg of purified uranyl nitrate. Other sources of ^{238}Pu have been studied and found negligible.

PACS numbers: 23.40.Bw, 14.60.Gh, 27.90.+b

There has recently been much interest in the rate at which certain nuclei decay by changing their nuclear charge by two units, a process usually considered to be the simultaneous emission of two beta particles (double beta decay). The rate of this process is of interest in considerations of the numbers and properties of neutrinos, of fundamental interactions and conservation laws, and of the existence of other than presently known particles. Such decays also test our understanding of the properties of excited states of nuclei since these are involved in calculating the rates of double beta decay. At present, there are about thirty experiments, worldwide, which study various aspects of this process. However, it is one of the slowest spontaneous decays in nature ($\lambda\lesssim10^{-20}$ yr^{-1}) and there have been, to date, only five systems for which experimental evidence has been obtained for its actual existence. In addition to the geochemical work on ^{82}Se, ^{130}Te, and ^{128}Te, some of which goes back about forty years, recently there have been published counter experiments on ^{76}Ge [1,2], ^{82}Se [3], and ^{100}Mo [4]. Recent reviews of the experimental situation are to be found in Refs. [4] and [5]. Theoretical reviews of double beta decay have been made by Haxton and Stephenson [6], Doi *et al.* [7], and Muto, Bender, and Klapdor and Tomada [8]. Compared to the other systems that have been studied, the decay of ^{238}U to ^{238}Pu by double beta decay has several special features. The Q value of the decay (1.1 MeV) is the lowest of the systems presently being studied. This leads to theoretical predictions of half-life in the 10^{23}-yr range. The low-Q value, however, allows competition by other than standard 2ν processes (such as 0ν or Majorana emission) to benefit from phase-space considerations. Thus, Staudt, Muto, and Klapdor-Kleingrothaus [9] predict that a neutrino with a value of the Majorana mass parameter $\langle m_\nu\rangle$ equal to 3 eV would lead to a zero neutrino double beta decay rate ($t_{1/2}=3\times10^{22}$ yr) faster than that calculated (5.2×10^{22} yr) for the conventional two-neutrino mode using a favorable value of the nuclear pairing parameter (g_{pp}).

A special feature of geochemical and milking experiments, such as the present one, is that they measure the total transformation rate of a nucleus ^{A}Z to $^{A}(Z+2)$ irrespective of the particular mechanism or particles involved. This consideration may be pertinent in view of recent [10] indications of the participation of 17-keV neutrinos in single beta decay.

The only previous experimental study of such a decay of ^{238}U is early work by Levine, Ghiorso, and Seaborg [11]. They used a radiochemical technique that was similar to the one used in the present work and set a lower limit of 6×10^{18} yr for the half-life. Theoretical estimates on the half-life based on conventional two-neutrino emission have ranged from 2.2×10^{19} [12] to greater than 5×10^{22} yr by Staudt, Muto, and Klapdor-Kleingrothaus [9]. This last estimate reflects the consideration [13] that cancellations by different contributions to the nuclear matrix elements can strongly suppress the rate of the conventional two-neutrino double beta decay process.

A favorable feature of the ^{238}U system is the high Z of the nuclei involved. This and the strong fission competition minimize the possibility of competing nuclear reactions forming ^{238}Pu. The most important such reaction is ^{238}U$(p,n)^{238}$Np. The ^{238}Np then decays with a 2.117-d half-life to ^{238}Pu. As part of the studies of competing reactions, the cross section for this process was determined at Los Alamos.

A serious practical consideration in dealing with the ^{238}U$(\beta\beta)^{238}$Pu system is the worldwide fallout of ^{238}Pu from atmospheric tests and satellites that have had ^{238}Pu power sources and then, on reentry, burned out in the atmosphere. A typical fallout concentration is 10^6 atoms of ^{238}Pu per cm^2 of surface all over the world [14]. Since the present experiments produce only $\sim10^5$ atoms of ^{238}Pu, such fallout introduction must be avoided. The present work used uranium nitrate that had been purified and isolated before much ^{238}Pu had been introduced into the atmosphere.

Haxton, Cowan, and Goldhaber [15] revived interest in the ^{238}U system and the present work was started as a result of their publication. The experiment involves the extraction of the accumulated plutonium from uranium salt that had been purified and isolated from fallout for 33 yr. This amount of uranyl nitrate (1.02×10^{25} atoms of ^{238}U) produces 2.3×10^5 atoms of ^{238}Pu in 33 yr if the half-life for double beta decay is 10^{21} yr. The chemically

isolated and purified plutonium is examined in low-background alpha counters. In the region of 5.5 MeV only ^{241}Am and ^{222}Rn can interfere with the ^{238}Pu alpha particles. The former was not prevalent in 1956 and is excluded by the chemistry. The latter, and its parents, should also be eliminated by the chemistry. In addition, its presence can be identified by the simultaneous presence of the daughter alpha radioactivities.

Chemical operations.—The uranyl nitrate used in this experiment had been purified by Shattuck Chemical Co. of Denver on about 1 July 1956. At the time there had not been much ^{238}Pu introduced into the atmosphere by large explosions and the large-scale use of ^{238}Pu for power sources on space vehicles had not started. Moreover, the chemical purification of the uranyl nitrate at the time probably used ether extractions which should have removed any fallout plutonium. The uranyl nitrate had been kept in a plastic bag inside a sealed cardboard container in Chicago until the summer of 1989. Several smaller samples of this material had previously been examined for ^{238}Pu content with negative results [16,17]. In August 1989, 8.47 kg of the salt were dissolved in deionized water and a known amount of ^{239}Pu tracer solution was added to this acid solution. This tracer had been prepared by neutron irradiation of uranium in a low enough flux so that the ratio of ^{238}Pu alpha particles to those of ^{239}Pu was less than 10^{-5}.

This solution was allowed to stand for a few days and then solid $NaHCO_3$ was added. A precipitate formed as neutrality was approached and then completely dissolved as the pH neared 7.0. The final approach to the desired pH of 7.1 was made by the addition of a saturated Na_2CO_3 solution. A total of 4.3 mole of CO_3^{2-} was used per mole of U and the final solution was about 40 L.

The isolation and purification of the plutonium involved primarily relatively standard chemical procedures. The most novel was the first step that concentrated the plutonium from the large mass of uranium by extracting the cupferron complex [18] into chloroform from the slightly alkaline carbonate solution. There followed chromatographic column removals of ^{234}Th and Fe^{3+}, column purifications of the plutonium according to the procedure of Hoffman [19], and then evaporation of the nearly-mass-free plutonium-containing solution on a platinum disc.

The final sample was measured on three different silicon alpha detectors. The first two were rather large-area detectors and showed 5 and 2 times the counting rate of ^{239}Pu of the third small detector. This was due to the rather wide dispersal of the sample over the platinum disk. The smaller detector (MCA1) had more than 10 times lower background and gave the most definitive results. Three different counters were used to minimize the uncertainty in the stability of the background rates when dealing with measurement times (many months) as long as those involved in this experiment.

Results.—Over the last three years, successively larger samples (experiments A-3, A-4, and A-5) of the 1956 uranyl nitrate have been worked up. Preliminary results [16] from A-3, giving a double beta decay half-life limit of 5×10^{19} yr, were presented in 1988. The value from our work on sample A-4, presented by Moe [17], was a limit of $> 10^{20}$ yr.

The data obtained from the present experiment (A-5), using the University of Chicago counter MCA1, are presented in Fig. 1. Shown are the number of events as a function of the energy of the alpha particles. The large peak on the left of Fig. 1 is from the ^{239}Pu tracer added at the start of the experiment. This served as a measure of the chemical recovery of the plutonium and of the particular counting efficiency.

Just to the right of this main group are the small number of alpha particles that can be identified by their energy as ^{210}Po (T_a=5.3 MeV), always present as contamination. Adequately separated from the ^{210}Po alpha particles, in the energy region between 5.35 and 5.55 MeV, are fourteen events (the correct region to be alpha particles of ^{238}Pu). Their distribution with energy is consistent with that expected from ^{238}Pu.

At somewhat higher energy, up to 6 MeV, are a small number of events whose origin is not completely understood. They all appeared in the first 70 d of measurement on this counter (months after sample preparation) and do not appear to be bunched up enough at 6 MeV for all to be ^{218}Po. In contrast, six of the fourteen observed 5.5-MeV events appeared in the last 78 d. There were no events registered between 6 and 7 MeV, thus setting adequate limits on other possible contaminants.

Table I summarizes the experimental data on the sample from experiment A-5 as obtained on three different alpha counters. All indicate excesses at the 5.5-MeV region. The excesses, corrected for the counter efficiencies, are consistent in indicating about 2.6 dis/d in the 8.47-kg uranyl nitrate sample. Interpreted as ^{238}Pu, they corre-

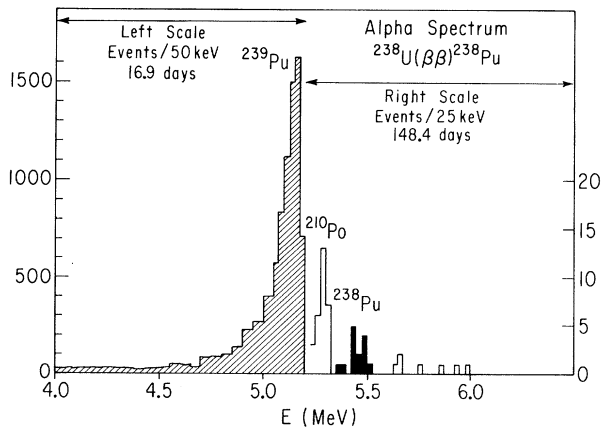

FIG. 1. A sample of the results from experiment A-5 on the University of Chicago counter MCA1 on the production of ^{238}Pu from ^{238}U. Shown are the numbers of alpha particles as a function of energy.

spond to the production of $(1.2\times0.4)\times10^5$ atoms of ^{238}Pu, or a half-life of 2.0×10^{21} yr for the decay of ^{238}U to ^{238}Pu. This rate is a factor of 40 longer than our previously published limit.

The possible contamination by ^{238}Pu fallout of the procedure or by chemicals has been mentioned. Evidence against this are the results from two previous experiments (A-3 and A-4) that, although starting with smaller amounts of uranyl nitrate, had, after the first steps, used mostly the same chemicals and procedures as experiment A-5. The net counting rates were 0.03 ± 0.04 and 0 ± 0.04 event/d on counter MCA1. After the A-5 experiment a blank run involving $NaHCO_3$ from the same large batch of salt and comparable in amount was put through almost the same chemistry. The final sample, corrected to the A-5 yield, measured 0.006 ± 0.014 event/d. Finally, a sample of the La^{3+} solution and HF (the only chemicals not used in previous experiments or the blank run) was tested and found to have no ^{238}Pu. These experiments make it very unlikely that the ^{238}Pu measured in A-5 is the result of fallout contamination of the chemicals or procedure.

As a final test, the measured sample of A-5 was removed from the platinum and subjected to an additional column purification that is specific for plutonium [19]. Within statistics, the resulting sample had the same ratio of ^{238}Pu to ^{239}Pu alpha particles.

The production rate of ^{238}Pu indicated by the present experiment is so low (~10 atoms/d) that reactions other than double beta decay for producing them must be considered. Since the sample analyzed contained no atoms heavier than ^{238}U, this nucleus would have to be the target of competing processes. The most serious competing reaction is the (p,n) process on ^{238}U leading to ^{238}Np. The protons could originate either from the intrinsic radioactivity of the sample or from external radiation (cosmic rays or other external particles). Because the (p,n) cross section on ^{238}U was not known, it was determined at the Los Alamos Van de Graaf accelerator. This cross section rises from negligible values at 6 MeV to ~5 mb at 12 MeV. A literature [20] value of 4 ± 2 mb at 14 MeV is consistent with this behavior. Using these data, the conversion of protons born inside a large $UO_2(NO_3)_2\cdot6H_2O$ sample into ^{238}Np was calculated to be only $\sim10^{-6}$ at 12 MeV. This is in contrast to the 50–100 times larger (p,n) probability in large samples of C_2Cl_4 or $8M$ $GaCl_3$ solution [21].

In spite of the high radioactivity of the sample (several mCi of alpha and beta activity and $\sim10^3$ spontaneous fissions per minute) the charged particles from this radioactivity are either too low in energy or too few at high energy to convert the ^{238}U to significant amounts of ^{238}Pu directly or indirectly.

The neutrons emitted in spontaneous fission are somewhat more serious because of their larger numbers. However, less than 1% are above 8 MeV, only a few percent of these will give rise to protons above this energy, and the low (p,n) yield per proton in $UO_2(NO_3)_2\cdot6H_2O$ leads to negligible numbers of ^{238}Pu. Thus no internal source for producing anywhere near 10^5 atoms of ^{238}Pu from 10^{25} atoms of U in 33 yr has been identified.

The possible production of ^{238}Np by cosmic rays during the long sea-level storage of the sample was estimated using data furnished by Davis [21]. In a large tank of C_2Cl_4, Davis found a conversion into ^{37}Ar of 6.6×10^{-23} per atom/yr due to cosmic rays and a similar number for the conversion of ^{71}Ga to ^{71}Ge in $8M$ $GeCl_3$. Interpreting these numbers as due to protons born inside the sample due to the different components of cosmic rays, and taking into account the different cross sections and stop-

TABLE I. Alpha measurements on sample A-5.

Counter	Chicago MCA2A	Los Alamos TA-48, No. 84	Chicago MCA1
Efficiency [a]	0.123	0.045	0.022
Background rate [b]			
Events observed (d)	12 (43.2)	14 (86)	4 (370)
Events/d	0.28 ± 0.08	0.16 ± 0.05	0.011 ± 0.006
Sample A-5			
Events observed (d)	16 (37.7)	14 (36.8)	14 (148)
Net events/d	0.14 ± 0.14	0.22 ± 0.11	0.084 ± 0.027
^{238}Pu produced (dis/d)	1.1 ± 1.1	4.9 ± 2.5	3.8 ± 1.2
Average dis/d [c]		2.6 ± 0.8	
Average atoms ($\times10^{-5}$)		1.2 ± 0.4	
$t_{1/2}(\beta\beta)\times10^{-21}$ yr [d]		2.0 ± 0.6	

[a]The efficiency is the product of the chemical recovery of the added ^{239}Pu tracer (5.76 dis/min) and the counter efficiency.

[b]All event rates are applicable to the 5.5-MeV region of ^{238}Pu. The tabulated background value is the result of 4 events in 298 d before experiment A-5 and 0 event in 72 d after A-5.

[c]This is the weighted average; the errors are the statistical (1σ) errors.

[d]Calculated from the number of atoms (1.02×10^{25}) and the decay time (33 yr).

ping powers involved, we calculate a production of only 200 atoms of ^{238}Np during the 33-yr storage of our sample.

An additional possible contaminating nuclear reaction must be considered in our case because, for about 14 of the 33 yr, the sample was stored near the control room of the 450-MeV University of Chicago cyclotron. A check of the radiation exposure records of operating personnel suggests a very conservative upper limit of 1 neutron/cm^{2}sec of 15–30-MeV neutrons during this period, which would lead to a production of less than 10^4 atoms of ^{238}Pu in our sample.

Thus no source of contaminating nuclear reactions has been identified that would produce even 10% of the 5.5-MeV alpha particles that have been observed.

Discussion.—The observation of a half-life of 2×10^{21} yr for the transformation of ^{238}U to ^{238}Pu is only the sixth nuclear system for which definitive evidence has been obtained for the occurrence of a double beta decay process. This half-life can be considered in comparison with the half-lives of other cases, with theoretical predictions, and in relation to current uncertainties about the numbers and types of neutrinos.

The comparison with other cases of double beta decay is best made via the implied matrix elements for the relevant nuclear transformation on the assumption that the process responsible is the most conventional type of two-neutrino, two-electron decay. The removal of the phase-space factor takes care of the approximately seventh-power dependence of the rate on the Q value of the transition. Using the phase-space factors of Boehm and Vogel [22] leads to M^2 values for the ^{238}U transition in the same range as deduced for ^{130}Te and ^{76}Ge. On the other hand, the latest theoretical estimates [9] give an upper limit that is 10 times lower. This large discrepancy implies either a defect in the calculations or the presence of a faster path than the standard two-neutrino mode in this case.

In considering possible alternate scenarios, the rate of transformation of uranium to plutonium as determined by this experiment has special features. The rate determined is the sum of all possible paths to the final state. The low-Q value enhances the relative importance of processes with small numbers of emitted particles. The large-Z environment also favors the concentration of the available energy into smaller numbers of particles. It is thus important to confirm or resolve the present discrepancy between the observed and calculated rates of transformation of ^{238}U to ^{238}Pu by the standard two-neutrino process.

We are pleased to acknowledge the help of W. Haxton in early discussions, of F. Lawrence and S. Knight of Los Alamos in the development of the plutonium chemistry, of G. Butler and the counting facilities at Los Alamos, and of E. Blume and K. Wielgoz in the chemistry and counting operation at Chicago. J. Wilhelmy of Los Alamos kindly arranged the measurements of the ^{238}U(p,n) cross section at the Los Alamos Van de Graaf accelerator. A. Staudt and H. V. Klapdor kindly furnished the results of their calculation before publication. This work was supported by the U.S. Department of Energy, Grant No. DE-FG02-88ER40450.M003.

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