REPORT

LOW ENERGY ACTIVATION ANALYSIS
for
GEOCHEMICAL EXPLORATION

Submitted By

Howard University, Washington, D.C. 20001

to

National Aeronautics and Space Administration

Washington, D.C. 20546

Dr. George A. Ferguson, Principal Investigator

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INTRODUCTION

The National Aeronautics and Space Administration has demonstrated a continued interest in techniques suitable for geochemical exploration of lunar and planetary surfaces.(1) Diagnostic techniques to assist the astronaut in sample selection or "insitu" analysis have been the subject of intensive study in recent years.(2) One such technique is the artificial excitation of the specimen and its identification by the measurement of the radiation spectrum produced by its subsequent characteristic de-excitation. This method is popularly called activation analysis. The artificial excitation may be achieved by a variety of means, for example, by gamma, beta, alpha, neutron or Bremstrahlung sources. The major problem in the identification of samples by activation analysis is the determination of the radiation spectrum incident upon the detector by an analysis of its experimentally measured response.

Three basic types of detectors have been used in the measurement of radiation. They are gas-filled counters, scintillation counters and solid-state counters. When radiation counters are employed during the explorations alluded to above, their characteristics should include ruggedness, high efficiency and good resolution. No one of the three basic detector types optimizes all of these desirable characteristics. Gas-filled counters may be made rugged but are slow in their response, thereby rendering them relatively inefficient. The solid-state counters exhibit excellent resolution, but in its present state of development it is cumbersome and also relatively inefficient.

The scintillation counter offers a satisfactory compromise of the required characteristics of detectors for exploration. For this reason, this investigation will concentrate upon the response of scintillation counters.

Identification of unknown samples by activation analysis may be achieved either by a direct comparison of the spectrum of the unknown with that obtained from an identical sample of known composition, or the unknown spectrum may be decomposed into its monoelemental components. While the former technique has some application, the later offers a more fundamental approach to the problem of identification. The process of decomposing a complex radiation spectrum into its monoelemental components, may be accomplished by fitting the upper end of the observed spectrum with the appropriate distribution function of a monoenergetic type of radiation (say, monoenergetic gamma rays). This specific spectrum, including its Compton tail (produced by scattering within the scintillator), is then unfolded. The remaining spectrum is operated on in the same manner until the original complex spectrum is completely analyzed.

To accomplish the unfolding, one must have available the pulse height spectrum produced by a monoenergetic gamma ray over the range of interest, in the same scintillation detection system as used to record the spectrum being analyzed. Modern digital computers may then be employed in the analysis. The procedure utilizes a large matrix whose elements describe the response of the detector to monoenergetic gammas. The unfolding of the observed spectrum, to obtain the incident spectrum, requires operations with the inverse of this matrix. Problems are encountered in performing this inversion due to the statistical variance in detector response which forms the elements of the matrix.(3) More importantly, this method of analysis is limited by the available known monoenergetic pulse height spectra. The complete success of this approach requires a complete library of carefully measured spectra which covers the entire energy range of interest.

Pulse height spectra of a large number of gamma ray sources have been tabulated (4), however, these spectra have generally involved radiation sources of high energy. There exists a scarcity of such information at energies below a few hundred kilovolts although many important and useful sources emit radiation with energies in this region. One of the purposes of this study is to investigate the response of a selected scintillation detector to incident monochromatic radiation with energies below one-hundred kilovolts (100 kv).

DESCRIPTION OF RESEARCH.

(a). Review of Techniques:

A review of existing techniques of the measurement of complex radiation spectra was conducted. From this review, and the helpful preliminary discussions of the proposed research program with Dr. S. Singh (Howard University), Dr. R. Deslattes (National Bureau of Standards), Dr. W. Johns (Lawrence Radiation Laboratory) and Prof. D. A. Lind (University of Colorado) it was decided that the technique of bent crystal spectrometry (DuMond) would be employed for monochromatization of the radiation source.

This source, for low energies (below 100 kv), was a modified Flaxitron X-Ray System (Model 8050-311) purchased for this purpose. (For energies above 100 kv in future studies, the use of the University Cockcroft-Walton accelerator is anticipated.)

(b). Production of Radiation:

The Flaxitron X-Ray System (Model 8050-311) was used as a source of variable energy radiation. This self rectified system operates on a continuous duty cycle with a variable output voltage within the range 10-110 kv. The 0.5 mm emission source emits radiation through a 0.025" beryllium window which is directed through a two meter long lead tunnel onto a single crystal of germanium serving as a monochromator.

(c). Monochromatization:

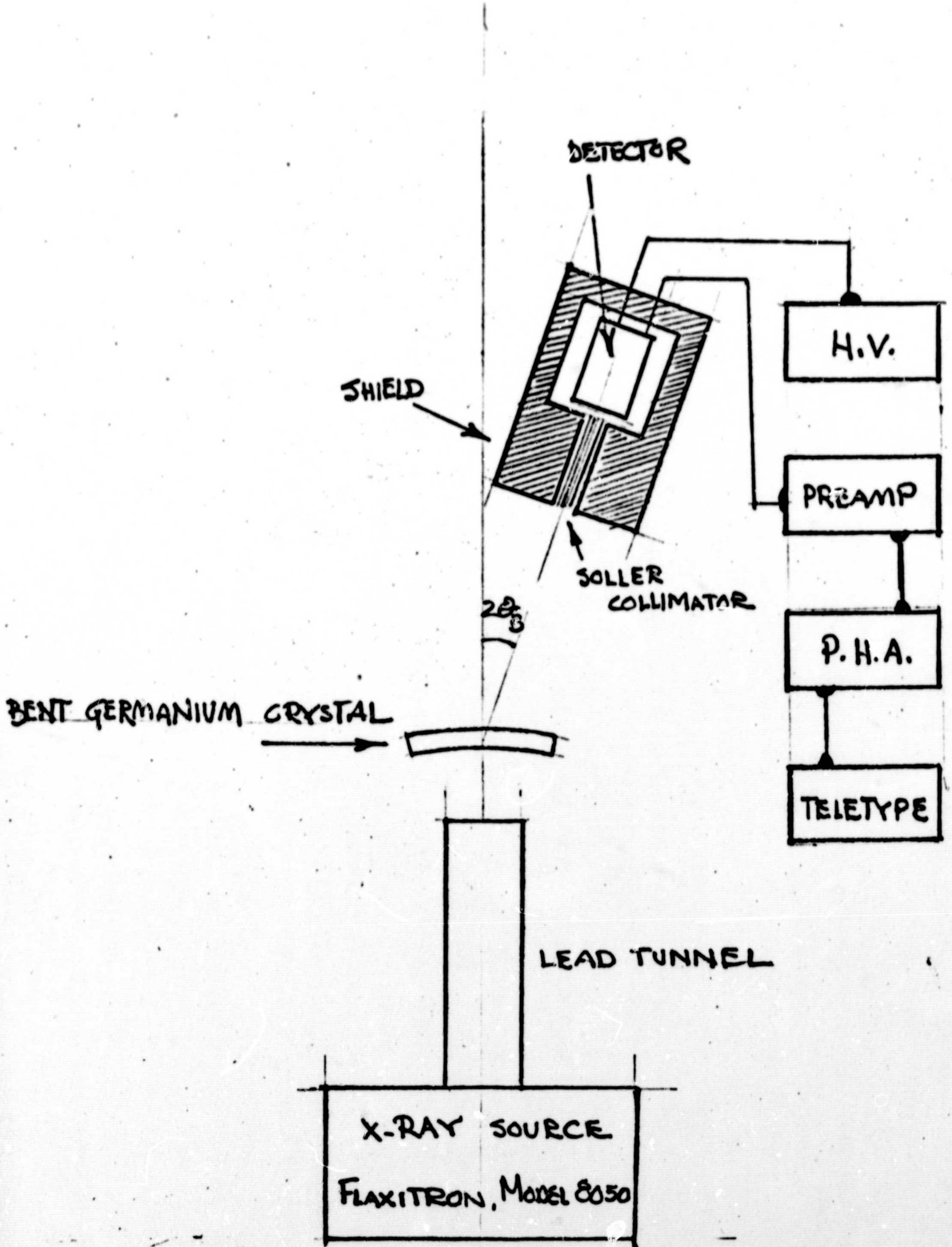
Descriptions of the interaction of gamma rays with matter, as well as the dynamical theory of diffraction of gamma rays by a crystal, have been extensively discussed [5]. It is known [6] that only for the incident angle, Θ_B, and spacing, d, of a given set of crystal planes, will coherent diffraction of wavelength λ be observed according to $n\lambda = 2d \sin \Theta_B$. Now when a crystal slab is elastically bent to a cylindrical surface such that the diffracting planes are parallel to and intersect the cylinder axis an increase diffracted intensity is observed [7]. DuMond [8] described and constructed such a bent crystal spectrometer. The essential features of the arrangement of source, crystal and detector used by DuMond are given in Figure 1. The DuMond geometry has been employed to construct the bent crystal spectrometer used for monochromatizing the x-ray source in this study. A single crystal of germanium was used as a monochromator.

(d). Detection:

The monochromatic radiation diffracted by the germanium crystal was directed onto the scintillation detector under study after passage through a sixteen inch long Soller slit collimator (see Figure 1). Collimation of the diffracted intensity was required to accurately define the scattering angle (and hence the energy of the diffracted beam) as well as to reduce background radiation resulting from incoherent scattering. The scintillation detector used for this study was a ruggedized "INTEGRAL LINE" Assembly, Type 12SPF12-x, serial # DQ-376 supplied by the Goddard Space Flight Center. The assembly consisted of a 3-inch NaI (Tl) scintillation crystal optically coupled to a 10-stage head-on type photomultiplier (RCA # C31009E)

(e). Recording:

Pulse height spectra were collected using a 512-channel analyzer (Kicksort Model 701) and stored on paper tape by memory read-out through a Model 33TZ Teletype. These data are available for future analysis.



BENT CRYSTAL SPECTROMETER

FIGURE 1

EXPERIMENTAL RESULTS:

Preliminary pulse height spectra have been obtained using radio-isotopic calibration sources and monochromatic radiation produced by the bent crystal spectrometer.

(a). Cs^{137} pulse height spectra.

Figure 2 is the pulse height spectrum obtained from a 0.5 microcurie Cs^{137} source. The identifying features of this spectrum have been labeled. The measured resolution of the 662 kev photopeak is 12% (which compares favorably with the value 8.8% measured when the scintillator was originally tested at Harshaw Chemical Company prior to shipment). The measured resolution of the 32% kev barium x-ray produced by this source is 60%.

(b). Ba^{133} pulse height spectra.

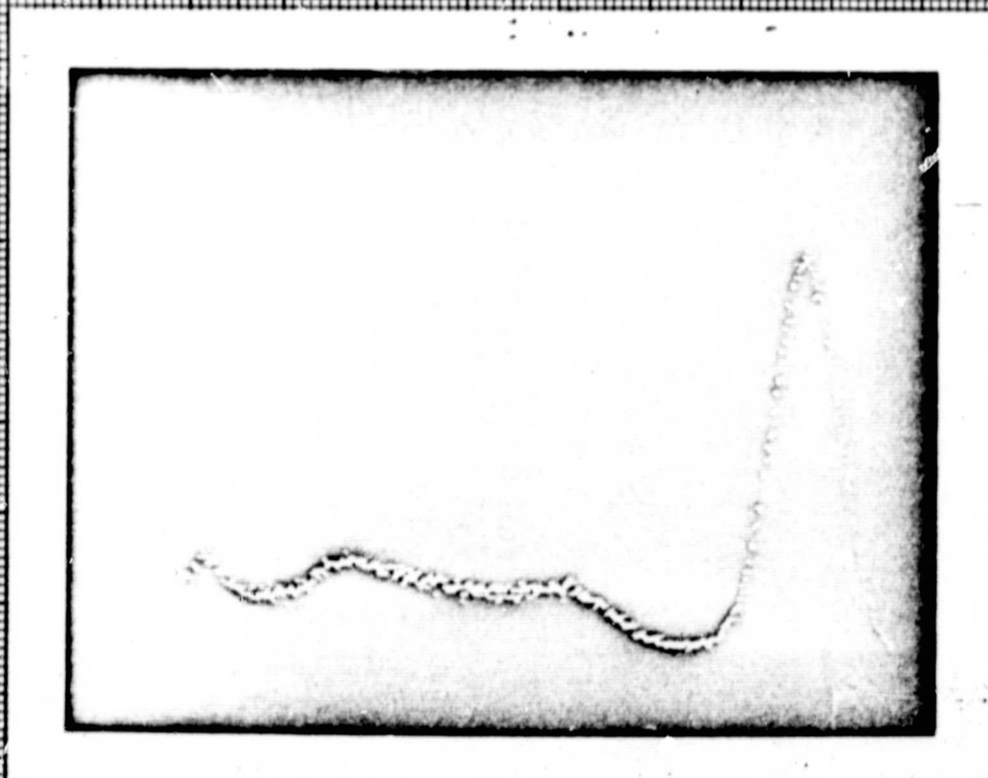
Figure is the pulse height spectrum obtained from a 0.5 microcurie Ba^{133} source. This source produces radiation with two discrete energies, namely 32 kev and 85 kev. The response of the scintillator, as shown in this figure, clearly distinquishes these two energies. Analysis of these data (and subsequent measurements) indicated that the response of the detection system was essentially linear with approximately 0.3 kev per channel.

(c). Monochromatic Radiation.

In Figure 4 is shown a detailed study of the 32 kev Ba x-ray line (from Cs^{137}) together (for comparison) with the pulse height spectrum from the detector when exposed to 33 kev monochromatic x-rays obtained with the spectrometer by diffraction from the (220) crystal planes of germanium.

PULSE HEIGHT SPECTRA

Cs^{137}



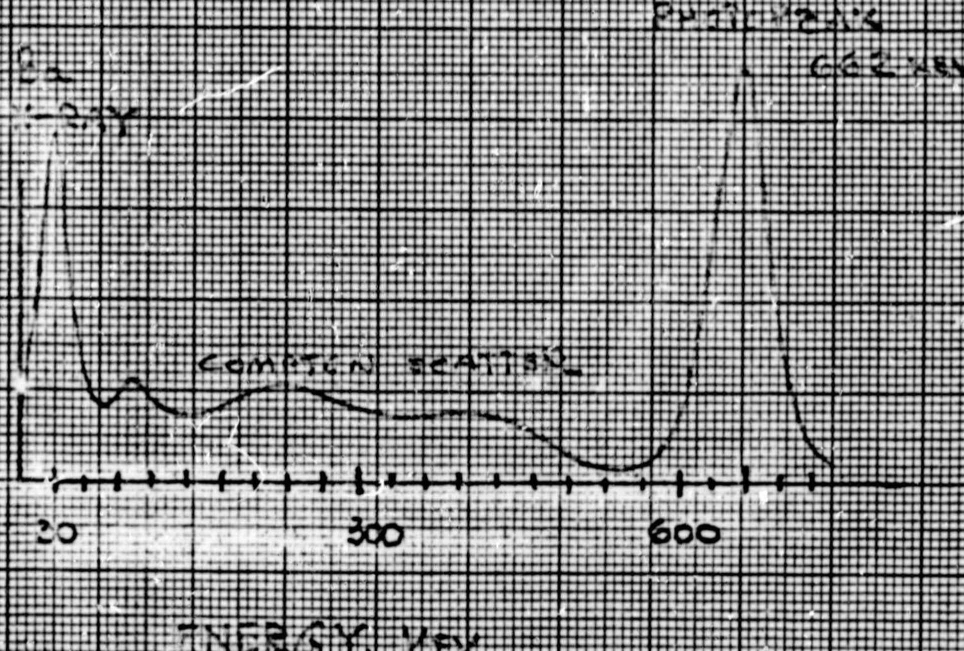
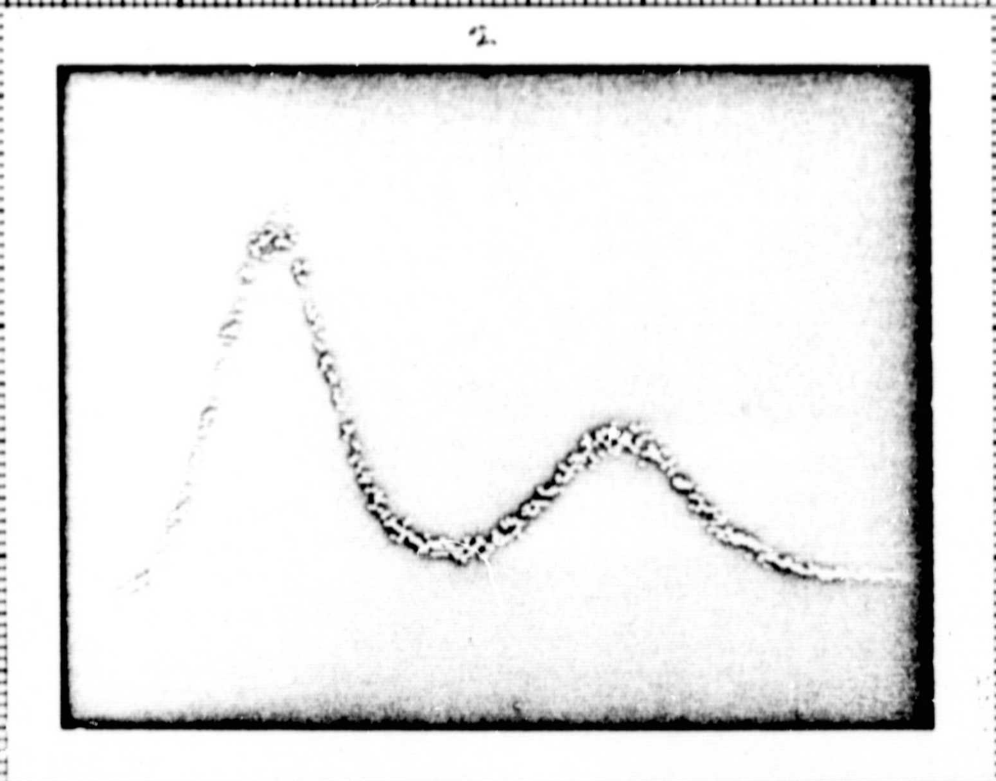


FIGURE 2

PULSE HEIGHT SPECTRA

Ba^{133}



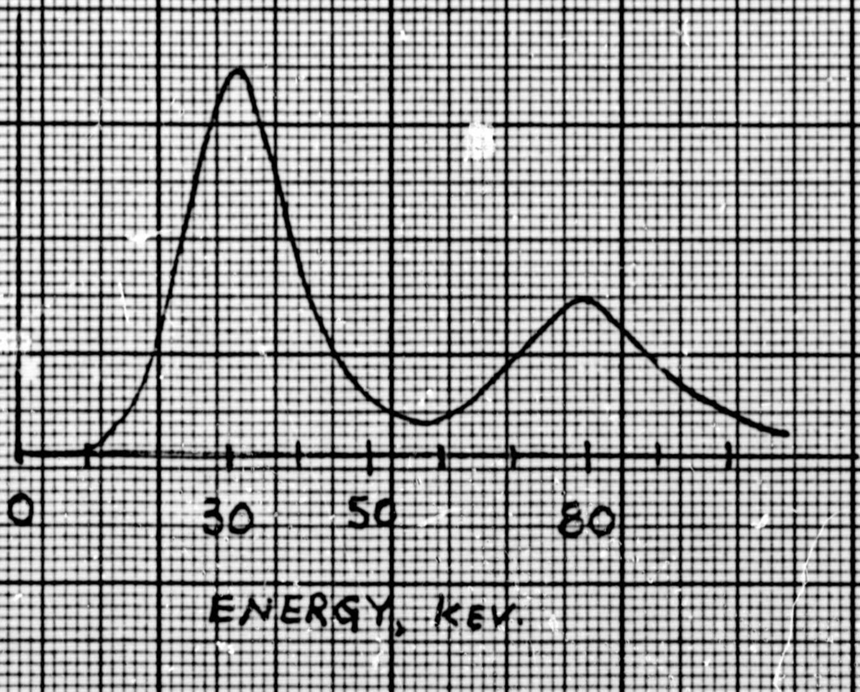
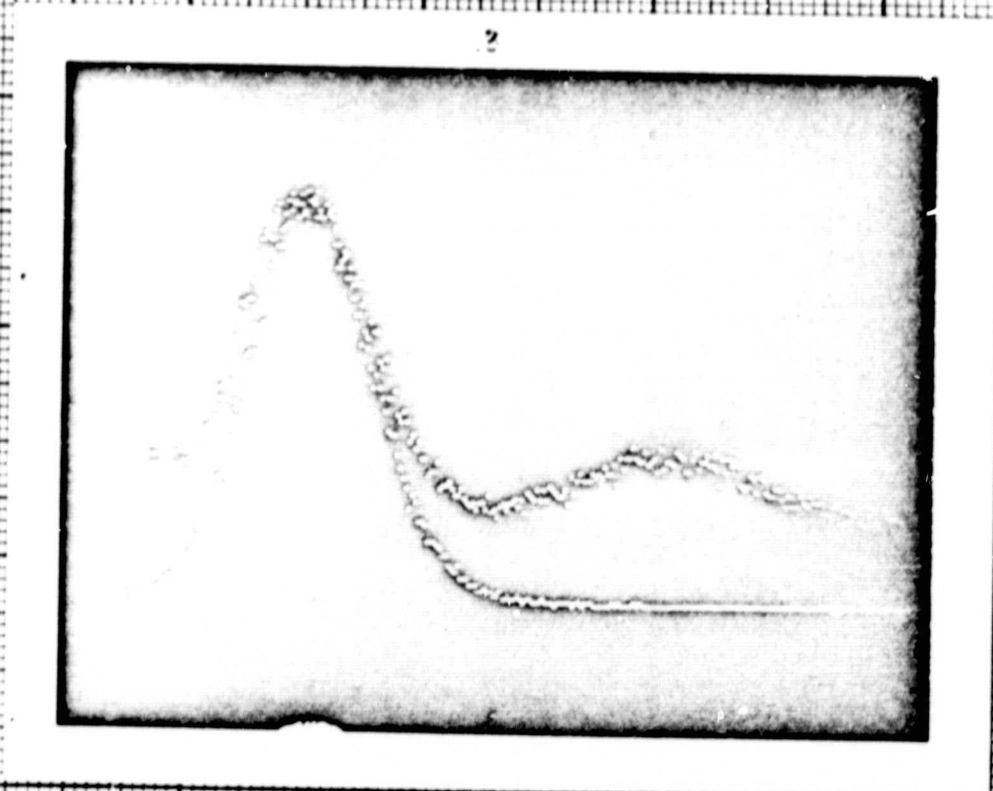


FIGURE 3

PULSE HEIGHT SPECTRA

33 KEV



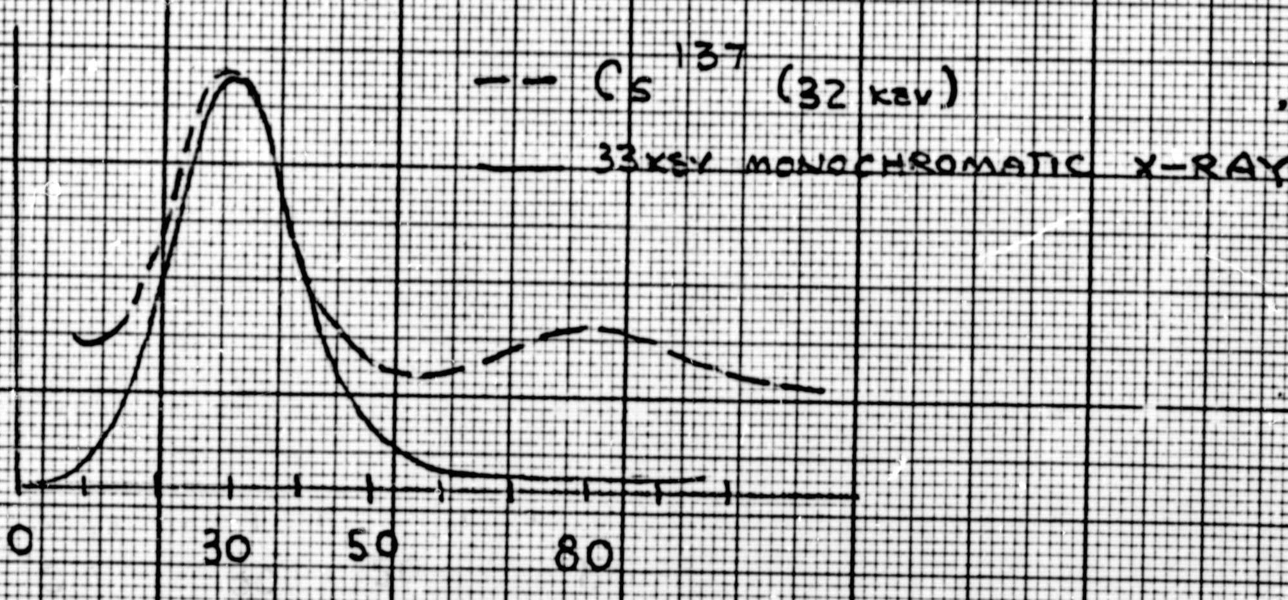


FIGURE 4

Satisfactory agreement in the signal-to-noise ratio and crystal angle calibration was obtained.

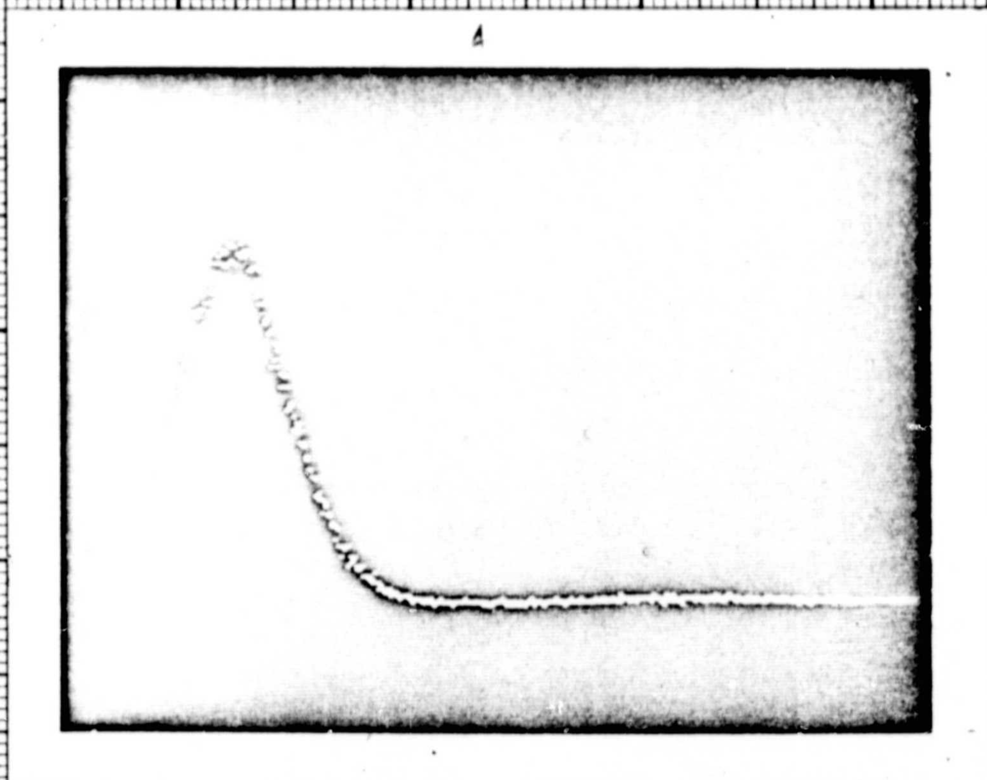
Figures 5 and 6 are pulse height spectra obtained when the detector is exposed to 25 kev and 32 kev monoenergetic x-rays, respectively. These spectra are seen to be sharp with peak shifts in agreement with calculated values. Energy differences of 5 kev (in this case) are easily resolved.

Figure 7 shows the response of the detector to monoenergetic x-rays of 40 kev and 80 kev (obtained by diffraction by the (220)-and (440)-planes, respectively, of the crystal). These data illustrate the ability to clearly resolve two neighboring energies (32 kev and 40 kev, in this case) alluded to above.

This preliminary list of pulse height spectra may now be easily expanded to obtain a complete tabulation of the response of the detector to monoenergetic x-rays for 20 kev $< E <$ 100 kev with $\Delta E = 1$ kev. This complete tabulation would form the library of response functions needed for the analysis described in the INTRODUCTION.

PULSE HEIGHT SPECTRA

25 KEV



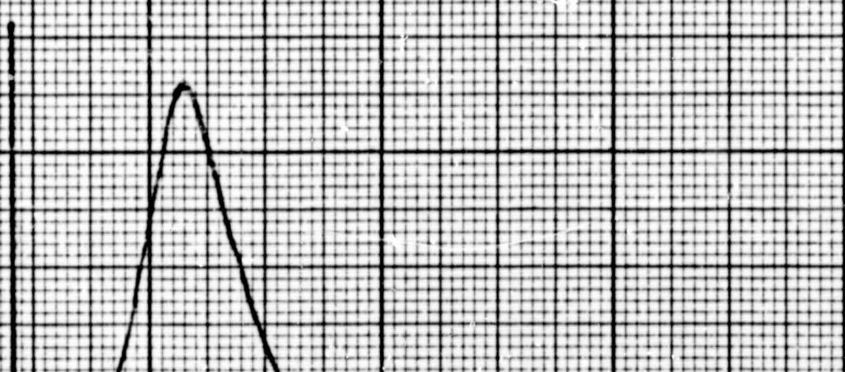


FIGURE 5

PULSE HEIGHT SPECTRA

32 KEV

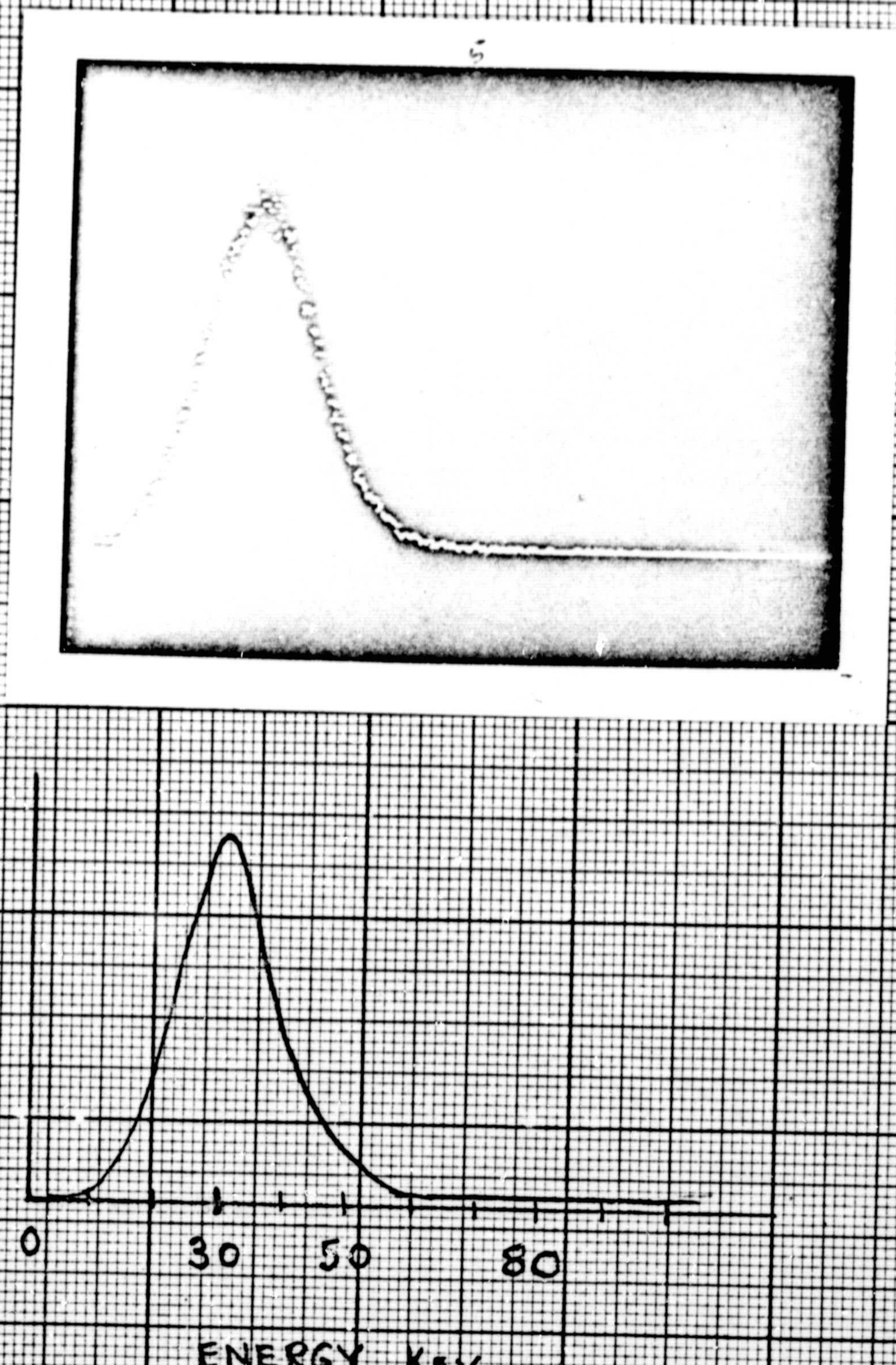


FIGURE 6

PULSE HEIGHT SPECTRA

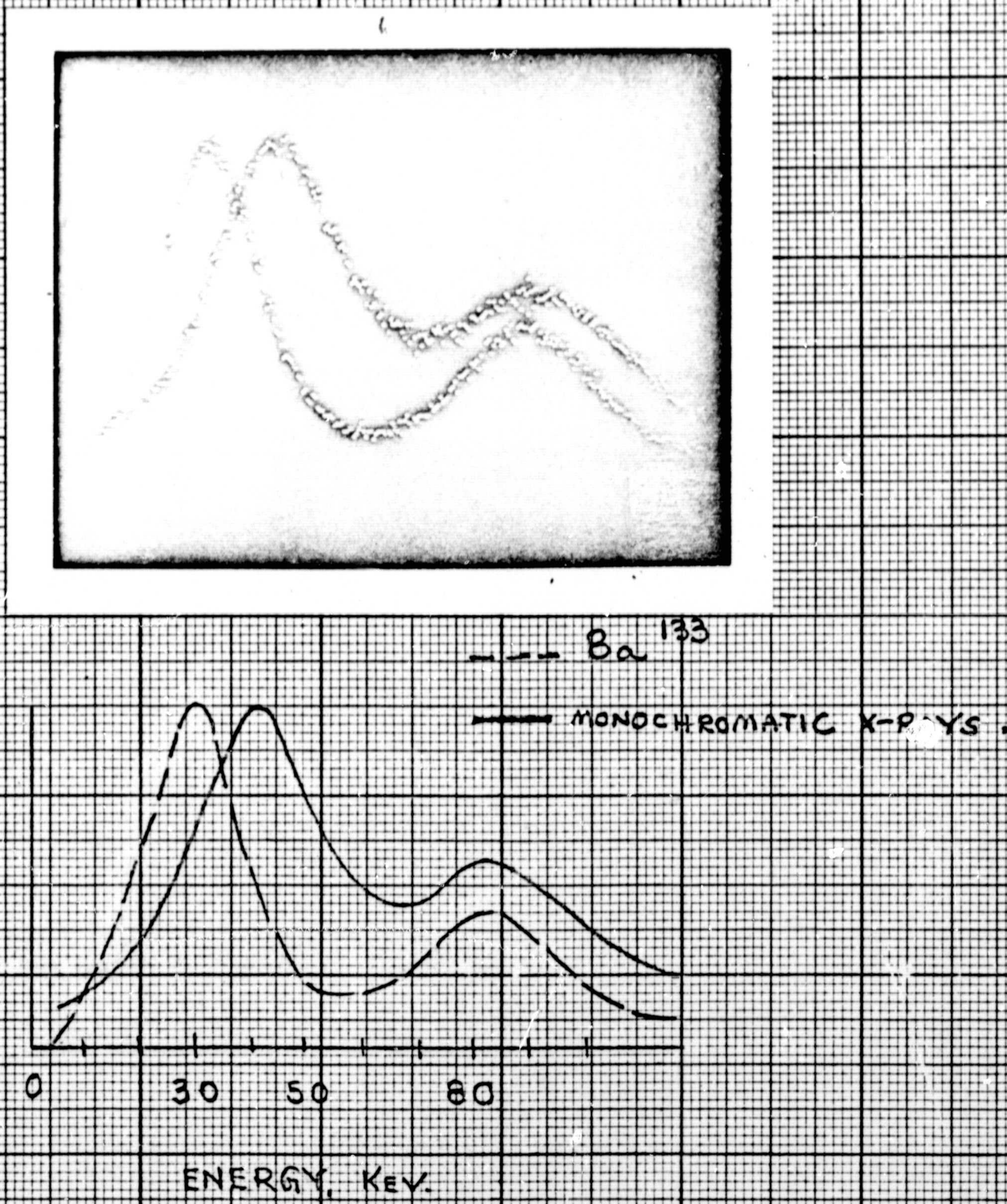


FIGURE 7

NEED FOR FURTHER INVESTIGATION.

(a). Additional measurements are necessary for the completion of this project. The satisfactory preliminary data, given in this report, and the ease with which a complete tabulation of these important pulse height spectra may now be obtained (with the equipment now assembled) makes the continued support of this project essential. It is clear that this program will yield results which are required for the research program of NASA.

(b). The measurement of the response of the detector system for monochromatic incident radiation in the energy range 20 kev $< E <$ 100 kev should be completed in order that shape functions may be generated. The analytical techniques which have been developed by these (and other) investigators should be employed to unfold complex spectra into its mono-elemental components.

(c). This research program was funded under Grant NGR 09-011-022 ($6000) and ended 1 August 1970. An interim request for continued support during the period 1 August 1970 - 30 September 1970 has been filed under NASA Control # 09-011-022 (032). A research proposal to support further investigation is being made.

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BUDGET REPORT

	Requested	Expended
Salaries*		
Research Assistant	1500	
Secretarial Assistance	600	1722.16
Overhead		
(32.2% of salary related costs)	676	557.06
Equipment and Expendible Supplies*	3224	4090.78
	6000	
University Cost Sharing	360	
	6360	6360

*Approval for deviations in these items was received (see attached correspondence)