10
3-25-92 JSD

LBL-31601
UC-413

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

**Investigation of the Effect of Electric Fields on
the Rate of Alpha Decay**

R.F. Gaylord, D.M. Lee, and D.C. Hoffman

July 1991

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. Neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or The Regents of the University of California and shall not be used for advertising or product endorsement purposes.

This report has been reproduced directly
from the best available copy.

Available to DOE and DOE Contractors
from the Office of Scientific and Technical Information
P.O. Box 62, Oak Ridge, TN 37831
Prices available from (615) 576-8401, FTS 626-8401

Available to the public from the
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, VA 22161

Lawrence Berkeley Laboratory is an equal opportunity employer.

LBL--31601

DE92 009497

Investigation of the Effect of Electric Fields on the Rate of Alpha Decay

R.F. Gaylord, D.M. Lee, and D.C. Hoffman

Department of Chemistry
University of California

and

Nuclear Science Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

July 1991

This work was supported by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics, Nuclear Physics Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

INVESTIGATION OF THE EFFECT OF ELECTRIC FIELDS ON THE RATE OF ALPHA DECAY

R.F. Gaylord, D.M. Lee, D.C. Hoffman

July 1991

Possible alteration of the alpha decay rate of a ^{249}Cf sample by application of an electric field of the order 10^7 V/m was investigated. Gamma spectroscopy of the 388 keV level in the ^{245}Cm daughter was used to eliminate the uncertainty inherent in absolute alpha counting. Ratios of the intensities of the 388 keV photopeak of ^{249}Cf to photopeaks from both beta and electron-capture decay of ^{152}Eu incorporated in the same sample were measured to eliminate variability due to changes in either sample geometry, or possible physical erosion of the samples. No change in any of the decay rates was seen within the estimated limits of reproducibility of the measurements.

INTRODUCTION

Immediately after the discovery of radioactivity, investigators began looking at the possibility that the rate of nuclear decay could be altered by environmental changes. In 1901, both Becquerel¹ and Curie² studied the effect of temperature on the emanations of radium. Temperatures from -255°C to 1300°C had no measurable effect on the half-life of a ^{226}Ra preparation. Rutherford³ measured the decay of radon in a cordite bomb, subjecting the sample to a pressure of 1200 atm and a temperature of 2500°C , and saw no perturbation. Other researchers applied such physical stresses as acceleration⁴ to 970,000 g, and magnetic fields⁵ of 83,000 Gauss, but were unable to change the nuclear decay rate. The apparent imperturbability of half-life led these early researchers to conclude that radioactivity was a nuclear, not atomic, phenomenon.

Interest in half-life variability was rekindled in 1947 by Segre's prediction⁶ that the rate of electron-capture (EC) decay could be altered by changes in the chemical state of the radioactive atom, and Daudel's similar hypothesis⁷ for internal conversion (IC). Electron-capture is the process by which a neutron-deficient nucleus decays through the capture of an orbital electron, with the subsequent conversion of a proton, plus the captured electron, into a neutron. Internal conversion is a decay mode exhibited by long-lived excited states of nuclei whereby the nucleus directly transfers its excitation energy to an orbital electron, which is then ejected from the atom. Both EC and IC decays involve extranuclear electrons, and their rates are therefore sensitive to the probability of finding an electron near the nucleus, hence the chemical form of the decaying atom can affect the half-life. Both theories were soon proven experimentally⁸: the EC-decaying nuclide ⁷Be has a 1% longer half-life when in the compound BeF₂ compared to the metallic state, and the decay of the isomeric state of ⁹⁹Tc has a 0.3% change in half-life between the forms KTcO₄ and Tc₂S₇. Similar effects have been seen in such nuclides as ^{197m}Au(IT), ⁶⁴Cu(EC), ^{57m}Fe(IT), and ^{90m}Nb(IT). The largest effect⁹ seen yet is a 5.6% increase in half-life for ^{235m}U between the 4⁺ oxidation state and the 6⁺ oxidation state. Uranium-235m decays by internal conversion from a very low energy (73 eV) isomeric state with a 26 minute half life¹⁰. To date, no alteration has been seen in the half-lives for alpha or β⁺ or β⁻ decay due to environmental effects, although Alder et al.¹¹ predict that a 0.2% change in decay constant should be observed in weak alpha emitters if the screening potential, V_S, is changed through chemical effects.

It has been recently proposed by Barker¹² that electric fields, applied to alpha-emitting radionuclides bound in a dielectric host, are capable of altering alpha decay rates. The proposed mechanism is a coupling of the external field to the nuclear Coulomb barrier. The Coulomb barrier is a potential energy barrier created by the repulsion of the positively-charged nucleus, and the positively charged alpha particle (a nucleus of ⁴He). Barker has reported that a positive applied voltage raises the barrier,

and therefore slows alpha decay, while a negative applied voltage decreases the barrier, so that the probability of particle emission is increased, and the sample decays faster. He has seen no change in decay rate in samples that decay by β - emission.

Barker cites two experiments which give evidence for accelerated alpha decay¹³. The first involved exposing an uncovered ^{230}Th source (in the form of ThO_2) to a voltage of 150 kV, supplied by a Van de Graaff generator, for approximately 14 hours. Barker calculates that at the molecular diameter this gives a field of order 9.6×10^7 V/m, and at the nuclear diameter, a field of order 10^{20} V/m, or ten percent of the nuclear barrier potential. The samples were counted with a Si surface barrier detector before and after the treatment. Barker measured a 44% decrease in alpha peak count, and a 125% increase in full width at half maximum after the treatment. He interpreted this as indicating that the applied voltage had changed the barrier for alpha-particle emission, and therefore decreased the half-life of the ^{230}Th .

In the second experiment, Barker exposed sources of ^{241}Am to voltages from +20 kV to -25 kV. The sources were counted with a Geiger-Mueller tube before and after treatments of approximately 10-20 hours. Counts were taken both with and without a paper cover over the detector to count gamma rays alone, and alpha and gamma radiations together (alpha particles interact strongly with matter, and can be stopped by a sheet of paper). At an applied voltage of +20 kV, he observed a 25% decrease in count, while a voltage of -25kV produced an astonishing 828% increase¹³ in count rate. In all cases, Barker saw no change in the paper-filtered count, which is from gammas alone. (85.2% of the ^{241}Am decays populate an excited level at 59.5 keV in the ^{237}Np daughter, which then decays by emission of a gamma ray.) A copy of Dr. Barker's americium results is included as table 1.

In June 1990, Dr. Barker spoke to us about his results. In order to verify the effect he measured, we designed an experiment to determine if electric fields could indeed alter alpha decay rates.

PROCEDURE

We electroplated sources of the two mixed activities, ^{152}Eu and ^{249}Cf , onto platinum foils. Californium-249 is a pure alpha emitter, and 66% of its decays are accompanied¹⁰ by a 388.1 keV gamma ray. Europium-152 decays by both EC (72.1%), and β -decay (28.9%). It has three intense gamma rays: 344.3 keV from β -decay, and 121.8 keV and 244.7 keV from EC decay¹⁰. Information about these nuclides is included in table 2. In addition to measuring the absolute disintegration rate of the ^{249}Cf from its known 388.1 keV photopeak, we also planned to measure the ratios of the intensities of the ^{152}Eu gamma lines to each other and those from the ^{249}Cf . Using ratios minimizes the error inherent in efficiency calculations, and also reduces the sensitivity of the intensities to possible source erosion or geometry changes. If high electric fields really change the decay rate of alpha emitters, we would expect that the ratios of the ^{152}Eu EC and β -photopeak intensities to the ^{249}Cf photopeak intensity would change after treatment. We decided to use gamma, rather than alpha spectroscopy in order to ensure a more accurate absolute result, since absorption in the source is not a problem. Isotopes of europium and californium were chosen because they share similar chemical properties¹⁵, in particular, volatilities, so that any process that caused a physical loss of Cf from the source would also result in a loss of Eu. Both nuclides also have strong, well-separated gamma lines to allow ease of gamma counting.

Source Preparation. To prepare the sources, 100 λ aliquots of ^{249}Cf were mixed with 50 λ aliquots of ^{152}Eu . The resulting solutions were evaporated to dryness, the residues were picked up in isopropyl alcohol, and the alcohol solutions were electroplated onto platinum foils. The foils were mounted on aluminum counting cards, and covered with 1 mil Mylar film. Our first source contained 63.5 nCi of europium, and 30.0 nCi of californium, while the second contained 76.3 nCi of Eu and 33.6 nCi of Cf.

A representative spectrum of one of the sources is shown in figure 1.

The sources were counted immediately after preparation, and again three months later, after they had been remounted and immediately before they were subjected to high voltage. The gamma spectra were taken with a 148.5 cm³ high-purity germanium detector, run with a Canberra System 100 multi-channel analyzer, and spectral analysis was done using the SAMPO gamma spectroscopy program which was run on the LBL VAX 8600. The SAMPO method of spectral analysis described by J.T. Routti was used¹⁶. The detectors were calibrated before each count with a nine peak gamma ray standard (Amersham Laboratories. Source number: 1780QB).

Treatment. The two sources were sent to the Altran corporation in Sunnyvale, CA in November of 1990. There, the samples were subjected to high voltage fields generated by a Cockroft-Walton generator. The applied voltages ranged from -270 kV to -500 kV, and the total treatment time was in excess of 1000 hours. Barker's theory indicates that -500 kV is the optimum voltage to perturb alpha decay in californium¹⁴. Dr. Barker counted the samples with an NaI(Tl) detector before and after each treatment. He reported that his pre-treatment counts agreed within 3%. On May 8, 1991, after the full treatment, he reported that the intensity of the 388 keV photopeak of ²⁴⁹Cf had decreased by $9.0 \pm 2.0\%$ for both sources, and that the intensity of the 121.8 keV peak of Eu had also decreased by $1.5 \pm 1.0\%$ in count for both sources. The samples were returned to us on May 16, 1991.

Upon receipt, we reanalyzed both sources with the same gamma ray spectroscopy system that the pre-treatment data had been taken on. Unfortunately, during the intervening time, the detector had to be repaired, and the Lucite shelf arrangement that holds the sample cards was removed and subsequently replaced. This changed the detector geometry, affecting the efficiency calibration of the detector. However, the use of ratios of photopeak intensities from two different nuclides should eliminate the

variability due to geometry changes, since the errors will cancel each other. We experimented with several arrangements, comparing the detector efficiencies in order to recreate the pre-treatment geometry as closely as possible. A comparison of our pre- and post-treatment efficiencies is included as table 3. A total of 22 sample counts and 10 standard counts were taken at distances ranging from 1.5 to 11.5 cm from the detector face.

RESULTS

We looked at both the absolute intensity of the 388 keV ^{249}Cf photopeak, as well as the ratios of the intensities of the 121.8 keV Eu line to the 344.3 keV Eu line, the 121.8 keV Eu line to the 388.1 keV Cf line, and the 344.3 keV Eu line to the 388.1 keV Cf line. For our first sample, the ^{249}Cf 388.1 keV peak count rate was $42,065 \pm 3.5\%$ min^{-1} before treatment, and $42,743 \pm 2.5\%$ min^{-1} after treatment, an increase of 1.61%, well within our experimental error of $\pm 4.3\%$. The europium peak at 121.8 keV showed a decrease of 1.71%, again within our uncertainty of $\pm 4.9\%$. For our second sample, we measured an increase of 1.38% in the 388.1 keV Cf peak count rate, with an error of $\pm 4.9\%$. A summary of the absolute disintegration rate results for both samples is given in table 4. The absolute gamma ray intensities indicate that there was no significant change in the activity of the samples measured before and after exposure to the electric field.

As for the results of the ratios of gamma line intensities, in our first sample, the pre-treatment ratio of the 121.8 keV Eu line to the 344.3 keV Eu line remained unchanged at 1.055, compared to the literature value of 1.067. These two gamma lines arise from different decay modes of ^{152}Eu , so it appears that the electric field affected neither electron capture nor β -decay. The ratio of the intensity of the 344.3 keV Eu gamma line to the 388.1 keV Cf line decreased by 3.2% during the treatment. This is a larger change than expected, since the use of ratios should give a more accurate result

than the use of absolute disintegration rates, because the errors due to geometry changes and efficiency calculations should cancel. We have discovered that this ratio is very geometry-dependent, since the *relative* efficiencies of detection of the 344 keV and the 388 keV gammas change with distance from the detector face. This is due to the fact that, with increasing distance from the detector face, the higher energy gamma rays have a smaller probability of interacting with an annular segment of the detector crystal, and thereby only depositing a portion of their energy in the detector. In addition, the 121.8 keV ^{152}Eu line is subject to summing, which cannot be corrected for by calibrating with a standard. Beyond these sources of uncertainty, the cause for the overly-large error is unexplained. This demonstrates the difficulty in quantitative and reproducible nuclear spectroscopy. A summary of the results for the ratios of gamma line intensities is given in table 5.

CONCLUSION

Within the limit of experimental error and reproducibility, we have seen no change in the decay rates of either the alpha emitting nuclide ^{249}Cf or the EC and β -decaying nuclide ^{152}Eu after exposure to the electric field produced by an electrostatic generator. In order to eliminate the difficulties in highly accurate alpha spectroscopy, we looked at the gamma rays from excited levels in the ^{245}Cm daughter to determine the decay rate of the ^{249}Cf sample. In addition to the absolute disintegration rate of the ^{249}Cf , we looked at ratios of the californium gamma ray to gamma lines from ^{152}Eu in the same sample to eliminate variability due to possible detector-sample geometry changes, and possible physical loss of the nuclide. The results for ratios of the gamma ray intensities of the two nuclides are more variable than expected from the statistics of the measurements alone. This probably results from the inability to reproduce sample placement exactly, which will cause slight variations of the measurement when one or

more of the nuclides emits gamma rays that sum. Despite the variability in the results, we feel confident that external electric fields of order 10^7 V/m are not capable of perturbing the nuclear Coulomb barrier, and therefore changing the probability of alpha particle emission. The apparent imperturbability of nuclear decay rates is once again demonstrated.

ACKNOWLEDGEMENT

This work was supported by the Office of High Energy and Nuclear Physics, Division of Nuclear Physics, U. S. Department of Energy under Contract DE-AC03-76SF00098.

REFERENCES

1. H. Bequerel, Compt. Rend. Acad. Sci. **133**, 199 (1901).
2. P. Curie, Soc. de Physics, Marz, 1901.
3. E. Rutherford and J. E. Petavel, Brit. Assoc. Advancement of Sci. Rep. A, 456 (1907).
4. S. Freed, A. H. Jaffey, M. L. Shultz, Phys. Rev., **63**, 12 (1943).
5. A. Piccard and G. Volkart, Arch. Soc. Phys. et Nature, **3**, 543 (1921).
6. E. Segre, Z. Physik, **71**, 274 (1947).
7. R. Daudel, Rev. Sci., **85**, 162 (1947).
8. H. P. Hahn, H. J. Born and J. I. Kim, Radiochimica Acta, **23**, 23 (1976).
9. M. Neve de Mevergnies and P. Del Marmol, Phys. Letters, **49B**, 428 (1974).
10. *Table of Isotopes*, C. M. Lederer and V. S. Shirley, ed., J. Wiley and Sons, New York, 1978, 7th ed.
11. K. Alder, G. Baur and U. Raff, Phys. Letters, **34A**, 163 (1971).
12. Barker, W. A., submitted for publication in Physical Review.
13. Barker, W. A., Altran Corporation, Sunnyvale, CA, unpublished results.
14. Barker, W. A., Altran Corporation, Sunnyvale, CA, personal conversation, 1991.
15. *The Chemistry of the Actinide Elements*, Seaborg, Katz, Morss, Ed., Chapman & Hall, New York, 1986, p. 1791.
16. Routti, J. T., UCRL report 19452, Oct. 20, 1969.

TABLE 1**Barker's Data: May 1991**

Copy of Dr. William Barker's data which gives evidence for accelerated alpha decay. Source was windowless americium-241, in form of Am_2O_3 . Source was exposed to electric field in a Van de Graaf generator for 14 hours. The paper cover was used to filter out alpha particles. All counts were five minutes in length.

Voltage (kV)	No cover Count	Paper cover Count	Net Alpha Count
0	1447	1136	311
-5	1571	1105	466
-10	1660	1151	509
-15	2005	1109	896
-20	2900	1139	1761
-25	4056	1170	2886

TABLE 2**Nuclides used in our experiment**

NUCLIDE	HALF-LIFE	DECAY MODE	GAMMA ENERGIES	INTENSITY
^{249}Cf	350.6 years	Alpha	388.1 keV	66.0%
			333.4 keV	15.5%
^{152}Eu	13.2 years	EC(72.1%)	121.8 keV	28.4%
			244.7 keV	7.5%
		β -(28.9%)	344.3 keV	26.6%

TABLE 3

Comparison of our pre- and post-treatment detector geometries, showing close agreement.

EFFICIENCY CALIBRATIONS USED 1990 MIXED GAMMA STANDARDS
SOURCE COUNTED ON DETECTOR 5 SHELF 1 FOR 20. MIN.
AT 90 8 15 9

ISOTOPE	ENERGY	COUNTS/MIN.	EFFICIENCY	UNCERTAINTY(%)
CD-109	88.00	2321.6	0.0572	3.60
CO-57	122.10	2534.6	0.0691	2.50
CE-139	165.80	2916.3	0.0644	3.10
HG-203	279.20	4357.6	0.0412	2.60
SN-113	391.70	3623.7	0.0290	4.50
SR-85	514.00	4947.7	0.0225	2.50
CS-137	661.60	2788.9	0.0175	3.20
Y-88	898.00	4651.5	0.0127	3.20
CO-60	1173.20	2079.3	0.0096	0.80
CO-60	1332.50	1868.8	0.0086	0.80
Y-88	1836.10	2566.9	0.0066	2.10

EFFICIENCY CALIBRATIONS USED 1990 MIXED GAMMA STANDARDS
SOURCE COUNTED ON DETECTOR 5 SHELF 1 FOR 30. MIN.
AT 91 5 30 10

ISOTOPE	ENERGY	COUNTS/MIN.	EFFICIENCY	UNCERTAINTY(%)
CD-109	88.00	1529.1	0.057999	3.60
CO-57	122.10	1222.1	0.069494	2.50
CE-139	165.80	678.0	0.063848	3.10
HG-203	279.20	0.0	0.000000	0.00
SN-113	391.70	640.0	0.029029	4.50
SR-85	514.00	230.5	0.022834	2.50
CS-137	661.60	2764.1	0.017684	3.20
Y-88	898.00	725.9	0.012907	3.20
CO-60	1173.20	1910.4	0.009808	0.80
CO-60	1332.50	1702.0	0.008726	0.80
Y-88	1836.10	392.2	0.006542	2.10

TABLE 4

Results for absolute disintegration rates of ^{152}Eu (gamma energy: 121.8 keV) and ^{249}Cf (gamma energy: 388.1 keV) in our samples 1 and 2. Data analysis was done using SAMPO gamma spectroscopy program.

RESULTS FOR EU-CF SAMPLE #1

GAMMA PEAK (keV)	NOV. 1990 Pre-treatment	MAY 1991 post-treatment	PERCENT CHANGE	TOTAL UNCERTAINTY
388	42,065 $\pm$ 3.5% dpm	42,743 $\pm$ 2.5% dpm	1.61%	4.3%
121	41010 $\pm$ 3.5% dpm	40,310 $\pm$ 2.5% dpm	-1.71%	4.3%

RESULTS FOR EU-CF SAMPLE #2

388	48,630 $\pm$ 3.5% dpm	49,300 $\pm$ 3.5% dpm	1.38%	4.9%
-----	-----------------------	-----------------------	-------	------

TABLE 5

Results for the ratios of gamma line intensities, pre- and post-treatment, for our Eu-Cf sample #1. The 121.8 keV line is from electron capture of Eu-152. The 344.3 keV line is from β - decay of Eu-152, and the 388.1 keV line is from alpha decay of Cf-249.

<u>RATIO</u>	<u>PRE-TREATMENT</u>	<u>POST-TREATMENT</u>	<u>CHANGE</u>
121/344	1.055	1.055	none detected
121/388	.975	.943	-3.3%
344/388	.924	.894	-3.2%

FIGURE CAPTION

Figure 1: Gamma spectrum of our Eu-Cf sample #1, taken on EG&G high-purity germanium detector. The peaks we measured are marked.

Figure 1

NOPB101.00 BEGIN: 512.37 DAY LT= 60.0MIN.

NOPB101.00 BEGIN: 512.37 DAY LT= 60.0MIN.

END

**DATE
FILMED
4 128192**