The Neutron Capture Cross Section of ^{208}Pb

by

Miriam Lambert

A PROJECT

submitted to

Oregon State University

University Honors College

in partial fulfillment of

the requirements for the

degree of

Honors Baccalaureate of Science in Physics (Honors Scholar)

Presented May 24, 2000

Commencement June 2000

AN ABSTRACT OF THE THESIS OF

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Abstract approved:

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Kenneth Krane

In this project we measure the neutron capture cross section of ^{208}Pb. We began by calibrating our silicon detector with ^{204}Tl. Two experiments were then performed, the first in December of 1999 and the second in March of 2000. Both experiments were performed in the nuclear reactor of the Radiation Center at Oregon State University. The first was done in the rabbit fast transfer facility, with a neutron flux of 1.1×10^{13} n/cm^2/s and the second was done in the thermal column of the reactor, with a flux of 7.35×10^{10} n/cm^2/s. The December sample was prepared by rolling a thin foil between steel rollers, and had a large number of gamma and beta ray contaminants. It was not successful. The March sample was not rolled out, and was irradiated with a far lower flux and no fast neutrons. Its gamma spectrum was clean, except for two contaminants. However, this sample showed considerable internal absorption of low energy beta particles. We used the March data to calculate the cross section of ^{208}Pb, and found that $\sigma = 0.091 \pm 0.018$ millibarns. This is considerably smaller (54%) than the previously published value of about 0.2 millibarns.

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Mentor, representing Physics

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Committee Member, representing Physics

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Dean of the University Honors College

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Miriam Lambert, Author

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For my father, who taught me to think,
and for my mother, who taught me to believe.

The Neutron Capture Cross Section of ^{208}Pb

Introduction

This project concerns data taken at the reactor at the OSU Radiation Center in an effort to measure the neutron capture cross section of ^{208}Pb. Two experiments were performed, the first in the rabbit fast transfer facility and the second in the thermal column of the reactor. The first experiment took place in December of 1999 and was performed by placing a sample of ^{208}Pb in the reactor and then irradiating it with both thermal and high-speed neutrons. The second experiment was in March of 2000 and involved a bombardment of only thermal neutrons.

Each atom within the irradiated sample then formed a target, which may or may not have been hit by a neutron. Neutron capture occurs when an atom absorbs a neutron. The probability that a neutron will be captured is referred to as the **neutron capture cross section**. In other words, the cross section is a measurement of the effective target area of the sample's atoms. A larger cross section indicates a higher probability that a neutron will be captured. It thus indicates the likelihood that an element will be excited by thermal neutrons in a reactor. A known cross section permits one to predict the excitability of an element in nuclear experiments.

The captured neutron changes the mass number of the atom, forming a different isotope. In the case of ^{208}Pb, the additional nucleon changes the element to ^{209}Pb, which is radioactive. ^{209}Pb decays by emitting electrons and neutrinos in a process known as **beta decay**.[1] In this experiment we seek to measure the neutron capture cross section of ^{208}Pb by exciting it to ^{209}Pb and then measuring the intensity of the emitted beta radiation.

^{208}Pb is in itself an interesting element. It is **doubly stable**, meaning that there are closed shells of both protons and neutrons (Z = 82, N = 126).[2] Moreover, once

activated to ^{209}Pb it decays to ^{209}Bi, the heaviest naturally occurring stable atom, and the end point of S-process stellar nucleosynthesis.[3] This means that the study of this process may lead to greater understanding of the nuclear formation and decay processes within stars.

However, this experiment is motivated by more than the theoretical benefits of such knowledge. We are aware of only one previous measurement of the cross section of ^{208}Pb, at Oak Ridge National Laboratory, where was published only in an internal laboratory report there. Published values show a large range, typically around 0.2 millibarns.[4] Related experiments at Oak Ridge National Laboratory have suggested that this value may be in error, and so we undertook to re-measure this cross section.

Experimental Procedure

Calibration

In order to obtain accurate measurements, we first had to calibrate our detector. In both the December and March runs we took data with a 1 mm silicon detector. We used a thallium (^{204}Tl) source to calibrate the detector. Thallium has a beta spectrum very similar to that of ^{209}Pb, with the similar end point energies and a known activity ($a = 2.87 \times 10^3\ s^{-1}$). The end point energy of thallium is 763 keV and that of lead is 645 keV, with a difference of 118 keV.[2]

Since we know the activity of the ^{204}Tl source, we can determine the efficiency of the detector by measuring the total energy of the decay process. When plotted on an energy vs. number of decays axis, beta radiation has a continuous spectrum, entirely unlike the sharp peaks found in a gamma ray spectrum. A typical gamma ray spectrum will show a continuous level of background radiation, punctuated by peaks corresponding to photons emitted at discrete energies. In contrast, a beta ray spectrum will show a continuous curve ranging from low to high energies (see appendix A, figure 1). Thus one cannot determine the total activity of the decay process by simply measuring discrete peaks, as is done in gamma ray studies. Instead, the area of the entire curve must be found.

We found the total number of counts in the ^{204}Tl spectrum using the method of energy divisions described in the data analysis section. This in turn allowed us to find the efficiency of the detector, as shown in equation 4. When the detector was calibrated, we could measure the neutron capture cross section of ^{208}Pb.

^{208}Pb measurements

There are several possible methods for measuring the cross section of an isotope. We chose to use the activation method, in which we irradiated the sample and then measured the radiation emitted as the excited isotope decayed.

The measurement was complicated, however, by the method by which ^{209}Pb decays. The excited isotope emits electrons and neutrinos through beta decay. As the electrons are emitted through a continuous spectrum ranging from low to high energies, we must be able to stop and record electrons with a wide variation in energy in order to obtain an accurate measurement. Most importantly, however, the electrons must reach the detection equipment in the first place. There was a danger that the emitted beta particles would be re-absorbed within the sample before we could detect them. For this reason we had to irradiate as thin a sample as possible.[5]

There were several possible ways of achieving this. We briefly considered using a powdered form of ^{208}Pb, but the difficulty inherent in making an even sample depth, as well as working with radioactive powder, soon precluded that possibility. This left two options which we considered in preparing the lead samples.

One possibility would have been to dissolve the lead with nitric acid and then precipitate it in sulfuric acid, finally evaporating the solution on a thin slice of mylar.[6] This would have the advantage of giving a very thin sample, thus minimizing the risk of the absorption of beta particles within the sample. However, the risk of contamination would mean that the sample could not be treated before irradiation, since the acid would pollute the data obtained. We considered dissolving the sample after it was irradiated, but this would mean extensive handling of a radioactive source. Although the activity was expected to be low, it is generally best to minimize contact with radioactive materials. In addition, the very short half-life of ^{209}Pb made it preferable to measure the activity immediately after the sample was irradiated. For these reasons we chose to use a thin

piece of foil instead of a dissolved sample. We performed two data runs, the first in December of 1999 and the second in March of 2000.

December 1999 experiment

In order to minimize scattering within the sample, it was necessary to ensure that the foil was as thin as possible. To prepare the first sample we rolled out 114 mg of ^{208}Pb to an area of 2.56 cm^2 and 0.04 mm thickness.[3] The steel rollers were cleaned with HCl acid and then washed with acetone and ethyl alcohol before the lead foil was inserted.

Our first sample irradiation took place in December of 1999 at the rabbit facility in the OSU reactor with a neutron flux of 1.1×10^{13} $n/cm^2/s$. About 90% of the neutrons were in the thermal region and 10% were epithermal or above. The foil was irradiated for approximately 1.5 times its half-life, or 4 hours. We then measured the sample's activity using a silicon electron detector of thickness 1 mm.

Despite the precautions taken in cleaning the rollers the data obtained showed a great many gamma ray peaks, which indicated the presence of impurities. ^{24}Na and ^{204}Pb were the strongest gamma ray contributors (see Appendix B, chart 1). This led us to conclude that the beta spectrum was too heavily contaminated to permit useful analysis. Another measurement was necessary.

March 2000 experiment

The next measurement was taken in March of 2000. For this run we did not roll out the foil. Although this may have permitted a greater number of electrons to lose energy within the sample than would occur in a thinner foil, the threat of contamination from the rollers or the handler made internal scattering the more acceptable risk.

The March sample was irradiated for two hours in the thermal column of the OSU reactor. The thermal flux was much smaller than that of the rabbit facility, 7.35×10^{10} $n/cm^2/s$ and had very few fast neutrons.

This data proved to have a very clean gamma spectrum, with only very weak peaks corresponding to ^{24}Na (a common contaminant) and ^{187}W (see Appendix B, chart 1). The sodium has a half-life of 15 hours, but at a given start time there were only 69 counts to 350 counts of tungsten, a difference of 281 counts. Thus we considered the sodium contaminant to be negligible.

Without the contaminants that clouded the December data, an analysis of the beta spectrum was possible. However, the other isotopes that were present decayed partially through beta decay, which masked the ^{209}Pb spectrum. We isolated the lead spectrum by examining the number of decays at a given energy range in our data and comparing this to the number expected given the short half-life of ^{209}Pb (3.25 hours). By estimating the half lives of the contaminants, we were able to eliminate those spectra and extract the lead data.

Data Analysis

We used two spectra in analyzing the March data. We had the experimental data, that is the beta spectrum which we measured (Appendix A, figure 2), and we also generated a theoretical plot of the ideal beta spectrum (Appendix A, figure 1). The experimental data showed a great deal of scattering in the lower energies, hence we were unable to find the total activity of that plot.

The theoretical plot was used to determine the total expected activity of the decay. We divided the plot into bins 50 keV wide, that is from 0 to 50 keV, 50 to 100 keV, and so on. Each bin had continuous, smooth slope along its energy interval. This permitted us to find an average point to estimate the height of each bin, which we then used to find the area of the bin. We summed these areas to find the area of the total curve, that is, the total activity which we expected to measure from the decay.

We divided the experimental spectrum into corresponding energy bins. We then found the area of each bin in the experimental curve and divided it by the total expected value from the theoretical curve. This gave us a normalized value for the activity at each energy bin. Due to extreme scattering of the low energy particles in the experimental curve, we neglected energies below 250 keV in this analysis.

Beta spectra analysis include a statistical factor, a shape factor, and a coulomb correction factor.[5] For the thallium beta spectrum we used a shape factor derived from the wave function of the electrons and neutrinos to correct the shape of the spectrum curve. However, for energies greater than 200 keV the shape factor shows no deviation from the allowed form for ^{209}Pb, and so we were able to neglect it in our analysis of this spectrum.[2] Hence the thallium curve shows the effects of this factor, while the lead curve is unaffected (see appendix A, figure 3).

Shape factor:

$$c(W) = k\left(1 + a\,W + \frac{b}{W} + c\,W^2\right) \qquad [1]$$

Statistical factor:

$$\frac{dn}{dW_o} = 16\,\frac{\pi^2\, m_o^{\,5}\, c^4\, \eta^2\, (W_o - W)^2}{h^6} \qquad [2]$$

where η is the electron's momentum in units p/m_0c and W is the total energy (including rest energy m_0c^2). The statistical factor $\eta^2(W_0 - W)^2 d\eta$ gives a good representation of the allowed beta-ray spectra in nuclides of low mass number. [6]

The number of counts as a function of energy depends upon the shape factor C, the momentum of the electron and the neutrino (p and q, respectively), the end point energy W_0, and the F function, a statistical factor derived from tables.[7]

$$N(W) = C\,p^2\,(W_o - W)^2\,(p^2 + q^2)\,F \qquad [3]$$

where the total energy is denoted by W and the F function was obtained from tables.[8] The area of each bin was divided by the total area as determined from the theoretical spectrum to give a normalized constant, which was then multiplied by the efficiency of the detector as known from our calibration, the activity, and the branching ratio of the decay to give the count rate as shown below.

$$C = \frac{a\,b\,\varepsilon \int_{W_i}^{W_f} N(W)\,dW}{\int_{0}^{W_o} N(W)\,dW} \qquad [4]$$

where C represents the counting rate,a is the activity of the sample, b is the branching ratio, and ε is the efficiency of the detector. The neutron capture cross section is calculated from the number of atoms in the sample N_{208}, the activity a, the neutron flux in the reactor Φ, and an exponential factor.

$$a_{EOB} = N_{208}\,\sigma\,\Phi\,(1 - e^{(-\lambda t)}) \qquad [5]$$

where $\lambda = \ln(2)/T_{1/2}$ and t is the length of time that the sample was irradiated, 2 hours in this instance.

Results

Our results show considerable scattering of the lower energies, thus we neglect values below 250 keV (appendix A, figure 2). A graph of the cross sections measured at each energy bin shows a nearly linear curve in the lower energies, which then levels off at higher energies (appendix A, figure 4). Extrapolation to higher energy levels would probably give a more constant value. However, due to the small recorded activity in the upper energy bins, we were unable to obtain statistically valid data at those points. We therefore focused on the measurements of the cross section from the last two energy bins, where the curve leveled off.

Upon averaging the values from 350 to 450 keV, we obtain a value of 0.091 millibarns for the average neutron capture cross section of ^{208}Pb. We estimate an error in this value of less than 20%, or 0.018 millibarns. See Appendix B, chart 2 for a summary of experimental procedures and results.

$$\sigma = 0.091 \text{ +/- } 0.018 \text{ millibarns} \qquad [6]$$

Recommendations for Future Analysis

A silicon detector was used in taking our measurements. This had no provisions to compensate for backscattering of electrons, nor was it thick enough to entirely stop high energy electrons. We therefore experienced some difficulties in using it to detect beta decay. As we were only sampling from one side of the sample, some beta particles clearly escaped the detector entirely. By itself, this would not be a difficulty, because our calibration would account for it. But it is also likely that at least some electrons lost only part of their energy upon striking the detector, and were then backscattered without recording all of their energy. It is also likely that some fast electrons passed entirely through the detector without stopping. The detector was only about one millimeter thick and may have been insufficient to stop the higher energy electrons. These concerns were accounted for in our detector efficiency calibration. However, a more accurate measurement may be obtained by using a detector specifically designed to measure beta radiation.

We propose the use of a beta detector made from plastic scintillator material.[3] This would be both cheaper than the material used for gamma ray detection and easier to cut. Hence we could use large blocks of the material, which would effectively absorb the high energy electrons that currently escape the silicon detector.

Electrons that enter the material will excite the atoms to create a flash of light. This signal could be measured in an electronic circuit connected to the scintillator. By creating a sandwich of the material into which a sample would be placed, radiation would be measured on both sides of the sample. Also, electrons which scattered from one plate would be absorbed in the other. Signals recorded from both plates and would add together. In this manner the problem of backscattering would be solved, for any energy unrecorded by one plate would be absorbed by the second half of the sandwich.

To prevent contamination from background light the scintillator would have to be wrapped in a light absorbent material. This process might cause a delay before the sample could be measured--a difficulty in regards to the ^{209}Pb sample with its 3.25 hour half life. However, the process would be facilitated by having the scintillator blocks ready before hand. One could then insert the sample and wrap it quickly before taking data. This should not be a significant delay in comparison to the time needed to process the sample at the radiation center before measurement.

Discussion

Despite limitations in our detecting equipment, we have obtained a reasonable and accurate value for the neutron capture cross section of ^{208}Pb. Our value of 0.091 millibarns is considerably smaller (54 %) than the previously measured value of approximately 0.2 millibarns. It is our hope that this new data will prove useful, as related experiments have suggested that the previous value was too large.

The size of our measurement is striking. ^{208}Pb has the smallest neutron capture cross section of all the heavier elements.[9] A more typical cross section might be about 1 barn, with some nuclei ranging as large as 10^5 barns. To examine the area of a "target" presented by a ^{208}Pb atom, we assume a radius r = 7.1 fm [10]. Then the target area is given by

$$A = \pi r^2 = 1.6 \times 10^{-24} \text{ cm}^2 = 1.6 \text{ barns.} \qquad [7]$$

In addition to the this tiny target area, it is possible that the small cross section is related to the closed shells of ^{208}Pb, as an incoming neutron may be weakly absorbed if the target atom's neutron shells are full.

Another factor that affects neutron capture is the possibility of resonance between the incoming neutron's wave function and that of the target nucleus. Should the two waves be in phase, the neutron would be more easily absorbed. ^{208}Pb, however, has no low energy resonances.[10] This diminishes the probability of neutron capture.

Finally, capture of high energy neutrons is less probable due to speed of such neutrons. In simple terms, fast neutrons move too quickly past the target nucleus to be absorbed. Slower neutrons are within range of the nucleus longer, which increases their chance of being captured.

All of these factors contribute to the tiny cross section of ^{208}Pb. Due to its small size and the beta decay of ^{209}Pb, the measurement of the neutron capture cross section of

^{208}Pb continues to be a challenge to experimentalists. Our results indicate that previous measurements may have been too large. With the use of our proposed scintillator detector, and more detailed analysis, this value may be made even more precise. In any event, the problem merits further study.

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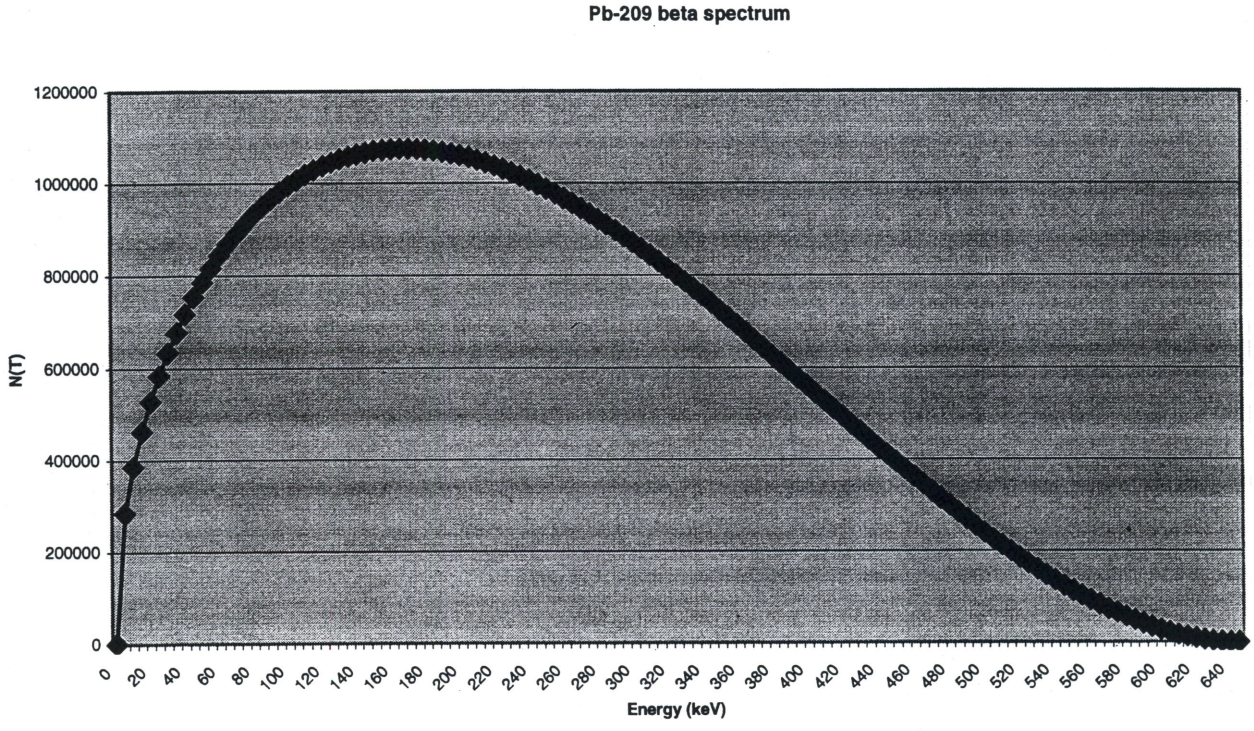
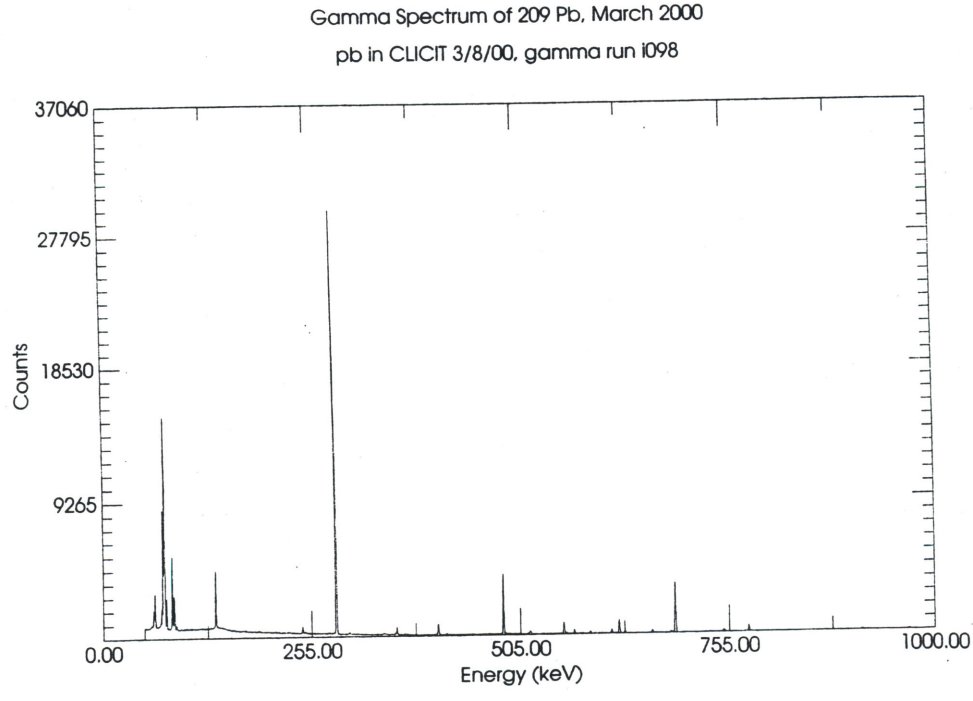
APPENDIX A

Appendix A, figure 1: A comparison of an ideal beta spectrum with a gamma ray spectrum.

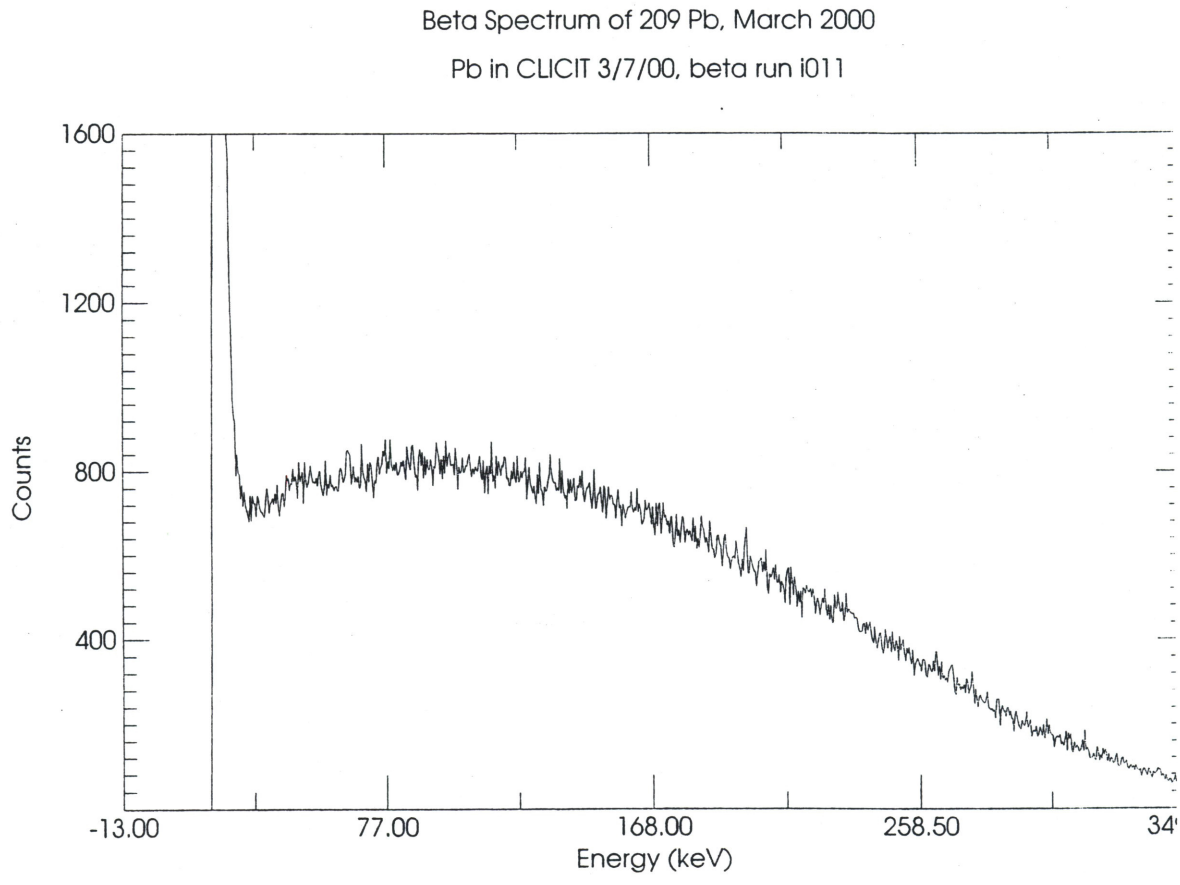
Ideal Beta Spectrum for ^{209}Pb.



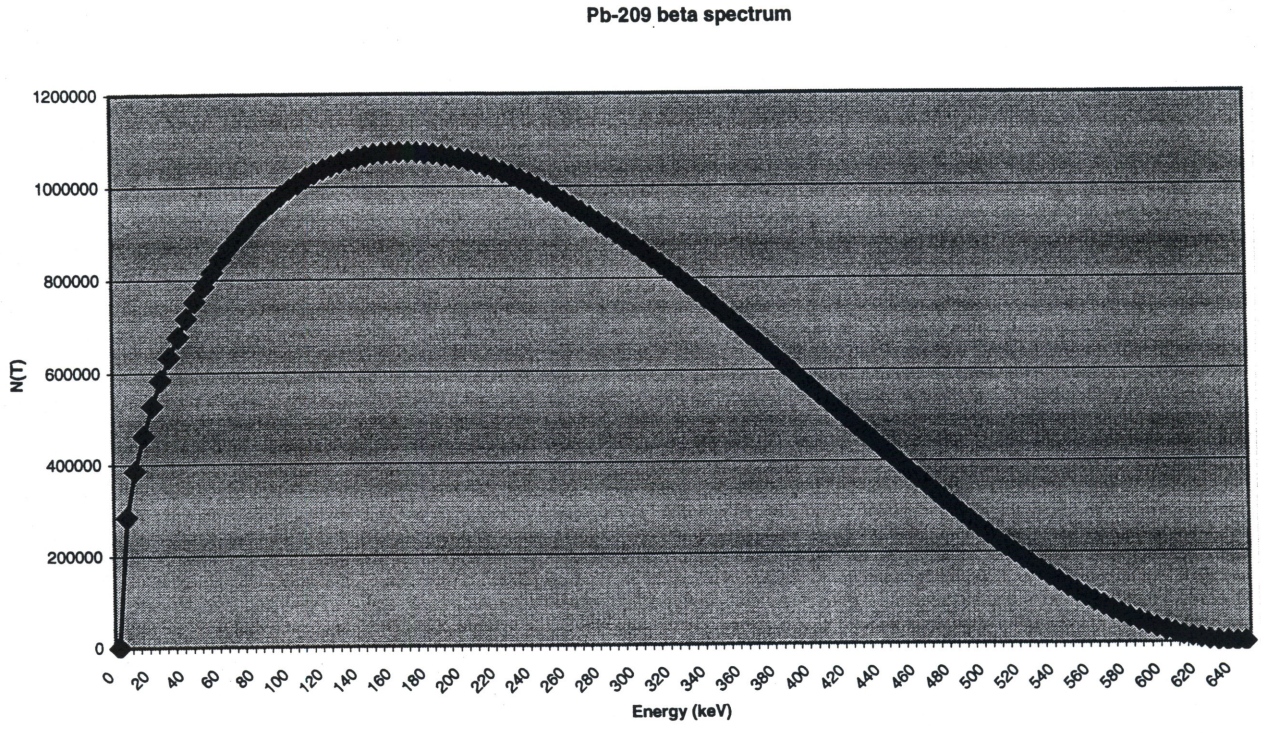
Experimental Gamma Spectrum of ^{209}Pb, March 2000.

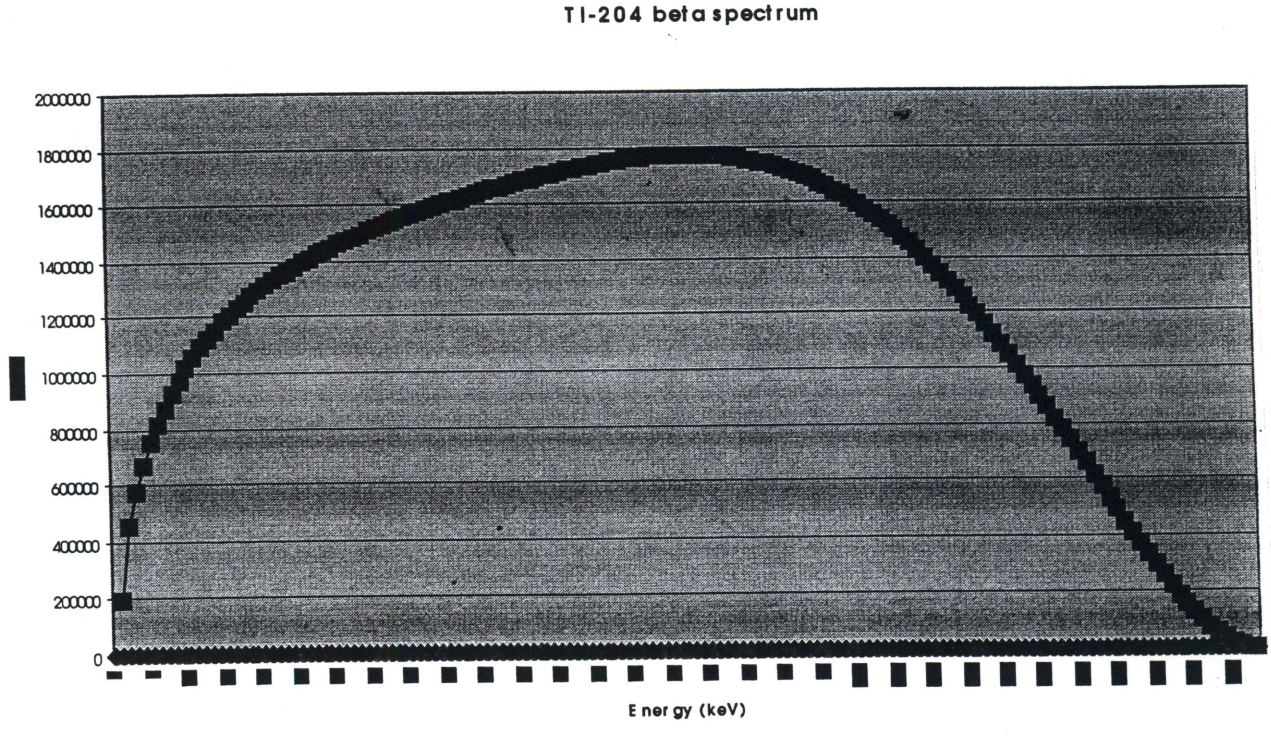


Appendix A, figure 2: Experimental Beta Spectrum of ^{208}Pb.

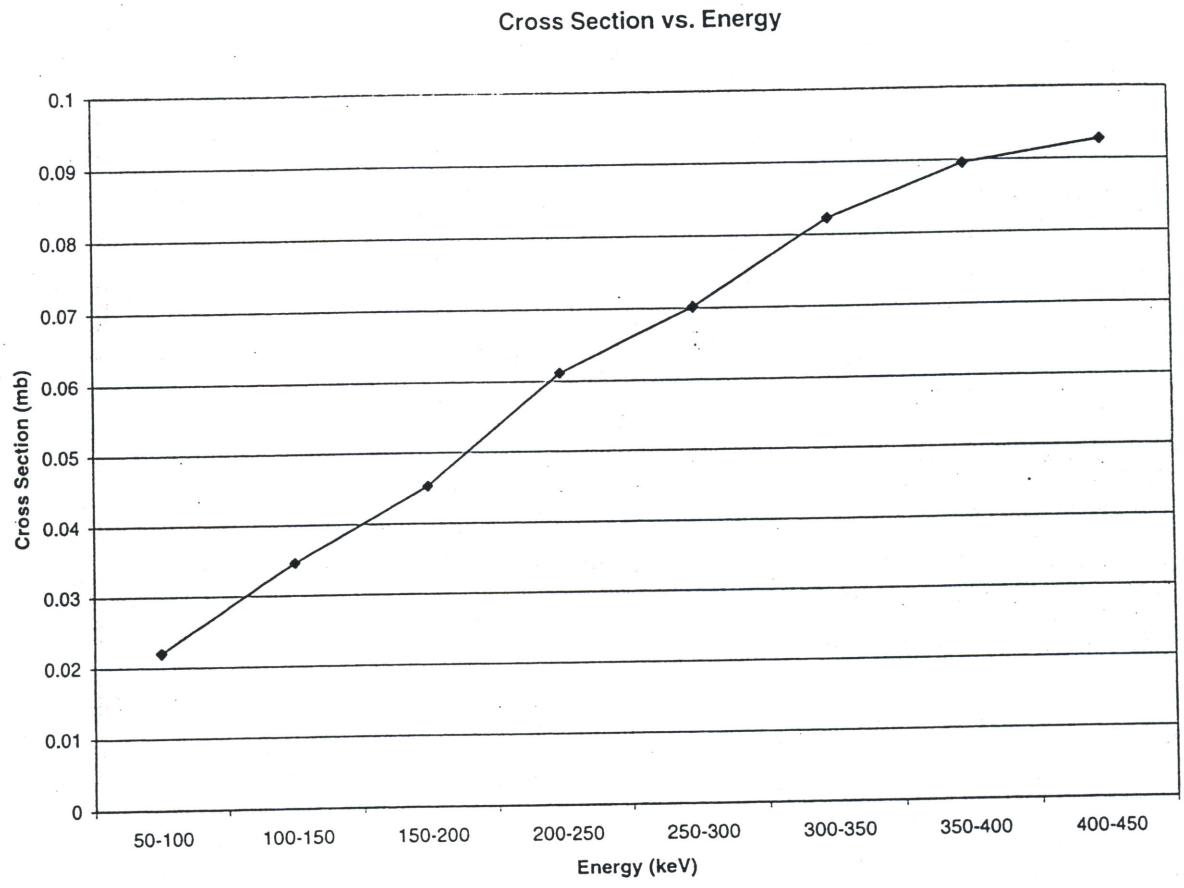


Appendix A, figure 3: Effects of the Shape Factor on Theoretical Plots of the Beta Spectrum of ^{208}Pb and ^{204}Tl.

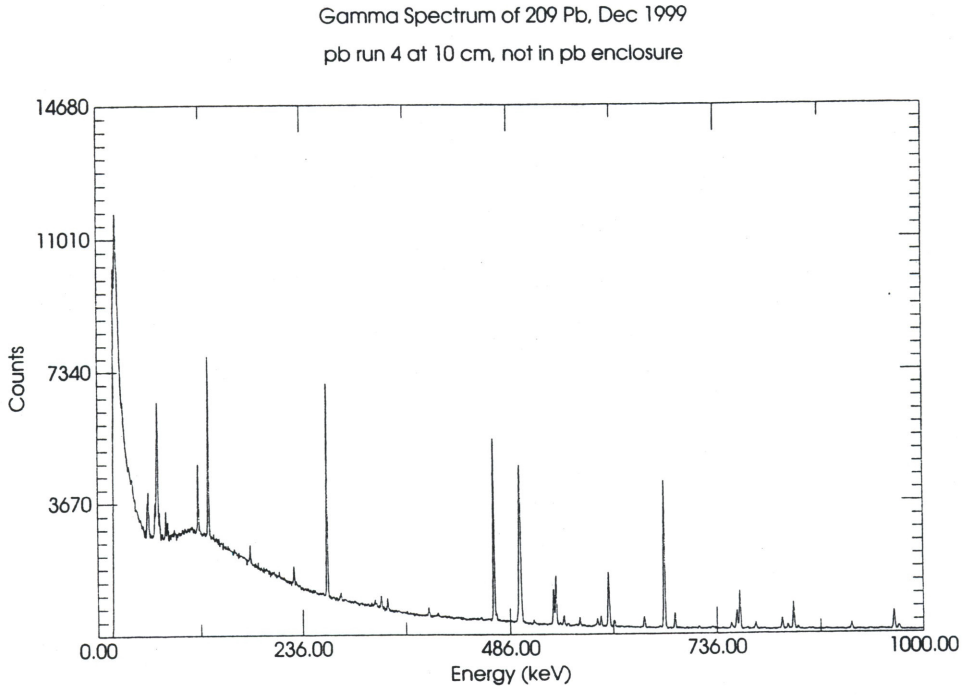


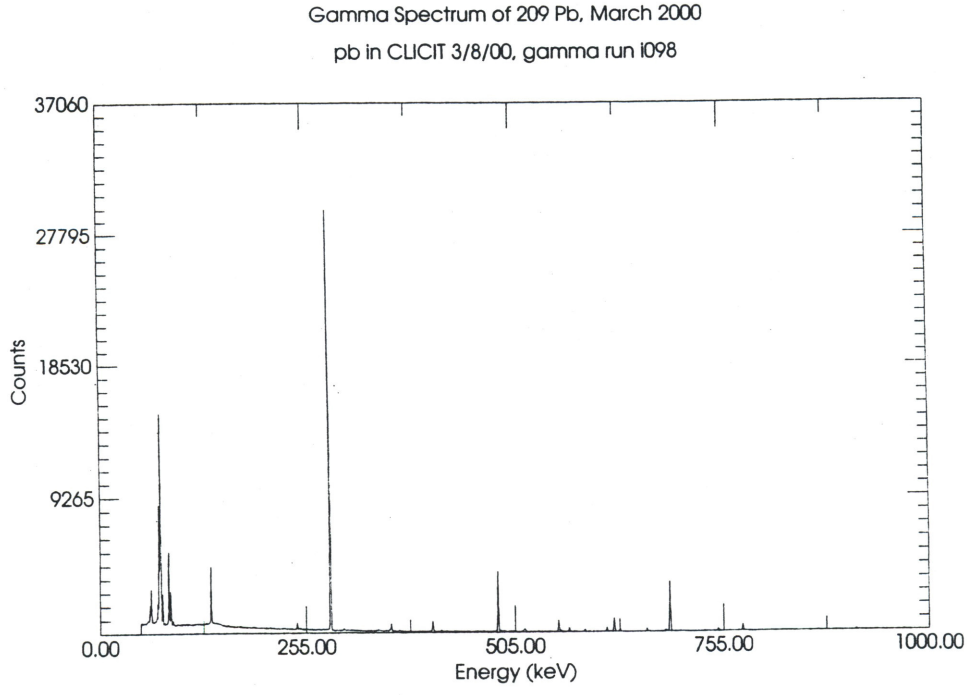


Appendix A, figure 4: The Cross Section of ^{208}Pb Measured at Various Energy Levels.



Appendix A, figure 5: The Gamma Spectra of the December 1999 Data and the March 2000 Data. Peaks indicate the presence of contaminants.





APPENDIX B

Appendix B, Chart 1: Impurities Present in the December 1999 and March 2000 Data.

December 1999
(partial list)

Energy (keV)	Intensity	Isotope
121.8	295	^{152}Ru
134.2	41	^{187}W
138.3	3	^{116}In
374.4	90	^{204}Pb
344.3	400	^{152}Eu
416.9	32	^{116}In
479.5	100	^{187}W
551.6	23	^{187}W
554.3	85	^{82}Br
619	52	^{82}Br
618.3	29	^{187}W
685.8	125	^{187}W
841.7	595	^{152}Eu
846.8	100	^{56}Un
899.2	100	^{204}Pb
911.9	97	^{204}Pb
2754.1	100	^{24}Na

March 2000

Energy (keV)	Intensity	Isotope
479.5	302 +/- 29	^{187}W
685.8	342 +/- 31	^{187}W
1368.8	69 +/- 16	^{24}Na
2753.9	45 +/- 7	^{24}Na

Appendix B, Chart 2: Summary of the Experimental Procedures and Results.

Isotope studied:	Half-Life	Decay method	End-point Energy	Activity	Cross-section
208 Pb --> 209 Pb	3.25 hrs.	beta decay	645 keV		0.084 +/- 0.017 millibarns
Calibrated With:					
204 Tl	3.78 yrs.	beta decay	763 keV	0.868 x 0.0892 uCi	

Experiment:	Facility:	Neutron Flux:	Neutrons	Time irradiated:	Rolled foil?	Successful?
Dec. 1999	RABBIT	1.1 x 10^13 n/cm^2/s	90% thermal, 10% fast	4 hours	Yes	No
March, 2000	Thermal Column	7.35 x 10^10 n/cm^2/s	thermal only	2 hours	No	Yes