

AN ABSTRACT OF THE THESIS OF

Seung-Hyuk Baik for the degree of Master of Science in Radiation Health
presented on August 8, 1991.

Title : Determination of $^{90}\text{Sr}/^{90}\text{Y}$ Contamination Using Beta-Particle
Spectroscopy with Active Gamma-Ray Discrimination

Abstract approved : Redacted for Privacy
/ Jack F. Higginbotham

Strontium-90 is a biologically significant radionuclide because of its bone seeking characteristic, its long radiological half-life(29.1 y) and its high cumulative decay energy. In the event of a catastrophic accident at a nuclear power facility a more rapid analysis method for determining ^{90}Sr concentration in the environmental matrices would be required to implement post-accident recovery operations. A beta-particle spectrometer system has been developed for prompt evaluation of ^{90}Sr activity in the presence of gamma-ray and low energy beta-particle components of mixed beta/gamma radiation field. Two detectors were used in this work. A gas-flow proportional counter which is sensitive only to beta-particles is used to gate a spectrometer to accept interactions induced by beta-particles. The proportional counter is located in the front of a scintillator so that beta-particles must traverse the first detector before interacting with scintillator. The $^{90}\text{Sr}/^{90}\text{Y}$ detection limits and instrument response parameters were determined. The sensitivity of the spectrometer depended on the background activity, the counting time, the mass or volume of the sample, and the interfering elements' activity. The evaluations show that a count time of 2 hours

yields a sensitivity value of $300 \text{ BqL}^{-1} \text{ } ^{90}\text{Sr}$ in about 1 liter of water sample. The spectrometer based analysis method is 10-300 times faster than the chemical separation method.

Determination of $^{90}\text{Sr}/^{90}\text{Y}$ Contamination Using Beta-Particle
Spectroscopy with Active Gamma-Ray Discrimination

by
Seung-Hyuk Baik

A THESIS
submitted to
Oregon State University

in partial fulfillment of
the requirement for the
degree of

Master of Science

Completed August 8, 1991

Commencement June 1992

APPROVED:

Redacted for Privacy

Assistant Professor of Nuclear Engineering/Radiation Health Physics

Redacted for Privacy

Head of Department of Nuclear Engineering

Redacted for Privacy

Dean of Graduate School

Date thesis is presented August 8, 1991

Type by Seung-Hyuk Baik for Seung-Hyuk Baik

ACKNOWLEDGMENTS

I would like to express my gratitude to my advisor Dr. Jack H. Higginbotham. His intensive help and encouragement to the completion of this thesis are gratefully appreciated and will be remembered. I am very thankful to my thesis committee, Dr. Brian Dodd, Dr. Roman A. Schmitt and Dr. Charles W. Drake.

I would appreciate the Korean Government, the Department of Nuclear Engineering at Oregon State University and the Oregon Department of Energy for the financial support.

I would also thank Mr. Mike Conrady of the Oregon State University Radiation Center for his kindly help in preparing the radiation sources.

I would like to acknowledge my parents, parents-in-law, sister and brothers. I can't complete acknowledgement without mention of my lovely wife, Yun-Kyung, and my lovely daughter, Jang-Mee, who never lost faith to my abilities and patiently offered the encouragement to me. I do not know how express my thanks to these two ladies.

Finally, I wish to dedicate this thesis to my deceased grand parents, Woon-Yi Baik and Oh-Chae Lee, who are always remained in my memory with love.

TABLE OF CONTENTS

	<u>Page</u>
1. INTRODUCTION	1
2. DETERMINATION OF STRONTIUM ACTIVITY BY ION EXCHANGE METHODS	4
2.1 Introduction	4
2.2 Reagents and Apparatus	5
2.2.1 Milk Sample	5
2.2.2 Grass Ash Sample	5
2.3 Experimental Procedures	7
2.3.1 Milk Sample	7
2.3.2 Grass Ash Sample	8
2.4 Results and Discussion	11
2.4.1 Milk Sample	11
2.4.2 Grass Ash Sample	13
2.4.3 Discussion	15
3. DETERMINATION OF STRONTIUM USING A BETA-PARTICLE SPECTROMETER	16
3.1 Introduction	16
3.2 Methodology	17
3.2.1 Characteristic of $^{90}\text{Sr}/^{90}\text{Y}$ and Interfering Emitters	17
3.2.2 Theoretical Study	25
3.2.3 Sensitivity Measurement	28
3.3 Experimental Equipment	32
3.3.1 Gas-Flow Proportional Counter	32

3.3.2 Plastic Scintillator Based Beta-Particle Spectrometer	37
3.3.3 Spectrometer System	40
3.4 Experimental Procedures	48
3.4.1 Preparation of Radiation Sources	48
3.4.2 Spectrometer Energy Calibration	53
3.4.3 Gamma-Ray Discrimination Capability of Spectrometer	56
3.4.4 Determination of Instrumental Parameters	58
3.5 Results	59
3.5.1 Energy Calibration of Spectrometer	59
3.5.2 Gamma-Ray Discrimination Capability	62
3.5.3 Determination of Instrumental Parameters	65
3.5.4 Measurement Sensitivity	67
4. CONCLUSIONS	72
5. FUTURE WORK	74
6. REFERENCES	75

LIST OF FIGURES

<u>Figure</u>	<u>Page</u>
Fig. 3.1 Side and End View of Gas-Flow Proportional Counter.	35
Fig. 3.2 Beta Plateau Measurement for Gas-Flow Proportional Counter Using ^{99}Tc with P-10 Gas.	36
Fig. 3.3 Components of Plastic Scintillator Assembly.	39
Fig. 3.4 Block Diagram of the Beta particle Spectrometer with Active Gamma-Ray Discrimination.	44
Fig. 3.5 Pulse Timing Characteristics of the Spectrometer System.	45
Fig. 3.6 Timing Diagram for the Linear Gate Stretcher Operating in the Gamma-Ray Discrimination Mode.	46
Fig. 3.7 ^{207}Bi Pulse Height Distribution.	47
Fig. 3.8 Side and End View of the Instrumental Measuring Sources Container.	52
Fig. 3.9 Energy Calibration Line of Spectrometer System.	61
Fig. 3.10 Minimum Detectable ^{90}Sr Specific Activity as a Function of Sample Mass(or Volume) M(in convenient units).	69
Fig. 3.11 Minimum Detectable ^{90}Sr Specific Activity as a Function of Counting Time.	70
Fig. 3.12 Minimum Detectable ^{90}Sr Specific Activity in Higher Efficiency Detector Systems than This Work.	71

LIST OF TABLES

<u>Table</u>	<u>page</u>
2.1 Results of the Strontium Determination from Milk	12
2.2 Results of the Strontium Determination from Grass Ash Sample	14
3.1 Beta Particle Emitting Radioisotopes. Fission Products with Yield ≥ 0.05 %, Cosmogenic, and Primordial Elements	20
3.2 Mixed Beta Particle and Gamma Ray Emitting Radioisotopes Fission Products with Yield ≥ 0.05 %, Cosmogenic, and Primordial Elements	22
3.3 The Concentrations of Nuclides in Different Environmental Matrices	24
3.4 Specifications of the Gas-Flow Proportional Counter	34
3.5 Physical Characteristics of BC-400 Plastic Scintillator	38
3.6 Spectrometer Equipment and Typical Instrument Settings	43
3.7 The Energy Calibration Sources of the Spectrometer System	50
3.8 The Instrumental Parameter Measuring Sources of the Spectrometer System	51
3.9 Energy Calibration Data	60
3.10 Gamma-Ray Discrimination Capability of a Spectrometer System	64
3.11 Instrumental Parameters of Spectrometer System	66

DETERMINATION OF $^{90}\text{Sr}/^{90}\text{Y}$ CONTAMINATION USING BETA-PARTICLE
SPECTROSCOPY WITH ACTIVE GAMMA-RAY DISCRIMINATION

Chapter 1
INTRODUCTION

In post accident conditions at nuclear power facilities, prompt analysis of soil and vegetation samples is important for the determination of protective action recommendations. One of the radionuclides of primary concern during such an event is ^{90}Sr . A significant fraction of the fission product inventory of a nuclear power reactor is ^{90}Sr and in the case of very severe accidents it can be released into the environment. Strontium-90 is biologically very significant because it is a bone-seeking nuclide, with a long radiological half-life (29.1 year) and high cumulative decay energy. ^{90}Sr decays by emission of beta particles with a maximum energy of 546 keV to ^{90}Y which decays 99.98+ % of the time by emission of β particles with maximum energy of 2281 keV [1]. The half-life of ^{90}Y is 64 hours and it is in equilibrium with ^{90}Sr .

The usual analytical techniques for qualifying ^{90}Sr contamination on soil, vegetation and in milk involve a chemical separation process which isolates the ^{90}Sr from the mixed fission products and a counting ^{90}Y after a 2 week ingrowth period. Various chemical analysis techniques have been used to measure ^{90}Sr in environmental samples [2-7]. Several authors report the use of liquid-scintillation counting for measuring ^{89}Sr and ^{90}Sr , including various techniques for unfolding mixed beta spectra of

^{89}Sr , ^{90}Sr , and ^{90}Y [8-10]. Bremsstrahlung radiation spectroscopy of a Ge(Li) detector has also been proposed [11-12]. Cerenkov counting has been used after separation of Sr followed by Y separation [13-14]. Most recently, a multi-detector system has been introduced that consists of a plastic scintillator beta particle detector and a guard ring of Bismuth Germanate (BGO) [15]. These method needs a 2 week count time to identify the concentration of ^{90}Sr in environmental matrices. This would be highly undesirable in a emergency condition. Thus a more rapid analysis method would be required for responding to the needs of a post-accident recovery operations.

In this work, a beta-particle spectrometer system has been developed which directly measures the concentration of ^{90}Sr and ^{90}Y in environmental samples. The system actually discriminates against the gamma-ray and low energy beta particle components of mixed beta/gamma radiation fields which would be emitted by fission product radionuclides also deposited on samples. To achieve active gamma-ray discrimination, a two detector system was used; the gas-flow counter and scintillator. The first detector, sensitive only to beta particles, was located in front of a scintillator so that beta particles must traverse the first detector before interacting with the scintillator. The gas-flow counter was used to gate the spectrometer to accept interactions induced by beta particles. The objective of this work is to determine if this spectrometer system can significantly decrease the length of time required to perform an analysis.

This thesis is presented in two major parts. The first part describes the chemical separation of Sr from milk and grass to determine the length of time required to complete the process and the method's separation yield. The two detector beta-particle spectrometer system and data analysis methodology are depicted in part two. A comparison of the two methods is then made.

Chapter 2

DETERMINATION OF STRONTIUM ACTIVITY BY ION EXCHANGE METHODS

2.1 Introduction

Chemical separations of stable Sr from milk and grass ash were performed to identify the method's separation yield and to determine the length of time required to process.

A known amount of stable Sr, in the form of $\text{Sr}(\text{NO}_3)_2$, was added to a milk or grass ash sample as a carrier to determine percent yield. A cation exchange resin column was used to separate Sr from other elements. The Sr was eluted from the column, precipitated as SrCO_3 , dried, and weighed to determine amount of Sr recovered.

The milk sample used in this work was obtained from a local grocery store and the grass sample was collected from the south field of the Oregon State University Radiation Center.

2.2 Reagents and Apparatus

2.2.1 Milk Sample

The equipment and supplies needed to perform the Sr separation include:

Cation exchange resin, Dowex 50W-X8, 50-100 mesh, Na⁺ form, or equivalent, a one liter separatory funnel and ion exchange column to contain 40 ml resin, and metrical DM-800 membrane filters or equivalent, 0.8 micrometer pore size and suction filter apparatus.

Prepare Sr carrier (20 mg of Sr⁺²/ml) by dissolving 48.3 g of anhydrous Sr(NO₃)₂ in 900 ml of water and 1 ml of 16 M HNO₃, and dilute to 1 liter. Prepare Ba carrier (20 mg of Ba⁺²/ml) by dissolving 38.1 g of Ba(NO₃)₂ in 900 ml of water and 1 ml of 16 M HNO₃ and diluting to 1 liter.

Prepare pH 5.0 and 5.2 CH₃COONH₄ buffer by dissolving each 153 g of CH₃COONH₄ in 700 ml of water and adjusting pH 5.0 and 5.2 with glacial CH₃COOH, and dilute to 1 liter.

Prepare 3 % Na₂EDTA by dissolving 33.3 g of Na₂EDTA in 900 ml water and diluting to 1 liter. Prepare EDTA complex solution by dissolving 216 g of Na₂EDTA in 2500 ml water and adding 20 ml each Sr and Ba carrier. Then add 200 ml CH₃COONH₄ buffer, pH 5.2 and adjust pH to 5.65 with 15 M NH₄OH. Dilute to 3 liters.

2.2.2 Grass Ash Sample

The equipment and supplies needed to perform the Sr separation include:

Nickel crucible, 250 ml volume and lid, a convection oven, blast burner and other needed apparatus are almost same with milk sample.

Prepare resin and Sr carrier (20 mg of Sr^{+2}/ml) by same procedure as outlined in section 2.2.1.

Prepare 6 % and 2 % Na_2EDTA by dissolving each 66.6 g and 22.2 g of Na_2EDTA in 900 ml water and diluting to 1 liter.

Prepare Ba carrier(5.0 mg Ba^{+2}/ml) by dissolving 9.5 g $\text{Ba}(\text{NO}_3)_2$ in 900 ml of water and 1 ml of 16 M nitric acid and dilute to 1 liter.

Prepare pH 4.6 CH_3COONa buffer by dissolving 200 g of CH_3COONa in 500 ml water, adjusting pH to 4.6 with glacial CH_3COOH and diluting to 1 liter.

2.3 Experimental Procedures [6]

2.3.1 Milk Sample

Filter a 1 liter sample of milk through cheese cloth, add 300 ml of the complex solution to the milk sample and mix well. Adjust milk mixture to pH 5.2 with 15 M NH_4OH using pH meter.

Pour the milk sample into a graduated separatory funnel and attach to the top of the ion exchange column. Open the stopcock on the reservoir to column and allow the milk to flow through the resin by gravity at 20 ml per minute. After all the milk has passed through the column, rinse sides at top of column with 50 ml water and wash with approximately 200 ml water until the effluent runs clear. At this time, do not let column run dry.

Add 800 ml of 3 % Na_2EDTA (pH 5.2) to the reservoir and elute through the column at 10 ml per minute. Add 200 ml of water to reservoir and run through column at 10 ml per minute. Add 200 ml of 1.5 M NaCl to reservoir and pass solution through the column at a flow rate of 5 ml per minute. Add 1000 ml 4 M NaCl to reservoir and pass through the solution the column at 10 ml per minute. Collect the first 400 ml of elute, which contains Sr and Ba, and allow the remaining 600 ml NaCl to pass through the column to regenerate the resin. Wash all excess NaCl from resin with water and check elute for chloride with 0.1 M AgNO_3 before reusing resin.

Add 1 ml of 6 M NaOH to the 400 ml of elute. Stir and slowly add 10 ml of 1.5 M Na_2CO_3 to precipitate SrCO_3 . Continue vigorous stirring for 30 minutes using magnetic stirrer. Transfer one half of the contents to a 250 ml centrifuge bottle. Centrifuge at 2000 rpm for 10 minutes and carefully pour off and discard supernate. Add the remaining solution to the precipitate in the 250 ml centrifuge bottle. Centrifuge at 2000 rpm

for 10 minutes, pour off and discard supernate.

Place the centrifuge bottle containing the precipitate in a hot water bath held at 70°C. Add 5 ml of 1 M HNO₃ to centrifuge bottle to dissolve precipitate. Transfer solution to a 50 ml centrifuge tube. Wash the 250 ml bottle with 5 ml of CH₃COONH₄ buffer, pH 5, and add the wash to the dissolved precipitate in the 50 ml centrifuge tube. Heat the 50 ml tube at 70°C in water bath with stirring and slowly add 1 ml of 0.25 M Na₂CrO₄ to precipitate BaCrO₄. Cool the tube in ice bath, centrifuge and decant supernate containing strontium into another 50 ml centrifuge tube. Discard BaCrO₄ precipitate.

Add 2 ml 15 M NH₄OH to the supernate with stirring and precipitate SrCO₃ by adding 2 ml 1.5 M Na₂CO₃. Stir precipitate for ten minutes. Centrifuge for five minutes. Pour off and discard supernate. Add 20 ml of water to the tube to wash the precipitate. Centrifuge and discard wash solution. Take up precipitate in 10 ml water. Gently swirl to break up precipitate.

Filter sample through a membrane filter. Wash sequentially with three 10 ml aliquots each of water and then three 10 ml aliquots C₂H₅OH. Dry sample for one hour in a desiccator. Weigh filter paper with precipitate, calculate weigh of precipitate, and compute chemical yield and the elapsed time.

2.3.2 Grass Ash Sample

Dry the sample at 110°C to constant weight. Ash the dried sample at 550°C for no less than 72 hours. Place 5 g of ash in a 250 ml nickel crucible. Add 2 ml of Sr carrier and 1 ml Ba carrier to the ash. Add 1 ml

of 2 M $\text{Ca}(\text{NO}_3)_2$ solution. Add 25 g of NaOH pellets, mix and fuse over a blast burner for 15 minutes. Slowly add 3 g of anhydrous Na_2CO_3 , swirl to mix, and heat the clean melt for 20 minutes. Transfer the crucible from the heat to a cold water bath in order to crack the fusion mixture. Transfer the sample to a 250 ml centrifuge bottle and centrifuge 5 minutes. Discard supernatant solution. Wash the residue twice with 200 ml portions of hot water, discarding the supernate each time.

Dissolve the residue in 200 ml 6 M HCl by gently boiling until the solution is transparent. Add 100 ml of water. Filter. If insoluble residue (silica) is present, wash the residue twice with 100 ml portions of water, and add wash solutions to the filtered solution. Discard residue. Add the filtrate to 500 ml of 6 % Na_2EDTA solution in a 2 liter beaker. Adjust pH to 3.8 with the pH meter using approximately 10 ml of 15 M NH_4OH . Stir vigorously for 75 minutes using a magnetic stirrer to precipitate the magnesium salt of EDTA. Filter off any MgEDTA and adjust filtrate to pH 4.6 (pH meter) with approximately 2 ml of 15 M NH_4OH . Add 20 ml of CH_3COONa buffer solution, pH 4.6 and readjust solution to pH 4.6 (pH meter) with approximately 4 ml of 15 M NH_4OH . Dilute to 1 liter.

Transfer solution to column reservoir and let flow through the cation resin at flow rate of 10 ml per minute. Stop the flow when just enough solution remains to cover top of resin in column. Adjust pH of 600 ml, 2 percent Na_2EDTA to 5.1 with 6 M NH_4OH , place in reservoir, and let flow through column at 10 ml per minute. Wash the column with 200 ml of water at a flow rate of 10 ml per minute. Discard all effluents. Place 460 ml of 1.5 M HCl in reservoir and elute at a flow rate of 8 ml per minute. Discard the first 60 ml of effluent. Collect the next 400 ml,

which contains the strontium fraction.

To the Sr fraction, add 200 ml of 15 M NH_4OH , and stir with a magnetic stirrer. Slowly add 10 ml of 1.5 M Na_2CO_3 solution, and stir 30 minutes. Collect SrCO_3 on a tared membrane filter. Wash with three 10 ml aliquots each of water and $\text{C}_2\text{H}_5\text{OH}$. Transfer to a planchet and allow sample to dry one hour in a desiccator. Weigh the precipitate and calculate the chemical yield and the elapsed time.

2.4 Results and Discussion

2.4.1 Milk Sample

Four analyses of milk samples were performed to compute chemical yield and measure the length of time required to perform the separation. The results are presented in Table 2.1. The chemical yields averaged 75.5 percent and ranged between 60 and 90 percent. The yield is systematically high because of the presents of stable Sr in milk. The average stable Sr value of milk is 0.86 mg per liter [16]. It creates an analytical error of 9 to 10 percent.

The results of this work agree well with the values of cited in the literature. The average yield of a variety of food samples was $70 \pm 3\%$ in literature [2]. The elapsed time for performing these chemical separations has been between 4 to 5 days.

Table 2.1 Results of The Strontium Determination from Milk

Sample	Weight of SrCO ₃ (mg)	Yield(%)	Elapsed Time(Days)
Milk # 1	60.7	90	5
Milk # 2	43.2	64	4
Milk # 3	49.2	73	5
Milk # 4	50.6	75	5
Mean		76 ± 9%	4.8 days

2.4.2 Grass Ash Sample

Two ash samples were performed to determine the chemical yield and the elapsed time for analysis. The yields averaged 49 percent and ranged between 42 and 56 percent. Due to the presence of stable strontium in vegetation, the yield is overestimated by 0.5 to 10 %. The range of stable strontium in vegetation is 0.2 to 4 mg [2].

In this work, the chemical yields are less than the literature cited values. The average yield of a variety of food samples was 70 ± 3 % in literature [2]. These may result from the errors during the ash preparation step. The times required are 9 to 10 days including 4 days sample preparation time. The results are presented in Table 2.2.

Table 2.2 Results of The Strontium Determination from Grass Ash Sample

Sample	Weight of SrCO ₃ (mg)	Yield(%)	Elapsed Time(Days)
Ash # 1	28.3	42	10*
Ash # 2	37.7	56	9*
Mean		49±7%	9.5 Days

* It includes 4 days sample preparation time.

2.4.3 Discussion

In this work only stable strontium was used to determine the strontium yield. Once the chemical yield is known additional information on the beta particle background activity, the detection yield of the detector and the ingrowth of ^{90}Y are needed to calculate the ^{90}Sr concentration of a sample. Most radiostrontium separation methods need a 2 week waiting period to allow for ^{90}Y ingrowth for counting.

Chapter 3

DETERMINATION OF STRONTIUM ACTIVITY USING A BETA-PARTICLE SPECTROMETER

3.1 Introduction

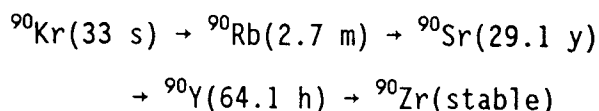
A beta-particle spectrometer system has been developed which directly measures the energy distribution of beta particles from a $^{90}\text{Sr}/^{90}\text{Y}$ source while actively discriminating against the gamma-ray and low energy beta-particle components of mixed radiation fields [19]. A two detector configuration was used, where the first detector was a gas-flow counter, positioned in the front of the entrance window to the second detector, a BC-400 plastic scintillator. Since the gas-flow counter was very insensitive to gamma-rays, it was used as a trigger detector to gate the spectrometer to accept only interactions induced by high energy beta particles.

The objective is to determine how applicable the above spectrometer system would be to quantify the amount of $^{90}\text{Sr}/^{90}\text{Y}$ in environmental samples. This is done by first determining what the expected radioisotopic composition in environmental samples is in post accident conditions at a nuclear power facility. This is done primarily by looking at the release data from the Chernobyl accident. Then radioisotope standard sources are prepared to evaluate the response parameters of the detection system. Specifically, this work will examine the detection limit as a function of sample count time, sample mass or volume and interference from other radioisotopes.

3.2 Methodology

3.2.1 Characteristics of $^{90}\text{Sr}/^{90}\text{Y}$ and Interfering Emitters.

^{90}Sr has a half-life of 29.1 year and is a pure beta-particle emitter with a maximum beta energy of 546 keV. It is the daughter product of the fission product ^{90}Kr (fission yield of approximately 5 %). The complete decay scheme of ^{90}Kr is



Within a couple of weeks ^{90}Sr is in secular equilibrium with ^{90}Y . ^{90}Y is also a pure beta emitter, having a maximum energy of 2.28 MeV. A few months of reactor operation is sufficient to achieve equilibrium of the ^{90}Sr and ^{90}Y in a few percent. At the Chernobyl reactor facility 36.5 kg of ^{90}Sr and 9.37 g of ^{90}Y were in the core at the time of the accident. Roughly 4 % of the ^{90}Sr was released according to the Soviet evaluations [17].

Through the complicated model of atmospheric transport and ground deposition both in Soviet and in other countries [17], it can be assumed that ^{90}Sr and ^{90}Y followed the same distribution pattern, and consequently, the ratio of radioisotope activities stays approximately the same. Also it can be assumed that strontium and yttrium should not be significantly separated.

Post accident environmental samples will show not only $^{90}\text{Sr}/^{90}\text{Y}$ but also fission products, cosmogenic β emitters or elements belonging to the natural ^{238}U , and ^{232}Th decay schemes. Table 3.1 shows radioisotopes which decay by β emission (yield ≥ 20 %). They are broken down into three categories fission products with fission yields ≥ 0.05 %, cosmogenic β

emitters and the primordial β emitters. This list of possible radioisotopes can be reduced by determining which isotopes would not be present at the time of sample analysis. The criteria for elimination of radioisotopes includes [15];

- 1) Short-lived radionuclides will have decayed away if their parents are also short-lived ($T_{1/2}$ = no more than a few hours).
 - 2) Exclude all emitters with energy lower than 1.5 MeV.
- and
- 3) Only natural background and Chernobyl inventory radioisotopes have been considered as a typical accident condition.

Using this approach, only a few interfering beta emitters remain: ^{144}Pr and ^{106}Rh are short-lived ($T_{1/2} < 1$ h), and their parents (^{144}Ce and ^{106}Ru) are much longer-lived ($T_{1/2} \sim 10^4$ h). Therefore they may be assumed to be in secular equilibrium when released. Similarly, for $^{234\text{m}}\text{Pa}$ and ^{210}Tl that equilibrate with ^{238}U (4.5×10^9 y) and ^{226}Ra (1620 y). ^{210}Tl is a negligible presence because of its low formation rate. ^{212}Bi also belongs to a natural family (^{232}Th). ^{32}P is a cosmogenic radionuclide having an environmental concentrations too low to be meaningful for consideration in this work. It can be inferred that only two β emitters having energies over 3 MeV can interfere with ^{90}Y : ^{106}Rh deriving from ^{106}Ru and ^{144}Pr deriving from ^{144}Ce .

When considering there exist β - γ interference, a similar procedure is for the fired field sources. A listing of possible radionuclides is given in Table 3.2. To obtain a list of possible radionuclides, the same method as above is applied. The possible interfering isotopes present in

the Chernobyl inventory or in the natural background are : ^{106}Rh , ^{132}I , ^{140}La , ^{208}Tl , ^{212}Bi , ^{214}Bi , and ^{228}Ac . Only ^{106}Rh has a higher energy than 2.2 MeV, and the last four elements belong to the natural series besides being produced by Pu isotopes. Hence the possible radioisotopes are listed in Table 3.3 for different environmental matrices which strongly depend on geographical parameters. The transuranic radionuclides are much lower concentration (several orders of magnitude lower than isotopes in Table 3.3) [17]. It can be assumed that the most possible radioisotopes among the β - γ interfering emitters are three: ^{106}Rh , ^{140}La and ^{132}I .

In conclusion, the possible interfering emitters having beta energies greater than 1.5 MeV are:

- Two β emitters: ^{106}Rh (78.8 %, $\epsilon_{\beta \text{ max}}=3.53$ MeV) and ^{144}Pr (97.7 %, $\epsilon_{\beta \text{ max}}=2.98$ MeV)
- Three β - γ emitters: ^{106}Rh (21.2 %), ^{140}La and ^{132}I
- A natural background due to two β emitters ($^{234\text{m}}\text{Pa}$; ^{212}Bi , 48%) and four β - γ emitters (^{208}Tl ; ^{212}Bi , 15.6 %; ^{214}Bi ; ^{228}Ac).

Table 3.1 Beta Particle Emitting Radioisotopes. Fission Products with Yield ≥ 0.05 %, Cosmogenic, and Primordial Elements.

	Isotope	$T_{1/2}$	ϵ_{\max} (MeV)	β percent
1	Pd107	6.5×10^6 y	0.04	100
2	Sm151	90 y	0.076	99.1
3	Zr93	1.5×10^6 y	0.09	100
4	Se79	6.5×10^4 y	0.15	100
5	Cs135	2.3×10^6 y	0.21	100
6	Rb87	4.8×10^4 y	0.282	100
7	Rh105	35.4 h	0.566	75
8	Ce141q	32.5 d	0.581	30
9	Kr85	10.73 y	0.687	99.6
10	Tel27(Sb127-93h) ^a	9.4 h	0.69	98.8
11	Sm153	1.93 d	0.69	21
12	Pr143	13.57 d	0.933	100
13	Br83	2.4 h	0.97	98.6
14	Pm149	2.21 d	1.072	96.6
15	Mol102	11.3 m	1.2	-
16	Se81	18.3 m	1.59	100
17	Tel129m	34.1 d	1.6	31
18	Ce145	3.0 m	1.7	-
19	Pr145	5.98 h	1.80	98
20	Zr97	16.8 h	1.92	86
21	Ba139	1.40 h	2.27	72
22	As79	9.0 m	2.3	100
23	La141	3.90 h	2.43	97
24	Sr91	9.5 h	2.7	30
25	Br85	2.87 m	2.87	100
26	Y93	10.2 h	2.88	90
27	Ru107	3.8 m	3.2	-
28	La143	14.1 m	3.3	93.7
29	Nb99	15.0 s	3.5	-
30	Y92	3.54 h	3.64	85.7
31	Kr87	1.27 h	3.9	30
32	Xe137	3.82 m	4.1	-
33	Tc102	5.3 s	4.2	-
34	Cs139	9.3 m	4.2	-
35	Br84	31.8 m	4.65	31.5
36	Y94	18.7 m	4.92	22.5
37	Kr89	3.15 m	4.93	-
38	Rb88(Kr88-2.8h) ^a	17.7 m	5.31	77.8
39	Rb91	58.0 s	5.85	-
40	Rb90	2.6 m	6.6	-
41	H3	12.3 y	0.019	100
42	Ru106	1.02 y	0.039	100

Table 3.1 (Continued)

Isotope	$T_{1/2}$	ϵ_{\max}	β percent
43 Pm147	2.62 y	0.224	100
44 Tc99	2.13×10^5 y	0.292	100
45 Ce144	284.6 d	0.318	76
46 Cs137	30.17 y	0.514	100
47 Sr90	29.1 y	0.546	100
48 Mo99	2.75 y	1.21	82
49 Sr89	50.52 d	1.49	100
50 Y91	58.5 d	1.545	99.7
51 Y90	2.67 d	2.281	100
52 Pr144(Ce144- 6.8×10^3 h) ^a	17.28 m	2.996	97.7
53 Rh106(Ru106- 8.8×10^3 h) ^a	30 s	3.54	78.8
54 Cl14	5730 y	0.157	100
55 Si32	1.0×10^2	0.221	100
56 P33	25.3 d	0.249	100
57 Be10	1.6×10^6 y	0.556	100
58 Ar39	269 y	0.565	100
59 Cl36	3×10^5 y	0.709	100
60 K40	1.28×10^9 y	1.33	89.3
61 Si31	2.62 h	1.48	100
62 S35	87.2 d	1.67	100
63 P32(Si32- 3.9×10^6 h) ^a	14.28 d	1.71	100
64 Th234(U238- 3.9×10^{13} h) ^a	24.1 d	0.198	72.5
65 Ac227(Pa231- 3×10^8 h) ^a	21.77 y	0.045	54.2
66 Pb210	22.3 y	0.061	20
67 Bi210(Pb210- 1.95×10^5 h) ^a	5.01 d	1.16	100
68 Pb211(Ra223-281h) ^a	36.1 m	1.38	93
69 Bi213(Ac225-240h) ^a	45.6 m	1.42	65
70 Tl207(Ra223-281h) ^a	4.77 m	1.44	100
71 Tl206(Bi210-120h) ^a	4.2 m	1.53	100
72 Pa234m(U238- 3.9×10^{13} h) ^a	1.17 m	2.29	98.3
73 Tl210(Rn222-92h) ^a	1.30 m	2.3	100
74 Bi212(Pb212-10.6h) ^a	1.0 h	2.25	48.4
75 Pu241	14.4 y	0.0208	100
76 Ra225(Th229- 6.4×10^7 h) ^a	14.9 d	0.32	32.7
77 Pb209(Ac225-240h) ^a	3.25 h	0.645	100

^a The parent element is given within parenthesis when longer lived than daughter and half-life greater than a few hours.

Note: Radionuclides 1 to 40 are fission products.
 Radionuclides 41 to 53 are Chernobyl fission products.
 Radionuclides 54 to 63 are cosmogenic isotopes
 Radionuclide 64 is primordial isotope.
 Radionuclides 65 to 74 are primordial isotopes present in Chernobyl fall-out.
 Radionuclides 75 to 78 are ²⁴¹Pu beta emitters daughters.

Table 3.2 Mixed Beta Particle and Gamma Ray Emitting Radioisotopes.
Fission Products with Yield ≥ 0.05 %, Cosmogenic, and
Primordial Elements.

Isotope	$T_{1/2}$	ϵ_{\max}	β - γ percent
1 I129	1.57×10^7 y	0.15	100
2 Ce144	284.6 d	0.318	25
3 Sr92	2.71 h	0.54	96.4
4 Rh105	35.4 h	0.566	25
5 Sb129	4.40 h	0.65	90
6 Sm153	1.929 d	0.69	79
7 Pm151	1.183 d	0.84	89
8 Xe135	9.1 h	0.91	100
9 Sb127	3.84 d	1.10	100
10 Ru105	4.44 h	1.187	100
11 Nd151	12.4 m	1.2	100
12 I133	20.8 h	1.24	100
13 Nb97(Zr97-17h) ^a	1.23 h	1.27	100
14 I135	6.57 h	1.3	100
15 Ce143	1.38 d	1.40	100
16 Nd149	1.72 h	1.42	100
17 Te129	1.16 h	1.45	100
18 Te129m	34.1 d	1.6	69
19 Se83	22.3 m	1.8	100
20 Ag113	5.3 h	2.01	100
21 Te131(Te131m-30h) ^a	25.0 m	2.1	100
22 Ba139	1.396 h	2.27	27.4
23 Te131m	30 h	2.46	74
24 Sr91	9.5 h	2.7	70
25 Kr88	2.84 h	2.9	86
26 Kr87	1.27 h	3.9	70
27 La142	1.54 h	4.52	93
28 Br84	31.8 m	4.65	68.5
29 Y94	18.7 m	4.92	77.5
30 Rb88(Kr88-2.8h) ^a	17.7 m	5.31	22.2
31 Nb95(Zr95-1560h) ^a	34.97 d	0.16	100
32 Te132	3.26 d	0.215	100
33 Ru103	39.27 d	0.223	93.6
34 Cs136	13.16 d	0.341	100
35 Xe133	5.243 d	0.348	100
36 Zr95	64.02 d	0.4	100
37 Ce141	32.5 d	0.581	70
38 I131	8.04 d	0.606	100
39 Na147	10.98 d	0.805	100
40 Kr85m	4.48 h	0.839	100
41 Ba140	12.75 d	1.02	100
42 La140(Ba140-307h) ^a	1.678 d	1.67	100

Table 3.2 (Continued)

Isotope	$T_{1/2}$	ϵ_{\max}	β - γ percent
43 I132(Te132-77h) ^a	2.28 h	2.16	100
44 Ra228(Th232-1.2x10 ¹⁴ h) ^a	5.76 y	0.039	100
45 Ac227(Pa231-3x10 ⁸ h) ^a	21.77 y	0.045	44.4
46 Pb210	22.3 y	0.061	80
47 Th234(U238-3.9x10 ¹³ h) ^a	24.1 d	0.198	27.5
48 Pa233(Np237-1.9x10 ¹⁰ h) ^a	27.0 d	0.256	100
49 Th231(U235-6.2x10 ¹² h) ^a	1.06 d	0.305	100
50 Ra225(Th229-6.4x10 ⁷ h) ^a	14.9 d	0.32	67.3
51 Pb212(Ra224-88h) ^a	10.64 h	0.569	90
52 Pb214(Rn222-92h) ^a	27 m	0.73	100
53 Fr223(Ac227-1.9x10 ⁵ h) ^a	21.8 m	1.15	100
54 Bi213(Ac225-240h) ^a	45.6 m	1.42	35
55 Bi214(Rn222-92h) ^a	19.9 m	1.54	82.3
56 Tl208(Pb212-10.6h) ^a	3.05 m	1.80	100
57 Ac228(Ra228-50370h) ^a	6.15 h	2.1	100
58 Np239	2.355 d	0.438	100
59 Cs134	2.065 y	0.658	100
60 Tl209(Ac225-240h) ^a	2.2 m	1.8	100
61 Bi212(Pb212-10.6h) ^a	1.0 h	2.25	15.6
62 Rh106(Ru106-8.8x10 ³ h) ^a	30 s	3.54	21.2

^a The parent element is given within parenthesis when longer lived than daughter and half-life greater than a few hours.

Note: Radionuclides 1 to 30 are fission products.

Radionuclides 31 to 43 are Chernobyl fission products.

Radionuclides 44 to 57 are primordial isotopes present in Chernobyl fall-out.

Radionuclides 58 to 62 are ²⁴¹Pu beta emitters daughters.

Table 3.3 The Concentrations of Nuclides in Different Environmental Matrices [17].

Isotope	North Italy Rain On May 2, 1986	Air Close to Chernobyl at 3 m altitude on May 20, 1986	Soil Sample within 30 km from Accident on May 17, 1986
Ru-106	~ 7 Bq/kg	0.1 - 2 Bq/l	192 Bq/g
La-140	26 Bq/kg	0.6 - 1.7 Bq/l	1.8×10^3 Bq/g
I-132	170 Bq/kg	1.3 Bq/l	3.4×10^2 Bq/g

3.2.2 Theoretical Study

Two kinds of detectors are needed to measure the beta-activity in mixed beta/gamma-ray radiation fields. The first is an electron detector to provide a gate signal and to ignore input from all radiation types. The second is a plastic beta spectrometer. Beta particles and gamma-rays are successfully separated through this detector system. Analysis of spectra is simplified using the method of Brini [15], the energy range of spectra is divided into two regions: above an energy threshold ϵ_0 (≈ 2.2 MeV) and the range $\Delta\epsilon$ (≈ 0.7 MeV) below ϵ_0 ($\epsilon_0 - \Delta\epsilon < \epsilon \leq \epsilon_0$).

Let A_1 be the β -emitter activity with maximum energy less than or equal to the threshold energy, ϵ_0 . Similarly A_2 will indicate the activity of a β -emitter in the energy range greater than ϵ_0 . A_3 will indicate the activity of β - γ emitter with the maximum beta energy $\epsilon \leq \epsilon_0$ and finally, A_4 is the activity of β - γ emitter where the beta energy is $\epsilon > \epsilon_0$.

The counting rates of β and γ -ray in the $\Delta\epsilon$ window and the integral counting rates for energies greater than ϵ_0 will be:

Beta particle count rates

$$\Delta\nu_1 = \nu_1 - \nu_{o1} \quad (\epsilon_0 - \Delta\epsilon < \epsilon \leq \epsilon_0) \quad (3.1)$$

$$= K_{11}A_1 + K_{21}A_2 + K_{31}A_3 + K_{41}A_4,$$

$$\Delta\nu_2 = \nu_2 - \nu_{o2} \quad (\epsilon > \epsilon_0) \quad (3.2)$$

$$= K_{22}A_2 + K_{42}A_4,$$

Gamma ray count rates

$$\begin{aligned}\Delta\nu_3 &= \nu_3 - \nu_{o3} && (\epsilon_o - \Delta\epsilon < \epsilon \leq \epsilon_o) \\ &= K_{33}A_3 + K_{43}A_4,\end{aligned}\tag{3.3}$$

$$\begin{aligned}\Delta\nu_4 &= \nu_4 - \nu_{o4} && (\epsilon > \epsilon_o) \\ &= K_{44}A_4.\end{aligned}\tag{4}$$

Where

ν_{oi} are the background rates (instrumental + environmental).

K_{11} is the instrumental parameter of A_1 at energy $\epsilon_o - \Delta\epsilon < \epsilon \leq \epsilon_o$ which includes the intrinsic efficiency of detector system and geometric factor at this energy range.

K_{21} is the instrumental parameter of A_2 at energy $\epsilon_o - \Delta\epsilon < \epsilon \leq \epsilon_o$.

K_{31} is the instrumental parameter of the β activity of A_3 at energy $\epsilon_o - \Delta\epsilon < \epsilon \leq \epsilon_o$.

K_{41} is the instrumental parameter of the β activity of A_4 at energy $\epsilon_o - \Delta\epsilon < \epsilon \leq \epsilon_o$.

K_{22} is the instrumental parameter of A_2 at energy $\epsilon > \epsilon_o$.

K_{42} is the instrumental parameter of the β activity of A_4 at energy $\epsilon > \epsilon_o$.

K_{33} is the instrumental parameter of the γ activity of A_3 at energy $\epsilon_o - \Delta\epsilon \leq \epsilon_o$.

K_{43} is the instrumental parameter of the γ activity of A_4 at

energy $\epsilon_0 - \Delta\epsilon < \epsilon \leq \epsilon_0$.

K_{44} is the instrumental parameter of the γ activity of A_4 at energy $\epsilon > \epsilon_0$.

From equations (3.1), (3.2), (3.3) and (3.4), the following activities can be obtained:

$$A_4 = \frac{1}{K_{44}} \Delta v_4 \quad (3.5)$$

$$A_3 = \frac{1}{K_{33}} \left(\Delta v_3 - \frac{K_{43}}{K_{44}} \Delta v_4 \right) \quad (3.6)$$

$$A_2 = \frac{1}{K_{22}} \left(\Delta v_2 - \frac{K_{42}}{K_{44}} \Delta v_4 \right) \quad (3.7)$$

$$A_1 = \frac{1}{K_{11}} \left[\Delta v_1 - \frac{K_{21}}{K_{22}} \left(\Delta v_2 - \frac{K_{42}}{K_{44}} \Delta v_4 \right) - \frac{K_{31}}{K_{33}} \left(\Delta v_3 - \frac{K_{43}}{K_{44}} \Delta v_4 \right) - \frac{K_{41}}{K_{44}} \Delta v_4 \right] \quad (3.8)$$

Equation (3.8) means: if the nine values of the K parameters are initially determined with known sources, only two measurements need to be made in β and γ counts in two regions.

Thus, if A_1 is the assessed activity of an environmental sample with mass or volume M , then the corresponding concentration A_1^* is:

$$A_1^* = A_1/M \quad (3.9)$$

where M is measured in convenient units.

3.2.3 Sensitivity Measurements

The minimum detectable level of activity A_1 depends on many factors. The primary one is the acceptable maximum error of the measurement result. For this work it shall be:

$$A_1 > 3\sigma_1$$

where σ_1 is the measurement standard deviation.

The minimum detectable counting rate of the spectrometer system is calculated by assuming that T and T_o are the count times of $\nu_1, \nu_2, \nu_3, \nu_4$ and $\nu_{o1}, \nu_{o2}, \nu_{o3}, \nu_{o4}$, respectively. The standard error of counting rates is obtained:

$$\begin{aligned} \sigma_1^2 = & \frac{1}{K_{11}^2} \left[\frac{\nu_1}{T} + \frac{\nu_{o1}}{T_o} + \left(\frac{K_{21}}{K_{22}} \right)^2 \left(\frac{\nu_2}{T} + \frac{\nu_{o2}}{T_o} \right) \right. \\ & + \left(\frac{K_{21}}{K_{22}} \right)^2 \left(\frac{K_{42}}{K_{44}} \right)^2 \left(\frac{\nu_4}{T} + \frac{\nu_{o4}}{T_o} \right) \\ & + \left(\frac{K_{31}}{K_{33}} \right)^2 \left(\frac{\nu_3}{T} + \frac{\nu_{o3}}{T_o} \right) \\ & \left. + \left(\frac{K_{31}}{K_{33}} \right)^2 \left(\frac{K_{43}}{K_{44}} \right)^2 \left(\frac{\nu_4}{T} + \frac{\nu_{o4}}{T_o} \right) \right] \end{aligned}$$

$$+ \left(\frac{K_{41}}{K_{44}} \right)^2 \left(\frac{\mathbf{v}_4}{T} + \frac{\mathbf{v}_{o4}}{T_o} \right)] \quad (3.10)$$

$$= \frac{1}{K_{11}^2} \left[\frac{\mathbf{v}_1}{T} + \frac{\mathbf{v}_{o1}}{T_o} + G_1 \left(\frac{\mathbf{v}_2}{T} + \frac{\mathbf{v}_{o2}}{T_o} \right) \right. \\ \left. + G_2 \left(\frac{\mathbf{v}_3}{T} + \frac{\mathbf{v}_{o3}}{T_o} \right) + G_3 \left(\frac{\mathbf{v}_4}{T} + \frac{\mathbf{v}_{o4}}{T_o} \right) \right] \quad (3.11)$$

where

$$G_1 = \left(\frac{K_{21}}{K_{22}} \right)^2$$

$$G_2 = \left(\frac{K_{31}}{K_{33}} \right)^2$$

and

$$G_3 = \left(\frac{K_{21}}{K_{22}} \right)^2 \left(\frac{K_{42}}{K_{44}} \right)^2 + \left(\frac{K_{31}}{K_{33}} \right)^2 \left(\frac{K_{43}}{K_{44}} \right)^2 + \left(\frac{K_{41}}{K_{44}} \right)^2.$$

In terms of activity A, it can be written

$$\sigma_1^2 = \frac{1}{TK_{11}^2} \left[\mathbf{v}_1 + \frac{T}{T_o} \mathbf{v}_{o1} + G_1 \left(\mathbf{v}_2 + \frac{T}{T_o} \mathbf{v}_{o2} \right) \right. \\ \left. + G_2 \left(\mathbf{v}_3 + \frac{T}{T_o} \mathbf{v}_{o3} \right) + G_3 \left(\mathbf{v}_4 + \frac{T_o}{T} \mathbf{v}_{o4} \right) \right] \quad (3.12)$$

$$\sigma_1^2 = \frac{1}{TK_{11}^2} [\Delta v_1 + G_1 \Delta v_2 + G_2 \Delta v_3 + G_3 \Delta v_4 + \frac{T+T_o}{T} (v_{o1} + G_1 v_{o2} + G_2 v_{o3} + G_3 v_{o4})] \quad (3.13)$$

then

$$\sigma_1^2 = \frac{1}{TK_{11}^2} [K_{11}A_1 + (K_{21} + G_1K_{22})A_2 + (K_{31} + G_2K_{33})A_3 + (K_{41} + G_1K_{42} + G_2K_{43} + G_3K_{44})A_4 + bv_o] \quad (3.14)$$

where

$$b = \frac{T + T_o}{T}$$

$$v_o = v_{o1} + G_1 v_{o2} + G_2 v_{o3} + G_3 v_{o4} .$$

When $T = T_o$, the following relation in terms of concentrations is given [18]:

$$A_1 > \frac{9}{2TK_{11}} \left\{ 1 + \sqrt{1 + \frac{4}{9} T(MW + 2\nu_o)} \right\} \quad (3.15)$$

Or in terms of specific concentration

$$A_1^* > \frac{9}{2TK_{11}M} \left\{ 1 + \sqrt{1 + \frac{4}{9} T(MW^* + 2\nu_o)} \right\} \quad (3.16)$$

where

$$W^* = (K_{21} + G_1K_{22})A_2^* + (K_{31} + G_2K_{33})A_3^* + (K_{41} + G_1K_{42} + G_2K_{43} + G_3K_{44})A_4^*$$

From the recent concentration data of the Chernobyl accident [17], it roughly can be assessed that $A_2^* \approx A_3^* \approx 10A_4^*$. In most cases, the last term under the square root, $\frac{4}{9} T(MW^* + 2\nu_o)$, is much greater than 1. Then equation (3.16) can be simplified:

$$A_1^* \geq \frac{3}{K_{11}} \sqrt{\frac{1}{TM} \left(W^* + \frac{2\nu_o}{M} \right)} \quad (3.17)$$

Equation (3.17) shows that the minimum detectable activity concentration depends on four factors:

1. The counting time T , which is limited by the circumstance,
2. The sample mass or volume M , in convenient units,
3. The background term ν_o , which can be reduced either with appropriate shielding or through the specific improvement of the detector system, and
4. The interfering isotopes term W^* .

3.3 Experimental Equipment

Two detection systems, a gas-flow proportional counter and a plastic scintillator, were combined to form the beta-particle spectrometer. The gas-flow proportional counter was designed to be insensitive to gamma-ray interaction. The gas counter and scintillator were connected to a gating unit which routed the analog output of the spectrometer to a multichannel analyzer (MCA) only when the gas counter sensed the passage of a beta particle.

3.3.1 Gas-Flow Proportional Counter [19]

Because of its fast response and charge amplification capabilities, the gas-flow proportional counter was selected for use as the gating detector instead of an ionization chamber or Geiger-Müller counter.

The gas counter was a 10 mm high aluminum cylinder with a cross-sectional area sufficient to cover the window area of the plastic scintillator, $\approx 2000 \text{ mm}^2$. An aluminum guide tube was attached to the gas counter to provide a means of positioning the scintillator and photomultiplier tube adjacent to the gas counter exit window. This guide tube also provide a means of shielding the scintillator from off axis beta particles. The counter, which was fabricated by LND Inc. (13230 Lawson Boulevard, Oceanside, Long Island, NY) with model number of 49561, is shown in Fig. 3.1. A complete list of specifications is given in Table 3.4.

A beta plateau was measured to determine its operating voltage. This measurement was made using a ^{99}Tc beta particle source with P-10 fill gas (10 % methane, 90 % argon). From this data, presented in Fig. 3.2, the operating voltage of gas counter was determined to be + 1800 V.

Table 3.4 Specifications of the Gas-Flow Proportional Counter[20].

<u>General Specifications</u>	
Operating Temp. Range	0 to + 50° C
Path Length	10.0 mm
Anode Material	Gold Plated Tungsten
Diameter	0.050 mm
Cathode Material	Aluminum
Maximum Length	118.1 mm
Effective Length	101.6 mm
Maximum Diameter	120.7 mm
Effective Diameter	101.6 mm
Connector	MHV
<u>Window Specifications</u>	
Areal Density	0.8 mg/cm ²
Material	Aluminized Mylar
Diameter	101.6 mm
<u>Electrical Specifications</u>	
Operating Voltage	1200 - 2100 volts
Capacitance	14 pf

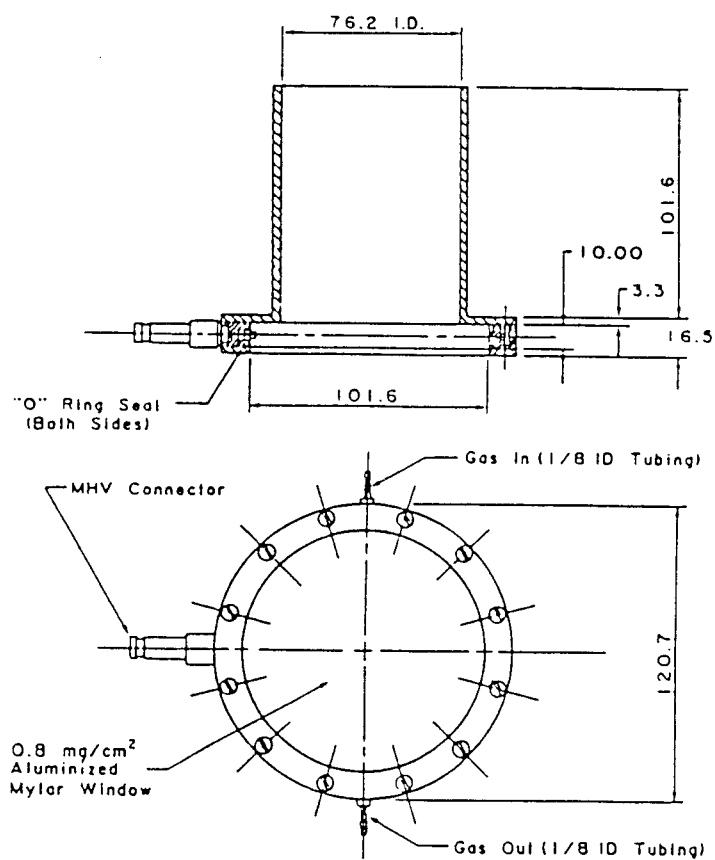


Fig. 3.1 Side and end views of gas-flow proportional counter[20]
(dimensions in millimeter).

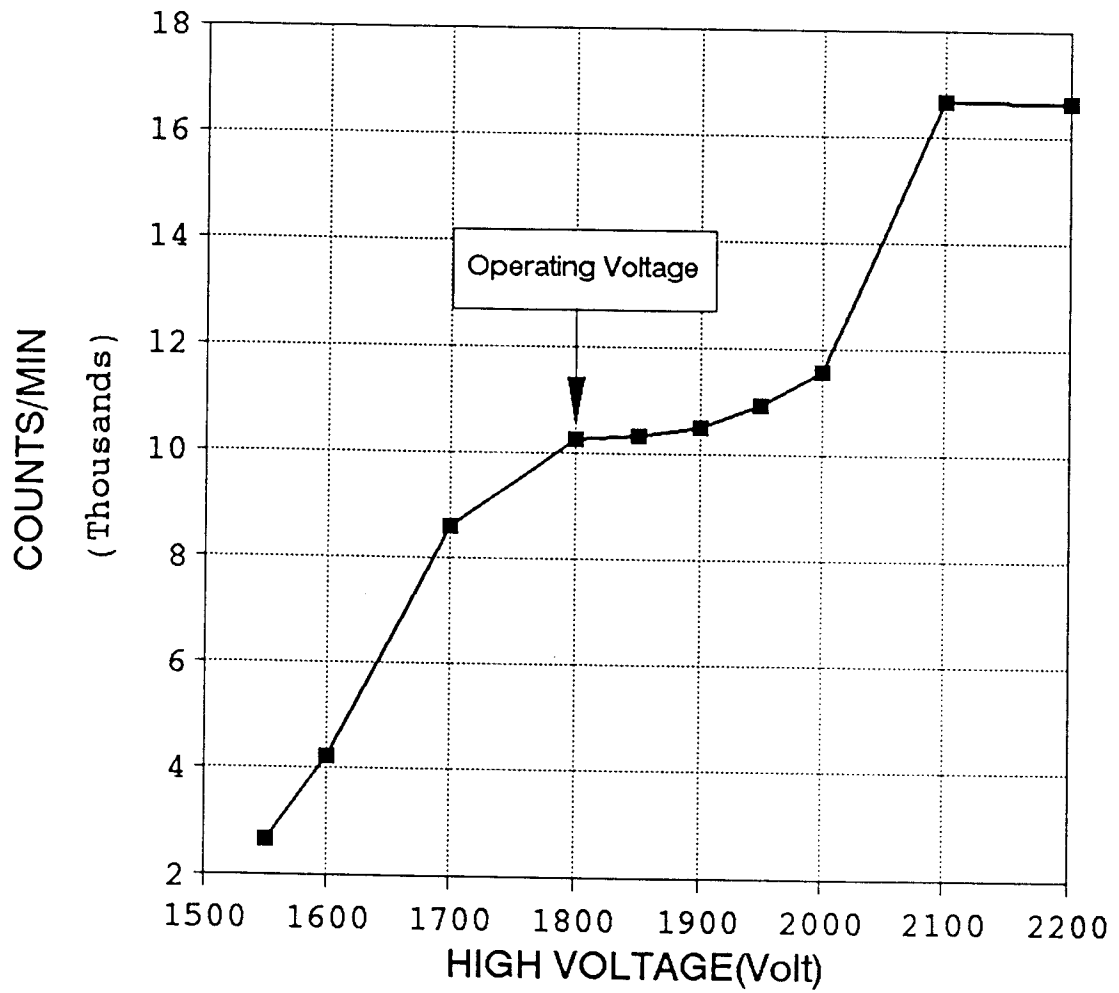


Fig. 3.2 Beta plateau measurement for gas-flow proportional counter using ^{99}Tc with P-10 gas.

3.3.2 Plastic Scintillator Based Beta-Particle Spectrometer [19]

Because the pulse height distributions measured with a spectrometer system based on a BC-400 plastic scintillator were more representative of theoretical expectations than those measured with systems based a solid state detector [19], a BC-400 plastic scintillator (Bicron Corp., 12345 Kinsman Road, Newbury, OH) was selected in this work. The physical characteristics of BC-400 plastic scintillator are outlined in Table 3.5. The scintillator used in this work was a polished cylinder with diameter of 50.8 mm and thickness of 25.4 mm.

The BC-400 scintillator was coupled to the PMT window using optical grease. A scintillator entrance window consisting of a light tight 0.96 mg/cm² aluminized Mylar sheet was glued to a graphite impregnated Nylon ring. The ring acted as a support for the Mylar to prevent flexing or creasing that could result in light leaks. Once the Mylar side of the entrance window ring was laid on top of the scintillator, a pre-cut sheet of light tight aluminized Mylar, with a thickness of 0.96 mg/cm², was wrapped around the sides of the scintillator to prevent light loss through this surface. This radial reflector extended from the top of the entrance window support ring to several millimeters beyond the PMT surface. The Mylar was held in place with a short strip of black vinyl tape along the overlapping seam of the radial reflector. A light cap of graphite-impregnated Nylon was lowered over the scintillator and attached to the PMT using black vinyl tape. The purpose of the light cap was to shield the scintillator from outside light sources. The location of components is shown in Fig. 3.3.

Table 3.5 Physical Characteristics of BC-400 Plastic Scintillator[20]

Description	Value
Density	1.032 g/cm ³
Refractive Index	1.581
Melting Point	75 °C
Light Output	65 % of Anthracene
Decay Constant	2.4 ns
Wavelength of Max. Emission	423 nm
H/C Atomic Ratio	1.104
Coefficient of Linear Expansion	- 7.8 x 10 ⁻⁵ Below 67 °C

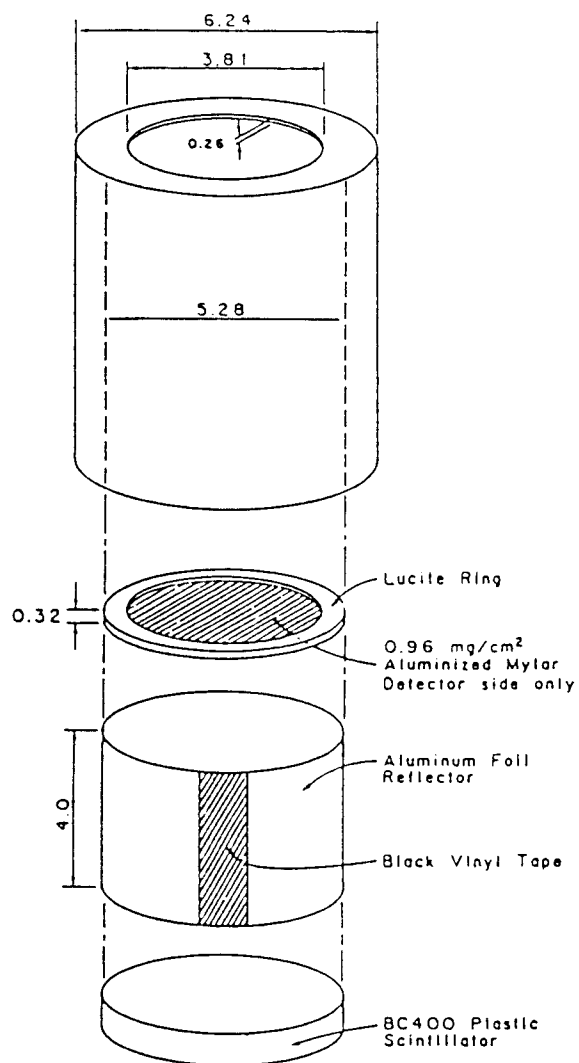


Fig. 3.3 Components of plastic scintillator assembly [20]
(dimensions in centimeters).

3.3.3 Spectrometer System

The gas detector was positioned upstream from the scintillation spectrometer using clamps and an ancillary stand. The scintillator and photomultiplier tube were positioned on the inside of gas counter guide tube to be adjacent to the gas counter exit window.

The spectrometer system consisted of two radiation detection channels: one based on a gas-flow proportional counter and the other on a BC-400 plastic scintillator. The gas-flow channel supplied timing information which was used to determine whether beta particles or gamma rays initiated the linear voltage pulses.

Pulse shaping, timing and gating were performed by commercial nuclear instrument modules which were interconnected as shown in the block diagram of Fig 3.4. To operate both detectors, each detector required a high voltage power supply, a preamplifier and a linear amplifier. The preamplifier served to match the impedance between the detectors and the linear amplifiers and the linear amplifiers performed pulse shaping and amplification. In the gas-flow counter channel, the pulses from the linear amplifier were sent to a single channel analyzer which generated a standard 5 V logic pulse provided the gate signal for the linear gate stretcher. A delay amplifier was used in the scintillator channel to ensure that the linear signal arrived at the linear gate stretcher after the gate signal.

The linear gate stretcher accepted a linear signal, qualified by gating requirements, and produced a reshaped wave form which retained the relative peak amplitude. The gating requirements established three modes of operation for this unit: NORMAL, COINCIDENCE and ANTICOINCIDENCE.

To ensure proper operation of the COINCIDENCE and ANTICOINCIDENCE mode, the leading edge of the stretched gate signal must precede the linear signal and the width of the stretched gate must be large enough to overlap the linear signal. Fig 3.5 shows how these timing requirements were applied to the spectrometer system.

In the NORMAL mode, the linear input signal was reshaped, internally gated to prevent pulse pileup, and then passed to the MCA regardless of the state of the gating signal.

For the COINCIDENCE operation mode, a reshaped linear signal was produced only when the incident linear signal was in coincidence with the supplied gate signal. The gate stretcher performed this function by using the leading edge of the stretched gate to trigger acceptance of the linear signal (see the timing diagram of Fig. 3.6). When the peak of the linear pulse was detected (or the time of effective gate period was exceeded), a busy output pulse was generated and the gate was inhibited. The incident linear pulse was continued to decay and when it dropped below the discriminator level, a reshaped, linear output pulse was produced. The busy signal continued until decay of the linear output pulse or until the discriminator had been reset. For proper operation in the COINCIDENCE mode, the gate signal supplied by the gas counter system had to arrive before the linear signal and the effective gate period had to overlay the peak of the linear input pulse.

In the ANTICOINCIDENCE mode, the linear gate stretcher reshaped the incident linear signal and passed it to the MCA unless it was inhibited by a signal supplied by the gate period generator. The gate then remained closed for the time of the gate period which was 5 μ s for this work.

Linear input signals arriving during this time interval were rejected. For the linear gate stretcher to operate properly in the ANTICOINCIDENCE mode, the gate period had to totally overlap the time that the linear signal was above the discriminator level.

The three modes of operation allowed the experimenter to select the type of particle interactions to be measured. In the COINCIDENCE mode, the signals from the gate stretcher unit were the scintillator response to beta particle interactions. For the ANTICOINCIDENCE mode, the signals from the gate stretcher unit were the result of gamma ray interactions in the scintillator. In the NORMAL mode, the spectrometer responded to both beta particle and gamma ray interactions.

Table 3.6 lists the settings of the nuclear instrument modules and other components used in this work. Fig. 3.7 shows a pulse height distribution of ^{207}Bi measured with this spectrometer system.

Table 3.6 Spectrometer Equipment and Typical Instrument Settings

Component	Gas counter system	Scintillator system	Gating system
High Voltage Power Supply	<u>Canberra 3002</u> + 1800 V	<u>Tennelec Tc904A</u> - 1500 V	
Preamplifier	<u>Ortec 142</u>	<u>Ortec 113</u> Input Cap. 0	
Amplifier	<u>Tennelec TC241</u> cg 10.0 fg 5.0 input + shaping 1.0 μ s BLR on	<u>Ortec 571</u> cg 50 cg 6.64 input + shaping 0.5 μ s BLR auto Delay in	
Shaping and Delay	<u>Canberra 2036A</u> ΔE 10.0 E 0.02 Delay 1.0 Shaping 0.5-0.2 μ s	<u>Ortec 427A</u> 1.0 μ s in	
PMT		<u>RCA 8575</u>	
PMT Base		<u>RCA AJ2207A</u> (voltage divider string)	
Gate			<u>Ortec 542</u> BLR low Gated Coinc
Multichannel Analyzer			<u>ACE III computer system</u>

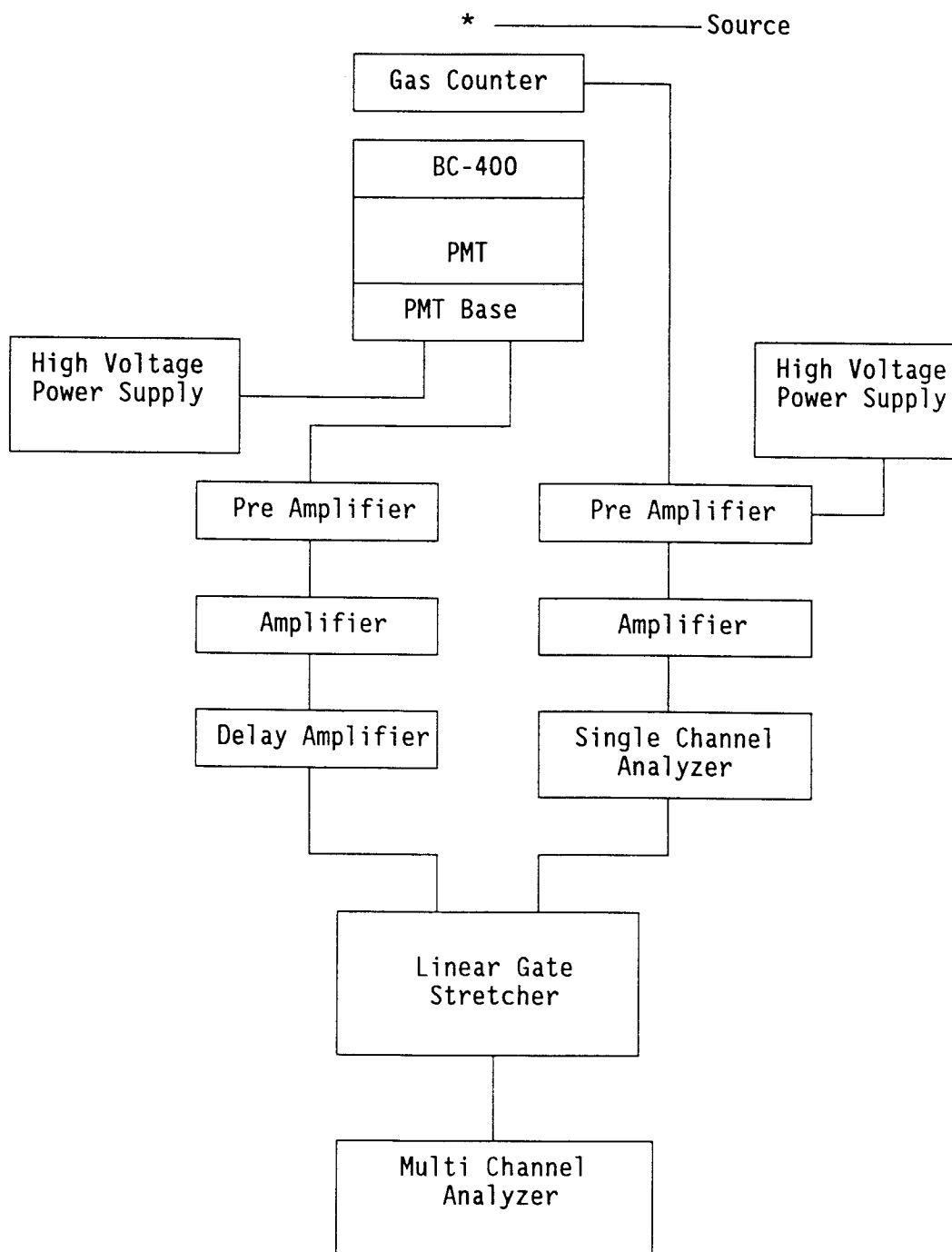


Fig. 3.4 Block diagram of the beta particle spectrometer with active gamma-ray discrimination.

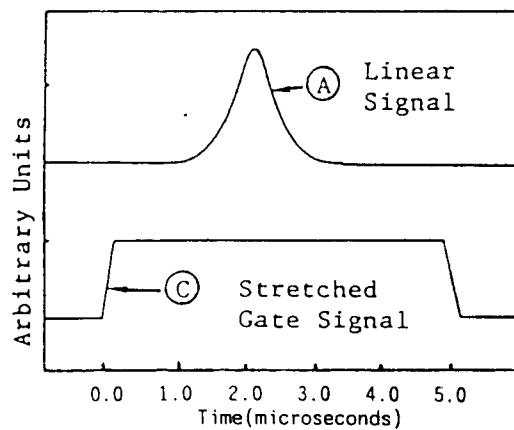
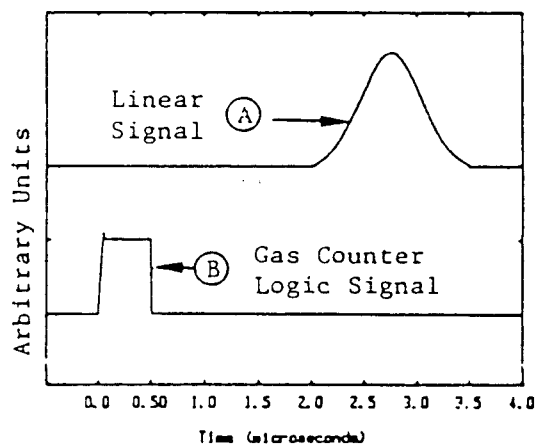
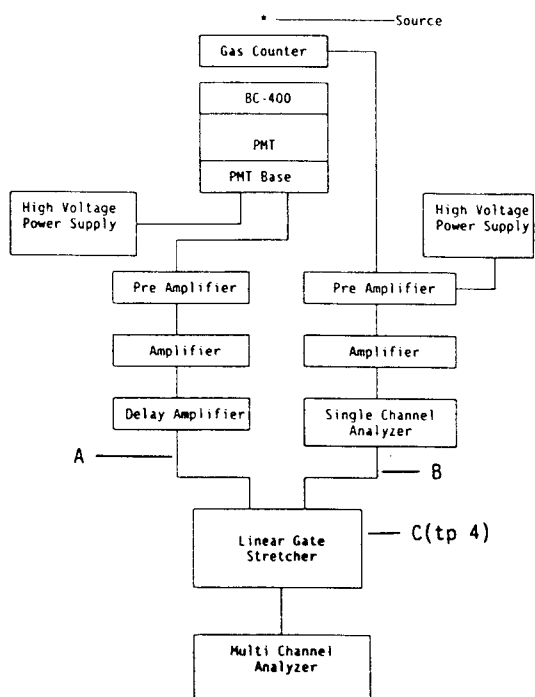


Fig. 3.5 Pulse timing characteristics of the spectrometer system.

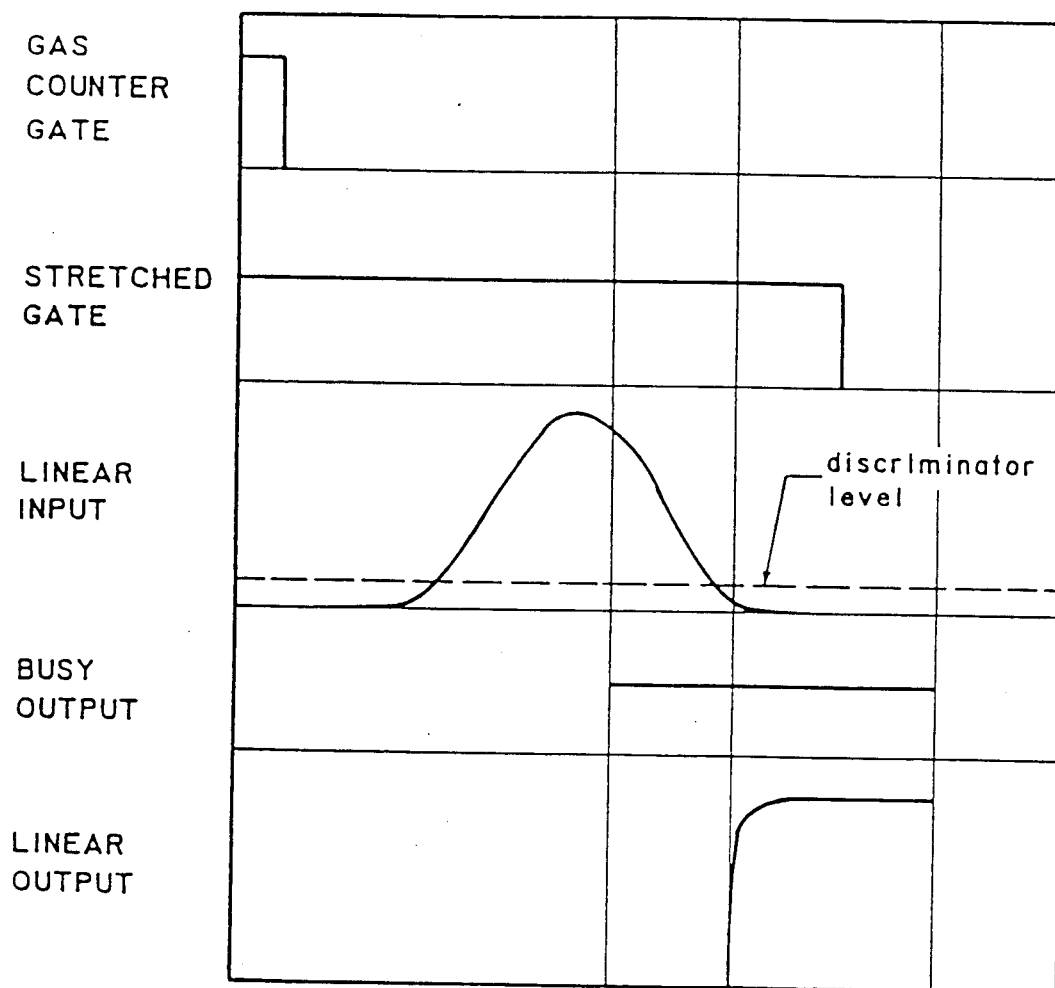


Fig. 3.6 Timing diagram for the linear gate stretcher operating in the gamma-ray discrimination mode.

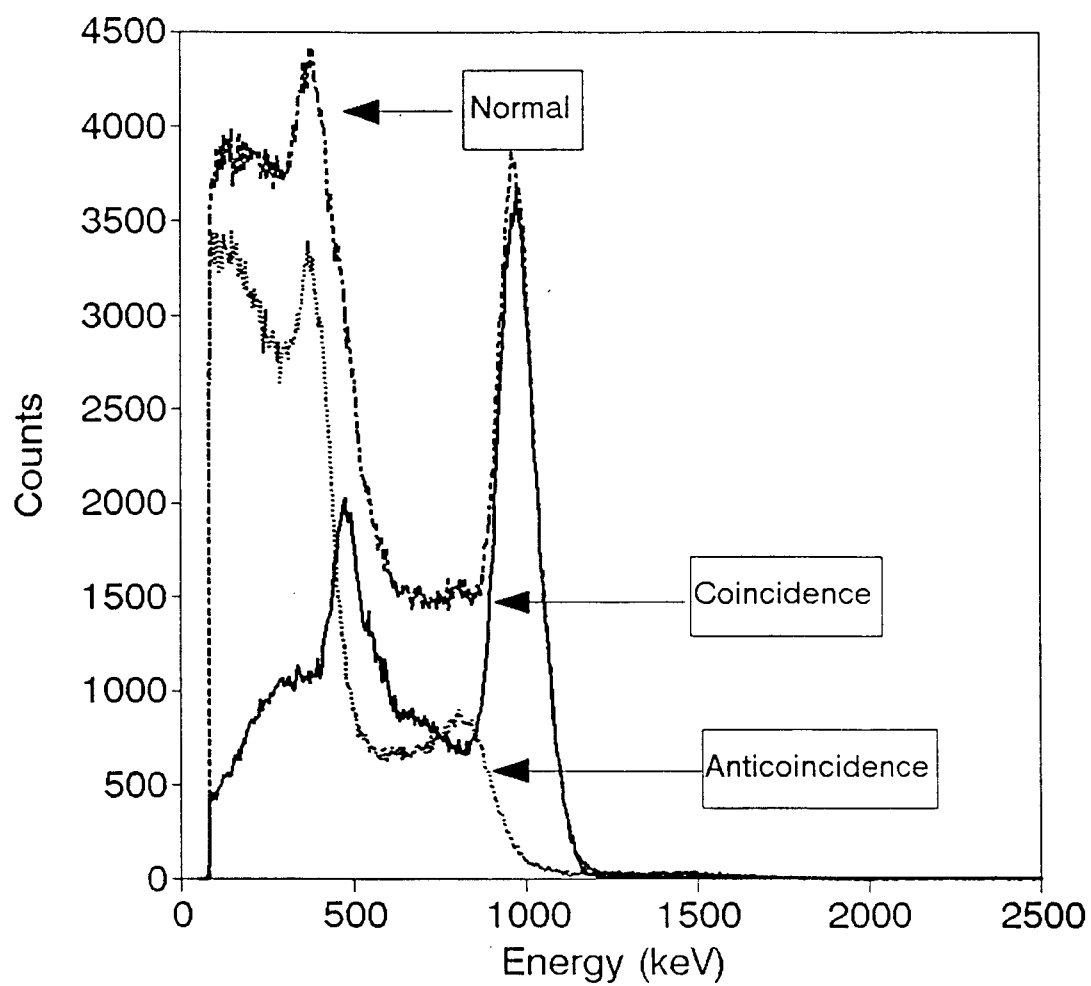


Fig. 3.7 ^{207}Bi pulse height distribution.

3.4 Experimental Procedures

3.4.1 Preparation of Radiation Sources

Several beta particle, gamma-ray and conversion electron radiation sources were used in this work. The majority of these sources were purchased from commercial suppliers. Only one source, ^{140}La , was prepared by neutron activation of reagent chemical, $\text{La}(\text{NO}_3)_3$, in the OSU TRIGA.

The sources which were used in this work were divided into two categories by their applications: the energy calibration sources and the sources used for measuring instrument parameters. The five sources listed in Table 3.7 were used in the energy calibration of the spectrometer system and the four sources in Table 3.8 were used in the determination of the instrument parameters of the spectrometer system.

The isotopes, ^{147}Pm , ^{99}Tc , ^{36}Cl and $^{90}\text{Sr}/^{90}\text{Y}$, had been purchased from Dupont NEN Products [21]. The activities of these isotopes were 0.103, 0.041, 0.02 and 0.0202 μCi , respectively. Each isotope source was encapsulated between two thin sheets of aluminized Mylar. The ^{207}Bi had been purchased from Isotope Product Laboratories [22] and was composed of 1.0 μCi of ^{207}Bi deposited on one side of an aluminized Mylar sheet.

The $^{106}\text{Ru}/^{106}\text{Rh}$ and $^{144}\text{Ce}/^{144}\text{Pr}$ standard solutions were purchased from Amersham Corporation [23]. These two isotope solution activities were 7.90 and 6.57 μCi , respectively. The $^{90}\text{Sr}/^{90}\text{Y}$ solution had been purchased for a previous research from Isotope Product Laboratories [22] with 10.03 μCi activity.

The ^{140}La was produced from neutron activation of a $\text{La}(\text{NO}_3)_3$ solution (5.0 mg La/ml) which $\text{La}(\text{NO}_3)_3$ dissolved in 0.1 N HNO_3 solution. 0.88 mg of La was activated in OSTR for 20 seconds. The activity of ^{140}La was 0.12

μCi at the end of irradiation as measured by a Ge gamma ray spectrometer.

In order to keep the counting geometry constant for the measurements of the instrument parameters a sample encapsulation was designed. This consisted of a plastic petri dish with diameter of 100 mm and a height of 15 mm into the top of which a 50 mm diameter hole was drilled (see Fig. 3.8). A circular sheet of 0.2 mg/cm^2 aluminized Mylar was centered inside this hole and attached to the plastic with "Krazy Glue". The irradiation solution or standard solution was removed from the polyethylene or glass vials and placed in the bottom of petri dishes. 50 ml of water was added to the bottom of dishes to increase the volume of material to that expected for actual environmental sampling. A silicon sealant was laid around the outside edge of the bottom of dishes. The top was pressed onto the bottom and allowed to dry to complete the encapsulation.

Table 3.7 The Energy Calibration Sources of the Spectrometer System.

Source	E_{\max} (keV)	$T_{1/2}$	Transmission(%)
^{147}Pm	224.7	2.62 y	67
^{99}Tc	292	214000 y	78
^{207}Bi	481.7, 975.6	33.4 y	
^{36}Cl	708.9	301000 y	94
$^{90}\text{Sr}/^{90}\text{Y}$	545/2245	28.5 y(64.1 h)	93(100)

Table 3.8 The Instrumental Parameter Measuring Sources of the Spectrometer System.

Source	E_{\max} of beta particle(keV)	$T_{1/2}$
A ₁ ⁹⁰ Sr/ ⁹⁰ Y	545/2245	28.5 y(64.1 h)
A ₂ ¹⁴⁴ Ce/ ¹⁴⁴ Pr	318/2996	284.6 d(17.28 m)
A ₃ ¹⁴⁰ La	2175	1.678 d
A ₄ ¹⁰⁶ Ru/ ¹⁰⁶ Rh	39/3540	1.024 y(30 s)

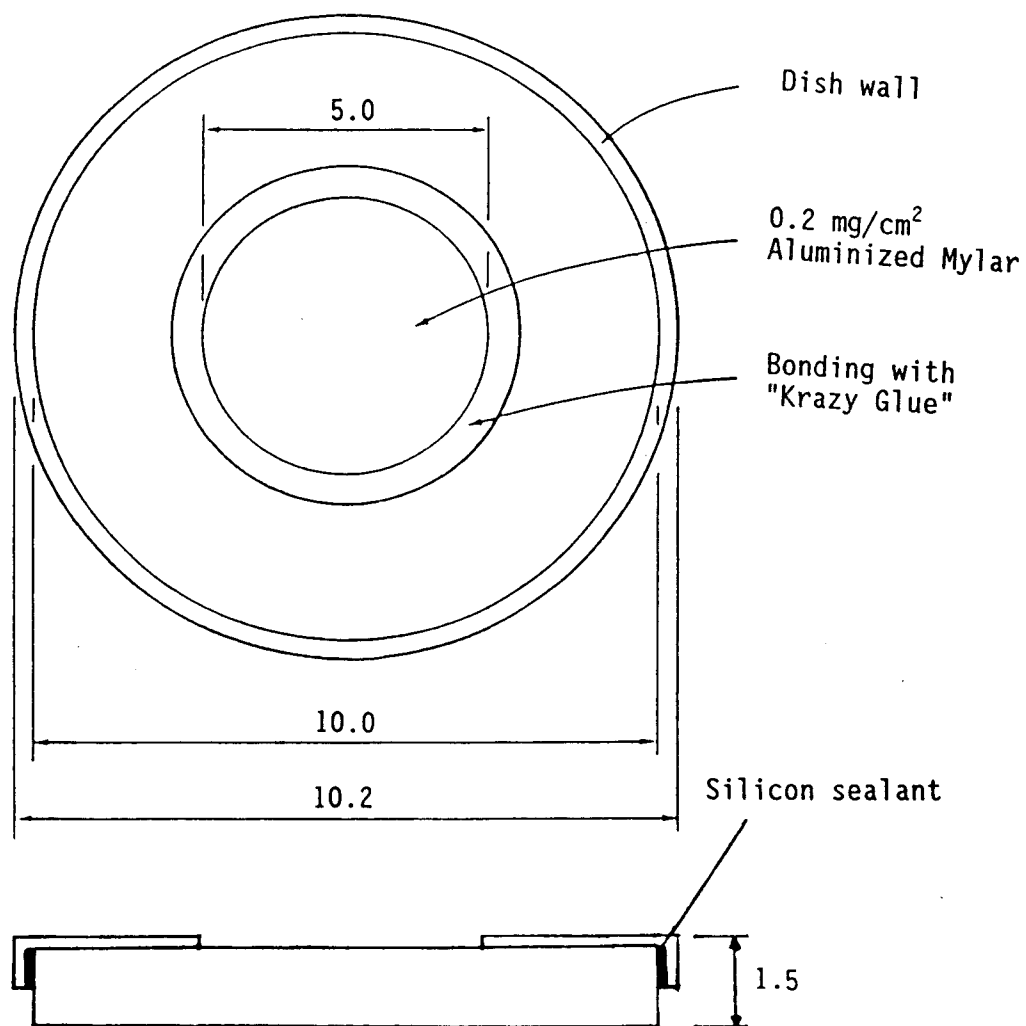


Fig. 3.8 Side and end view of the instrumental measuring source container (dimensions in centimeters).

3.4.2 Spectrometer Energy Calibration

Conversion electron and beta particle radiation sources were used to perform the energy calibration. Calibration with a conversion electron source was preferable because the monoenergetic electron yielded an easily defined peak in the measured pulse height distribution. A beta particle energy distribution is characterized by its endpoint value. The determination of the channel number corresponding to this endpoint was accomplished by approximating the usual Fermi plot function with a linearization function developed by Cramer [24].

A Gaussian function was fit to the peak in a conversion electron pulse height distribution. The centroid of this Gaussian function was the channel number corresponding to the energy of the incident electron. The fit procedure [25] began by describing the Gaussian function as

$$N(x) = N_0 \exp\left(-\frac{(x-x_0)^2}{2\sigma^2}\right) \quad (3.18)$$

where $N(x)$ is the amplitude of the Gaussian at channel x , N_0 is its maximum which occurs at channel number x_0 and σ is the function's standard deviation. By defining the quantity

$$Q(x) = \frac{N(x-1)}{N(x+1)} = \exp\left(\frac{2(x-x_0)}{\sigma^2}\right) \quad (3.19)$$

and taking logarithms

$$\ln Q(x) = \frac{2(x-x_o)}{\sigma^2} \quad (3.20)$$

A linear function of x is derived. A linear least-square fit to the data expressed in term of equation (3.20) yielded a line whose slope, m , and intercept, b , were related to the Gaussian centroid and standard deviation as

$$\sigma = \sqrt{\frac{2}{m}} \quad (3.21)$$

and

$$x_o = -\frac{b}{m} \quad (3.22)$$

The end point channel number of a beta particle spectrometer was calculated by linearizing the pulse height distribution using the expression [24].

$$Y(x) = \left(\frac{N(x)}{x^k} \right)^{1/2} \quad (3.23)$$

where $N(x)$ was the number of counts in channel number x and k is a constant equal to 1.4. A linear least-squares fit to $Y(x)$ versus x provided the intercept corresponding to the endpoint channel number. This value corresponds to the endpoint energy of the incident beta particle

distribution.

Pulse height distributions were measured for radiation sources shown in Table 3.7. From these distributions, the endpoint and centroid channel numbers corresponding to the characteristic electron energies were determined and an energy calibration curve was calculated. The results of these calculations are presented in section 3.5.1

3.4.3 Gamma-ray Discrimination Capability of Spectrometer

The degree to which the spectrometer is capable of actively discriminating against gamma rays is very important for this work. In order to measure this capability, a unit of measure, called the rejection ratio, is used. This is defined as the ratio of the number of gamma ray induced spectrometer pulses which are rejected to the number of leakage pulses recorded [19]. Leakage pulses are gamma ray induced events which, for whatever reason, are recorded in the beta particle spectrum. From this definition it follows that as the rejection ratio increases, the spectrometer's gamma ray rejection capability improves.

To measure the rejection ratio, the gating capabilities of the linear gate stretcher were utilized. In the NORMAL mode, output pulses from the scintillator were the result of beta particle or gamma ray interactions. These beta particles and gamma rays could originate at the source or from the natural background emissions. In COINCIDENCE mode, pulses from the scintillation channel were the result of beta particle interaction in the scintillator or the leakage pulses. If a radiation source was shielded with plastic so that only the gamma rays were incident on the spectrometer, then the only events recorded in the COINCIDENCE mode would be leakage pulses. The rejection ratio, R_γ , was expressed as

$$R_\gamma = \frac{\Sigma Cts(NORM) - \Sigma Cts(BKG)}{\Sigma Cts(COIN)} \quad (3.24)$$

where $\Sigma Cts(COIN)$ is the sum of the counts of the spectrum as measured in the COINCIDENCE mode, $\Sigma Cts(NORM)$ is the sum of the spectrum measured in the NORMAL mode and $\Sigma Cts(BKG)$ is the sum of the spectrum measured in the

NORMAL mode without the source being present.

The gamma-ray discrimination capability of the spectrometer could have been a function of the energy of gamma rays and the interaction rate. In this work, two β - γ emitter sources, ^{140}La and $^{106}\text{Ru}/^{106}\text{Rh}$, were considered to study the effect of incident gamma-ray energy. The thickness of lucite plate used in each measurement was greater than the range corresponding to the highest beta particle endpoint energy. Thus the beta particle component of the source's radiation field was eliminated and the system response to the gamma rays was used to calculate the discrimination ratio. The results of these measurements are presented in section 3.5.2.

3.4.4 Determination of Instrumental Parameters

To develop a useful spectrometer system the instrumental parameters, K_{11} , K_{21} , K_{22} , K_{31} , K_{33} , K_{41} , K_{42} , K_{43} , and K_{44} must be determined. These parameters are highly dependant on interfering radionuclides present in the sample. The three primary interfering isotopes were identified using Chernobyl data [15] as ^{106}Rh , ^{144}Pr , and ^{140}La .

The nine parameters were derived from equations (3.1) to (3.4) using the known activities of sources listed in Table 3.8 and the measured, energy dependant, count rates. Only one source was used in each evaluation of parameters. The parameters can be expressed as:

$$\begin{aligned} K_{44} &= \Delta\nu_4/A_4, \quad K_{33} = \Delta\nu_3/A_3, \quad K_{22} = \Delta\nu_2/A_2, \quad K_{11} = \Delta\nu_1/A_1, \\ K_{21} &= \Delta\nu_1/A_2, \quad K_{31} = \Delta\nu_1/A_3, \quad K_{41} = \Delta\nu_1/A_4, \quad K_{42} = \Delta\nu_2/A_4, \\ \text{and } K_{43} &= \Delta\nu_3/A_4 \end{aligned}$$

The results of these calculations are presented in section 3.5.3.

3.5 Results

3.5.1 Energy Calibration of the Spectrometer

To perform the calibration, pulse height distributions were first measured for the five sources listed in Table 3.4. The data reduction procedures described in section 3.4.2 were used to determine the endpoint or centroid channel numbers which corresponded to the energy of the characteristic electron of each spectrum. The results of these calculations are presented in Table 3.9. A linear least squares fit to these data resulted in the expression:

$$\text{Energy(keV)} = 3.93 \times (\text{Channel Number}) + 48.6$$

The correlation coefficient for the fit was 0.99996. The channel number verses energy data are shown in Fig. 3.9 along with the calibration line.

Table 3.9 Energy Calibration Data

Source	Energy(keV)	Endpoint or Centroid Channel Number
^{147}Pm	224.7	42.79
^{99}Tc	292	63.67
^{207}Bi	481.7	108.3
^{36}Cl	708.9	170.2
^{207}Bi	975.6	235.69
$^{90}\text{Sr}/^{90}\text{Y}$	2245	557.82

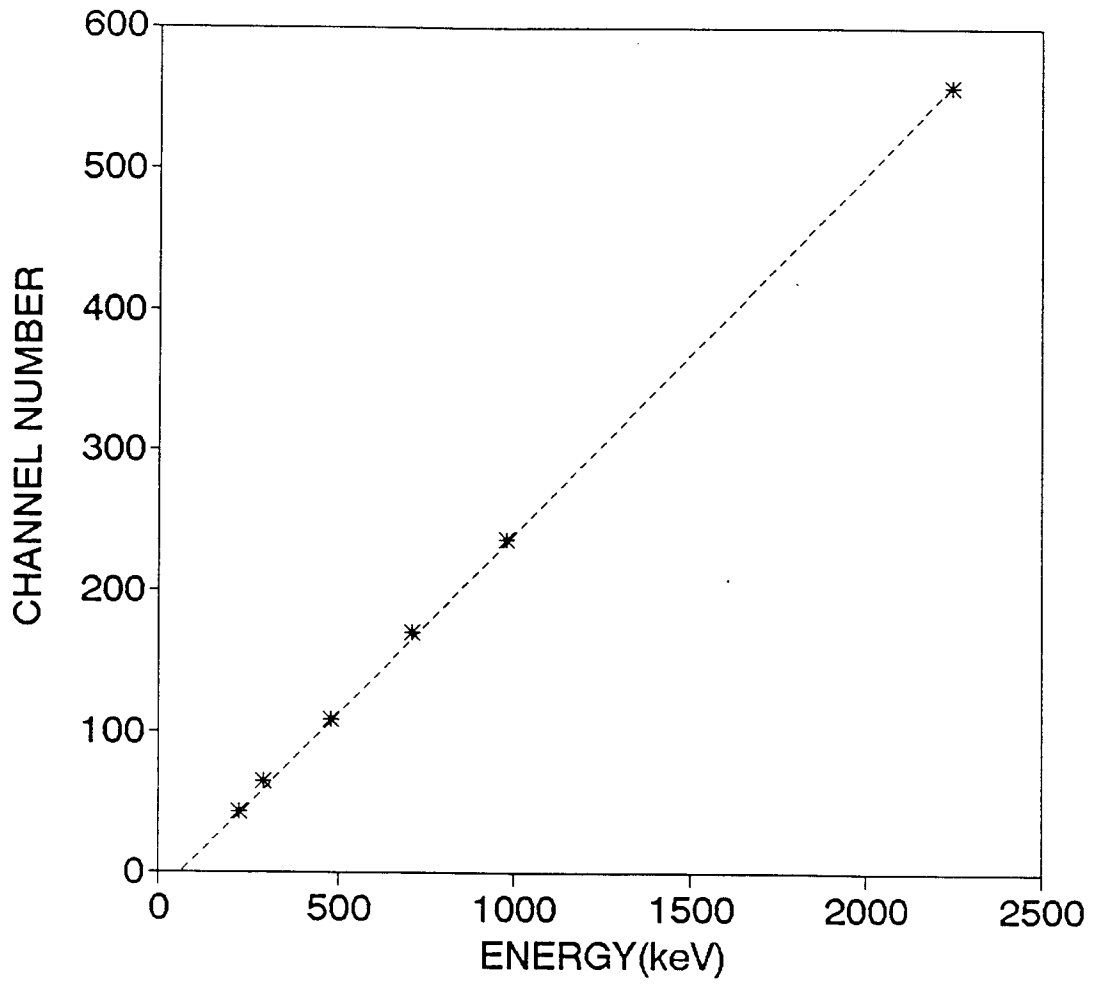


Fig. 3.9 Energy calibration line of spectrometer system.

3.5.2 Gamma-Ray Discrimination Capability

The spectrometer discrimination capabilities for two isotopes, ^{140}La and ^{106}Ru , were investigated and the results are presented in Table 3.10. The discrimination ratio was 157:1 for a gamma-ray field characterized by ^{106}Rh (512 and 622 keV) and 8.5:1 for ^{140}La (1596, 923, 815, 487 and 329 keV). The system discrimination capability decreased with increasing incident gamma-ray energy.

The measurement of the discrimination ratio assumes a radiation field consisting of only gamma rays was incident to the gas-flow counter and that once a gamma-ray passed through the counter there exists no interaction mechanism which would result in the triggering of the counter. Hence, the only method of producing leakage pulse would be direct gamma-ray interaction with the counter. However in actuality, the gamma rays are accompanied by an electron field which is produced by gamma-ray interaction in not only the gas counter but in the lucite beta particle shield positioned before the detector, in the entrance window of the scintillator and in the plastic scintillator itself. These electrons could interact in both the gas counter and the scintillator, resulting in the generation of a leakage.

Recoil electrons are produced in the scintillator as a result of gamma-ray interactions by the mechanisms of photoelectronic absorption, Compton scatter, or, if the gamma-ray energy is large enough, pair production. If the recoil electrons have sufficient energy and are traveling in the proper direction, they may exit the scintillator and enter the gas counter, thus inducing a leakage pulse. The region of the scintillator in which this backscattering can occur is limited to the top

portion of the scintillator corresponding to the range of the highest energy increased, the active "backscatter" volume also increased; thereby increasing the probability of gamma-ray interaction. By this argument, there must be a limit on the discrimination capability of a spectrometer system based on the use of detector geometries and the photon interaction probabilities of the scintillator. More study is needed about the actual determination of the limit on the discrimination ratio.

Table 3.10 Gamma-Ray Discrimination Capability of a Spectrometer System

Source	Gamma-Ray Energy(keV)	Rejection Ratio
^{106}Rh	512(21%), 622(11%)	157 ± 12
^{140}La	1596(96%), 923(10%)	
	815(19%), 487(40%)	8.5 ± 0.2
	329(20%)	

3.5.3 Determination of Instrumental Parameters

To determine the nine instrumental parameters, first beta and gamma counts were measured in two energy regions for the three interfering sources and $^{90}\text{Sr}/^{90}\text{Y}$ listed in Table 3.8. For evaluation of each parameter, only one source was used in this work. The $^{90}\text{Sr}/^{90}\text{Y}$ source was used for determining K_{11} , the ^{144}Pr source for K_{21} and K_{22} , the ^{140}La source for K_{31} and K_{33} , and the ^{106}Rh source for K_{41} , K_{42} , K_{43} , and K_{44} . Using equations in section 3.4.4, the nine parameters were estimated. The results of these estimations are presented in Table 3.11.

Because of the gamma-ray discrimination capability of the spectrometer system, β - γ emitters, ^{140}La and ^{106}Rh , may affect the β counts. Measured β counts will increase due to the leakage pulses which are mentioned in section 3.5.2. The effect of the discrimination ratio on the instrumental parameters needs further investigation and is left to future researchers.

Table 3.11 Instrumental Parameters of Spectrometer System

Parameter	Source	Radiation	Value
K ₁₁	⁹⁰ Sr/ ⁹⁰ Y	β	9.33×10^{-4}
K ₂₁	¹⁴⁴ Pr	β	6.28×10^{-4}
K ₂₂	¹⁴⁴ Pr	β	5.67×10^{-5}
K ₃₁	¹⁴⁰ La	β	1.17×10^{-5}
K ₃₃	¹⁴⁰ La	γ	3.36×10^{-5}
K ₄₁	¹⁰⁶ Rh	β	1.40×10^{-3}
K ₄₂	¹⁰⁶ Rh	β	3.06×10^{-4}
K ₄₃	¹⁰⁶ Rh	γ	1.48×10^{-4}
K ₄₄	¹⁰⁶ Rh	γ	2.97×10^{-4}

3.5.4 Measurement Sensitivity

The minimum detectable activity value for a spectrometer system depends on the sample counting time, the sample mass and geometry, the background radiation levels and the interfering radioisotopes. Representations of equation (3.16) and (3.17) are shown in Fig. 3.10 and Fig. 3.11, where A_1^* is represented as function of M, in a convenient units, and a function of counting time T.

Fig. 3.10 indicates that when $A_2^* \approx 100 \text{ BqL}^{-1}$ ($W^* = 1.60 \text{ BqL}^{-1}$) and 7200 seconds counting time, the sensitivity of sample of about 1 liter of water can reach about 300 BqL^{-1} . The ICRP detection limit of routine monitoring of Y class ^{90}Sr is 0.4 BqL^{-1} which is β counting of urine after chemical separation [26]. When the Derived Air Concentration (DAC) of class D ^{90}Sr is considered, 1 m^3 of air is needed to count the requested concentration. For class Y ^{90}Sr , about 6 m^3 samples are needed. The ICRP DAC (40h/wk) is 300 Bq/m^3 for class D ^{90}Sr and 60 Bq/m^3 for class Y ^{90}Sr [27]. At low volume or mass, the low activity interfering emitters do not affect the sensitivity. In high background count rates the sensitivities are lower than low background. But the sensitivities are almost same at large volume or mass. This means that higher sensitivity can be achieved with small volume or mass in a suitable background reduction, which can be reached either with appropriate shielding or through a more sophisticated of spectrometer system.

Fig. 3.11 shows that when $A_2^* \approx 100 \text{ BqL}^{-1}$ ($W^* = 1.60 \text{ BqL}^{-1}$), samples of about 10 m^3 of air need over 3000 seconds counting time to achieve a 60 Bq/m^3 detection limit. However, with interfering isotopes of low activity and/or a large volume or mass of sample, shorter counting times are

needed.

There are two methods for finding the optimal analysis parameters:

- 1) selecting a detection limit and determining the count time for a given mass or volume, or
- 2) selecting a detection limit and determining the mass or volume for a given time.

Either method gives a reasonable result. Finally, reducing the detection limit further calls for increasing the absolute efficiency of the detector. The instrumental parameters are a function of the efficiency of detectors. New higher efficiency detectors will achieve a higher sensitivity than that realized with this work. Fig. 3.12 represents the theoretical values of detection limits in higher efficiency detector systems than were used in this work.

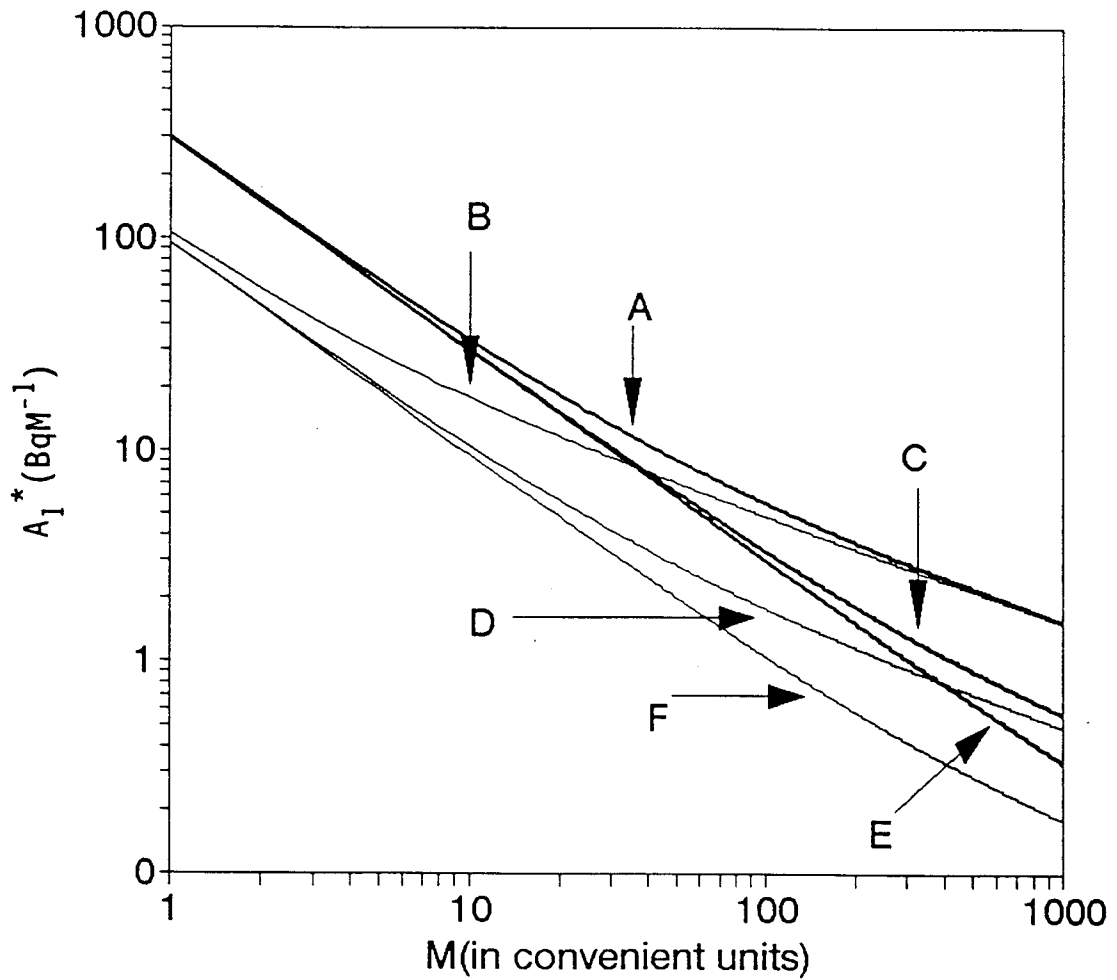


Fig 3.10 Minimum detectable ^{90}Sr specific activity as a function of sample mass (or volume) M (in convenient units).

Note $T = 7200$ sec

A : $W^* = 1.60$ Bq/l $\nu_o = 31.20$ c/sec

B : $W^* = 1.60$ Bq/l $\nu_o = 3.12$ c/sec

C : $W^* = 0.16$ Bq/l $\nu_o = 31.2$ c/sec

D : $W^* = 0.16$ Bq/l $\nu_o = 3.12$ c/sec

E : $W^* = 0.016$ Bq/l $\nu_o = 31.2$ c/sec

F : $W^* = 0.016$ Bq/l $\nu_o = 3.12$ c/sec

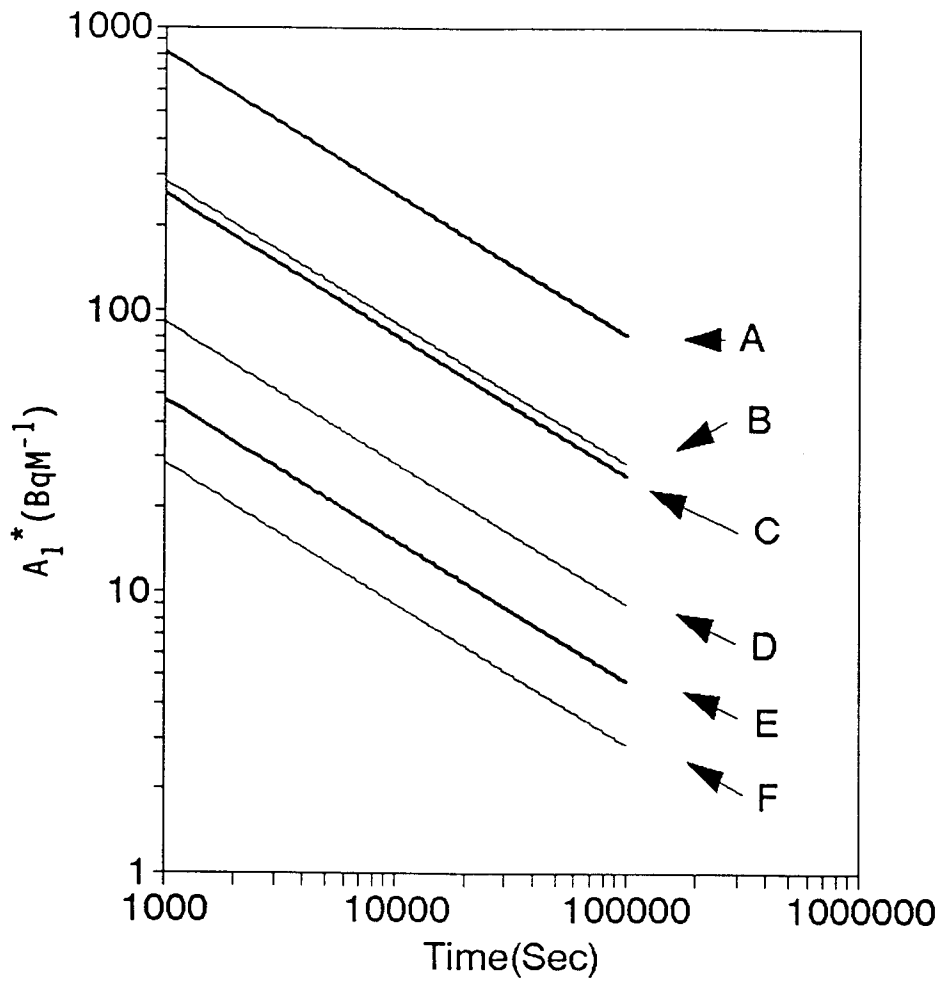


Fig. 3.11 Minimum detectable ^{90}Sr specific activity as a function of counting time T .

Note

$M = 1$

A : $W^* = 1.60 \text{ Bq/l}$ $\nu_o = 31.20 \text{ c/sec}$

B : $W^* = 1.60 \text{ Bq/l}$ $\nu_o = 3.12 \text{ c/sec}$

C : $W^* = 0.16 \text{ Bq/l}$ $\nu_o = 3.12 \text{ c/sec}$

$M = 10$

D : $W^* = 1.60 \text{ Bq/l}$ $\nu_o = 31.20 \text{ c/sec}$

E : $W^* = 1.60 \text{ Bq/l}$ $\nu_o = 3.12 \text{ c/sec}$

F : $W = 0.16 \text{ Bq/l}$ $\nu_o = 3.12 \text{ c/sec}$

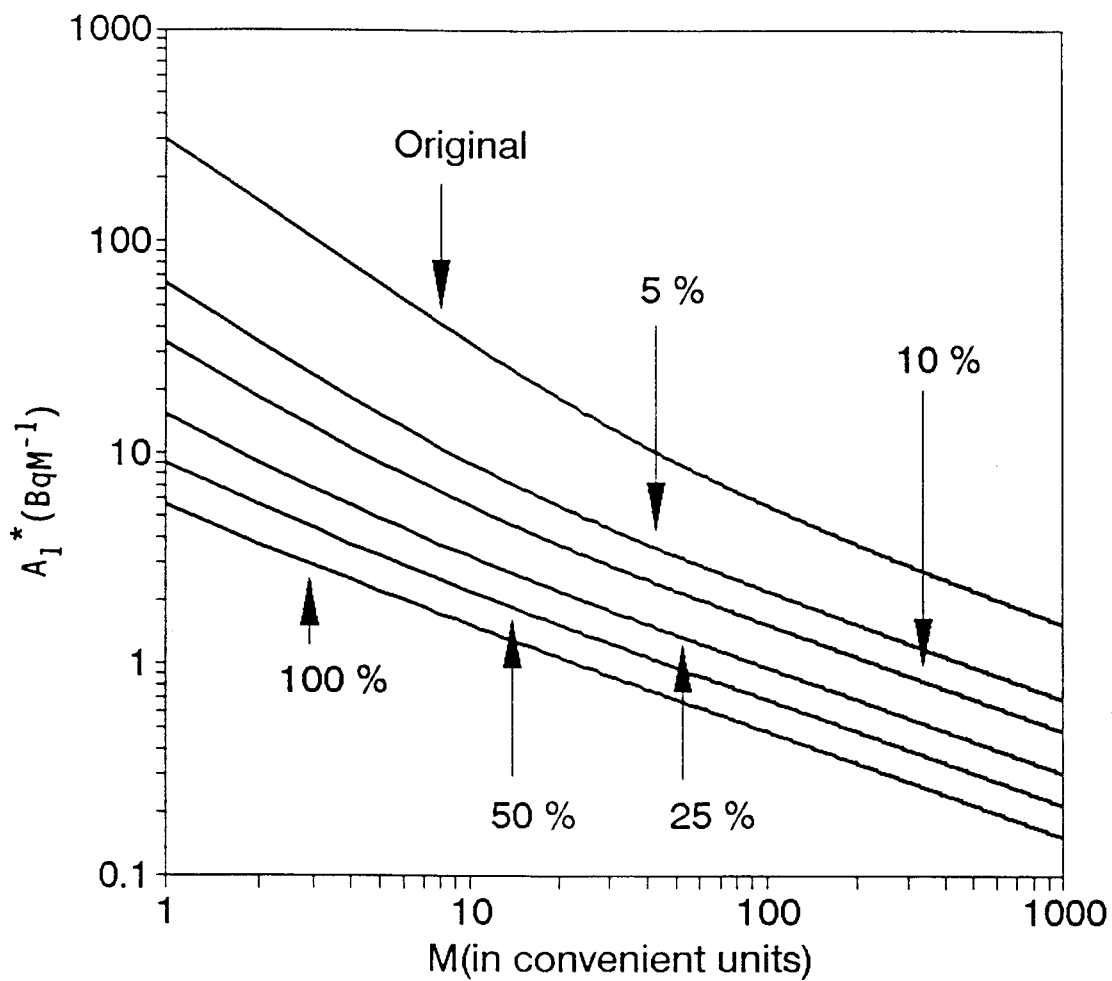


Fig. 3.12 Minimum detectable ^{90}Sr specific activity in higher efficiency detector systems than this work.

Note $A_2^* \approx 100 \text{ Bq/l}$
 $\nu_o = 31.20 \text{ c/sec}$
 $T = 7200 \text{ sec}$

Chapter 4

CONCLUSIONS

Two techniques for assessing ^{90}Sr contamination in environmental matrices have been investigated; the chemical separation method and the beta-particle spectrometer system. Using the chemical separation method, the required length of time needed to perform the analysis and the separation yield were determined. The required time length to measure the ^{90}Sr concentration is about 19 days for milk samples and 25 days for grass ash samples. The chemical yields are $76 \pm 9 \%$ and $49 \pm 7 \%$, respectively. The beta-particle spectrometer technique gives positive results for almost in-real-time measurements. The spectrometer method is approximately 10 - 300 times faster than compared to the chemical separation methods. Estimates concerning detection limit indicate that in a measurement time of about 2 hours, it is possible to assess contamination of about 300 Bq ($8.11 \times 10^{-3} \mu\text{Ci}$) in 1 liter of water sample. This determination is possible with reasonably small amounts of sample and can be performed even when β and β - γ emitters are present.

The measurement sensitivity of ^{90}Sr in environmental matrices depends on the counting time, the sample mass and geometry, the background radiation level and the interfering isotopes. The very low activities required larger and larger samples which cause self-absorption complications. The thickness of the sample must be limited according to the energy range of the electrons to be detected. The ^{90}Sr -measured concentration values could be affected by a systematic error which is not perfectly assessable in a simplified measurement system. More sophisticated equipment for spectral data analysis will limit the error to

the magnitude of the statistical fluctuations. The gamma-ray discrimination capability of the spectrometer affects the ^{90}Sr -measured concentration values. The magnitude of background radiation will also substantially affect the sensitivity. Therefore, it should be reduced by the use of proper shielding and/or more sophisticated detectors. The efficiency of the detector system also affects the detection limit. A higher efficiency detector system will give a lower detection limit than a low efficiency detector system.

In conclusion, this work looks very promising for in-real-time measurement of ^{90}Sr concentration. These optimal analysis methods will achieve a lower limit of detection with short measuring times and smaller samples than other methods involving chemical separation.

Chapter 5

FUTURE WORK

A few ideas which would be interesting to continue research on include:

1. The actual determination of the limit on the discrimination ratio. By the argument in the section 3.5.2, many factors affect the discrimination ratio. So each contribution to the discrimination ratio should be clearly identified in order to determine the limit.
2. The determination of correction factors for the instrumental parameters so that the influence of the discrimination ratio is considered in determining ^{90}Sr concentrations.
3. The improvement of the detection efficiency of the spectrometer system. A higher detector efficiency would result in a lower detection limit.
4. The testing of the spectrometer under "accident" conditions, by labelling milk and grass samples with $^{90}\text{Sr}/^{90}\text{Y}$ in various known concentrations and then using the spectrometer to quantify the $^{90}\text{Sr}/^{90}\text{Y}$ concentrations.
5. Confirm through experimentation that the minimum detectable activity of ^{90}Sr is related to the sample mass and count time as indicated in Figures 3.10 and 3.11.

Chapter 6

REFERENCES

1. F. W. Walker, J. R. Parrington, F. Feiner, Nuclides and Isotopes, 14th ed, GE Nuclear Energy, CA(1989).
2. C. R. Potter, et al., "Determination of Radiostrontium in Food and Other Environmental Samples," *Environmental Science and Technology*, 1, pp 745-750(1967).
3. C. Potter, et al., "Determination of Strontium-90 in Milk by an Ion Exchange Method," *Analytical Chemistry*, 33, pp 1306-1307(1961).
4. C. Potter and B. Kahn, "Improved Determination of Strontium-90 in Milk by an Ion Exchange Method," *Analytical Chemistry*, 36, pp 676-678(1964).
5. D. Cahill and G. Lindsey, "Determination of Strontium-90 in Urine by Anion Exchange," *Analytical Chemistry*, 38, pp 639-640(1966).
6. EPA - 520/5 - 84 - 006, Radiochemical Procedures Manual, US Environmental Protection Agency.
7. EPA - 600/4 - 76 - 011, Tentative Reference Method for the Measurement of Strontium-89 and Strontium-90 in Environmental Waters, US Environmental Protection Agency.
8. G. S. Vyesugi and A. E. Greenberge, "Simultaneous Assay of Strontium-90 and Yttrium-90 by Liquid Scintillation Spectroscopy," *Int. J. Appl. Radiat. Isot.*, 16, pp 581-587(1965).
9. H. V. Piltingsrud and J. R. Stencel, "Determination of Y-90, Sr-90 and Sr-89 in Samples by Use of Liquid Scintillation Beta Spectroscopy," *Health Physics*, 23, pp 121-122(1972).

10. R. B. Randolph, "Determination of Sr-90 and Sr-89 by Cerenkov and Liquid-Scintillation Counting," *Int. J. Appl. Radiat. Isot.*, 26, pp 9-16(1975).
11. R. L. Brondzinski and H. L. Nielson, " A Well Logging Technique for the in situ Determination of ^{90}Sr ," *Nucl. Instr. Meth.*, 173, pp 299-301(1980).
12. R. L. Brondzinski, "Portable Instrumentation for Quantitatively Measuring Radioactive Surface Contaminations, Including ^{90}Sr ," *IEEE Trans. Nucl. Sci.*, NS-31, pp 686-690(1984).
13. J. E. Martin, "Measuring of ^{90}Sr in Reactor Wastes by Cerenkov Counting of ^{90}Y ," *Int. J. Appl. Radiat. Isot.*, 38, pp 953-957(1987).
14. B. Carmon, "The Use of Cerenkov Radiation for the Assay of Radiostrontium in Aqueous Solutions," *Int. J. Appl. Radiat. Isot.*, 30, pp 97-100(1979).
15. D. Brini, et al., "A proposed Multi-detector Method for Prompt ^{90}Sr Detection in Post-accident Environmental Matrices," *Health Physics*, 58, pp 329-339(1990).
16. G. L. Rehnberg, et al., "Levels of Stable Strontium in Milk and the Total Diet," *Environmental Science and Technology*, 3, pp 171-173(1969).
17. USSR State Committee on the Utilization of Atomic Energy, The Accident at the Chernobyl Nuclear Power Plant and Its Consequences, Part II. Annexes 1,3,4,5,6(1986).
18. N. Tsoulfanidis, Measurement and Detection of Radiation, pp 67-68, Hemisphere Publishing Co., New York(1983).

19. G. G. Simons and J. F. Higginbotham, "Beta-Particle Spectroscopy with Active Gamma-ray Discrimination," Nucl. Instr. Meth., 293, pp 551-554(1990).
20. J. F. Higginbotham, Beta-particle Spectroscopy with Active Gamma-ray Discrimination, Ph.D. Dissertation, KSU(1987).
21. E.I. du Pont de Nemours & Co.(Inc.), 331 Treble Cove Road, North Billerica, MA 01862
22. Isotope Products Laboratories, 1800 N. Keystone Street, Burbank, CA 91504
23. Amersham Corporation, 2636 South Clearbrook Drive, Arlington, Heights, IL 60005
24. J. G. Cramer, B. J. Farmer and C. M. Class, "A Scintillation Spectrometer for High Energy Beta Decays," Nucl. Instr. Meth., 16, pp 289-300(1962).
25. G. F. Knoll, Radiation Detection and Measurement, 2nd ed, John Wiley and Sons, Inc., New York(1989).
26. ICRP, Individual Monitoring for Intakes of Radionuclides by Workers: Design and Interpretation, Pergamon Press, ICRP Publication 54, p 98(1988).
27. ICRP, Limits for Intakes of Radionuclides by Workers, Pergamon Press, ICRP Publication 30, Supplement to Part 1, p 85(1979).