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Ge(Li) Low Level In-Situ Gamma-Ray Spectrometer Applications*

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G. W. Huckabee, and D. L. Sawyer

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Ge(Li) LOW LEVEL IN-SITU GAMMA-RAY SPECTROMETER APPLICATIONS

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Summary

Currently we are employing a Ge(Li) spectrometer for in-situ measurements of radionuclides contained in soil. This is being done at nuclear reactor sites and in complex radionuclide fields at the Nevada Test Site. The methodology and precision of the in-situ spectrometric technique has previously been established for analysis of radionuclides in soil. Application of the technique to gaseous and liquid effluents containing radionuclides has shown a great deal of promise.

Introduction

The methodology and feasibility of in-situ Ge(Li) spectroscopy for radionuclides contained in soil has been explored at our laboratory and by others.¹⁻⁵ It is clear that in certain instances the method offers advantages over laboratory gamma-ray spectroscopy methods. Probably the most significant advantages of in-situ Ge(Li) spectroscopy are the very short counting times required (30 minutes compared to 1000 - 2000 minutes in the laboratory), and the fact that a very large quantity of soil is examined (several metric tons compared to a few hundred grams in the laboratory). A major drawback is the requirement to know the distribution of the radionuclides as a function of depth in the soil. However this usually is not a requirement for the natural occurring radionuclides which are commonly found to have a uniform distribution with depth. The most difficult case is for surface deposited radionuclides that have leached into the soil and where distribution is related to a complicated process involving the amount of rainfall, physical-chemical properties of the soil, vegetation, etc. Another cause of non-uniform distribution could be man-made working of the soil, i.e., agricultural and construction disturbances. The distribution must be taken into account when determining radionuclide distribution in soil; therefore, some initial soil sampling is necessary. We have found that large areas (several Km²) can probably be represented by 2-3 each, 0-30 cm soil profile measurements. We currently have a program under way to further evaluate to what extent these assumptions can be made. Measurements of radionuclide concentrations on the soil surface is a much simpler problem, since a depth distribution is not required in the calibration. Examples of this type of radionuclide distribution would be from fresh fallout from weapon tests or from a nuclear accident where radionuclides would be transported from a source and deposited downwind on the soil surface.

In addition to determining the radionuclide concentrations in soil from the field in-situ Ge(Li) spectral data, external gamma exposure rates may be calculated. This is particularly useful where the exposure rate contribution from a particular radionuclide must be determined. The methodology was developed by Beck.⁶⁻⁷ Anspaugh⁵ more recently has developed a computer program that has been successfully used for detailing the external gamma exposure

for a complex mixture of radionuclides (12 man-made, 3 natural occurring) contained in soil at the Nevada Test Site. A feature of the method for determining exposure rates is the fact that it is much less dependent upon knowing the distribution of radionuclides as a function depth.

Currently we have several projects within our laboratory, using the in-situ Ge(Li) spectrometric technique for quantifying radionuclides in soil. These include studying the distribution and redistribution of radionuclides at the Nevada Test Site, baseline studies for radionuclide distribution at nuclear power reactor sites, radionuclide inventories of tailing piles from uranium mills and radioactive hot-spot areas where higher than expected fallout has taken place.

The Ge(Li) in-situ method is most readily adapted to measuring radiation flux from radionuclides in the soil; however, we are investigating its use in gaseous and water effluents containing radionuclides. These have primarily been done at reactor sites.

Ge(Li) Detector Characteristics

For in-situ gamma-ray spectroscopy, we selected the closed-end type Ge(Li) detector. The choice was dictated by the wide angle photon acceptance requirement. Unlike laboratory gamma-ray spectrometers, where the radiation entrance is confined to the end surface of the detector, in-situ methods require high-efficiency on the sides as well. Photon acceptance angle approaches 90° with the detector 1 m above the soil surface.

Two Ge(Li) coaxial detectors have been evaluated. The housing and mounting configurations are shown in Fig. 1. An important criteria for good low energy response is to have a minimum of low Z material between the detector and entering photon flux. This includes the vacuum cap, heat shield, mounting hardware and attention to the Ge(Li) detector outer dead-layer surface. The detector efficiencies were found to vary by approximately a factor of two at 662 keV. A plot of detector efficiencies is given in Fig. 2.

The Ge(Li) detector identified as V-3 was found to have appreciably better low energy response at 60 keV than the detector listed as V-1. Vacuum caps and heat shields for both detectors are approximately the same, however, in processing the V-1 Ge(Li) detector an abnormal thick N layer resulted and hence the poor low energy response. We have used the V-1 detector for extensive field measurements,^{3,5} but the better lower energy response of V-3 as well as the much better overall efficiency makes it the detector of choice. Characteristics of the two Ge(Li) detectors are given in Table I.

Measurement System

The gamma-ray analysis system is shown in Fig. 3. Details of the system have previously been described⁴

and only a brief summary will be given in this paper. The heart of the system is the Ge(Li) detector, however a variety of detectors make up the total system. Besides the Ge(Li) detector, included are a high pressure ionization chamber, Fidler³ NaI(TL) detector and a 2" x 2" NaI(TL) scintillator. All the apparatus is housed in the van-type vehicle shown in Fig. 4. Electrical power is supplied by a small gasoline engine-driven electrical generator and a 120 ampere-hour battery/inverter supply.

Signal output from the Ge(Li) detector is fed to a 4096 channel pulse height analyzer and stored in core memory. Spectral data may be transferred from the core memory to either magnetic tape for future analysis or to a teletype printer for a printout of selected channels. The normal operation is to store the data on magnetic tape for later processing by existing computer programs. However, the teletype has proven invaluable since it permits on-the-spot reduction of photopeak information into radionuclide values. This is necessary for example where radionuclide concentrations must be determined as a guide to subsequent measurements. It is also a great aid for field calibrations.

The Fidler detector may also be used in conjunction with the pulse height analyzer. The instrument has been used primarily in our research efforts directed toward quantifying plutonium/mericium soil.⁹ Output from the high pressure ionization chamber is recorded on a strip chart recorder and also may be displayed on a time-scaler for a more accurate reading. A continuous recording of the relative gamma-ray background is provided from the 2" x 2" NaI(TL) scintillator which is permanently mounted on the inside wall of the van. A rate meter display is mounted near the vehicle's instrument panel.

Spectral Analysis Methods

Field measurements are made with the Ge(Li) detector mounted on a tripod with the detector 1 m above the soil surface. Care is taken to place the detector at least 20-30 m away from the support van or other obstructions.

The general methodology used to interpret field spectra follows that developed by Beck, Condon and Lowder;¹ further details are given by Anspaugh, et al., Refs. 3,5.

The equation used for the determination of the concentration of radionuclides in soil is

$$\frac{N_f}{S} = \frac{N_0}{\theta} \times \frac{N_f}{N_0} \times \frac{\theta}{S}$$

where N_f/N_0 is the counting rate in the photopeak of interest per unit of soil radioactivity; N_0/θ is the number of counts in the photopeak per incident photon/cm² when a point source is placed at 0° from the detector axis and sufficiently far away that the flux striking the detector can be considered parallel; θ/S is the flux of unscattered photons incident at the detector per unit of soil radioactivity; and N_f/N_0 is a correction factor that must be applied if the response of the detector is not uniform as a function of angle from the detector axis. All factors are energy dependent.

The calibration process consists of measurements

of N_f/θ and the detector response as a function of angle from its axis using point sources and calculations of θ/S and N_f/N_0 for any desired source characteristics. The distribution of radionuclides with depth must be known. In most cases we have assumed that natural occurring radionuclides are uniformly distributed with depth and that fallout radionuclides are distributed exponentially with depth.

The calibration process is quite time-consuming and cannot be repeated frequently without a severe loss in available field counting time. The only calibration parameter that is likely to change is N_0/θ , which relates directly to the intrinsic efficiency of the detector. The efficiency of the detector system is therefore checked daily by counting a small calibration source mounted in a jig and attached in a reproducible position directly underneath the detector. The calibration source contains 9 radionuclides producing 20 photopeaks. Counting time is 200 nsec.

Applications of In-Situ Ge(Li) Spectroscopy

Terrestrial Radiation Flux

Natural occurring radionuclides contained in soil consist mainly of those formed from the radioactive decay that takes place in the uranium-thorium (238U, 232Th) series and from potassium-40 (40K). In addition to those of natural origin, trace amounts of man-made radionuclides may be found in soil. These are primarily from fallout associated with previous atmospheric weapon tests. The most abundant gamma-emitting radionuclide from the latter source and found throughout the global environment is 137Cs.

A 180-minute in-situ count with the Ge(Li) detector one meter above the soil surface was performed. This was done in an area of the Livermore Valley, where the soil was known to have been undisturbed for at least the last 30 years. The resultant gamma-ray spectrum is shown in Fig. 5. Measurements from this site have previously been reported by Anspaugh,³ et al., who found the 137Cs in the soil to be distributed exponentially as a function of depth and with a slope of $-0.16 \pm 0.01 \text{ cm}^{-1}$. The photopeak at 662 keV for 137Cs as shown in Fig. 5 represents $39 \pm 2 \text{ pCi/m}^2$. Distribution of the natural radionuclides 238U, 232Th, 40K were found to be uniform as a function of depth. This is generally the case for the natural occurring radionuclides. Analysis of the spectrum of Fig. 5 has shown the concentrations in soil for 238U equal to $0.65 \pm 0.02 \text{ pCi/g}$; 232Th equal to $0.67 \pm 0.02 \text{ pCi/g}$; and 40K equal to $15.4 \pm 0.3 \text{ pCi/g}$. These values are in close agreement with laboratory measurements performed by Ge(Li) and mass spectroscopy.³

A significant advantage of in-situ counting is the reduction in counting time required as compared to a typical laboratory analysis. The in-situ example given here represents 180 minutes of counting time; however, a reduction in counting time to 30 minutes resulted in a small decrease in statistical accuracy and for most cases has proven adequate, i.e., the 137Cs concentration value had an accuracy of $\pm 11\%$. On the other hand, laboratory analysis by Ge(Li) spectroscopy requires at least 1000 minutes per sample for the same accuracy, not including time for sample collection and preparation.

Another advantage is the greatly increased area the in-situ Ge(Li) spectrometer views as compared to soil sampling and laboratory spectroscopy. The viewing area for 50% photon flux at the detector is approximately 300 m² for an exponentially distributed

source with a slope of -0.16 cm^{-1} . On the other hand, soil sampling usually represents a few hundred cm^2 of soil surface.

The in-situ method is also well suited for quantifying radionuclides contained in a complex mixture and deposited on the soil surface. An example of this is given in Fig. 6. Here 12 man-made radionuclides are identified as well as those occurring naturally. This in-situ measurement was made near the Sedan crater at the Nevada Test Site.⁵

Varied Geographical Sites

Measurements were conducted at a number of sites in California representing a wide variation in geographical regimes. These involved sites in a northern-inland coastal region, sites in the central valley extending from the northern to its southern boundaries, and regions of mountains and desert in southern California. A partial summary of the in-situ Ge(Li) measurements are given in Table II. It is of interest to note the variation of natural and man-made radionuclide levels throughout the state. The lowest levels of natural radionuclides (U-Th series, ^{40}K) were measured in the coastal area of Hayward, while the highest levels were seen in the central valley near Fresno. External exposure rates from the gamma-radiation associated with the natural radionuclides varied by a factor of 2.4 between the two sites. External gamma-ray exposure rates calculated from field spectrometric measurements were 10.7 $\mu\text{R}/\text{Hr}$ for Fresno and 4.5 $\mu\text{R}/\text{Hr}$ at Hayward.

At all sites, the man-made radionuclide ^{137}Cs was detected. The highest levels were seen at the Tehachapi Summit and in the northern coastal regions. The calculated maximum external gamma-ray exposure rate from ^{137}Cs was determined to be approximately 0.3 $\mu\text{R}/\text{Hr}$. Probably the most significant point of interest is the demonstration of extremely good sensitivity for detecting trace levels of recent fallout radionuclides. This was demonstrated by the detection of $^{95}\text{Zr}/^{95}\text{Nb}$ in Pleasanton, California, following an atmospheric nuclear weapons test conducted by the Chinese in March, 1972. The in-situ measurement followed approximately two months after the Chinese Test. Levels for ^{95}Zr were 1.6 nCi/m^2 and 1.8 nCi/m^2 for ^{95}Nb .

Geo-Physical Studies

In the case where the redistribution of radionuclides is related to a geo-physical phenomena, i.e., transport by rain or stream run-off and accumulation downstream, in-situ gamma-ray spectrometric techniques may be useful in plotting the movement and build-up of radioactivity. An area in the Amargosa Desert west of the Nevada Test Site was selected to investigate if a build-up of ^{137}Cs was taking place. Measurements were made in the Forty Mile Canyon wash area (near Hwy. 95) intermittently fed from rain run-off during seasonal storms. Measurements were also made north and south of the Canyon-wash. Results are given in Table III. Natural radionuclides were found to be of uniform concentration however the results suggested a definite build-up of ^{137}Cs in the wash terminus.

Measurement of Gaseous Emissions

In-situ detection of gaseous radionuclides is possible with the Ge(Li) field spectrometer. This is demonstrated in Fig. 5. The photopeak identified as ^{41}Ar at 1293 keV was a result of off-gassing of the LLL pool type reactor during its normal operation.

The in-situ Ge(Li) spectroscopy technique has also previously been used by us at the site of BMR type reactor.⁴ We could clearly identify the rare gases ^{135}Xe , ^{138}Xe , ^{135}Kr and ^{85}Kr all present in the plume.

It is suggested that external exposure rate contributions from low-levels of gaseous radionuclides may be determined from the combined use of a Ge(Li) detector and a sensitive high pressure ionization chamber. This may be accomplished by performing terrestrial gamma-ray measurements during the absence of gaseous radionuclides and calculating the exposure rates resulting from terrestrial U-Th, ^{40}K and fallout radionuclides. This is done by analysis of the photo-peaks present in the Ge(Li) spectrum.^{6,7} The combined cosmic and terrestrial external exposure rate are determined by the ion-chamber. This establishes an exact "signature" of the radionuclides and corresponding dose rates. In the case where gaseous radionuclides are present, and the previous measurements have been performed, the external exposure rate contribution from the gases would be done by subtracting the terrestrial and cosmic contributions previously determined. The Ge(Li) detector measurement would verify the presence of gaseous radionuclides. This is necessary since the ion-chamber measures total gamma flux only. The relationship between exposure rate fluctuations and variations in the terrestrial cosmic radiation or the gaseous source could also be determined with the Ge(Li) detector - ion chamber combination. We are presently conducting a series of experiments to fully evaluate the technique.

Reactor Liquid Waste Discharge

Calibration of the Ge(Li) detector was done for the measurement of terrestrial gamma radiation. However, we have on a demonstration basis used the system to study the feasibility of measuring gamma-ray emitting radionuclides in liquid effluents. We did this recently at the Humboldt Bay nuclear reactor located on the coast of Northern California. An on-line measurement of the discharge canal water was made during a planned rad-waste discharge. These planned discharges are carried out in such a way that accumulated liquid wastes contained in a few hundred gallons of water are mixed over a period of hours into the main reactor cooling water flow of 100,000 gallons per minute. This mixture of waste radionuclides and cooling water exit to a discharge canal and subsequently to the ocean.

The in-situ measurement was made by continuously pumping the water out of the canal, then flowing the water into a 100 liter container, where the Ge(Li) detector was located, and discharging water out of the container to a point downstream. The experimental layout is shown in Fig. 7. Counting times at the beginning of the experiment were 1000 seconds. This was extended to 4000 seconds during the latter part of the experiment. Prior to diluting the rad-waste with the cooling water, an aliquot was taken to determine the concentration of radionuclides. An estimation of concentration was then made for the discharge canal. These values are given in Table IV.

Counting was performed over a 14 hour period. Results clearly showed that the Ge(Li) detector, as used in this application had excellent sensitivity for ^{134}Cs and ^{137}Cs . The time history for the ^{137}Cs is plotted in Fig. 8. Maximum observed ^{137}Cs was 35 ± 3 cpm. Background level (^{137}Cs) for the discharge canal water was 2 ± 0.5 cpm.

Detection of ^{54}Mn , ^{60}Co and ^{65}Zn was more difficult at the levels shown in Table IV. Sporadic detec-

tion of these radionuclides was noted throughout the rad-waste release, however a reasonable concentration/time history of the ^{65}Zn and ^{60}Co could be reconstructed. The ^{54}Mn was observed four out of eighteen counting periods.

Acknowledgments

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Table I. Characteristics of two Ge(Li) detectors used for in-situ measurements.

	<u>Closed-end Coaxial</u>	
	V - 1	V - 3
Approx. Active Volume	70 cm ³	70 cm ³
diameter	40 mm	44 mm
cross section	14.5 cm ²	15 cm ²
length	68 mm	55 mm
Eff (662 kev)	1.4 $\frac{\text{counts}}{\gamma\text{-cm}^{-2}}$	2.9 $\frac{\text{counts}}{\gamma\text{-cm}^{-2}}$
Resolution, FWHM @ 1.33 MeV	3.1 KeV	3.0 KeV

Table III. Results of a Survey made over a Wide geographical area. The radionuclides $^{95}\text{Zr}/^{95}\text{Nb}$ are believed to be from a nuclear weapons test conducted by the Chinese, March 1972, (^{137}Cs , $\alpha = -0.16 \text{ cm}^{-1}$).

LOCATION	RADIONUCLIDE CONCENTRATION		LOCATION	RADIONUCLIDE CONCENTRATION	
	natural (n Ci/kg)			natural (n Ci/kg)	
Tehachapi	^{40}K	21.6		^{40}K	18.4
Summit	^{238}U	1.0	Manteca	^{238}U	0.9
Calif.	^{232}Th	1.3	Calif.	^{232}Th	1.1
3/25/72	man-made (n Ci/m ²)		3/24/72	man-made (n Ci/m ²)	
	^{137}Cs	95.5		^{95}Zr	3.3
				^{137}Cs	21.7
	natural (n Ci/kg)			natural (n Ci/kg)	
Barstow	^{40}K	22.1		^{40}K	24.7
Calif.	^{238}U	0.9	Fresno	^{238}U	1.2
3/25/72	^{232}Th	1.2	Calif.	^{232}Th	1.4
	man-made (n Ci/m ²)		3/24/72	man-made (n Ci/m ²)	
	^{137}Cs	66.0		^{95}Zr	3.5
				^{137}Cs	34.3
	natural (n Ci/kg)			natural (n Ci/kg)	
Hayward	^{40}K	11.0		^{40}K	16.9
Calif.	^{238}U	0.5	Bakersfield	^{238}U	1.2
3/29/72	^{232}Th	0.6	Calif.	^{232}Th	0.9
	man-made (n Ci/m ²)		3/25/72	man-made (n Ci/m ²)	
	^{95}Zr	1.1		^{137}Cs	22.8
	^{137}Cs	78.2			
	natural (n Ci/kg)			man-made (n Ci/m ²)	
Pleasanton	^{40}K	12.1	Pleasanton	^{95}Zr	1.6

Democracy	40_k	21.6		40_k	18.4
Summit	^{238}U	1.0	Manteca	^{238}U	0.9
Calif.	^{232}Th	1.3	Calif.	^{232}Th	1.1
3/25/72	man-made (n Ci/m ²)		3/24/72	man-made (n Ci/m ²)	
	^{137}Cs	95.5		^{95}Zr	3.3
				^{137}Cs	21.7
	natural (n Ci/kg)			natural (n Ci/kg)	
Barstow	40_k	22.1		40_k	24.7
Calif.	^{238}U	0.9	Fresno	^{238}U	1.2
3/25/72	^{232}Th	1.2	Calif.	^{232}Th	1.4
	man-made (n Ci/m ²)		3/24/72	man-made (n Ci/m ²)	
	^{137}Cs	66.0		^{95}Zr	3.5
				^{137}Cs	34.3
	natural (n Ci/kg)			natural (n Ci/kg)	
Hayward	40_k	11.0		40_k	16.9
Calif.	^{238}U	0.5		^{238}U	1.2
3/29/72	^{232}Th	0.6	Bakersfield	^{232}Th	0.9
	man-made (n Ci/m ²)		Calif.	man-made (n Ci/m ²)	
	^{95}Zr	1.1	3/25/72	^{137}Cs	22.8
	^{137}Cs	78.2			
	natural (n Ci/kg)			man-made (n Ci/m ²)	
Pleasanton	40_k	13.1	Pleasanton	^{95}Zr	1.6
Calif.	^{238}U	0.6	Calif. (cont.)	^{95}Nb	1.8
5/16/72	^{232}Th	0.7	5/16/72	^{137}Cs	96.1

LOCATION	^{137}Cs (nCi/m ²)	^{40}K (nCi/kg of soil)	ION CHAMBER (μR/Hr)
33n, south of dry wash	$60.2 \pm 11\%$	$30.8 \pm 2\%$	17.7
37s, north of dry wash	$49.2 \pm 13\%$	$29.8 \pm 2\%$	16.9
36s, north of dry wash	$54.0 \pm 13\%$	$31.6 \pm 2\%$	16.8
35, north of dry wash	$50.8 \pm 13\%$	$29.4 \pm 2\%$	17.0
Dry wash area	$105.2 \pm 7\%$	$29.1 \pm 2\%$	17.6

Table III. Results of in-situ measurements in the vicinity of a desert dry wash area and within the dry wash. The build-up of ^{137}Cs in the dry wash may be related to run-off from the surrounding mountains. It was assumed that ^{137}Cs had the same vertical distribution at all locations. Most of the external gamma exposure was from the natural emitters, ^{238}U , ^{232}Th , and ^{40}K .

Radionuclide	*Estimated Concentration (pCi/l)
^{54}Mn	5.0
^{60}Co	13.3
^{65}Zn	9.4
^{134}Cs	268
^{137}Cs	437

Table IV. Estimated concentration of radionuclides in discharge canal water during rad-waste release. *Estimates of concentrations are based on measured rad-waste values divided by cooling water through-put.

Figure Legends

Fig. 1 Cryostat and mounting details for two coaxial Ge(Li) detectors used in in-situ spectroscopy. The detector on the left is V-3 and on the right is V-1. Note the detector holder on V-3 is designed to minimize attenuation of gamma-rays entering through the side walls.

Fig. 2 Detector efficiencies for two Ge(Li) spectrometers used for in-situ counting.

Fig. 3 The field spectrometric system. The Ge(Li) detector, Fidler and ion-chamber are connected to the recording instruments by 30 meter long cables. Recording of background gamma radiation is accomplished by the 2" x 2" NaI(Tl) scintillator mounted on the inside wall of the vehicle.

Fig. 4 A van-type vehicle houses the complete field spectrometric system. Shown in the foreground is the tripod mounted Ge(Li) detector set up for a field measurement. Normally, the detector sets approximately 25-30 m away from the van during a measurement.

Fig. 5 Gamma-ray spectrum generated from a 180 minute in-situ measurement. Ge(Li) detector V-1 (see Table I) was used. The spectrum contains photopeaks from the natural occurring U-Th, 40K, global fallout ^{137}Cs and from ^{41}Ar , a gaseous by-product of a pool type reactor. See text for concentration values.

Fig. 6 This gamma-ray spectrum was recorded at the Nevada Test Site. The area is near the Sedan Crater. Counting time was 2000 sec. After Anspaugh, Ref. 3.

Fig. 7 Technique for measuring radionuclides in liquid effluent discharged from a BWR power plant. This was done during a planned rad-waste release.

Fig. 8 A time history of ^{137}Cs present in the discharge canal. The flat bars of the graph represent counting times. The measurement was performed as shown in Fig. 7.

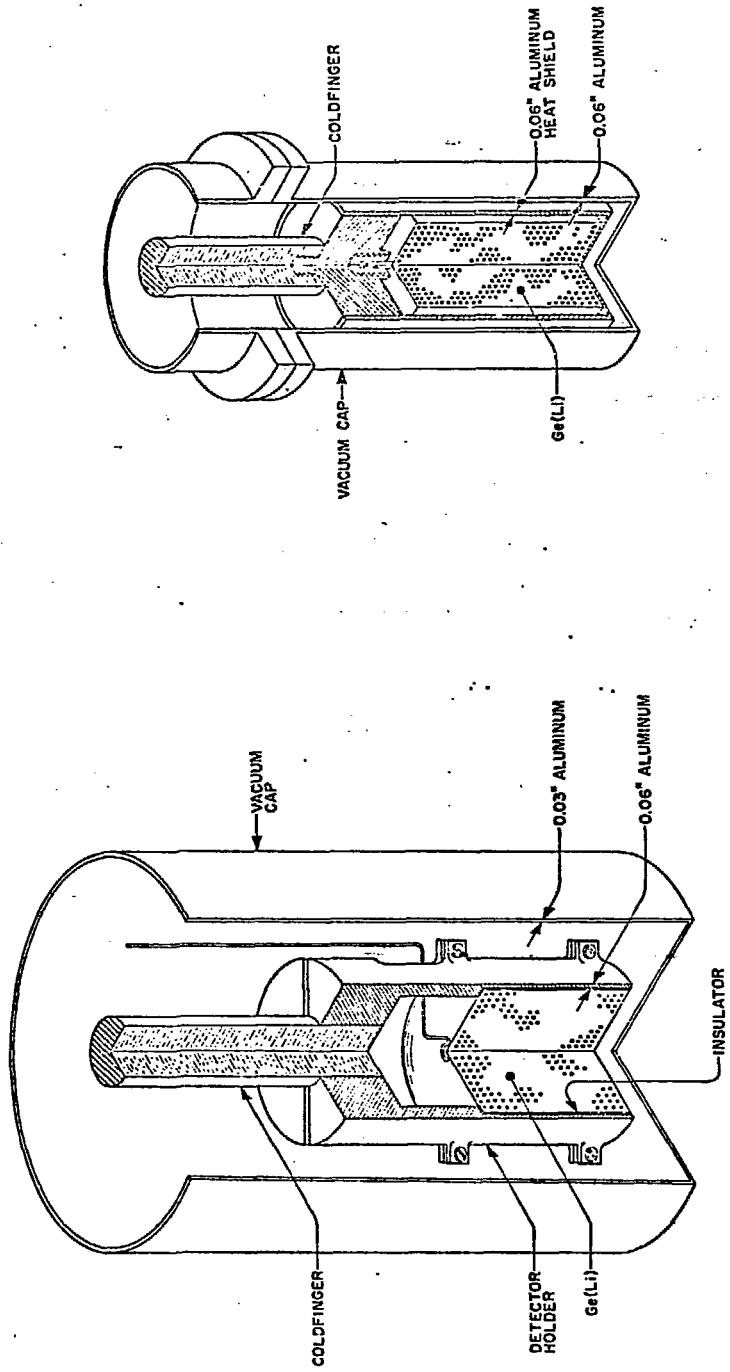


Fig. 1
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et al.

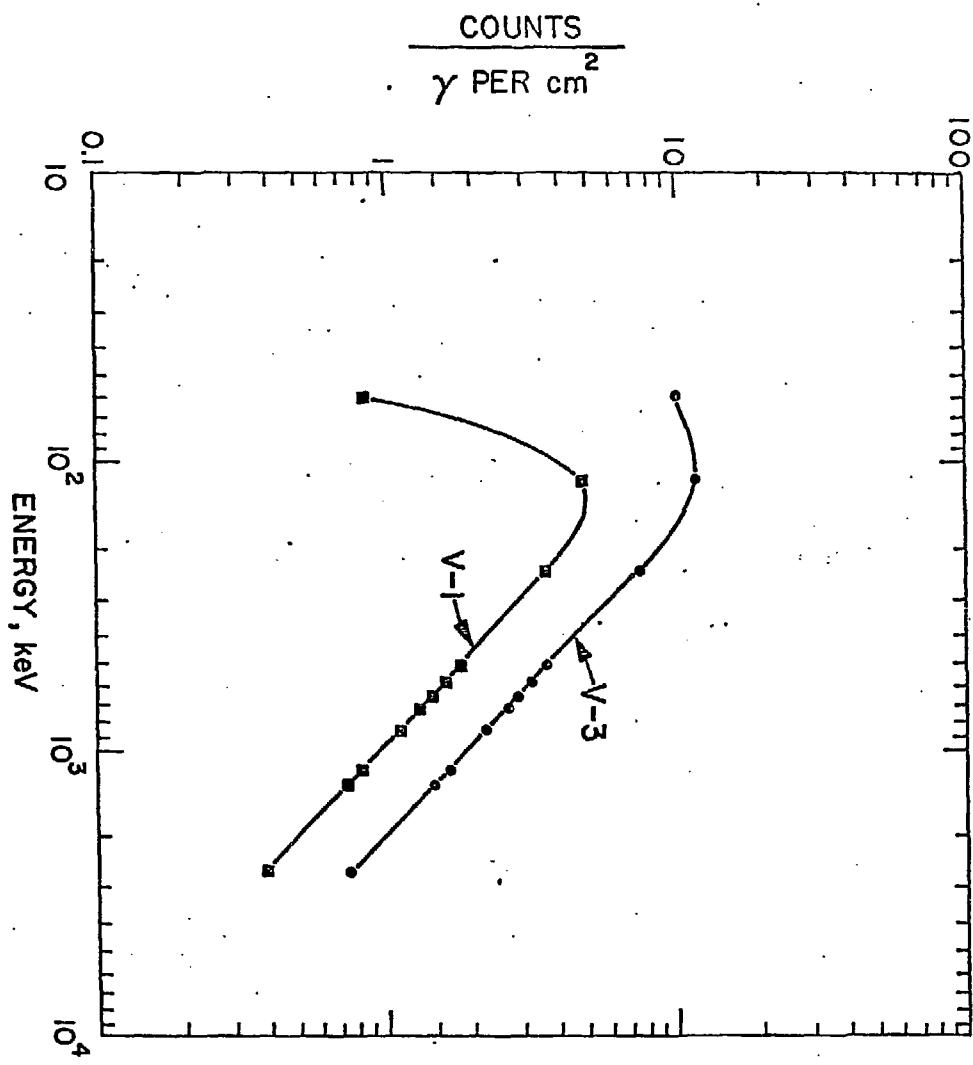


Fig. 2
P. Phelps,
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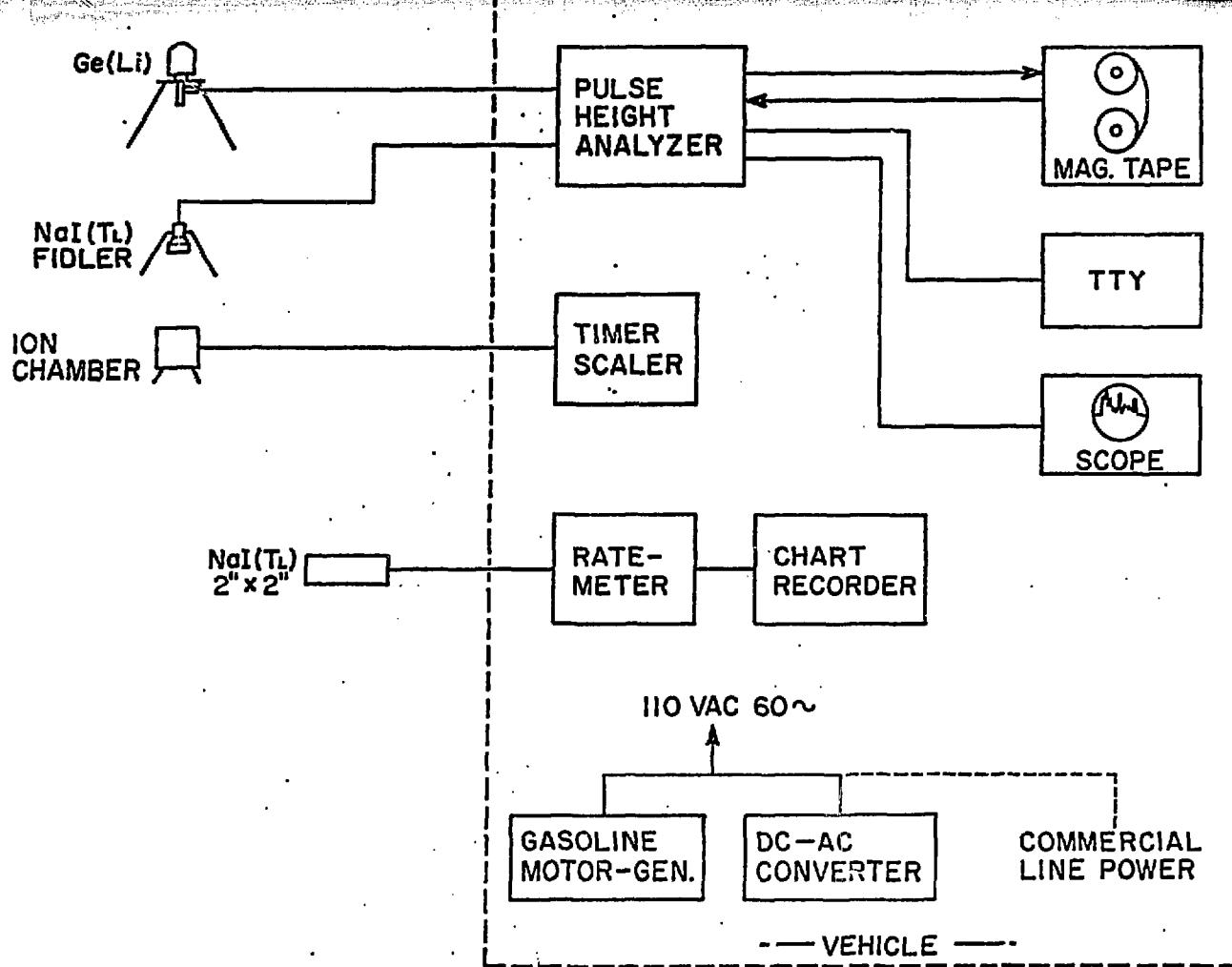


Fig. 3
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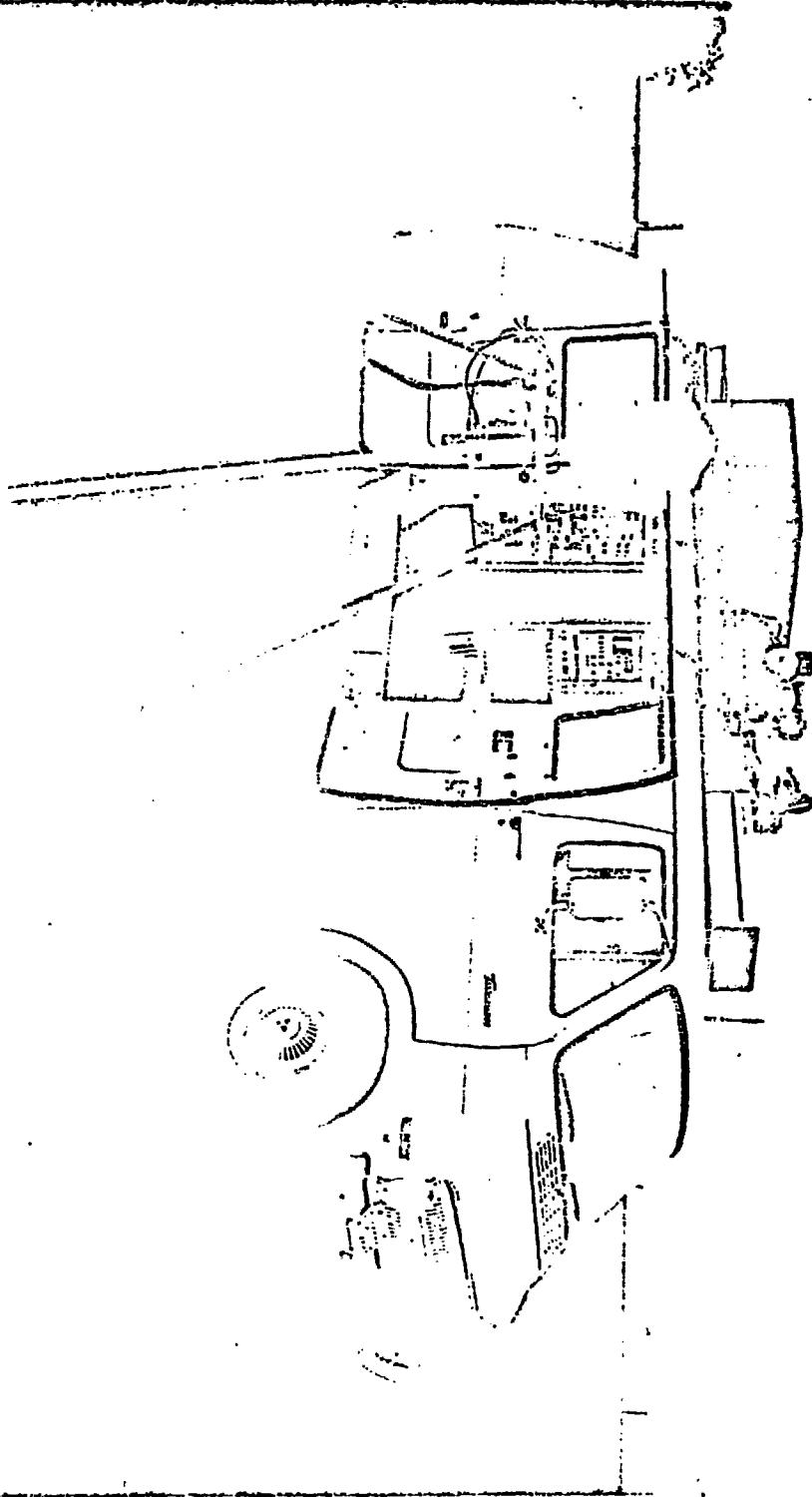


Fig. 4
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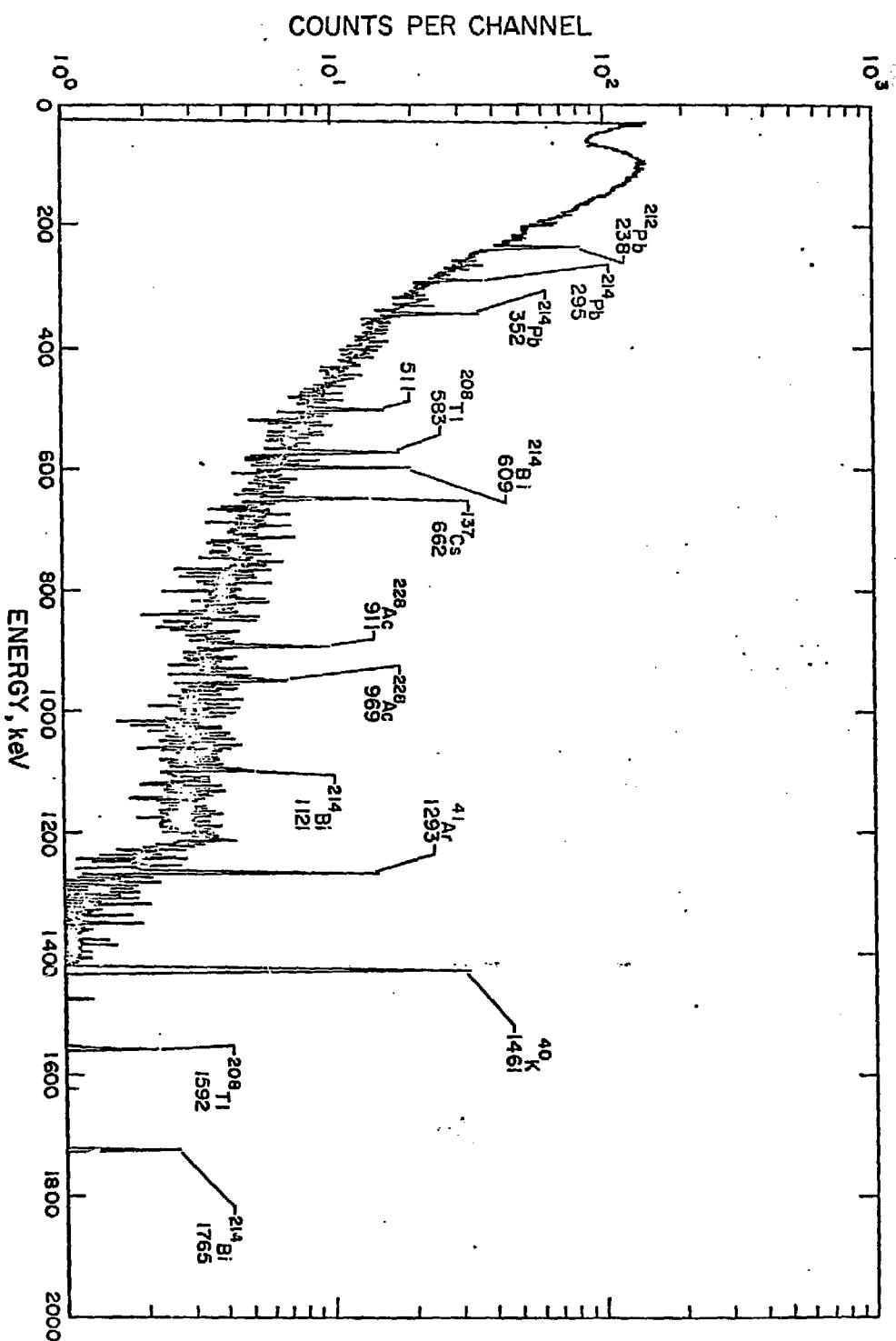


Fig. 5
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et al.

COUNTS / CHANNEL

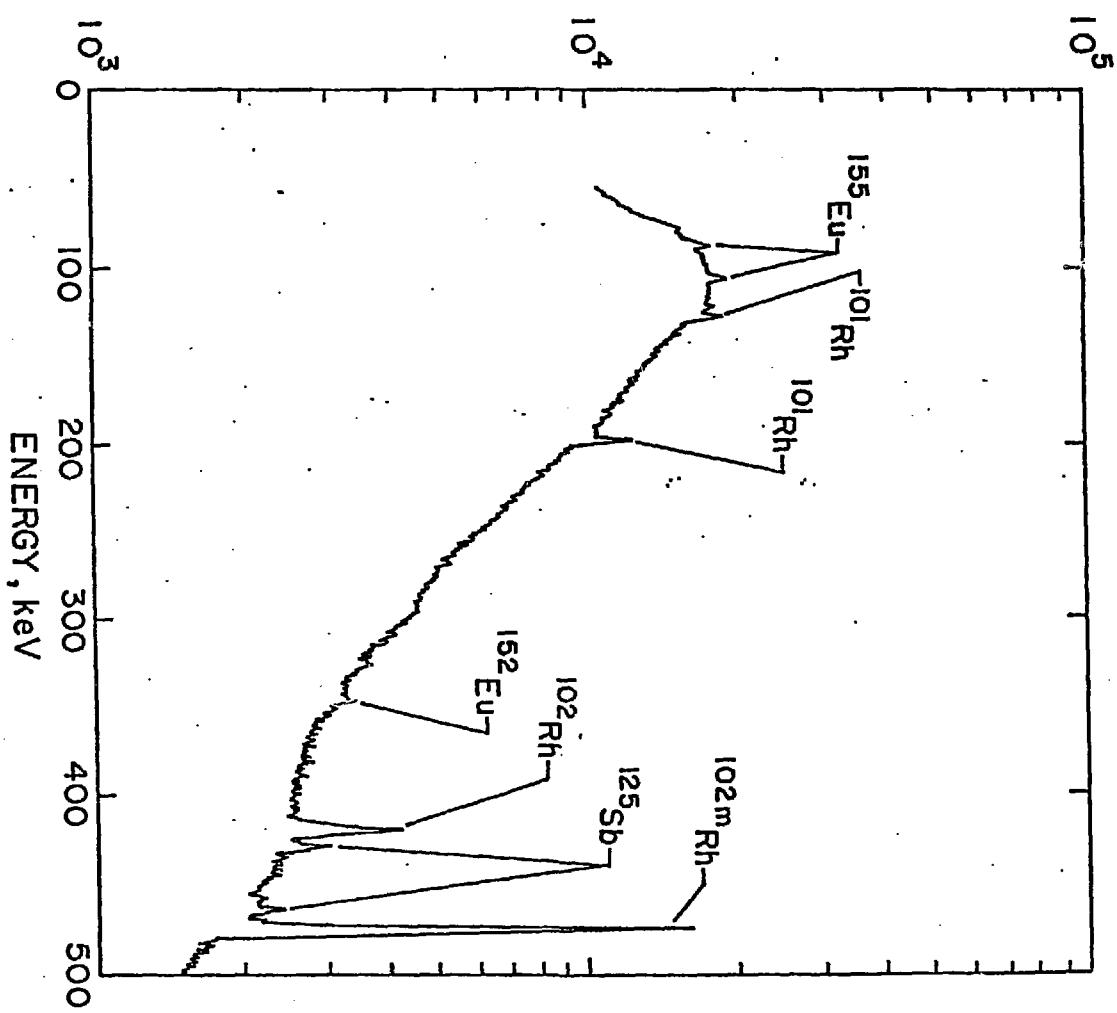
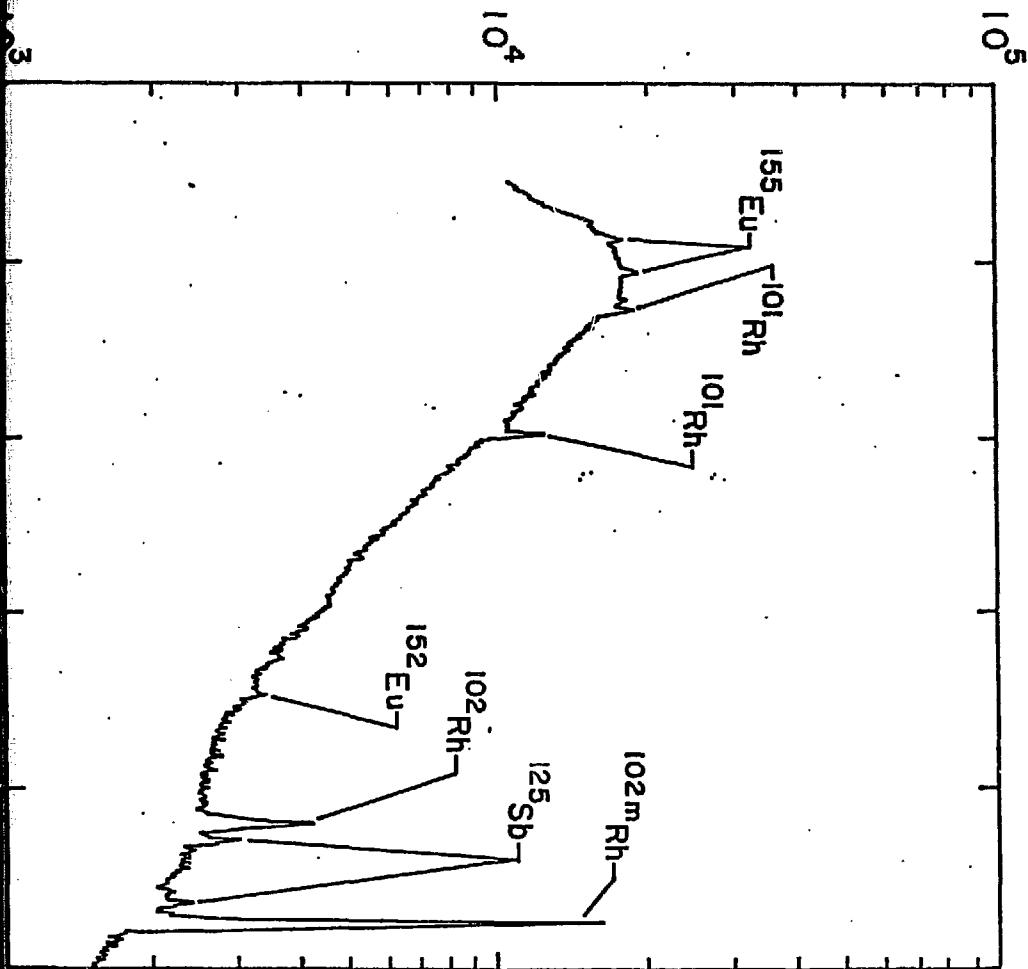


Fig. 6a
P. Phelps,
et al.

COUNTS / CHANNEL



2

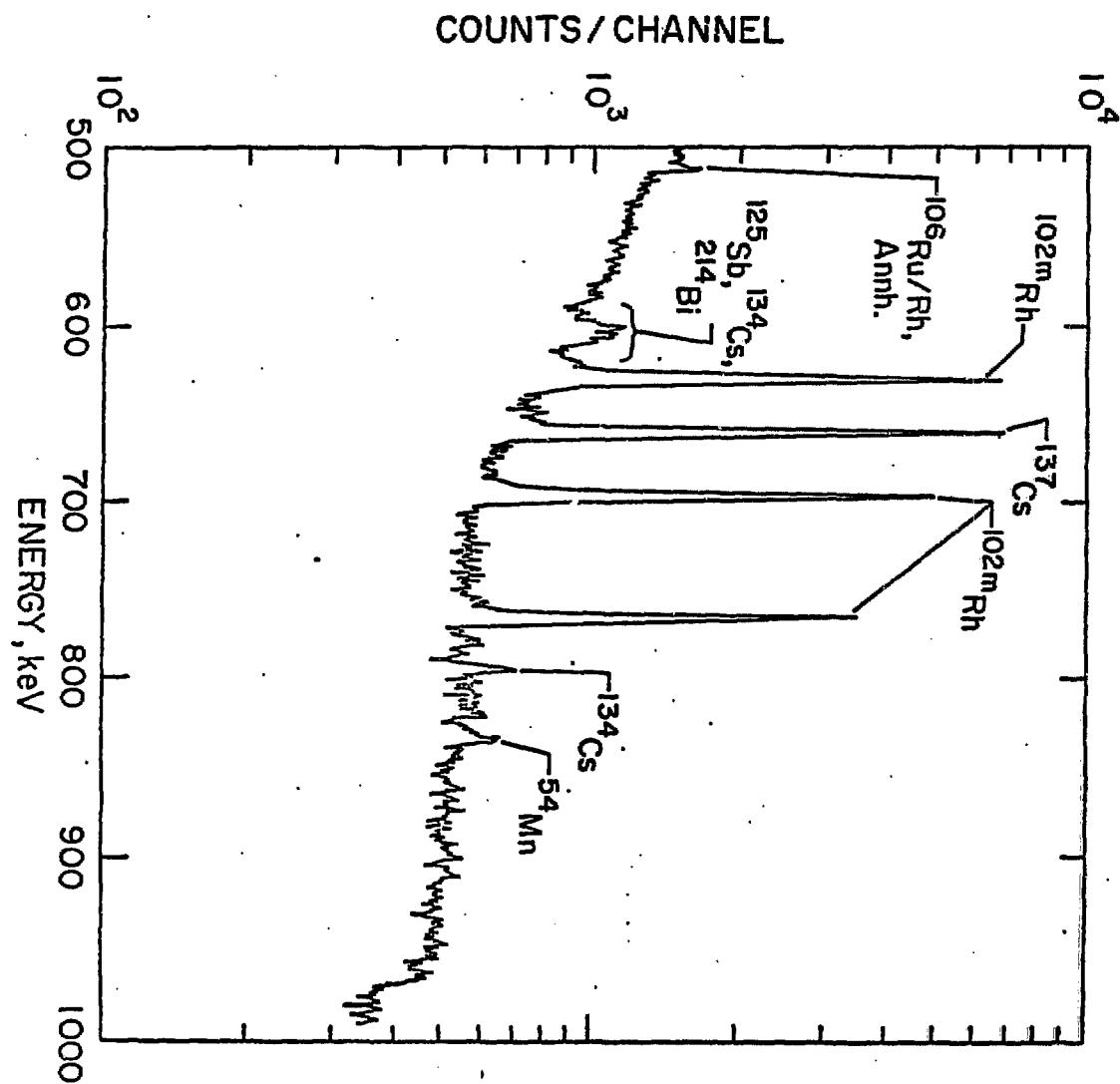


Fig. 6b
P. Phelps,
et al.

COUNTS/CHANNEL

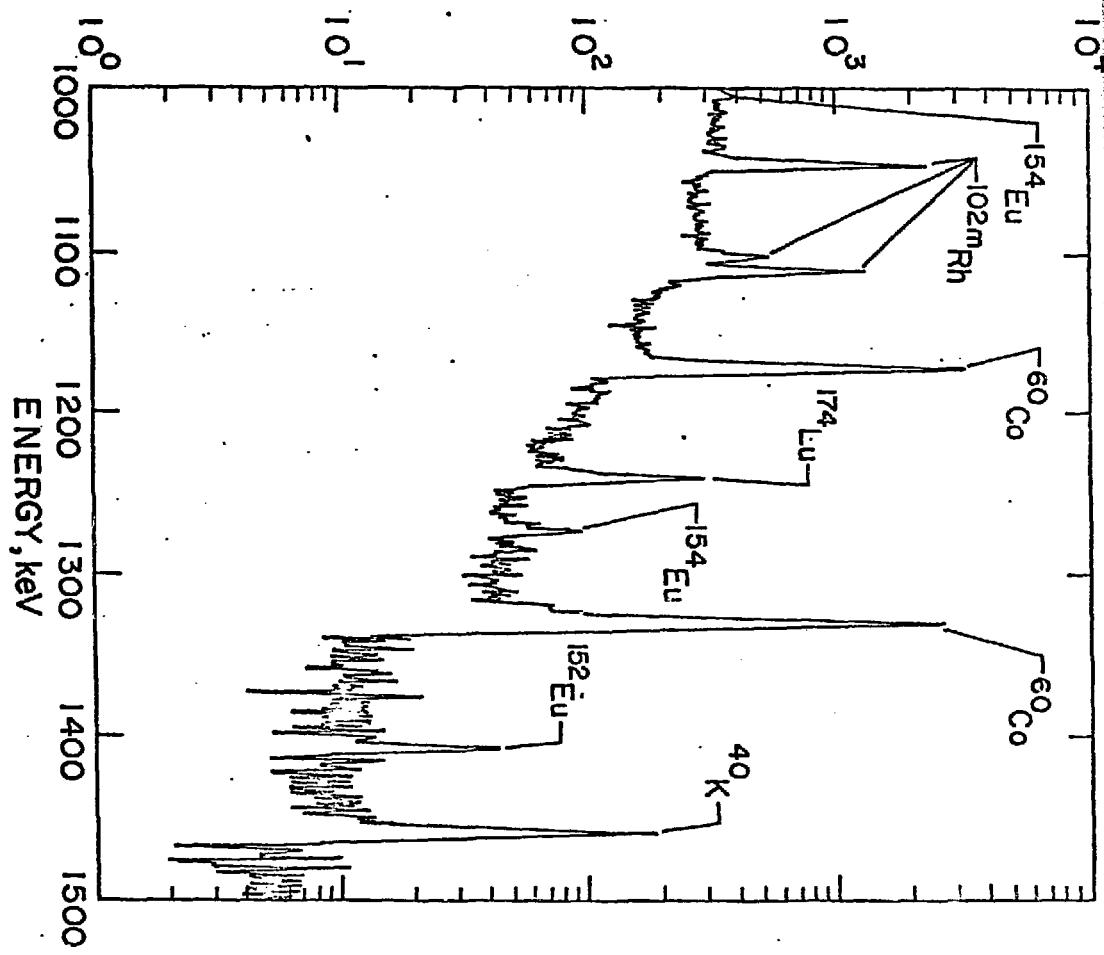
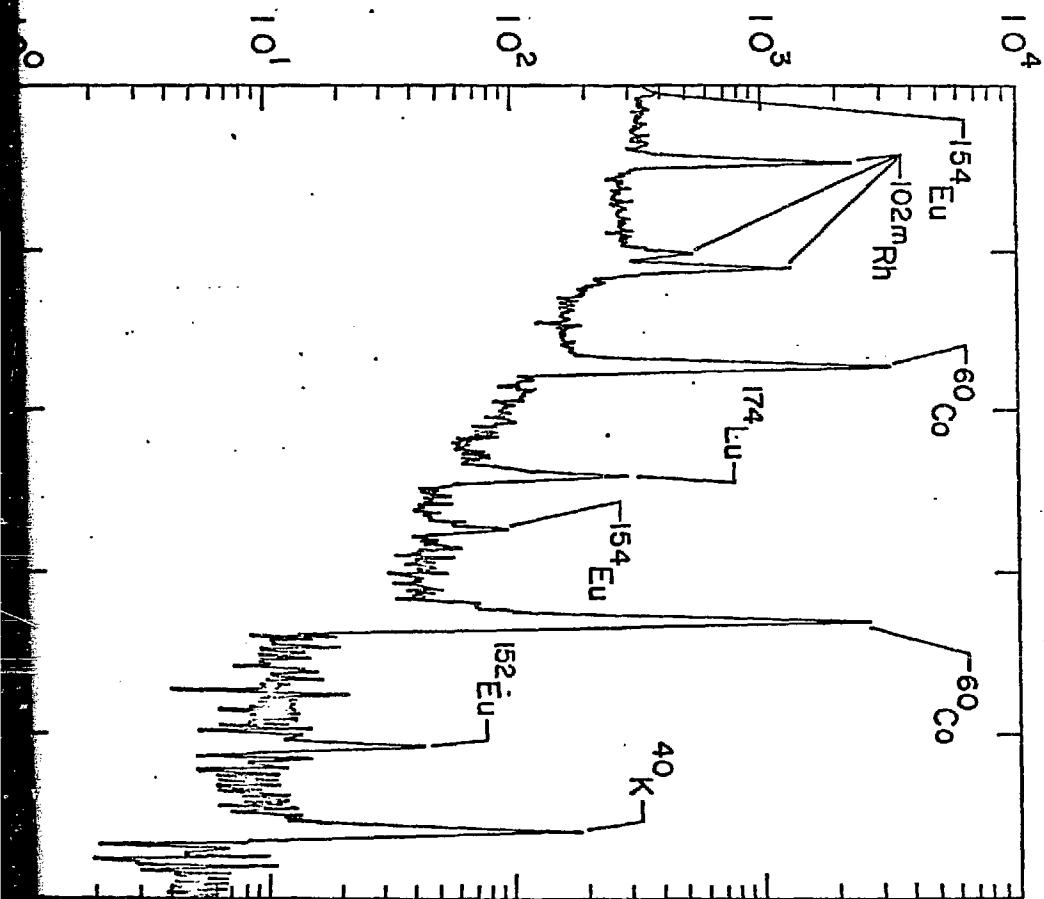


Fig. 6c
P. Phelps,
et al.

COUNTS/CHANNEL



2

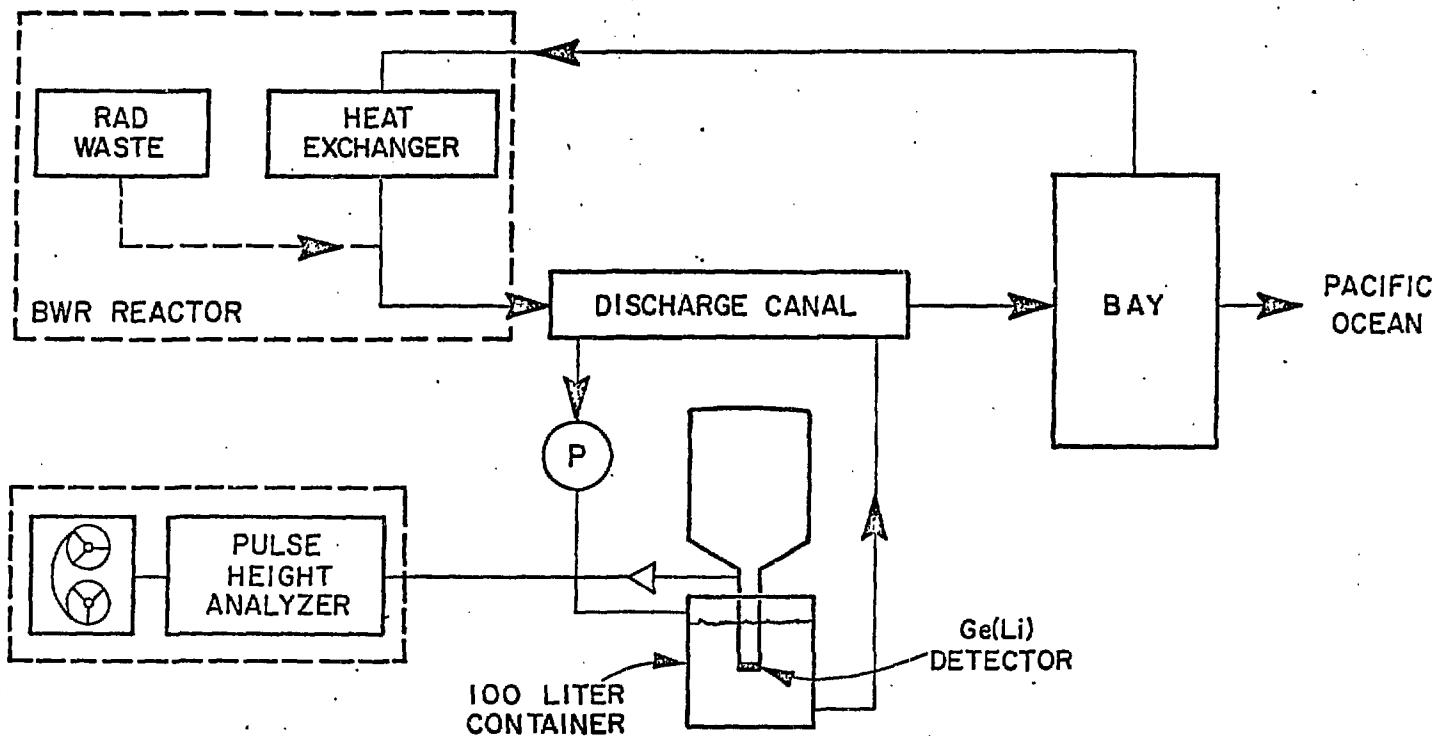


Fig. 7
 P. Phelps,
 et al.

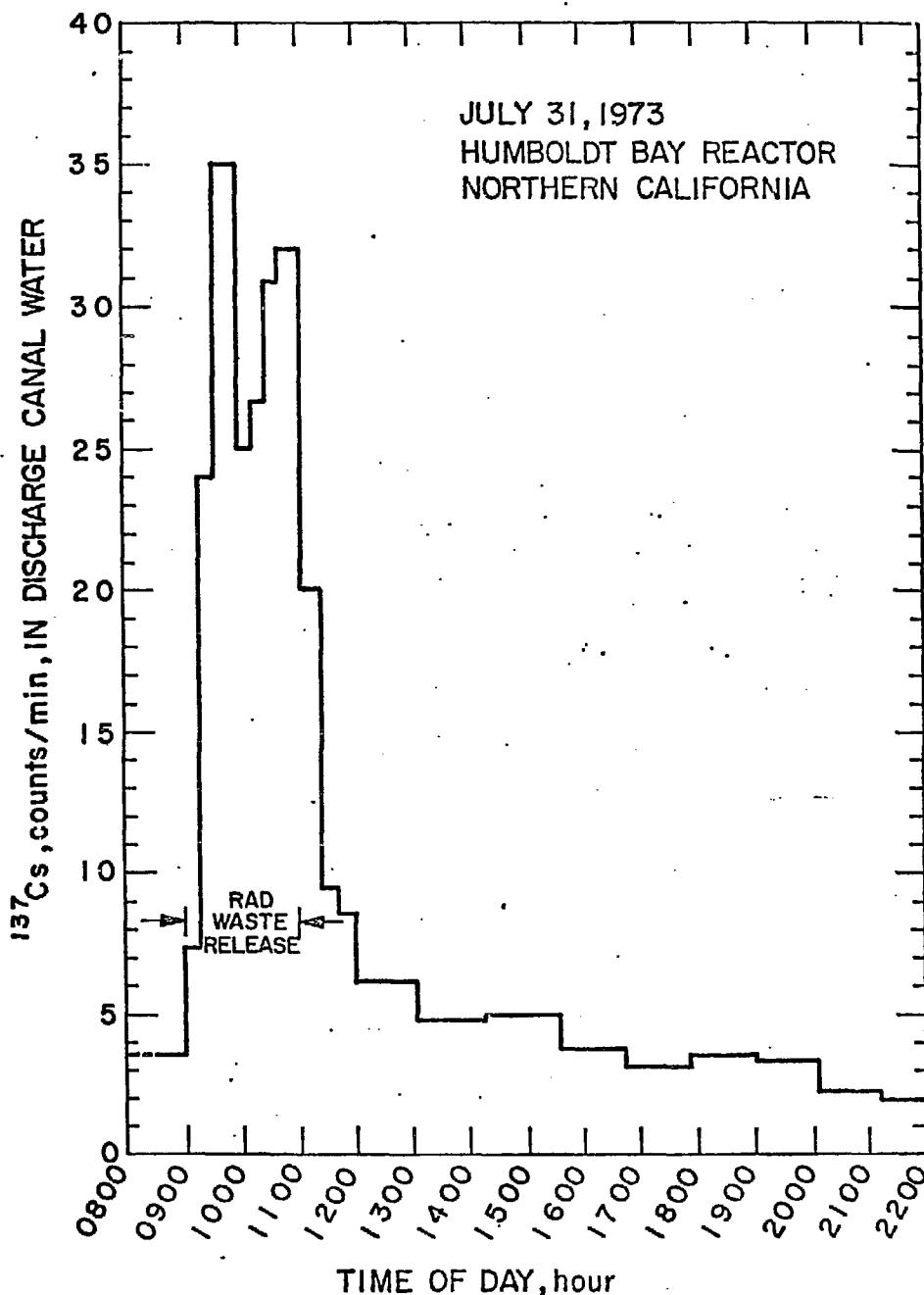


Fig. 8
P. Phelps.