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A GAS PROPORTIONAL COUNTER SYSTEM FOR THE DETECTION OF ^{13}N AND OTHER BETA EMITTING GASES

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Abstract:

A simple gas proportional counter system is described that has been developed for measurements of ^{13}N gases in the labeled effluent from gas chromatographs and in recirculating gas atmospheres from soil cores. The gas proportional counter is designed to detect the positrons from the decay while remaining fairly insensitive to the annihilation gamma-rays. A commercial high density amplifier-comparator was used in place of traditional NIM electronics. The general properties of the system are described and compared to those employing traditional low density NIM electronics. With this system a multiple detector array can be built at low cost and without concern for cross-talk.

1. Introduction

The most useful radioisotope of nitrogen for tracer studies is ^{13}N. This nucleus has a relatively short half-life, 9.96 minutes, and decays by positron emission to the stable ^{13}C. Because of the short half-life, ^{13}N must be produced by accelerator based nuclear reactions and be used immediately. The short half-life also dictates that batch production runs on the accelerator be short, on the order of 20 minutes, certainly no longer than 30 minutes. With such a short production cycle efficient use of the accelerator will require that a large number of replicate measurements be run simultaneously. Thus emphasis should be placed on developing a detection system that can measure a large number of ^{13}N samples simulataneously.

^{13}N can be traced through the chemical and biological transformations of interest by following the emission of the positrons, or their annihilation radiation. The measurement of the annihilation radiation can be done in a straightforward

manner with scintillation counters, such as NaI(Tl) or plastic [1]. These detectors can have a high efficiency but also have large backgrounds unless they are well shielded. The shielding has to be particularly good when a large number of measurements are being made in close physical proximity to one another in order to avoid cross-talk. The background can be surpressed by requiring the coincident detection of both annihilation gamma-rays. However, such coincidence systems are not well suited to large arrays of identical systems because they require a large number of NIM modules to make up each logic circuit.

Alternatively, the measurement of the positrons can be made with a simple gas proportional counter (GPC). These detectors are fairly insensitive to cross-talk. The walls of the detectors are generally thick enough to stop external electrons making such detectors only subject to cross-talk from gamma-rays that create photoelectrons or pair-produce in the detector walls. The annihilation radiation is below the threshold for pair-production by definition, thus greatly reducing the possibility of cross-talk between GPC's measuring positrons from ^{13}N. The efficiency of such detectors depends on the energy of the electrons being measured because of the absorption of low energy electrons in the source and the window. Solid sources have to be carefully prepared and the window has to be fairly thin. In this work the ^{13}N sources were always in the gas phase so that self-absorption of the positrons was minimal. The gas phase sources can be counted internally if the chemical makeup of the gas is suitable or externally through a window if not. In certain biological applications it is desireable to recycle the gas after counting and thus a separation of the source from the counting gas is required. The detector system that we have developed has this feature.

In this paper we report the development of a gas proportional counter coupled to a commercial high density amplifier-discriminator system. The counter system is particularly well suited to accelerator based experiments that use ^{13}N as a radiotracer. The GPC is insensitive to cross-talk and a large number of identical channels can be produced at a low cost.

2. Description

2.1 Proportional Counter

The characteristics of gas proportional counters are well known and have been broadly applied to assays of radioactivities [2]. Two different types of samples were measured with the GPC system, the effluent gas from gas chromatographs and the gases circulating in a closed loop containing a soil core. For the reasons given above the sources had to be external to the sensitive volume of the detector.

A schematic diagram of the detector is shown in Figure 1. Approximately 215 cm^3 of flowing counting gas (a mixture of 10% methane with argon) was contained in the cylindrical volume at the top. The detector body was standard pipe fittings, a 6.35 cm diameter copper pipe with a soldered copper end cap. The GPC was operated at atmospheric pressure with an aluminized mylar window

separating the counting gas from the radioactive source gas. The window was 828. $\mu g/cm^2$ aluminized mylar. These gas phase sources were forced to move past the window in a maze machined into the base plate. Several mazes were produced with different volumes; the volume was usually on the order of 2 - 10 cm^3. The maze depth for the gas chromatograph unit was adjusted so that the separated radioactive gases were not mixed during their transit through the GPC. The maze depth for those detectors connected in series with the soil cores was 1 cm which maximized the sensitivity of the detector by increasing the fraction of the volume of the closed system gas in front of the window. The window was glued to the base creating a sealed volume to contain the radioactive gas. The electrons from the multiplication of the primary ionization were collected on a 51 μm thick nickel wire in a 10 mm diameter loop near the center of the counting gas. The loop was attached to a panel-mount SHV connector secured to the copper end cap.

The detector operated in the voltage range of 1500 to 2800 volts as discussed below. The calculated profile of the equipotential surfaces with 2kV on the collecting loop is shown in Figure 2 [3]. The calculations were performed with a two dimensional grid with an assumed cylindrical symmetry. The GPC is not cylindrically symmetric and the true equipotential surfaces should lie between the the two limits presented in Figure 2. The upper limit to the collector voltage was found to be approximately 2800 V due to discharges through the gas. Twelve identical detectors were built and their voltage versus countrate characteristics were measured. As expected, the shape of the collector loop was found to cause the largest variation between detectors and was critical in determining the reproducibility of the detector response. If a collector loop was damaged by mishandling it had to be replaced to maintain uniformity among the detectors.

2.2 Electronics

The electronics used with the GPC's are indicated schematically in Figure 3. The characteristics of the GPC were determined at first with NIM electronics, i.e. a single HV supply, a low noise preamp (x100 gain) coupled to a linear amp (approximately x200 gain) followed by a single channel analyzer (SCA). The results of the measurement of countrate versus voltage as a function of threshold is shown in Figure 4A. A ^{137}Cs source was positioned approximately 5 cm below the window and the channeled base plate was replaced by an aluminum plate with a clearance hole.

The number of counts registered in the scaler from the GPC depends on the applied HV (and thus on the multiplication of the primary ionization) and on the lower level discriminator of the SCA. The lower level threshold of the SCA was varied from 50 to 250 mV with the upper level discriminator disabled. The signal to noise ratio at the output of the preamp was approximately 50:1. This ratio allowed the discriminator level to be as low as 50 mV while maintaining good noise rejection. A plateau can be seen in Figure 4A that extends over the region of 2200 to 2700

volts. The GPC began to spark when the collector voltage was above 2800 volts.

In this mode the pulse heights of the signals were not important, i.e., the number of counts being scaled was simply the number above a lower level discriminator. A much simpler circuit containing only a fast amplifier and a comparator before the scaler can perform this function. Such a circuit is presently available in a hybrid DIP package from LeCroy Research Systems [4]. This circuit can accept a positive or negative input having dual fast amplifiers before a differential comparator. The chip requires an external threshold level in the range of -1 to -16 V (converted internally into a comparator threshold) along with ±5 V power supplies. The internal comparator threshold is set in proportion to the external level and can be varied from ±0.20 to ±3.2 mV. The chip is available in a single package (MVL100TB) and in groups of 16 on a single printed circuit board (models 7791 and 7790N). In the initial work that we report here the single unit was used. However, our complete system uses a group of 12 chips out of the 16 available on the Model 7791 board.

The measurement of countrate versus voltage for the LeCroy chip is shown in Figure 4B. The same detector and source were used in conditions that were as identical as possible to the previous measurements utilizing the NIM electronics. The curves are labeled according to the threshold voltage. This internal threshold varies from 0.4 to 1.2 mV over this range as calculated with the nominal values of the chip calibration. The gain of the fast amplifier stage is x100. This electronic gain is significantly lower than that in the NIM electronics and requires a larger multiplication inside the detector to produce signals above the threshold. However, the response of the MVL100 circuit matches that of the NIM electronics in the region of 2600 to 2700 Volts with an internal threshold of approximately 0.5 mV. A larger gain in the fast amplifier is desirable but not available. In critical applications an additional fast amplifier (such as the LeCroy VV100B hybrid chip) can be inserted between the GPC and the MVL100. The signal to noise after the fast amplifier is approximately 10:1 at these voltages.

3. Discussion

We have used the GPC's to detect ^{13}N in two different applications, measuring the labeled effluent from a gas chromatograph to identify the chemical form of the radiotracer, and in monitoring the microbiological reduction of nitrate to dinitrogen gases by continuously circulating gas through a soil core to a GPC in a close loop [5]. The latter use of the GPC represents a novel approach to denitrification studies and will be described in a more detailed report. The coupling of GPC to a gas chromatograph is a more general application and is briefly discussed below.

A GPC was coupled to a Carle Instruments Model 8000 gas chromatograph for a calibration of its efficiency with a known activity of ^{85}Kr. The carrier free krypton was injected into the chromatograph operating at approximately 45°C with a helium carrier gas flowing through a Porapak Q column at approximately 25 ml/min. The ECL output of the MVL100 was shifted to a NIM

level and sent to a multichannel analyzer operating as a multiscaler. The dwell time of the multiscale was 1.0 seconds. Typical results from an injection of approximately 0.2μCi sample of ^{85}Kr are shown in Figure 5. The peak shape is nearly Gaussian, having a significant tail towards longer times. This tailing indicates turbulent flow past the window. The efficiency of the GPC's were found to be approximately 45 percent (maze volume of 2.1 cm^3). The background in the detectors was on the order of 1 to 2 counts per second. This created a lower limit to the sensitivity of the GPC of approximately 2 nCi.

We also tested the GPC's for their efficiency in counting ^{14}C as this isotope is the one most commonly used in biological experiments. $^{14}CO_2$ was injected into the gas chromatograph and the effluent peak measured in a GPC with the standard mylar window and was found to be approximately 15 percent. A second measurement was made with a thinner aluminized polypropylene window (100 $\mu g/cm^2$). The efficiency for detecting the lower energy beta particle was found to be essentially the same as that for the higher energy ^{85}Kr beta particle.

4. Conclusions

Our results show that modern high density electronics can be coupled to a simple gas proportional counter to create a ^{13}N counting system. We have measured the characteristics of the GPC with traditional NIM electronics and compared them to those found with the LeCroy MVL100 fast amplifier/comparator. The signal to noise ratio was approximately 50:1 and 10:1 in the two cases, respectively. With this and the fixed gain of the hybrid electronics the GPC had to be operated at higher voltage to match the efficiency obtained with the NIM electronics. The efficiency of the standard GPC's was found to be approximately 45 percent for moderate energy beta particles (^{85}Kr), dropping to approximately 15 percent for low energy betas (^{14}C).

The 12 detector system that we have described was developed for accelerator based experiments using ^{13}N as a radiotracer. However, these high density electronics can be applied to other situations where a large number of identical detectors are needed. For example, self-shielded low level proportional counters have a large number of internal elements that are operated in anticoincidence. Alternatively, a single LeCroy MVL100 fast amplifier/comparator chip coupled to a gas proportional counter is an attractive combination for measuring radiotracers in the effluent from gas chromatographs. Such a detector only requires a HV supply and an adjustable discriminator threshold. A similar detector could be coupled to a liquid chromatograph, but its efficiency would be limited by self-absorption in the liquid source. This system is much less costly than the commercial GPC units sold for use with gas chromatographs.

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References

1. e.g., M.B. Knickelbein, J.W. Root and C.R. Hurlbut, Nucl. Instrum. Meth. 216 (1983) 121.

2. e.g., P. Povinec, Nucl. Instrum. Meth. 176 (1980) 111, and references therein.

3. The code POISSON represents a continuing upgrade of the code TRIM by A.M. Winslow, J. Computational Phys. 2 (1967) 149. The former code is available at the National Superconducting Cyclotron Laboratory.

4. LeCroy Research Systems Corp., Spring Valley, New York

5. T. Parkin, H.F. Kaspar, A.J. Sexstone, and J.M. Tiedje, Soil Biol. Biochem. 15 in press (1983).

Figure captions

1. A schematic diagram of the gas proportional counter is shown. The counter gas is separated from the radioactive gas by a thin aluminized mylar window. The source gas enters a closed volume in the base and then either exits to an exhaust manifold when connected to a gas chromatograph or returns to the soil core.

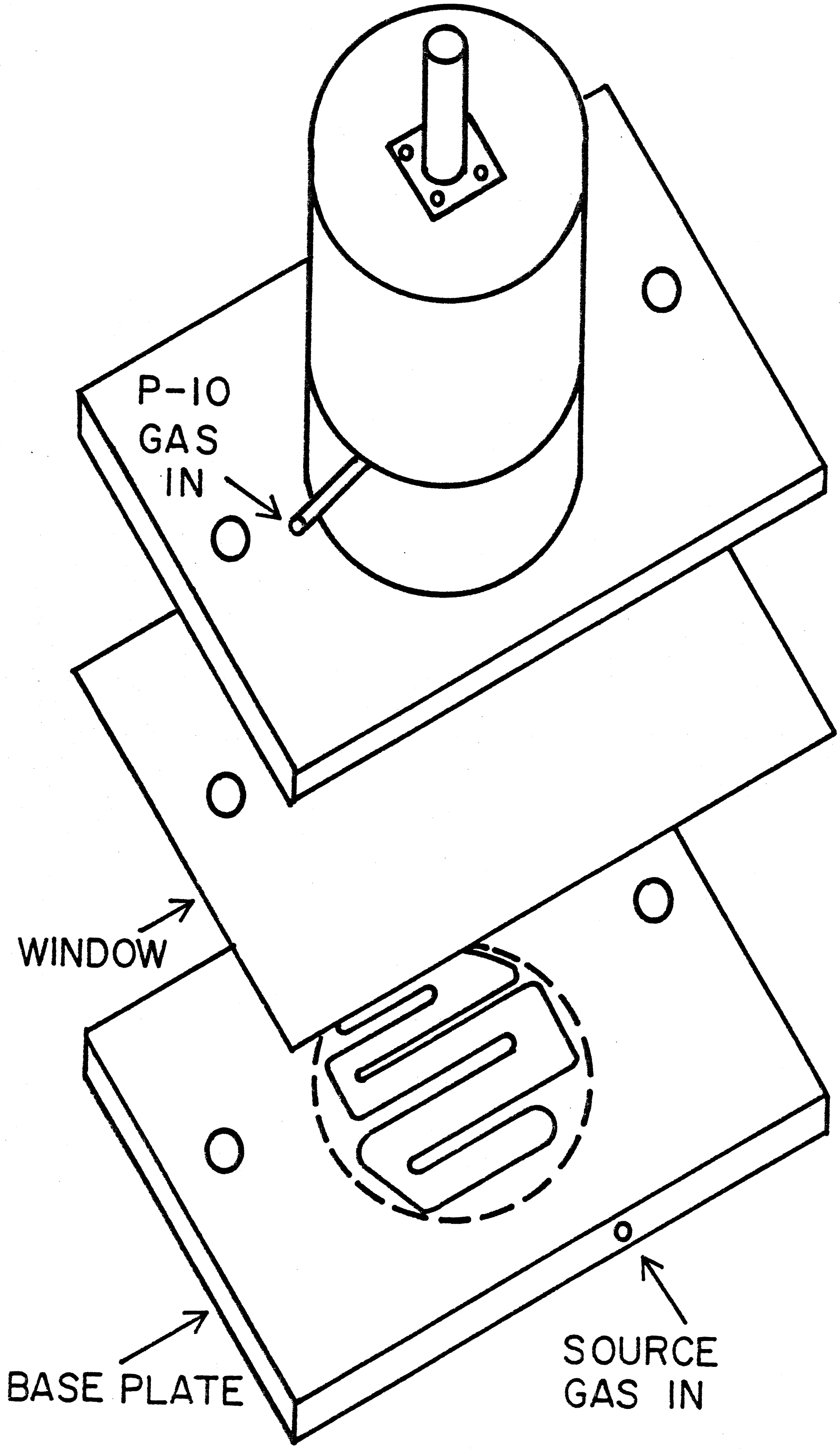
2. The equipotential surfaces are shown for two perpendicular sections through the GPC at a typical operating voltage of 2 kV. The surfaces were calculated with the computer code POISSON [2] on a two dimensional grid with an assumed cylindrical symmetry.

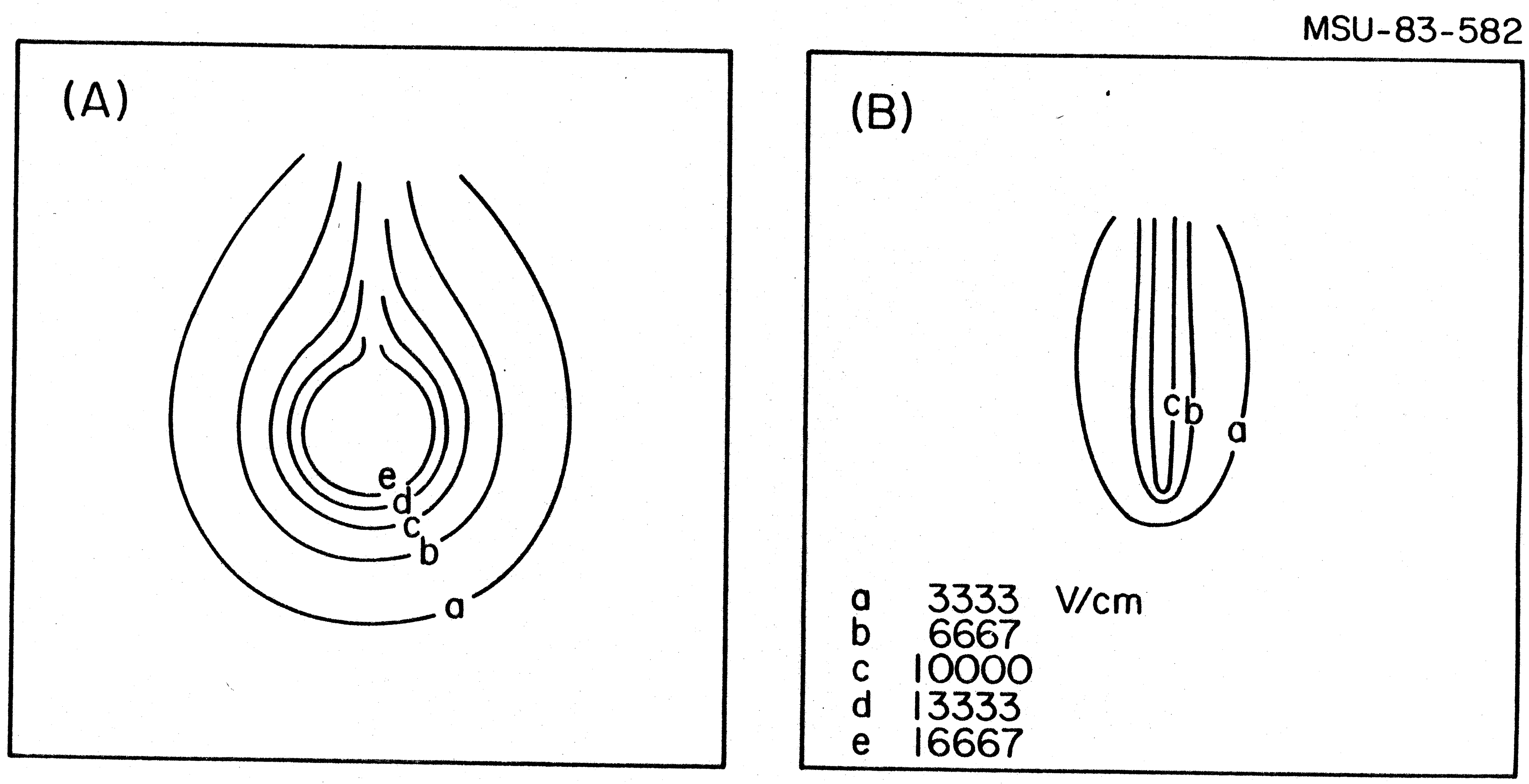
3. A schematic diagram of the electronics coupled to the GPC is shown. The traditional NIM electronics are indicated by the sequence A-B-C-D. The high density electronics is indicated by the sequence A-E-F-I. Alternate connective sequences are discussed in the text. The modules are labeled as follows: LA for linear amplifier; SCA for single channel analyser; DISC for discriminator; ECL/NIM for ECL to NIM level shifter; the two scalers are labeled according to their input logic level.

4. The voltage versus efficiency for a GPC obtained with traditional NIM electronics is shown in part A for a series of lower level discriminator settings. Similar measurements of the same detector with the high density amp/comparator are shown for comparison in part B.

5. A multiscale of the counts observed in a GPC connected to the output of a gas chromatograph. A sample of ^{85}Kr with an activity of approximately 0.2μCi was injected into the chromatograph.

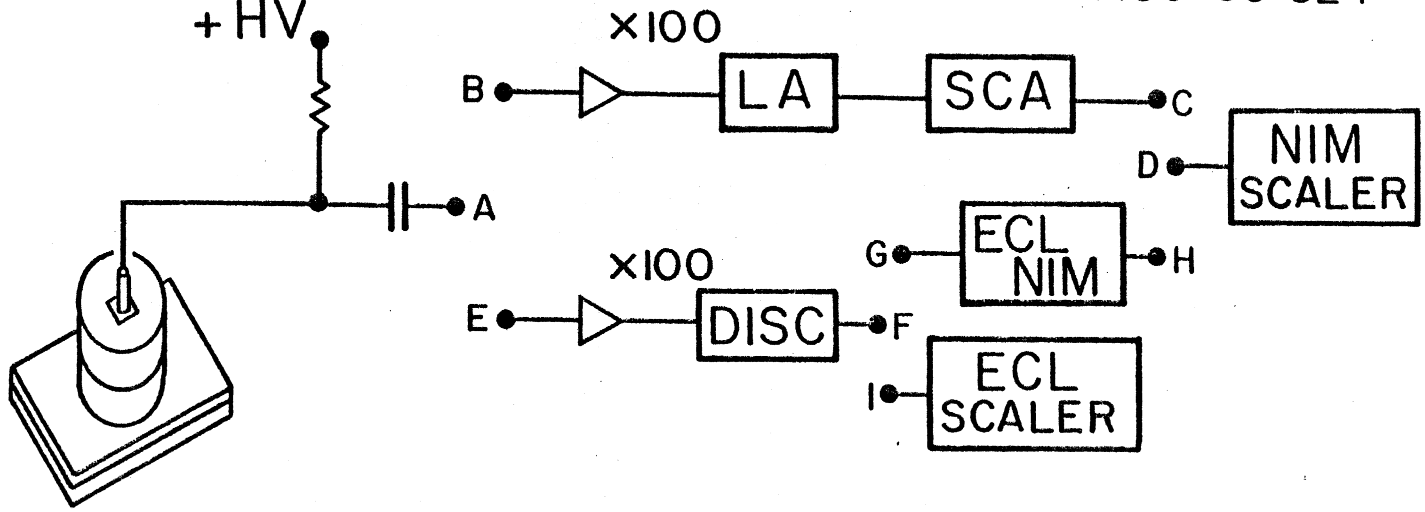
MSU-83-523





MSU-83-582
(A)
e
d
c
b
a
(B)
c
b
a
a 3333 V/cm
b 6667
c 10000
d 13333
e 16667

MSU-83-524



+HV
A
B
×100
LA
SCA
C
D
NIM
SCALER
×100
E
DISC
F
G
ECL
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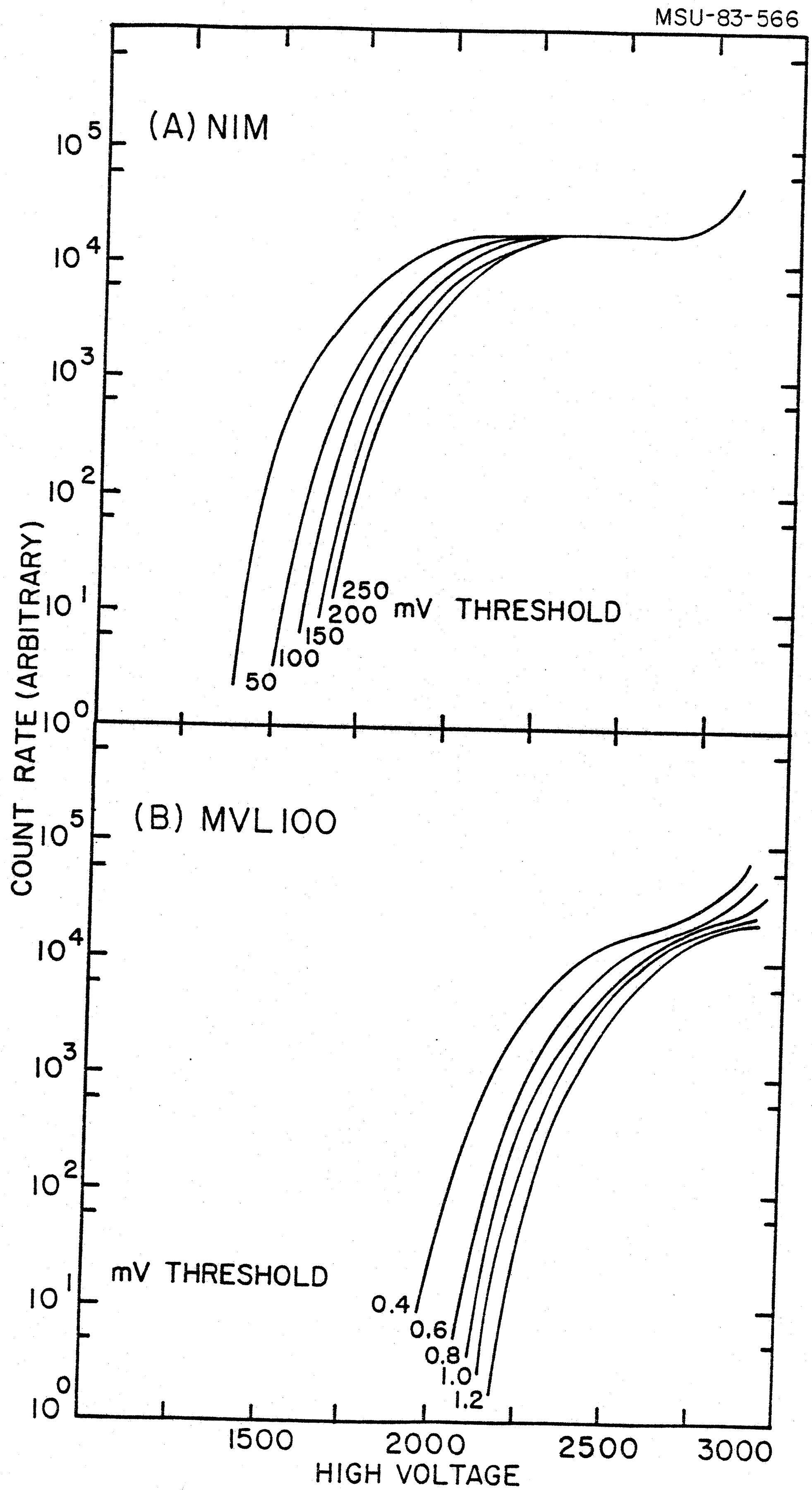
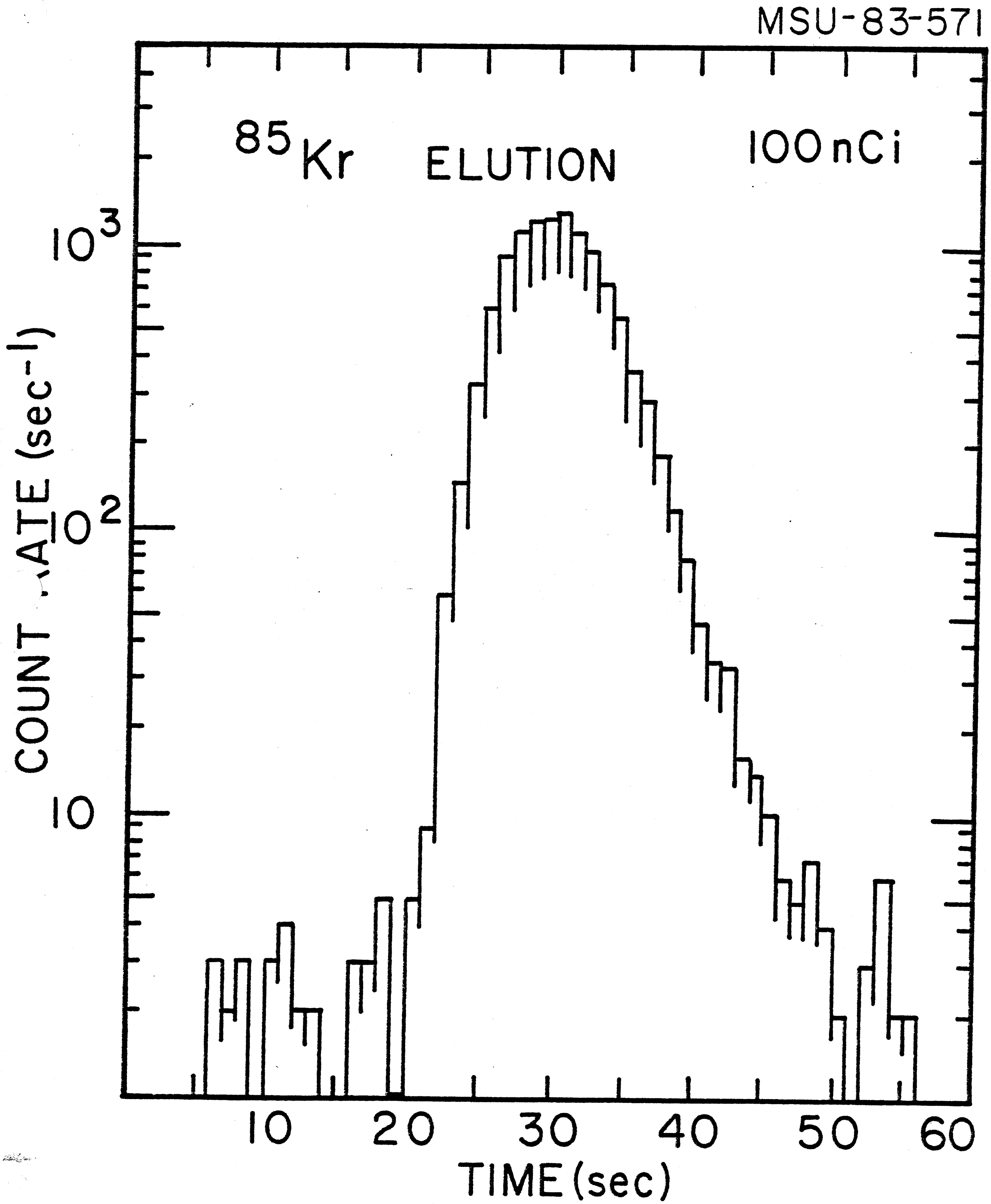
H
I
ECL
SCALER



MSU-83-566
(A) NIM
(B) MVL100
COUNT RATE (ARBITRARY)
HIGH VOLTAGE
1500
2000
2500
3000
50
100
150
200
250
mV THRESHOLD
0.4
0.6
0.8
1.0
1.2
mV THRESHOLD


MSU-83-571
85Kr ELUTION
100nCi
COUNT RATE (sec-1)
10^3
10^2
10
10
20
30
40
50
60
TIME (sec)