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FINAL REPORT

Double Beta Decay of Uranium-238;

Proton reactions of ^{238}U in 5-12 MeV Range

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ABSTRACT

This report is in two parts.

The first part reports on the experimental work determining the half-life for double beta decay of ^{238}U to ^{238}Pu to be $(2.0 \pm 0.6) \times 10^{21}$ years. This is the first evidence for a third mode of decay of this heaviest naturally occurring nucleus. This rate is about 10^6 times slower than spontaneous fission, which itself is about 10^6 times slower than alpha decay. The implication of this double beta decay to neutrino masses depends on uncertain theoretical calculations of the rate for such a heavy nucleus. This work has been published [*Phys. Rev. Lett.* **67**, 3211 (1991)].

The second part reports on yields of principal fission products from 5.6, 7.3, 9.4, and 11.5 MeV proton interactions with ^{238}U . The yields at 11.5 MeV are similar to those from 14 MeV neutron fission of ^{238}U . At the same time, the production cross sections of ^{238}Np at the same energies were determined. This nuclide is produced as often as fission at the lowest energy but only 3.8% as often at the highest energy.

PART I

A. Introduction

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 invoked 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. Double beta decay also tests our understanding of the properties of excited states of nuclei, since these are involved in calculating the rate of such decay. At present, there are about thirty experiments, worldwide, studying various aspects of this process. However, it is one of the slowest spontaneous decays in nature ($\lambda \lesssim 10^{-28} \text{ sec}^{-1}$) and there have been, to date, only five systems for which experimental evidence has been obtained for its actual existence.

The possibility of spontaneous nuclear decays with $\Delta Z = 2$, $\Delta A = 0$ was first treated, in terms of modern theory, by M. Mayer in 1935 (Ma-35), following a suggestion by E. Wigner. M. Mayer calculated a half-life for the simultaneous emission of two beta particles and two (anti) neutrinos to be greater than 10^{17} yr.

The first experimental evidence for the occurrence of such a process was the discovery of anomalies in the isotopic composition of old tellurium minerals by Inghram and Reynolds in 1949 (In-49). Since that time their geochemical technique has been used to study the phenomenon in ^{82}Se , ^{130}Te , and ^{128}Te . Recently (after completion of this contract work) this geo-

chemical technique has been used to measure carefully the rates of double beta decay in ^{130}Te and ^{128}Te at Washington University (Be-93), with important implications for the mass of the antineutrinos that are involved. The special feature of this geochemical approach is that it measures the total rate of transformation, irrespective of the mechanism.

Recently, counter experiments have achieved the sensitivity required to detect this very rare process. Results on ^{76}Ge , ^{82}Se , and ^{100}Mo have been published [see reviews by Moe (Mo-93) and Avignone (Av-88)], as well as lower limits on the rates in other nuclides.

Theoretical reviews of double beta decay have been made by Haxton and Stephenson (Ha-84), Doi et al. (Do-85), Muto, Bender, and Klapdor (Mu-89), and Tomada (To-91).

The recent revival in interest in the process of double beta decay has been sparked by the realization that this process probes the possibility of alternate modes of transformation, such as the emission of two electrons and *no* neutrinos. The standard formulation of such processes requires that neutrinos have a non-zero mass.

The possibility that neutrinos have such a non-zero mass has recently been invoked to explain the dearth of measurable neutrinos from the Sun (Da-90). Other suggestions that, even in a single beta decay, a neutrino of 17 keV may be involved (Si-85), have recently been discounted.

The theoretical calculations of the rates of double beta decay have had a checkered history. The early predictions were generally faster than the geochemical results that were the first experimental values available. About the time that the geochemical result on ^{82}Se was confirmed by the time projection chamber result of Elliott et al. (El-87), theoretical reasons were proposed by Vogel (Vo-86) to explain this suppression. A history of

these theoretical predictions for ^{238}U is presented in Table I. At the time that the present work was completed, the predictions of Staudt et al. (St-90) were the latest available. Since then a preprint has been received (Wu-92) reporting on a new theoretical approach that predicts a half-life close to that observed in this work, but in poor agreement with the experimental values at lower mass numbers.

Table I
Theoretical Calculations on the Double Beta Decay Rate of ^{238}U

1986	2.2×10^{19}	Grotz and Klapdor (Gr-85)
1987	1.3×10^{20}	Haxton et al. (Ha-83)
1990	1.5×10^{23}	Staudt, Muto, and Klapdor-Kleingrothaus (St-90)
1992	0.91×10^{21}	Wu et al. (Wu-92)

This large scatter in predicted values — a range of 10^4 — reflects the difficulty of calculating the nuclear matrix elements for standard two-neutrino double beta decay, particularly for a nucleus as heavy as ^{238}U .

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 one of the lowest of the systems presently being studied. The low Q value allows competition by other than standard 2ν processes (such as 0ν or Majoran emission) to benefit from phase-space considerations. Thus, Staudt, Muto, and Klapdor-Kleingrothaus (St-90) predict that a neutrino with a value of the Majoran mass parameter $\langle m_\nu \rangle$ equal to 3 eV would lead to a zero-neutrino double beta decay rate ($t_{1/2} = 3 \times 10^{22}$ 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}).

The only previous experimental study of such a decay of ^{238}U is early work by Levine, Ghiorso, and Seaborg (Le-50). 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.

Another 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 $^{238}\text{U}(p,n)^{238}\text{Np}$. The ^{238}Np then decays with a 2.117-day half-life to ^{238}Pu . As part of the studies of competing reactions, the cross section for this process was determined at Los Alamos (see Part II of this report).

A serious practical consideration in dealing with the $^{238}\text{U}(\beta\beta)^{238}\text{Pu}$ system is the worldwide fallout of ^{238}Pu from atmospheric nuclear tests and from 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 (Be-71). Since the present experiment produced only $\sim 10^5$ 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 (Ha-83) revived interest in the ^{238}U system and the present work was started as a result of their publication. The present 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.

An addition interest in the double beta decay of ^{238}U arises from the large Z environment in which the process occurs. The electron density in the uranium nucleus is about 10^{27} electrons per cm^3 . This is much higher than the electron density at the center of the Sun, or even in supernovae. Thus, any effect of electron density on the effective mass of the anti-neutrinos involved in beta decay would affect the rate of this process, or at least provide limits on its importance.

A final, historical comment is appropriate for this study of the double beta decay of ^{238}U . It is almost 100 years since Becquerel (Be-96) first discovered radioactivity in studying uranium salts. In 1939, Petrzhak and Flerov (Pe-40) discovered the spontaneous fission of ^{238}U with a half-life of 2×10^6 times longer than that of the alpha decay studied by Becquerel. The present study of the double beta decay of this nuclide establishes the rate to be 3×10^6 times longer than the age of the universe and is one of the longest half-lives for which experimental data are available. These historical notes document increases in experimental sensitivity of about 10^6 every fifty years.

A fourth, energetically possible mode of decay of ^{238}U is that of emission of complex nuclear fragments such as ^{14}C (Ro-84). Such modes of decay have been seen in lighter nuclides, but have not been reported yet for ^{238}U . Thus, at present, three spontaneous decay modes of ^{238}U have been estab-

lished experimentally: alpha decay, spontaneous fission, and double beta decay.

B. Plutonium tracer

An early decision was made to use ^{239}Pu to measure the efficiencies of the chemistry and measurement processes. A radioactive tracer was indicated because of the lengthy chemistry and lack of information about the levels of contamination to be encountered. *Since less than 10^6 atoms of ^{238}Pu were to be extracted from 10^{25} atoms of ^{238}U , unknown modes of decay of this nuclide in addition to those of impurities might be encountered.*

There were appreciable advantages in *not* using ^{239}Pu as a tracer. If none had been added at the start, the amount present at the end would have provided information about the nuclear history of the sample, such as the history of exposure to cosmic ray neutrons.

However, no other suitable plutonium tracer was available. Other tracers were either too short-lived or not available with certifiably low levels of ^{238}Pu content. Even the ^{239}Pu that was easily available had too much ^{238}Pu content. Special tracer was obtained by irradiation of uranium in the thermal column of the Omega West reactor at Los Alamos. The neutron flux at the irradiation position was too low to produce ^{238}Pu by secondary reactions (the main mechanism for ^{238}Pu production in production reactors). After irradiation of the uranium, the plutonium was separated by standard chemical techniques. (We are indebted to Dr. Knobeloch of Los Alamos National Laboratory, INC-11, for help in obtaining this tracer.)

In order to be sure that the tracer was suitable, samples were measured on the same counter as used in the main experiment. The ratio of

$^{238}\text{Pu}/^{239}\text{Pu}$ alpha particles was less than 10^{-5} — the limit of the counting statistics. In addition, several years after preparation, another test was made for the presence of 5.5 MeV alpha particles in the tracer. This might have occurred if ^{241}Am had somehow gotten into the tracer. This test confirmed the absence of such alpha particles in the tracer solution.

C. Chemical Operations

The first experiments in this program were performed on ~14 kg of depleted uranium (as uranyl nitrate) furnished by the Los Alamos National Laboratory. This sample was dissolved and converted into carbonate by the addition of solid NaHCO_3 . The pH was adjusted to about 7.1 by the addition of a saturated solution of Na_2CO_3 . After addition of a cupferron solution, the plutonium cupferrate was repeatedly extracted with chloroform. About 32 successive extractions of the plutonium cupferrate were made.

Sampling of these extracts showed that the original uranium was heavily contaminated with ^{238}Pu . Even after the large number of extractions, the levels of this contamination were too high to anticipate getting to the low levels needed for this experiment. Work with this sample was abandoned and the solution of uranium isolated.

Although this original experiment was unsuccessful, it provided good experience under the tutelage of F. Lawrence and S. Knight of the Los Alamos National Laboratory, who instructed the authors with their experience in plutonium chemistry.

The contamination of this sample was fortunately overcome by the discovery of ~15 lb of uranyl nitrate that had been bought in 1956 and kept stored in doubly sealed containers in Chicago during the intervening period. This material had been purified by the Shattuck Chemical Co. of

Denver in about July, 1956. At that 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.

This material had originally been bought with funds shared with the late Professor N. Sugarman under a contract with the U.S. Atomic Energy Commission. The purpose then was to look for heavy particle (other than alpha) decay of uranium. Such a decay of heavy nuclei by the emission of heavy particles such as ^{14}C has since been discovered in other trans-lead nuclei (Ro-84). It undoubtedly occurs in ^{238}U but has not been seen yet.

After the positive results with this sample were obtained (see below), it was confirmed that the isotopic composition of the original uranium was that of normal uranium. There was no evidence for the presence of ^{236}U , supporting the conclusion that this uranium had neither been in a reactor nor been subjected to isotopic distortion. We are indebted to E. Perrin of the INC Division of the Los Alamos National Laboratory for the careful isotopic analyses of these samples.

Successively larger samples of this 1956 material (Expt. A-3, 800 g; Expt. A-4, 1.6 kg) were put through the chemistry (see below) with negative results for ^{238}Pu . The results of those experiments have been reported (Tu-88 and Tu-90).

In August 1989, the remainder (8.47 kg) of the uranium nitrate was dissolved in deionized water and a known amount of ^{239}Pu tracer solution

(see above) was added to this acid solution. This experiment (A-5) provided positive evidence for the double beta decay of ^{238}U .

The principal chemical problem in this project was the first extraction of the plutonium from the large quantities of original uranium. This was complicated by the existence of many valence states of plutonium as well as the tendency of plutonium to hydrolyze and attach itself to colloidal particles.

Both of these problems were approached by working in a carbonate solution. This chemistry was suggested by Dr. G.A. Cowan (Co-84). Plutonium in the +4 valent state forms a very stable carbonate complex and the equilibrium constants favor the conversion of all other valence states to the tetravalent plutonium complex (Ca-83). In addition, the cupferron complex of this +4 valence state extracts well into cupferron from carbonate solutions of pH of ~7.1.

In preparing this slightly basic solution from a solution of uranyl nitrate by the addition of solid NaHCO_3 , the uranium first precipitates and then redissolves by the addition of more carbonate. Typically, a total of ~4.3 moles of CO_3^{--} were used per mole of uranium and the final solution was about 0.5M in uranium.

Most other cations do not extract with cupferron under these conditions. The slight basicity also enhances the stability of the cupferron, which tends to decompose in more acid solutions.

The extraction of the plutonium at this stage could be followed by monitoring the radioactive emissions of the daughters of ^{234}Th that follow the plutonium at this stage. Since the uranium in Expt. A-5 had several mc of ^{234}Th activity, this could easily be done, originally externally, and later by measuring the beta activity of aliquots of different phases.

The extractions were carried out in ~2-liter batches using about 200 ml CHCl_3 per extraction in ~2.5-l acid bottles using a mechanical shaker. This could shake 4 batches at a time for ~30 min. After phase separation, the aqueous layer was shaken with a second portion of CHCl_3 . In all cases the CHCl_3 used had first been saturated with gaseous CO_2 in order to maintain the pH of the aqueous phase.

A complication that showed up in the experiment starting with the large amounts of uranyl nitrate was the appearance of a large interface between the aqueous and chloroform layers. Radiation measurements indicated a significant amount of gamma activity in this phase and measurements on an aliquot showed ^{239}Pu concentration appreciably higher than in the bulk organic phase.

Because it was expected that this interface had contributed to the lower than expected yields in earlier experiments, this interface layer was worked up separately. The material was evaporated to remove the organic material, fused with Na_2CO_3 , and, after dissolving and addition of La^{+++} carrier, a LaF_3 precipitate made several times to concentrate the plutonium. The last LaF_3 was dissolved in H_3BO_3 , a hydroxide brought down several times and then taken up in HCl and added to the main extractant.

Meanwhile the main organic extractant had been filtered through paper soaked with CHCl_3 and evaporated to dryness in quartz, the organic matter burned off and the residue (obviously mostly Fe) taken up in 9N HCl . At this point all the UX activity of the original uranium was in the sample.

The subsequent chemistry had as objective the removal of UX (Th) and of the iron that had extracted with the plutonium. The first objective was achieved by passing the 9M HCl solution through 3 successive cation

columns (AG50-X4) that held on to the Th^{4+} ions while allowing the halide-complexed, negatively charged plutonium to pass through.

The thorium removal in these steps was very effective. Even the first column reduced the gamma activity below externally measurable levels. In spite of much preparation work, however, it is suspected that plutonium losses occurred at this stage, possibly due to inadequate valence state preparation.

The iron was removed by first adjusting the pH to 5 and then passing the solution through an anion column (AG 1×10 — 200–400 mesh) that adsorbed the more stable chloride complex of iron, allowing the plutonium to go through.

The plutonium was finally purified by the procedure of Hoffman (Ho-90). After adsorbing on a small column in the +3 valence state, it was eluted by oxidizing to the +4 state. This procedure was repeated before evaporating the small eluent on platinum for alpha counting. The samples were ignited and covered with a thin film before alpha measurements.

The final samples differed in quality in the successive experiments, primarily because of the presence of inert material and the area extent of the deposit. The sample from A-3 was thin and concentrated. That from A-4 had more inert material, leading to a broader ^{239}Pu alpha spectrum. The sample from A-5 was spread over a larger area of the platinum. It showed a very narrow ^{239}Pu alpha peak, but the larger extent of the deposit made the counting efficiencies very dependent on the size of the alpha detector (see below). The A-5 sample after additional purification was again rather thick.

The final sample was measured on three different silicon alpha detectors. The first two were rather large-area detectors and showed (for sample

A-5) 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. This last, smaller detector (MCA1) had more than 10 times lower background and gave the most definitive results. The 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.

D. Results

Over the last three years of this program successively larger samples (experiments A-3, A-4, and A-5) of the 1956 uranyl nitrate have been worked up. Preliminary results from A-3, giving a lower limit for the double beta decay half-life of 5×10^{19} yr, were presented in 1988 (Tu-88). The value from our work on sample A-4, presented by Moe (Tu-90) was a limit of $>10^{20}$ yr.

The data obtained from the last 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 (the correct region to be alpha particles of ^{238}Pu), are fourteen events. 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 days 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 days. There were no events registered between 6 and 7 MeV, thus setting adequate limits on other possible contaminants.

Table II 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 correspond to the production of $(1.2 \pm 0.4) \times 10^5$ atoms of ^{238}Pu in sample A-5, 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 20 longer than our previously published limit.

The possible contamination of the procedure or chemicals by ^{238}Pu fallout 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.

Table II
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 ($\times 10^{-5}$)		1.2 ± 0.4	
$t_{1/2}(\beta\beta) \times 10^{-21} \text{ yr}$ ^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 in counter MCA1.

^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).

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 (Ho-90). Within statistics, the resulting sample had the same ratio of ^{238}Pu to ^{239}Pu alpha particles as the original sample.

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 ~ 6 mb at 12 MeV (see Part II of this report). Using these data, the conversion of protons born inside a large $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ sample into ^{238}Np was calculated to be only $\sim 10^{-6}$ 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 (Da-85).

In spite of the high radioactivity of the sample (several mCi of alpha and beta activity and $\sim 10^3$ 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 $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ leads to negligible numbers of ^{238}Pu . Thus no internal source for producing anywhere near 10^5 atoms of ^{238}Pu 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 (Da-85). 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 stopping 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/cm²sec 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.

E. 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 (Bo-92) leads to M^2 values for the ^{238}U transition smaller than ^{76}Ge or ^{100}Mo , but larger than ^{82}Se , ^{128}Te , or ^{130}Te . On the other hand, the theoretical estimates of the 2ν process in ^{238}U (St-90) predict a half-life that is even longer than observed in this work. A more recent theoretical calculation (Wu-92) predicts a value for ^{238}U close to that observed but also predicts much too fast rates for ^{128}Te and ^{130}Te . These large discrepancies imply either continued problems 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 the present experiment has special features. All possible paths to the final state contribute to the rate determined by this experiment. The low Q value enhances the relative importance of processes with small numbers of emitted particles, such as zero neutrino or Majoran-involved decay. The large- Z environment also favors the concentration of the available energy into smaller numbers of particles. Thus, the present experimental result together with the calculations of Staudt, Muto, and Klapdor (St-90) would imply an effective mass of about 14 eV for the antineutrinos involved in ^{238}U double beta decay. This is a much larger mass for the antineutrino than limits placed by other experiments (Be-93, Mo-93), and such a conclusion must await confirmation of the experimental result of this work and clarification of the theoretical situation.

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

PART II

A. Introduction

In spite of the long-time interest in the nuclear properties and reactions of the heaviest nuclides there appears to be only sparse data on the simple (p,n) reaction on the common nuclide, ^{238}U . The cross section between 14 and 16 MeV has been determined by Ageev et al. (Ag-67) with approximate values of 7 ± 2 and 11 ± 2 MeV. Somewhat more precise values are available in the region 12–18 MeV from the work of Aaltoner et al. (Aa-90). Aside from interest relative to competition with the fission of excited ^{238}Np , as studied by Choppin et al. (Ch-63) and Bate and Huizinga (Ba-64), the cross section for this reaction is important in evaluating background effects in experiments on double beta decay of ^{238}U , as proposed by Haxton et al. (Ha-83) and recently measured by Turkevich, Economou, and Cowan (Tu-91).

In the present work the formation probability of ^{238}Np by protons reacting with ^{238}U in the energy range 5–12 MeV was determined by proton irradiation of thin uranium foils and comparison of the intensities of gamma rays from ^{238}Np with those of fission products. A cross section was calculated by using the fission cross section of Bate and Huizinga (Ba-64).

In addition, more detailed data were obtained on the fission product distribution of proton-induced fission in this energy range than was possible from the electronic measurements of fission products (Ch-63, Ba-64).

B. Experimental

Depleted uranium foils, 52 mg cm^{-2} , 27 mm in diameter, were sandwiched between 6.5 mg cm^{-2} Al foils and backed by a tantalum beamstop. The irradiations were performed at the Los Alamos Van de Graaf accelerator, whose proton energy was known to $\pm 30 \text{ keV}$. The beam was restricted

by a carbon collimator to the central 8 mm of the target foils. Single irradiations at incident proton energies of 6, 8, 10, and 12 MeV were performed for times up to 95 min at the lowest energy. At this lowest energy the target consisted of two uranium foils in order to determine the effects of background particles on the lowest cross sections expected here. The proton intensities were approximately 1 μ amp.

The incident proton energies at the Van de Graaf were accurately known. The average energies of the protons in the foils, taking into account the energy loss in the aluminum and uranium, are shown in column 1 of Table IV.

After irradiation and a short decay period, the foils were mounted in a standard fashion and the emitted gamma rays were measured on a Ge-Li detector at a position where the photopeak efficiencies were determined by comparison with NBS standards. The efficiency was about 0.5% at 1 MeV. At the lowest energies (below about 300 keV), the relative efficiencies were checked via the gamma rays associated with the decay of ^{238}U in the target foils. The self-absorption of the lower energy gamma rays in the uranium was corrected for by calculation.

Gamma measurements were made 1, 2, and 3 days after the irradiation. The gamma-ray spectra of the irradiated foils showed the expected lines from the ^{238}U decay products, fission product gamma rays, and, in addition, the 984.47 (27.8%), 1025.88 (9.6%), and 1028.5 (20.3%) gamma rays of 2.12-day ^{238}Np . The numbers in parentheses are the percentages of ^{238}Np decays (Le-78) giving rise to the particular gamma rays. The back-up uranium foil at 6 MeV showed no ^{238}Np or fission product gamma rays. The small contribution from the ^{132}Te chain to the 984 keV gamma ray of ^{238}Np could be neglected as confirmed by the intensity of the 22 times larger

954.4 keV branch of the same decay. At the highest energy of this work, the 1024.5 keV gamma ray of ^{97}Zr chain ($t_{1/2} = 16.9$ d) made the ^{238}Np estimate at this energy more uncertain at early times. At the lowest energy, long-lived contributors of unknown origin at 984 keV amounted to about 25% at 3 days. Otherwise, the estimates from the two gamma-ray regions agreed to better than 3%. The produced numbers of atoms in the separate irradiations (column 5 in Table IV) should therefore be good to at least 10%.

C. Fission Product Yields

The gamma-ray spectra of the irradiated uranium foils made possible the identification and estimates of the production probabilities of many fission products. The observed intensities were corrected for decay, for the fraction of the gamma-ray peak used, for the efficiency of detection, for the branching ratios of the gamma rays (Le-78), and for recoil losses. These latter were measured for many fission products in the upstream Al foil in the 11.8-MeV irradiation. The fractional recoil losses were assumed to be the same in both hemispheres, and independent of energy except at 4.8 MeV, where only upstream losses were assumed to occur. This was because of the large energy loss of the proton in the foil and the steep excitation function at this low energy. At higher energies the total recoil losses were about 12% for the light fragments and 7% for the heavy fragments.

Table III presents the results of the fission product measurements for the four irradiations. For each identified fission product the number of atoms produced is given (together with the number of gamma rays used, if more than one). The error in these numbers is estimated to be $\pm 5\%$ unless a larger number is indicated due to poorer statistics or poorer agreement between results from different gamma rays.

Table III
Fission Product Production
(Number of Atoms)

A	Nuclide	11.5 MeV ($\times 10^{-8}$)	9.4 MeV ($\times 10^{-8}$)	7.3 MeV ($\times 10^{-7}$)	5.6 MeV ($\times 10^{-6}$)
91	Sr	3.03 ± 0.15 (2)	1.30 ± 0.07 (2)	1.6 ± 0.2 (2)	
93	Y	5.0 ± 0.4 (2)	2.46 ± 0.25	2.8 ± 1.4	
95	Zr	4.72 ± 0.24	1.95 ± 0.18	3.8 ± 1.4	
97	Zr, Nb	5.39 ± 0.27 (2)	2.06 ± 0.11 (2)	3.33 ± 0.17 (2)	1.0 ± 0.1 (2)
99	Mo-Tc	6.15 ± 0.35 (4)	2.46 ± 0.13 (2)	3.89 ± 0.19 (2)	1.4 ± 0.1
103	Ru	5.43 ± 0.27	2.24 ± 0.12	4.2 ± 0.3	0.6 ± 0.3
105	Rh	4.13 ± 0.21 (2)	1.53 ± 0.08 (2)	2.49 ± 0.16	
109	Pd	2.1 ± 0.4			
111	Ag	1.8 ± 0.9	—		
112	Pd-Ag	1.03 ± 0.05 (3)	0.28 ± 0.02	0.47 ± 0.10	
115	Cd	0.81 ± 0.04	0.153 ± 0.016	0.17 ± 0.06	
125	Sn	0.9 ± 0.3	—	—	
127	Sb	1.78 ± 0.09 (2)	0.547 ± 0.027 (2)	0.59 ± 0.11 (2)	—
128	Te	1.38 ± 0.26 (2)	—	—	—
131	Te	1.48 ± 0.07 (6)	0.487 ± 0.013 (6)	0.76 ± 0.17 (3)	—
	I	3.28 ± 0.16	1.50 ± 0.08	2.67 ± 0.13	—
	Total	4.76 ± 0.24	1.99 ± 0.10	3.42 ± 0.19	—
132	Te	4.90 ± 0.24	1.72 ± 0.09	2.74 ± 0.14	1.0 ± 0.3
	I	4.12 ± 0.26 (6)	1.72 ± 0.09 (5)	2.8 ± 0.14 (5)	0.71 ± 0.09 (2)
133	I	5.99 ± 0.30 (2)	2.34 ± 0.12 (2)	3.69 ± 0.18 (2)	
	Xe	6.63 ± 0.33	2.61 ± 0.13	3.68 ± 0.18	1.39 ± 0.14
135	I	4.64 ± 0.23 (3)	2.45 ± 0.28		
	Xe	6.07 ± 0.37	2.47 ± 0.13	3.94 ± 0.20	0.98 ± 0.14
140	Ba	4.25 ± 0.24	2.01 ± 0.09	3.26 ± 0.52	
	La	4.46 ± 0.23 (4)	1.75 ± 0.09 (4)	2.88 ± 0.21 (4)	1.49 ± 0.40
141	Ce	4.50 ± 0.23	1.98 ± 0.12		
143	Ce	3.91 ± 0.20 (2)	1.58 ± 0.08	2.60 ± 0.13	0.54 ± 0.22
147	Nd	2.4 ± 3.5			
149	Pm	1.5 ± 0.1			
151	Pm	1.04 ± 0.06 (3)	0.40 ± 0.02	0.71 ± 0.14	

Since the fraction of the incident proton beam actually incident on the foils was not known, the following procedure was used to calculate fission yields. It was assumed that the absolute yields of the most prominent fission products were independent of the energy or nature of the initiating particles. Thus, using the 14-MeV neutron fission yields in ^{238}U (Be-74), the number of fissions in each foil was calculated using only those products whose yields are greater than 3% in the neutron case.

Table IV shows the results. For each irradiation there are listed the number (9–17) of fission products used in this procedure, the calculated number of fissions (N_F), and the root-mean-square of this calculated number of fissions (5–15%).

Table IV
Calculated Cross Sections of $^{238}\text{U}(p,n)^{238}\text{Pu}$

T_P (MeV)	n	N_F	rms (%)	$N(238)$	σ_F^*	$\sigma(p,n)^{**}$
11.5	17	1.0×10^{10}	12	3.8×10^8	150	5.7
9.4	16	4.1×10^9	9	3.7×10^8	32	2.9
7.3	16	6.3×10^8	5	5.1×10^8	1.0	0.80
5.6 ^b	9	2.1×10^7	8	2.3×10^7	.017 ^a	0.019

* σ_F from curve in Bate and Huizinga (Ba-64). Values in mb.

**Values in mb.

^a Taken as 60% of the $^{235}\text{U}(p,f)$ cross section (2.8×10^{-2} mb).

^b At the lowest energy, the *incident* proton energy is given.

Using these numbers for the total number of fissions, absolute yields for *all* of the fission products that were measured could be calculated. These are presented in Table V and plotted in Fig. 2. For reference, the figure also shows the 14-MeV neutron fission product distribution (Bl-74).

A limit to the fission background in the area of the irradiation was obtained from the lack of ^{91}Sr (9.5 h) activity in the back-up uranium foil in the 6 MeV run. In the 1.0243 MeV region of ^{91}Sr (10 ± 9) events in 10^4 sec were present, corresponding to 4×10^6 fissions. Thus, at most 20% of the fissions in the main foil at this energy were due to background particles.

Fig. 2 shows that the main features of the fission product distribution in the 11.5-MeV proton reaction with ^{238}U are similar to those of the neutron reference curve. Although there was not enough intensity in this experiment to completely explore the valley yields, the data are consistent with similar peak-to-valley ratio in the proton and neutron interactions. The observed peak-to-valley ratio of about 10 is much higher than the value of 2.5 measured by Bate and Huizinga (Ba-64). Presumably this is due to the better mass resolving power of this work compared with the kinetic energy technique used by Bate and Huizinga.

Table V
Fission Yields (%)

A	11.5 MeV	8.4 MeV	7.3 MeV	5.6 MeV
91	3.2	2.5	2.5 ± 0.3	—
93	5.0 ± 0.4	6.0 ± 0.6	4.4 ± 2.2	—
95	4.7	4.8 ± 0.4	6.0 ± 2.2	—
97	5.4	5.0 ± 0.3	5.3 ± 0.3	4.8 ± 0.5
99	6.15 ± 0.35	6.0	6.2	6.7 ± 0.5
103	5.4 ± 0.3	5.5	6.7 ± 0.8	2.9 ± 1.5
105	4.1	3.7	4.0	—
109	2.1 ± 0.4	—	—	—
111	1.8 ± 0.9	—	—	—
112	1.03	0.68 ± 0.05	0.75 ± 0.16	—
115	0.81	0.37 ± 0.04	0.27 ± 0.10	—
125	0.9 ± 0.3	—	—	—
127	1.8 ± 0.1	1.3	0.9 ± 0.2	—
128	1.4 ± 0.3	—	—	—
131	1.5	1.3	1.2 ± 0.3	—
	3.3	3.5	4.2	—
Total	4.8	4.8	5.4	—
132	4.9	4.2	4.3	4.8 ± 1.6
	4.1	4.2	4.5	3.4 ± 0.4
133	6.0	5.7	5.9	—
	6.6	6.4	5.9	6.6 ± 0.7
135	4.6	6.0 ± 0.6	—	—
	6.1	6.0	6.3	4.7 ± 0.7
140	4.3	4.9	5.2 ± 0.8	—
	4.5	4.3	4.6	7.1 ± 1.9
141	4.5	4.8	—	—
143	3.9	3.9	4.1 ± 0.2	2.5 ± 1.0
147	3.4 ± 3.5	—	—	—
149	1.5 ± 0.1	—	—	—
151	1.06	1.0	1.1 ± 0.2	—

Fig. 2. Mass yields in the proton fission of ^{238}U . The top curve is the 14 MeV neutron curve from (B1-74). The abscissae are mass numbers. The ordinates are displaced by decades for clarity.

D. ^{238}Np Cross Section

As indicated in section C, the ^{238}Np gamma rays at 984 keV and in the 1027 keV region could be seen in the uranium from all four irradiations. After correcting the observed intensities for decay, the fraction of the peak used, the contribution from interfering gamma rays, the branching ratios, and the gamma ray efficiencies, the number of ^{238}Np atoms produced in each irradiation could be calculated. These are shown in column 5 of Table IV.

Column 6 of Table IV gives the fission cross section from Bate and Huizinga (Ba-64) at the appropriate energies. From the ratio of ^{238}Np production to the number of fissions, the cross section for the (p,n) reaction on ^{238}U can be calculated. This is shown in the last column of Table IV.

The data indicate that the (p,n) cross section is slightly larger than the fission cross section at the lowest energy of this work. This energy corresponds to an excitation energy of 10.9 MeV in ^{239}Np and indicates $\Gamma_F/\Gamma_{\text{total}}$ is less than 0.5. This is consistent with the value of 0.4 at 11 MeV obtained by Gavron et al. (Ga-76) in $\text{He}^3(d,f)$ counter studies.

PART III

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C. Publications, Talks, and Reprint

1. Publications

- 1988 "Chemical and geochemical experiments on double beta decay", A. Turkevich, G.A. Cowan, T. Economou, F. Lawrence, and S. Knight. In *Fundamental Symmetries and Nuclear Structure*, ed. J.N. Ginocchio and S.P. Rosen. Santa Fe, New Mexico, 1988 October 12, p. 186. World Scientific Press.
- 1991 "Double beta decay of ^{238}U ", A.L. Turkevich, T.E. Economou, and G.A. Cowan. *Phys. Rev. Lett.* **67**, 3211 (reprint attached).
- 1992 "Double beta decay of ^{238}U ", G.A. Cowan, A. Turkevich, and T.E. Economou. Paper presented March 10 (to be published in *Proceedings*) at the Galileo Conference, Venice, Italy.

2. Talks

- 1987 American Chemical Society Meeting, New Orleans, Louisiana. "Double Beta Decay of U-238".
- 1991 American Chemical Society Meeting, New York, August 27. "Double Beta Decay of U-238". A.L. Turkevich, t. Economou, and G.A. Cowan.

3. Reprint removed. *ds*

Phys. Rev. Lett. (1991), attached.