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SAND2001-1763
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Final Report of LDRD Project: Accelerated Beta Decay for the Reduction of Legacy Wastes

Guillermo M. Loubriel, Fred J Zutavern, Lars D. Roose, Malcolm T. Buttram, Mark S. Derzon, Steven A. Wright, and David W. Vehar

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Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under Contract DE-AC04-94AL85000.

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FINAL REPORT OF LDRD PROJECT: ACCELERATED BETA DECAY FOR THE REDUCTION OF LEGACY WASTES

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Abstract

This report provides a summary of the LDRD project titled: Accelerated Beta Decay for the reduction of legacy wastes. The project met all its milestones even with a tight <1year schedule and total funding of $ 100 k. High level waste (HLW) comprises over 60% (500,000 cubic meters) of the DOE current inventory and projected generation of mixed waste over the next 5 years. HLW is generated and stored at four DOE sites (Hanford Site, Idaho National Engineering Laboratory, Savannah River Site, and the West Valley Demonstration Project, New York). The majority of HLW is comprised of radioactive materials that have forbidden beta (β) decays, like 137Cs and 90Sr. The lowest order transition probability for β decay in these materials is rigorously zero due selection rules. These rules are believed to be invalid, in a number of papers by H.R. Reiss, in a strong electromagnetic radio frequency (RF) or audio field, which penetrates to the nucleus. Thus, the isotopes can be induced to β decay if they are placed in a

RF/Audio field. The magnitude of the RF/Audio effect has been calculated (H. R. Reiss, Phys. Rev. C, Vol. 27, pp. 1199-1228, and pp. 1229-1243). The theory predicts that we may reduce the half-life of radioactive materials to allow for treatment of HLW on site. In particular, the theory shows that the strength of the effect depends on an intensity parameter, z, that is proportional to (E/f) squared where E is the electric field and f is the frequency of the RF/Audio field. The theory predicts a maximum in the decay rate in the range 10 kV/(cm-MHz) < E/f < 1000 kV/(cm-MHz). We designed an experiment to sweep through this range by independently varying E and f. Some experiments had been performed prior to this LDRD (by Dr. Reiss and others) that showed interesting, but were not definitive perhaps because they did not have sufficiently strong RF/Audio fields at sufficiently low frequency. We performed a key experiment intent on verifying the existence of the effect of RF/Audio-accelerated β decay with the required fields and frequencies.

To obtain high fields, we used a parallel plate transmission line to deliver electric fields above 10 kV/cm to a 137CsCl-impregnated Kapton dielectric that is 5 mils thick (8 Ω). This required >130 V and 2 kW (supplied by a reasonably common audio amplifier). We varied the frequency from 1 kHz to 40 kHz. The intensity parameter will be 10E6 larger than in previous experiments. We looked for: 1) a dramatically modified decay rate, and 2) the effect of electric field and frequency on the decay rate. The frequency range in these experiments was 5kHz-100kHz. The estimated fields per frequency were 100 kV/(cm-MHz) < E/f < 4×10^5 kV/(cm-MHz).

No strong RF/Audio-induced enhancement was observed. One possibility is that the enhancement was not observed because the electric field was screened by the electron cloud in the crystal and did not penetrate to the nucleus.

The ability of RF/Audio to penetrate the electronic cloud in the atom and reach the nucleus depends on the degree of shielding by the electrons. In this experiment using CsCl salt, with a singly ionized Cs atom (assuming that one electron is transferred to the Cl atom) some of the RF/Audio was expected to penetrate based on calculations carried out for a bare Cs nucleus (no shielding). Future experiments and calculations should address this shielding issue which could be mitigated by higher frequencies, using ionized atomic 137Cs, or ionized molecules such as 137CsCl.

CONTENTS

Section

1.0 BACKGROUND

High level waste (HLW) comprises over 60% (500,000 cubic meters) of the DOE current inventory and projected generation of mixed waste over the next 5 years. HLW is generated and stored at four DOE sites (Hanford Site, Idaho National Engineering Laboratory, Savannah River Site, and the West Valley Demonstration Project, New York). High-level wastes possess the feature that all of the radioactive nuclei exhibit the type of radioactivity known as forbidden beta decay, like 137Cs and 90Sr. The word forbidden is a misnomer in the sense that the beta decay is only strongly inhibited because of the violation of certain quantum-mechanical selection (symmetry) rules. Nuclear fragments arising from nuclear fission undergo the radioactive process of beta decay, wherein a neutron within the nucleus is converted to a proton with the accompanying emission of a beta particle (i.e., an electron). Ordinary beta decay (allowed beta decay) occurs sufficiently quickly and thus the half life of the isotope is so short that it poses no waste disposal problem. However, if the change in quantum numbers when the nucleus undergoes decay from the parent to the daughter state fails to satisfy quantum selection rules on angular momentum and nuclear parity, then the radioactive decay is slowed by a factor of about 10,000 as compared to allowed beta decay. It is for this reason that wastes such as Cesium-137 and Strontium-90 have half-lives of about thirty years rather than one day or so.

Considerable early work (summarized in reference 1) has been performed on non-nuclear means of altering radioactive processes, but the focus was on small alterations of rates, and in types of radioactivity that are not of consequence for the high-level waste problem. The idea being explored in this LDRD is to **strongly** accelerate decay rates of high-level wastes by providing, by inexpensive and external means, the angular momentum and parity needed by the nucleus to avoid violation of the quantum selection rules. This idea of RF/Audio-accelerated beta decay has been developed by Dr. Howard Reiss of American University (references 2 to 8) and may result in a much larger change in decay rates by the application of an intense electromagnetic field, which provides angular momentum and parity. Low frequency radiation is inexpensive, but the heart of the problem is to achieve coupling of the very small nucleus to the long wavelength of low frequency radiation. There are two mechanisms that make this coupling possible. Both these mechanisms are unique to the intense-field environment, and have no weak-field (perturbation theory) counterparts.

We emphasize the word strongly since this limited-scope LDRD will test for large effects only. We intend to test whether the decay rate can be made substantially larger (by a few percent). Whether RF/Audio fields affect, at all, the decay rate may be the subject of further sudy.

One of the strong-field mechanisms is associated with the application of a very strong, low frequency field. When influenced by the strong field, the nuclear state loses its usual property of being a pure angular momentum quantum state, but instead it acquires an admixture of other angular momentum states not normally present. Some portion of this admixture makes the allowed decay channels accessible, leading to much faster radioactive decay. The second strong-field mechanism involves the electron emitted in the beta decay. In a very intense plane-wave electromagnetic field, this electron is forced to undergo a violent motion that follows from the combined effect of the electric and magnetic fields. The magnetic field is often neglected in low-frequency problems, but it is well known that intensity effects alone are enough to make magnetic fields important. This intense-field motion introduces large amounts of angular momentum into the final state of the radioactive decay process, thus altering rates for the quantum-mechanical transition between initial and final states.

This RF/Audio-accelerated beta decay mechanism allows forbidden beta decay isotopes to beta decay if they are placed in a RF/Audio field and if the atom is ionized (as is the case in an alkali halide compound). The magnitude of the RF/Audio effect has been calculated (references 2, 3). The theory predicts that we may reduce the half-life of radioactive materials to allow for treatment of HLW on site. Some experiments have already been performed (by Dr. Reiss and others) that show some interesting, but are not definitive perhaps because they did not have sufficiently strong RF/Audio fields at sufficiently low frequency. Our experiment tested the effect of RF/Audio-accelerated beta decay with the required fields and frequencies.

The theory shows that measurable effects on the half-life of 137Cs can be accomplished in RF/Audio fields that are feasible to achieve. In particular, the theory shows that the strength of the effect depends on an intensity parameter, z, that is proportional to (E/f)2 where E is the electric field and f is the frequency of the RF/Audio field. We expect a maximum in the decay rate in the range 10 kV/(cm-MHz) < E/f < 1000 kV/(cm-MHz). We will design an experiment to sweep through this range by independently varying E and f. We will measure the effect of electric field and frequency on the half-life. To obtain the high field, we will use a parallel plate transmission line to deliver electric fields of up to 10 kV/cm to a 137CsCl-impregnated Kapton dielectric of dimensions 5 mils thick by 135 mils wide (8 Ω). This requires only ~130 V and 2 kW (a reasonably common audio amplifier). We will vary the frequency from 1 kHz to 40 kHz. The intensity parameter will be 10^6 larger than in previous experiments. We will look for: 1) a dramatically modified decay rate, and 2) the effect of electric field (E) and frequency (f) on the decay rate. We will study 137Cs because it is an important component of HLW and because the β decay is accompanied by gamma (γ) emission, which is much easier to detect than the β decay itself.

The present experiment builds on, but is markedly different, from previous experiments. The first test of the theory was carried out at the University of Arizona using the University-sponsored radio station and the RF frequency was fixed at about 1 MHz. Furthermore, the peak electric field was very low (1 kV/cm) because they used the coaxial cable (cylindrical transmission line) used by the radio station and were limited by the station's peak power. Finally, the effective power on target was very small in previous experiments since the CsCl was in a small pellet which was embedded in the transmission line's coax. Our approach is innovative in that we can increase the applied field transmission line and we can vary frequency (and get to lower frequencies). Our experiment is much better designed from the RF/Audio point of view.

	Task	Milestone
1.	Design and build RF/Audio system	3QFY00
2.	Measurement of decay rates in fields of varying intensity and frequency	4QFY00
3.	Final SAND report	4QFY00

2.0 EXPERIMENTS

2.1 General description of the experiment

The experiment consisted of applying an AC voltage of varying frequency and voltage to a CsCl sample and detecting the gamma particle that accompanies the beta decay. To obtain high fields, we used a parallel plate transmission line to deliver electric fields of up to 10 kV/cm to a 137CsCl-impregnated Kapton dielectric that is 5 mils thick (8 Ohm impedance). To generate this field we used a reasonably common audio amplifier. This is schematically represented below.

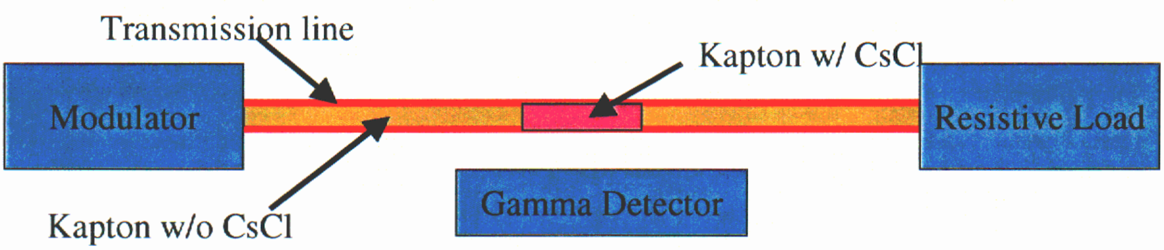

Figure 2.1. Schematic of the experiment

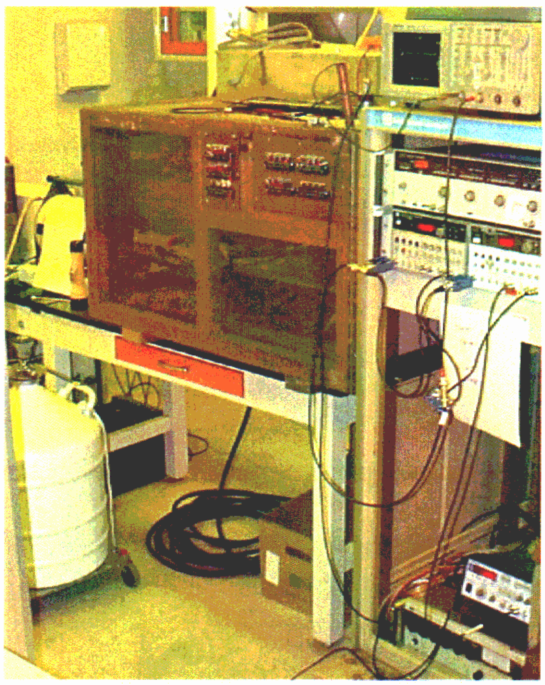

Figure 2.2 Photograph of the experimental setup. From left to right: the detector (behind the flashlight), the screen box (with the source, transmission line, resistive loads, and audio amplifier), and a rack with electronics (scope, pulse generator, two side-by-side Hewlett Packard sweep/function generators, and a Tektronix function generator).

2.2 Transmission line

The radioactive sources were purchased from North American Scientific, Inc. (Chatsworth, CA, http://www.nasi.net). North American Scientific manufactures and markets a line of low-level radiation sources and reference standards for medical, scientific, and industrial uses. We ordered a total of 6 calibrated sources. The sources were made by depositing a given quantity of 137CsCl onto a Kapton dielectric film and then the CsCl was covered by another layer of film producing a "sandwich" of Kapton, CsCl, and Kapton. The size of the sandwich was 1" by 10" but the size of the active area and its radioactivity varied from source to source. Three had active areas of 0.075" by 1 cm with 5 µC, 1 µC, and 0 µC (using non-radioactive Cs). The other three sources had active areas of 1" by 10" and the same radioactivity levels.

The sources were made into transmission lines by placing Cu tape on either side. The total thickness of the Kapton sandwich was 0.005" and the thickness after the tape was added was 0.008". Two configurations were used: 1) 0.5" wide Cu tape and 2) 0.163".

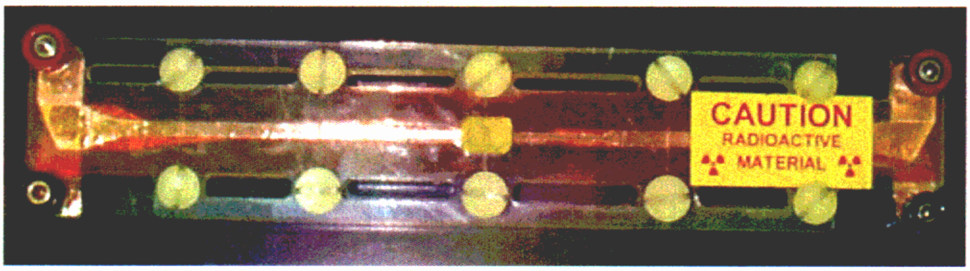

Figure 2.3 Photograph of the 137CsCl sample holding fixture. It consists of two plexiglass™ plates, Cu tape, and the Kapton sandwich that contains the 137CsCl. The plexiglass plates™ (2" top to bottom, 10" left to right, and 0.225" thick) serve to center the Cu conductors. In the center cut-out of the plate facing view, there is compressed foam to insure precise clamping of the sample area, so that the dielectric spacing retains its integrity. Slots on either side of sample strip allow for exact centering during assembly.

2.3 Audio systems used for generating RF/Audio fields

We used two Hewlett Packard model 3314A sweep/function generators (A and B), a Tektronix function generator (C), an audio amplifier (Mackie M2600 Fast Recovery Series, high current power amplifier), an oscilloscope, and a computer. The function generators "A" and "B" were gated on and off by the controlling computer, which also controls the detector. Diode inputs to each of the function generators "A" and "B" prevent loading down of the signal from the computer. The "A" and "B" outputs were mixed together with a Diode Mixer which hangs below Generators and feeds the mixed signal to the audio amplifier through a connection at the top of the screen box. The oscilloscope was used to measure the voltage across the resistive load inside the screen box, as well as the input to the amplifier at mixer output of the "A" and "B" function generators.

Figure 2.4 Photograph of the screen box showing approximately half of the load resistance used and the Mackie amplifier.

2.4 Counting electronics

The following block diagram shows the system used to obtain the measurements of Cs-137 gammas from the source (which accompany beta decay). With the exception of the ADC, all modules are manually set. The ADC is controlled by Canberra Genie software through an Acquisition Interface Module (AIM).

Data were collected in the form of 8192-channel spectra, from which the number of counts in the 662-keV line were determined using standard peak search routine built into the Genie software. This software displayed on a video terminal the peak centroid, net peak area, peak FWHM, and peak area uncertainty. It was not necessary to display the actual spectrum except during the initial setup of the system.

Genie console-level commands were used for all data acquisition and analysis functions. However, a number of measurement series were recorded to determine count rate as a function of time. For these, a script was written to automate the sequence of measurements and to ensure appropriate measurement intervals.

All measurements were made to a nominal peak area of 100000 counts, to provide counting uncertainties on the order of 0.3%.

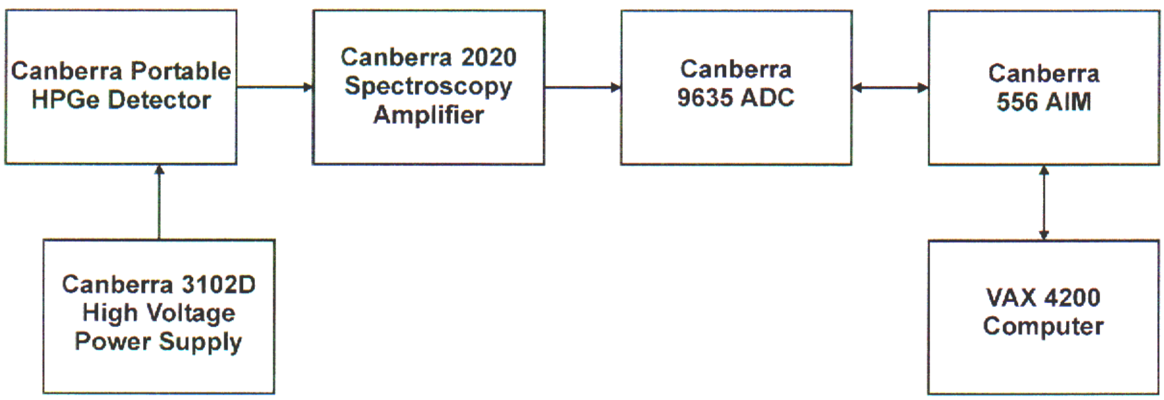

Figure 2.5 Block diagram of the counting electronics.

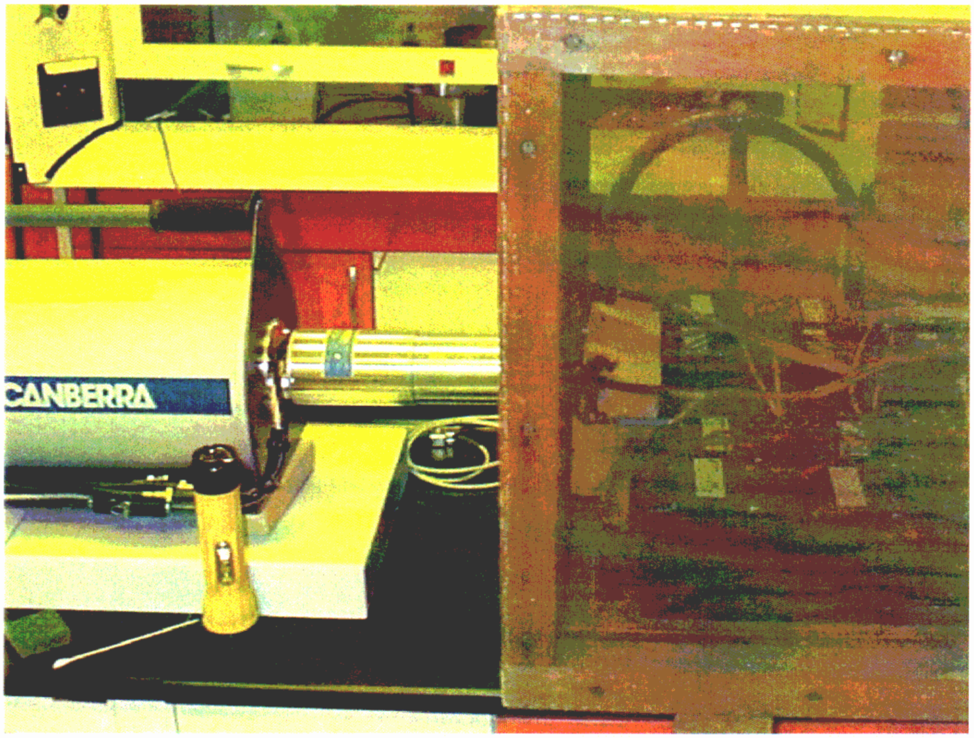

Figure 2.6 The gamma detector. On the left and the transmission line (inside the screen box). The connections to the transmission line and some of the step-up transformers are also visible.

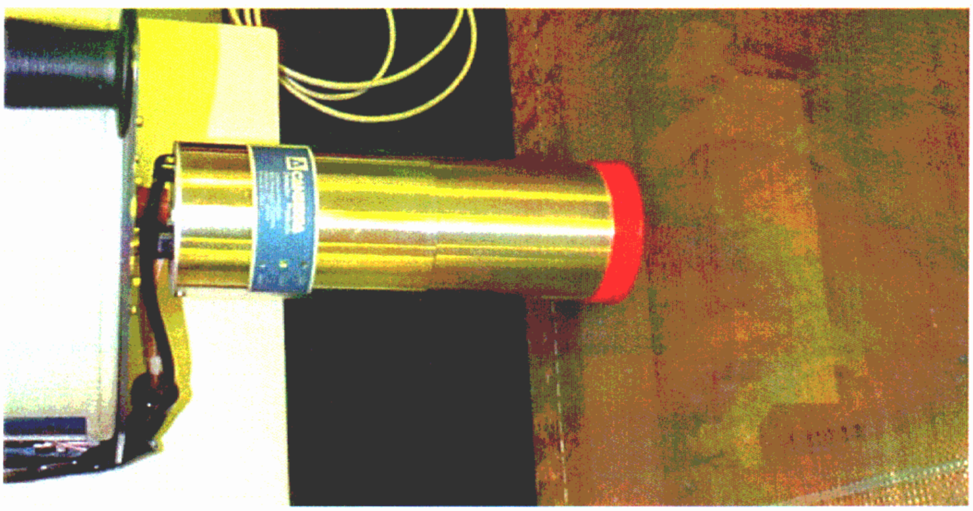

Figure 2.7 Close-up of the detector head outside of screen box, in close proximity to sample (inside screen box). The separation distance is ~1-5/8 inches and two sheets of copper screen (~1 inch).

2.5 Experiments

The initial experiments consisted in assembly of the system and dry runs with a non-radioactive sample to check that the system would work. The system was then moved to the facilities where the radioactive sample and counting electronics were located.

The first tests with the counting electronics revealed a noise problem for the counter at audio frequencies above 10 kHz with a voltage across the load of ~174 (peak to peak). We tried various shielding attempts, nothing helped, except for twisting input and output lines around each other. That fixed the problem at 10 kHz, but not at higher frequencies. We then installed a small screen-box and placed the amplifier, and transmission line/sample inside and kept counter outside box. Ran amp one-to-two minutes at each frequency full bore from 10, 30,40,50,60,70, 90KHz with Vout=130 v p-p @90KHz where amp auto-shut down to demonstrate that the noise problem was totally eliminated.

We obtained counts for a number of values of frequency and electric field which cover portions of the expected optimal regime for beta decay acceleration. This optimal range according to the theoretical work of Reiss was 10 kV/(cm-MHz) < E/f < 1000 kV/(cm-MHz). The parameter range for which we obtained counts is shown in figure 2.8. We show the space for which count data was acquired. Errors in field and frequency are assumed to be small. There were no significant variations in rate observed for any of the frequency field combinations.

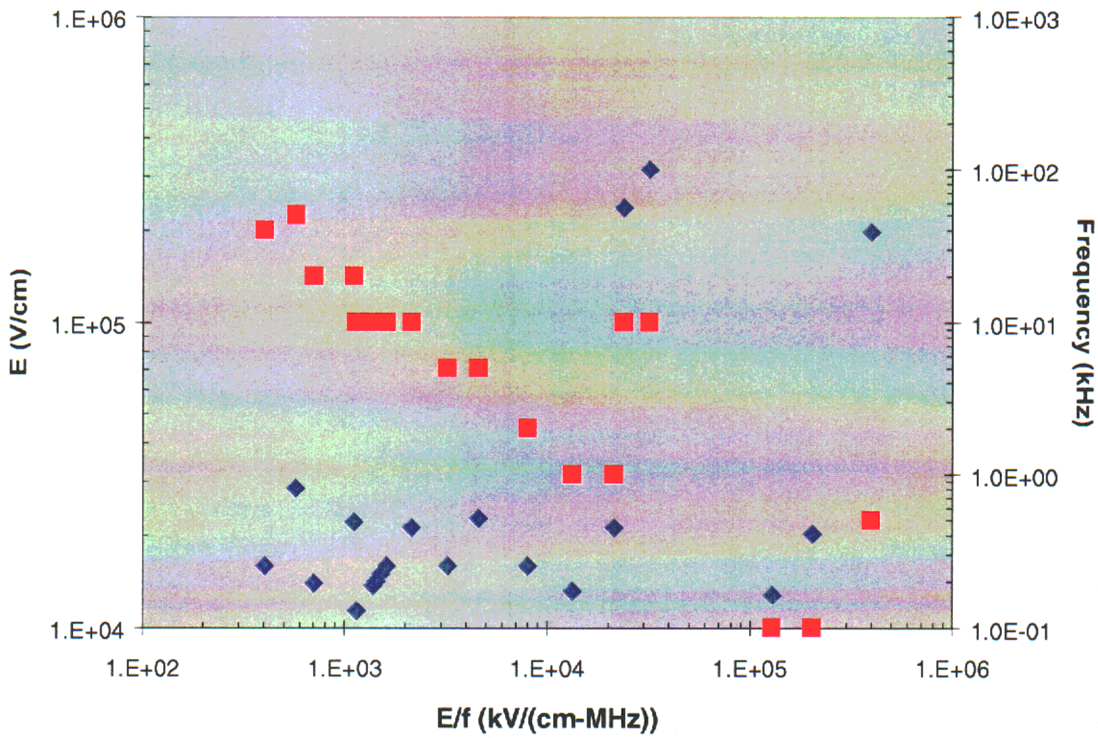

Fig. 2.8 Counting data was taken for each of the frequency and field points shown.

As an example of the typical count rates, we show two plots. The first, figure 2.9, shows some data taken at 10 kHz where the field is varied. The plot shows pairs of counts (counts with RF/Audio on and off) for varying conditions (including different counting times) as a function of electric field for a fixed frequency (10 kHz). The counts on (diamonds, blue) and off (squares, magenta) are within the error bars for most of the data, so much so that within a pair of data points, the "off" and "on" points are not discernable. To illustrate this, a subset of that data is plotted in figure 2.10.

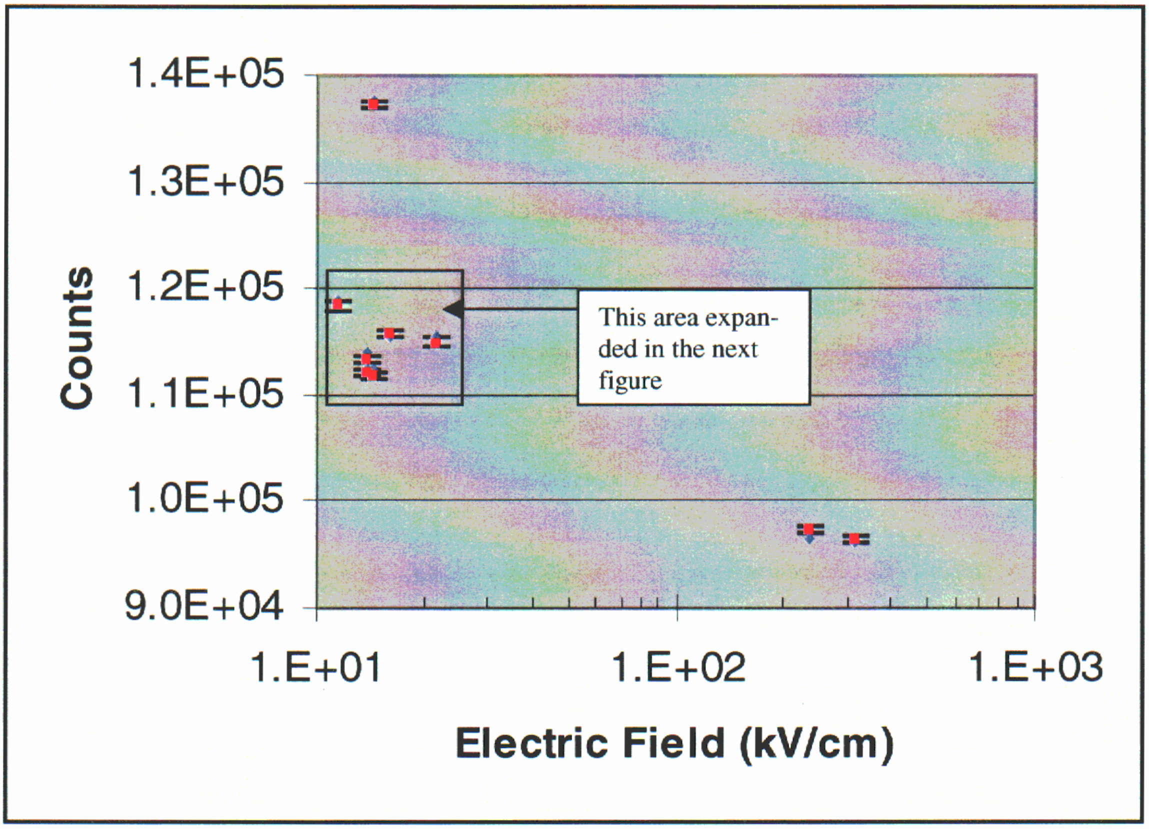

Figure 2.9 The plot shows pairs of counts (counts with RF/Audio on and off) for varying conditions (including different counting times) as a function of electric field for a fixed frequency (10 kHz). The counts on are shown as blue diamonds, blue and the counts off are shown as magenta squares.

Figure 2.10 shows data from electric fields of 11.3 kV/cm to 21.1 kV/cm. In this figure, we shifted the electric field values for the Counts off (squares, magenta) by 1 kV/cm.

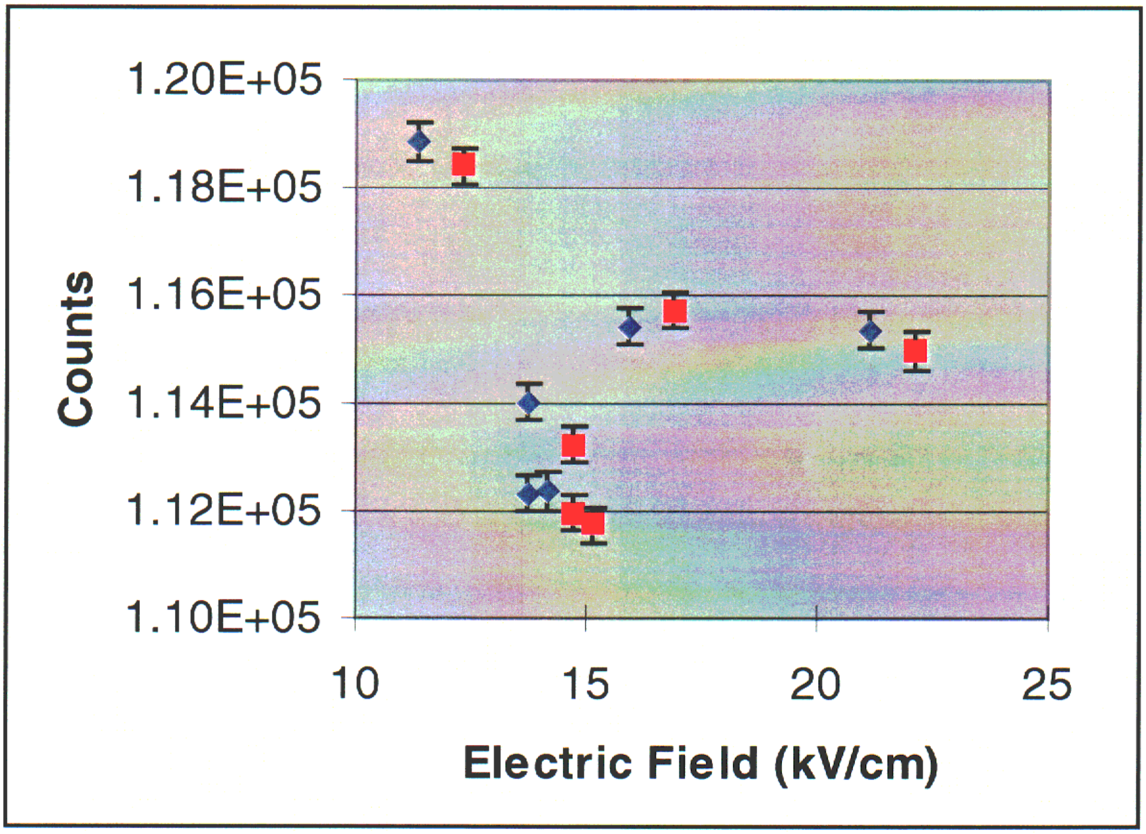

Figure 2.10 The plot shows pairs of counts (counts with RF/Audio on and off) for varying conditions (including different counting times) as a function of electric field for a fixed frequency (10 kHz). The counts on are shown as blue diamonds and the counts off are shown as magenta squares. The latter were shifted along the x axis by 1 kV/cm to allow for easier comparison of "on" and "off" values.

Perhaps the best way to look at the data is to show the ratio of counts on to counts off for a variety of conditions. This is illustrated in figure 2.1. Note that there are 12 data points above 1.0 and 12 below. Also note that, with the exception of five data points, all the data is consistent with a ratio of 1.0 (no effect of the electric field). Out of these 5 data points, 3 show enhancement of the decay rate (ratio greater than 1) and 2 show a decrease in the decay rate (ratio smaller than 1).

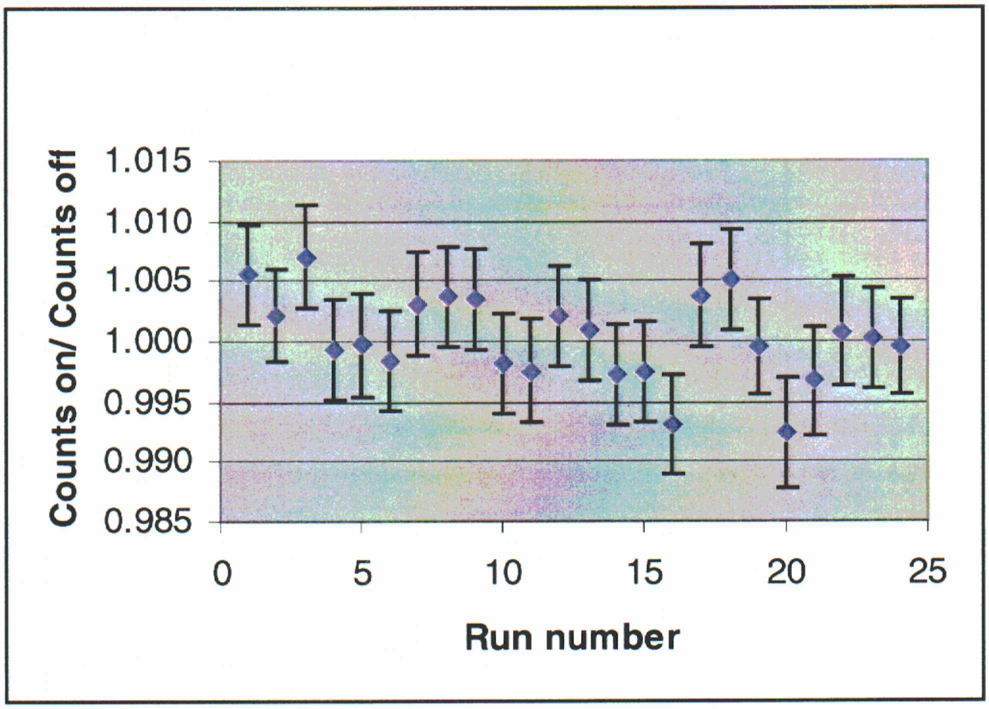

Figure 2.11 Counts on divided by counts off for a variety of conditions. Note that there are 12 data points above 1.0 and 12 below. Also note that, with the exception of five data points, all the data is consistent with a ratio of 1.0 (no effect of the electric field). Out of these 5 data points, 3 show enhancement of the decay rate (ratio greater than 1) and 2 show a decrease in the decay rate (ratio smaller than 1).

3.0 INTERPRETATION OF RESULTS/ CONCLUSIONS

The idea of the experiment was to look at large effects of ~10% enhancement. No RF/Audio-induced strong (>2%) enhancement was observed. At all the frequencies and fields, the count rate was approximately the same when the RF/Audio field was on as when it was off. We performed measurements with count rates that were, typically, in the order of 110,000 whose SQRT(N)/N error was 0.3%. In two instances, we counted for much longer times and achieved count rates of ~1,250,000 whose error was 0.08%. In those cases, the counts with the Audio field on were 0.04% and 0.01% smaller than with the Audio field off.

A possibility is that the strong enhancement was not observed because the electric field was shielded by the electron cloud in the crystal and did not penetrate to the nucleus. Another possibility is that this non-perturbation theory is wrong: that the overlap in time and space between such long wavelength "photons" (~100 m and 10^{-4} seconds) and either the size of the nucleus (~6 x 10^{-15} m) or Beta decay time (10^{-23} s) are too small to have any appreciable effect.

Whether RF/Audio fields affect, at all, the decay rate may be the subject of further study.

4.0 REFERENCES

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5.0 APPENDIX

This appendix is an experimental table that shows all the data collected.

Accelerated Beta-Decay Ce-137 tabulated results

Date	F (KHz)	V (p-p)	KV/cm (p-p)	LOAD Z (Ohms)	Comments	Count (off)	Count (on)	d-Count
8/10	10.	180	11.8	12.5	1x1cm 1uci sample (1/2" foils) (10-min runs,cnt last 2)	111743 (2-minute count	112364 (2-minute count)	+0.56%
8/17	10.	180	11.8	12.5	1x1cm 5uci sample (1/2" foils) (2-minute run)	137303 (2-minute count)	137599 (2-minute count)	+0.22%
8/17	10.	(?)	(?)	OPEN	1cmX.075" 5uci sample (0.163"w foils) –(8-ohm) (2-min run) (Load open connection)	114014 (2-minute count)	115497 (2-minute count)	+1.30%
	10.	174	11.4	8.6	(2-minute run)	113235 (2-minute count)	114029 (2-minute count)	+0.01%
	20.	177	11.4	8.6	(2-minute run)	113874 (2-minute count)	113791 (2-minute count)	-0.07%
8/21	1.0	167	11.0	8.6	Screen-box installed (15-minute run)	112183 (5-minute count)	112149 (5-minute count)	-0.03%
	0.1	162	10.6	8.6	Still on from above test) 15-minute run	112149 (using prev. count)	111965 (5-minute count)	-0.16%
	10.	174	11.4	8.6	Still on from above test 15-minute run	111965 (using prev. count)	112310 (5-minute count)	+0.31%
8/29	10.	144	9.4	5.4	Ckt brkr blew (approx. 5-min run)	118408 (3-minute count)	118838 (3-minute count)	+0.36%

9/13	10.	268	17.6	10M	1000:1 v-divider, 13-minute runs, (count final 3 minutes)	114971 (3-minute count)	115363 (3-minute count)	+0.34%
	1.0	268	17.6	10M		115363 (using prev. count)	115153 (3-minute count)	-0.18%
	0.1	256	16.8	10M		115153 (using prev. count)	114872 (3-minute count)	-0.24%
	50.	36 0	23.6	10M		114872 (using prev. count)	115107 (3-minute count)	+.20%
	20.	280	18.4	10M		115107 (using prev. count)	115207 (3-minute count)	+.09%
	5.0	288	18.9	10M		115207 (using prev. count)	114898 (3-minute count)	-.27%
	0.0	-0-	-0-		Power off, wait 10-minutes, begin count	114898 (using prev. count)	114918 Pwr Off (3-minute count)	+.02%
9/14	10.	202	13.3	404	13-minute runs, count final 3 minutes	115710 (3-minute count)	115405 (3-minute count)	-.26%
	5.0	202	13.3	404		115405 (using prev. count)	114613 (3-minute count)	-.69%
	2.0	202	13.3	404		114613 (using prev. count)	115049 (3-minute count)	+.38%
	40.	202	13.3	404		115049 (using prev. count)	115640 (3-minute count)	+.51%
9/20 to 9/22	10.	190	12.5	404	Foil width cut to .085"(16-ohms) 1cm X .075" 5uci sample RUN CONTINUOUS 50%	126998.8 5th average of 97.	126946.9 5th average of 97	-.04%

					duty (15-minutes on/15-minutes off) 97 CYCLES; count every 3-min.			
9/23 to 9/25	*	130 to190 fluctuating	8.5 to 12.5	404	RUN CONTINUOUS 50% duty 107 CYCLES count every 3-min *Mixed/beat Random Freq.	126358.3 5th average of 107	126342.9 5th average of 107	-0.01%
9/29	10.	3000.	196.8	10M	1000:1 v-divider, 13-minute runs, (count final 3 minutes) ** -Using Output TRANSFORMER -	97235 (3-minute count)	96493 (3-minute count)	+0.76%
	10.	4000.	262.4	10M		96493 (using prev. count)	96173 (3-minute count)	-0.33%
	0.5	2500.	164.0	10M		96173 (using prev. count)	96247 (3-minute count)	+0.08%

9/29	0.5	800 to 1250 ***	52.5	404	***Initially 800, drift to 1250, tripped amp seperate ckt brkr . approx. 5 minute run, begin count upon shut-down.	96247 (using prev. count)	94622 (3-minute count)	-1.69%
	-0-	None	-0-	404	Power off, 3-minute count	94622 (using prev. count)	94880 (3-minute count)	+0.27 %
	-0-	None	-0-	404	Power off, 3-minute count	94880 (using prev. count)	94821 (3-minute count)	-0.06%

- *On 9/29, Employed 3 generators, Generator A beat against Gen. B using a diode mixer. Both A & B 10VAC, and offset +1VDC
- Generator A Sweep time = 35.5-mS, Sweep 1KHz-10KHz, sine wave. Gen B. sweep time = 1.212-S, sweep 3KHz-18KHz, FM by Gen C.
- Generator C ~ 7 Hz
-
- **Output Transformer matrix consisted of 12 Audio output Transformers for Vacuum Tube Amps outputs.
- Used here in opposite orientation.
- Matrix of 12 Transformers combined, approx 6-ohms to 416-ohms for matching and ~ Gain of 70. Transformers (12) P/N: HAMMOND 1640E each.
-
- *** no precount performed. Possibility exists that sample may have been displaced before test.
- One or more transformer primary winding shorted during test.

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