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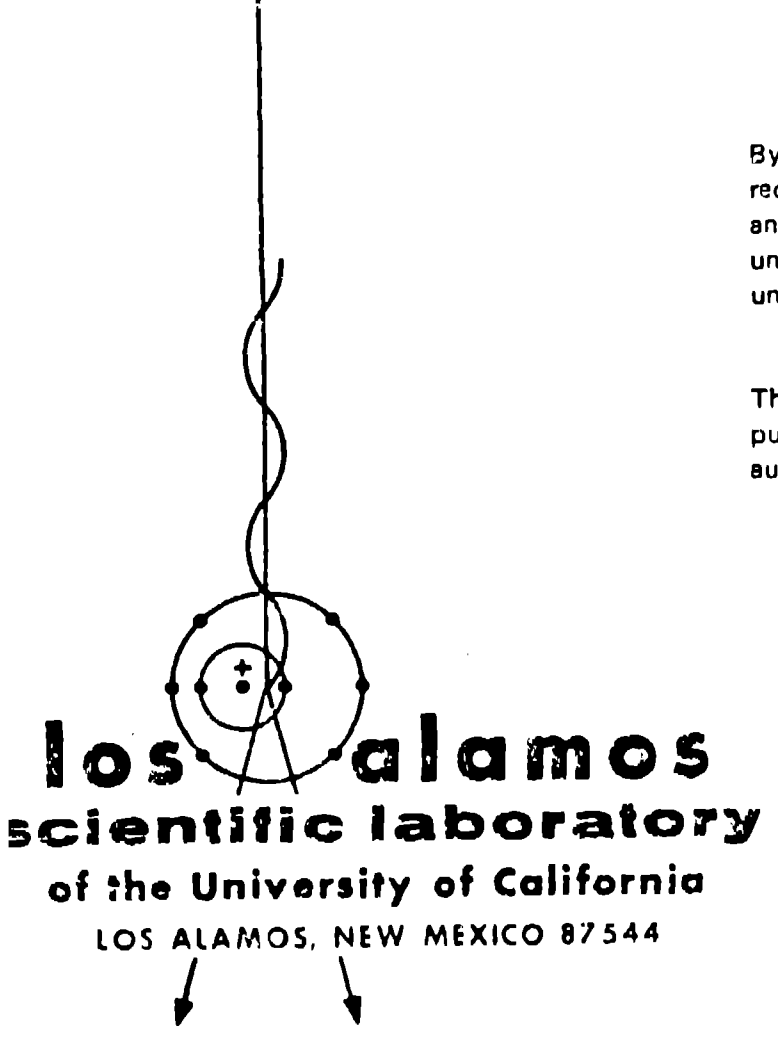

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FIELD ASSAY OF PLUTONIUM WITH A NEW COMPUTERIZED SEGMENTED GAMMA SCAN INSTRUMENT*

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ABSTRACT

An automated passive gamma scanner for NDA is described, engineered around a minicomputer and an automated scan table. The computer permits virtual hands-off operation and offers a segment-by-segment profile of fissile content, as well as an estimate of the propagated statistical measurement error. Since the measurement is transmission corrected on a segment-by-segment basis, a wide variety of container sizes and material matrices can be accommodated, permitting two-percent measurements on scrap and waste samples containing between five and five-hundred grams of fissile material. Field tests of the new instrument at ARHCO in Richland, Washington, are also described.

INTRODUCTION

Nondestructive assay of fissile content of material in various sample containers depends primarily upon detecting either neutrons or gamma radiations from the nuclei under measurement and relating the number of events detected to the number of nuclei present in the sample. Active interrogation systems employ some means to induce radiations in the nuclei under assay, while passive systems detect emanations emitted spontaneously by the sample.

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*Work performed under the auspices of the U. S. Atomic Energy Commission.

In the instrument to be described, we are concerned with detecting gamma radiation spontaneously emitted by nuclei of fissile material under assay. There are several serious problems associated with any quantitative assay by means of gamma detection; viz., gamma rays are attenuated by material between the nucleus and the detector, the detection system must be able to identify correctly only those gammas from the isotope under measurement, the detector and electronics may respond differently to different data rates, and the assay may take a prohibitively long time if system detection efficiency is not high enough. The attenuation problem is by far the most serious of the above, since gamma attenuation varies with transition energy as well as the density of the material through which the ray passes. Matrix inhomogeneities make this attenuation a very serious matter indeed, since in this case the attenuation factor relating detected counts to fissile content is varying in some unknown manner with sample configuration.

LASL Group A-1 has developed a passive segmented gamma detection scheme employed in two systems designed to overcome these and other problems. The first of these systems is sufficiently complex and difficult to operate that it remains strictly a laboratory research instrument for use by physicists and nuclear engineers.

The second of the two systems, which is to be described here, is a descendent of the above laboratory model, considerably redesigned and re-engineered, and with the operation difficulties greatly minimized. The use of a modern minicomputer permits automation of the scan table as well as automatic calculational capability, free from human error. In addition, it offers something never before available with the segmented gamma scan system: a propagated error calculation yielding an estimate of measurement error. The system, while not suitcase portable, is now at least readily transportable in a light truck. In addition, operation personnel need no longer be highly trained scientists.

Other methods of making NDA measurements with gamma radiation involve the use of sodium-iodide detectors to measure gamma peaks. Unfortunately, most of these methods make no reasonable provision for solving the most serious problem with gamma detection schemes mentioned previously, namely, the attenuation problem. In order to correct for matrix attenuation in the segmented gamma scan system, a separate transmission measurement is made, which provides a correction factor to compensate for gamma matrix attenuation. Since the sample being measured may be quite inhomogeneous, there is a large class of samples for which a single transmission measurement is not sufficiently accurate, and for this case we have segmented the assay in such a way that the sample is assayed in 1-cm slices, a separate transmission measurement being made for each one. Further than this, the sample is also rotated to wash out radial inhomogeneities. It should be noted here that such an attenuation correction is good only for the matrix material. That is, if there is considerable self-absorption of gammas in the fissile material itself, it will not be possible to

make a quantitative measurement by this gamma detection system, since no reasonable attenuation correction is possible except in very special cases.

In order to resolve the energy peaks of the gamma spectrum with sufficient precision to permit accurate transmission measurements, a Ge(Li) detector must be used. If the transmission peak used to measure attenuation is not sufficiently close in energy to the fissile peak being used for the assay, this attenuation measurement will not be accurate, since gamma attenuation varies with both matrix material and gamma energy, the variation becoming more severe at lower energies. The transmission measurement is made by utilizing a separate transmission source which is viewed through the sample being assayed and compared with a known transmission value obtained from a background run taken with no sample present in the system. In addition to the transmission correction a pile-up and live-time correction is made by counting a separate peak from a small source placed close to the detector crystal and viewed at all times by the detector. When the count rate from fissile material and transmission sources is high, this live-time source will vary in its counting rate in a manner which permits accurate measurement of live-time correction.

Since segmented scanning with a transmission source while rotating the sample can yield good results, why not simply replace the old NaI systems with Ge(Li) gamma scanners? Mainly, the problem is that a segmented scan system with separate transmission and live-time correction measurements is quite complex, and hence would normally be quite critical to adjust and difficult to operate. Unfortunately, there are some measurements for which the Ge(Li) resolution and transmission measurement are absolutely essential; therefore, the problem was to make the transition to the segmented gamma scanner as painless as possible.

In the present system, the complexity of both calculation and operation is hidden behind the panel of a minicomputer. These complexities have been made as transparent as possible to the operator. So far as the operator is concerned, the assay simply consists of placing the sample on the scan table and pushing the start button. Scanning is automatic, and the answer in grams of fissile material is printed out on a teletype at the end of the assay run, which requires ten seconds per centimeter of sample. Thus, a common 15-cm ash can sample is assayed automatically in about two minutes, complete with printout and error calculation.

THE MODERN MINICOMPUTER SCANNER

Figure 1 shows the complete segmented gamma scan system with all component parts except for the lead collimator in front of the detector. Obviously, it is not suitcase transportable but is quite readily movable by light truck from one fixed location to another. Setup time will be a few hours, so that it will find its usual applications where it can be set up as a fixed-base operation for routine assay applications over some period of time.

Since homogeneous samples do not require segmenting in order to make accurate gamma assays, the present system will find its widest application in the areas of scrap and waste measurements. It is, for example, ideally suited for measurement of ash cans containing ten to several-hundred grams of plutonium from waste, which may be in different density layers in the can. These cans can contain some real surprises occasioned by inhomogeneous matrix and loading, and the gamma scanner will measure these very accurately. As a general rule, any sample in which self-absorption is not a problem, and for which a transmission measurement is possible, can be accurately assayed by this instrument. The segmented gamma scan is suitable for assay of ^{238}Pu, ^{239}Pu, or even ^{235}U, but the plutonium assays provide a simpler case than uranium, since the former require only a single transmission peak due to their higher gamma energies. The uranium requires measurement of the 185-keV gamma line, where the attenuation coefficients are varying rapidly with energy. In this case, it is necessary to bracket the uranium line with two transmission lines, and the analysis is slightly more difficult, though the computer once again makes the additional complexity transparent to the user.

The operator control panel is shown in Fig. 2. We have adopted the philosophy that it is basically simpler for an operator to press an appropriately labelled pushbutton rather than having to type instructions into a teletype keyboard, and accordingly the panel as shown provides pushbutton control of all instrument functions. Not only can the various types of data runs be initiated by pushbutton control, but also assay windows can be entered and the interactive display wholly controlled from the same panel. The large display shown at the top of Fig. 2 is a memory oscilloscope which displays in 15-by-20-cm format the various spectra, either the entire 2K spectrum or any 256-channel segment of the total spectrum. The control panel immediately below the display provides manual control of the scan table, which is not normally used in routine operation but provides for setup and maintenance. The length of the scan (container height) in inches is determined by the thumbwheel switches on this panel, which allow containers up to 483 mm (19 inches) in height to be assayed. The scan table can accommodate containers up to 18 liters (5 gallons) in size and up to 68 kg (150 lb) in weight.

The bottom panel in Fig. 2 shows the operator function pushbuttons. In addition to setting in data windows and starting various types of runs (background and assay), the operator has the option of choosing either of two types of printout and of typing comments regarding the assay or sample number on the teletype.

Figure 3 shows an actual printout for an ash can sample assayed by the instrument at the ARHCO facility in Richland, Washington, where the AEC is field-testing the scanner. This illustrates the so-called "long" printout, where all data are typed out on a segment-by-segment basis. From this long printout, the operator can build up an actual fissile content profile of material in the sample. In this case it is clear that the plutonium

stops at about segment 11, so that in this 18-cm can material appears only in the bottom 14 cm. The transmission peak also affords some interesting information. Note that this value varies considerably from segment to segment, so that an average transmission, if it had been used for the entire assay, would have yielded 216 grams for the assay instead of the actual 277--a whopping 22% error! This illustrates the importance of a segmented transmission and fissile measurement for samples which are vertically inhomogeneous. By this method of correcting the measurement for attenuation losses, it is quite reasonable to assay materials for which the transmission may drop as low as 2-5%. Note also that using the long printout, the operator has the choice of which segments to use in the final calculation. This feature is useful for nonroutine samples where the fissile material may be concentrated in one part of the matrix. Eliminating those segments from the assay which do not include fissile material has the effect of improving the statistical accuracy of the overall measurement, since "empty" segments are not contributing their scattered counts to degrade the statistics. The estimated standard deviation printed out by the computer is based on a segment-by-segment calculation of statistical error, assuming Poisson statistics on the peaks and including all sources of such error. Note that in the example illustrated in Fig. 3, simply taking the square root of the corrected counts as an estimate of the error would yield 1.79 grams as the estimated error--a value low by more than a factor of two. Printing out the estimated standard deviation effectively assures that the operator is left without excuse for accepting statistically invalid answers.

The short printout mode suppresses the segment-by-segment data and is useful for reducing printout time on "routine" or repetitive assays, where the content profile is unimportant and where it is well known that the transmission measurement will be reasonable for the samples being assayed. In this mode, the operator does not choose which segments to use in the final calculation. The computer uses all segments for the final number.

Even initial setup of the data windows is greatly simplified through the large display coupled with an interactive cursor and the push-button switches. To set in the data windows, one has simply to make a setup run, which yields the spectrum (from plutonium) shown in Fig. 4. The first major peak on the left of the spectrum is the 356-keV barium live-time peak. The 400-keV transmission peak from ^{75}Se is next to the 414-keV photopeak from the ^{239}Pu. In the cursor mode, an intensified dot moves along this spectrum graph. The operator simply pushes the stop and start buttons as he moves the cursor along the graph to indicate where he wishes to set in the data windows.

Figures 5 and 6 show the main control chassis and the motor control chassis, respectively. The open construction in these units permits fast, easy troubleshooting and maintenance on a plug-in module basis and guarantees minimal downtime and reliable operation. In fact, experience to date with this unit in more than a thousand hours of operation indicates no downtime due to electronic component failure.

FIELD EXPERIENCE AT ARHCO

During the month of April this new gamma scan instrument was installed in the Plutonium Finishing Section of the Atlantic Richfield Hanford Co., Richland, Washington, at the request of the AEC Richland Operations Office. Uncrating, setup, and initial calibration of the instrument required about six hours, after which many assays were made and demonstrations held for the personnel at that facility. A wide variety of samples were assayed, including a plutonium oxide sample (for which the transmission was marginal), centrifuge sludge samples, solution samples (for which the segmentation is unnecessary), the complete set of ARHCO ash calibration samples, and about fifty cans of ash ready for shipment to LASL for plutonium recovery. Figures 7 and 8 show the instrument installed at ARHCO and ready for assay work. No particular precautions are necessary, except that about 4.5 m^2 of space and 110 V ac must be available.

Several surprises were encountered during the initial assay work at ARHCO. A centrifuge sludge sample which had been tagged with 60 grams of plutonium from a sodium iodide assay was found to contain actually about six grams. This illustrates the difficulty of correctly calibrating a NaI system, where the calibration standard must not only be geometrically identical with the samples to be assayed, but where it must also be similar in matrix attenuation characteristics. When this is not the case, large errors will result, as this centrifuge sludge sample strikingly demonstrates. Of perhaps the most significance was the resolution of a long-standing discrepancy between the measurements at ARHCO on ash cans being shipped to LASL for recovery, and the same measurements on the incoming shipments made at LASL. For about a year, it had been noted that LASL was recovering about 20% more plutonium from this ash than ARHCO claimed it was shipping. We assayed thirty of these ash cans ready for shipment with the new segmented gamma scan instrument and compared the results with the ARHCO assays of the same cans. The results are shown in Fig. 9, where the discrepancy between the ARHCO NaI measurement and the gamma scan measurement are plotted as a function of when the NaI measurements were made. Not only is the 20% discrepancy apparent, but it is also clear that this discrepancy is increasing with time, indicating that the NaI system is getting ever further out of calibration. Since the computer constantly updates the calibration each time a standard run is made with the gamma scanner, it will not become uncalibrated. Also, further research indicates that part of the low assay with NaI was due to different packaging between the ash cans and the standards used for calibration of that instrument. With a transmission correction being made for each segment, this cannot happen with the gamma scanner system. With the segmented gamma scan instrument, the only critical feature of the calibration standard is that the standard must be geometrically in the same configuration as the samples to be assayed, since the calibration factor in this system is truly a geometric one. The matrix materials need not be similar, providing only that a transmission measurement can be made for each sample and the standard.

Figures 10 and 11 illustrate the importance of having a system which is both rapid as well as accurate. As regulations requiring 100% assay of inventory become ever more stringent, the speed of making the assay will have important economic impact. Fissile material is not only a dangerous material which must be safeguarded from improper use by unauthorized persons but is a valuable economic commodity as well, which fully justifies the increasing emphasis on total dynamic accountability. Waste materials being shipped for plutonium recovery as illustrated in the two figures will increase in numbers as the industry expands. The technique of segmented gamma scanning offers a reasonable method of making reasonably rapid, very accurate assays.

REFERENCES

None

FOOTNOTE

Page

* Work performed under the auspices of the U. S. Atomic Energy Commission.

FIGURE CAPTIONS

Fig. 1. Automated segmented gamma scan instrument.

Fig. 2. Control panel.

Fig. 3. Sample assay printout.

Fig. 4a. Plutonium spectrum showing barium live-time peak.

Fig. 4b. Plutonium spectrum showing transmission and Pu peaks.

Fig. 5. Main control chassis.

Fig. 6. Motor control chassis.

Fig. 7. System being field-tested at ARHCO--scan table.

Fig. 8. System being field-tested at ARHCO--control panel.

Fig. 9. Ash can data--Ge(Li) vs NaI values.

Fig. 10. Ash cans to be assayed for plutonium.

Fig. 11. More ash cans to be assayed for plutonium.

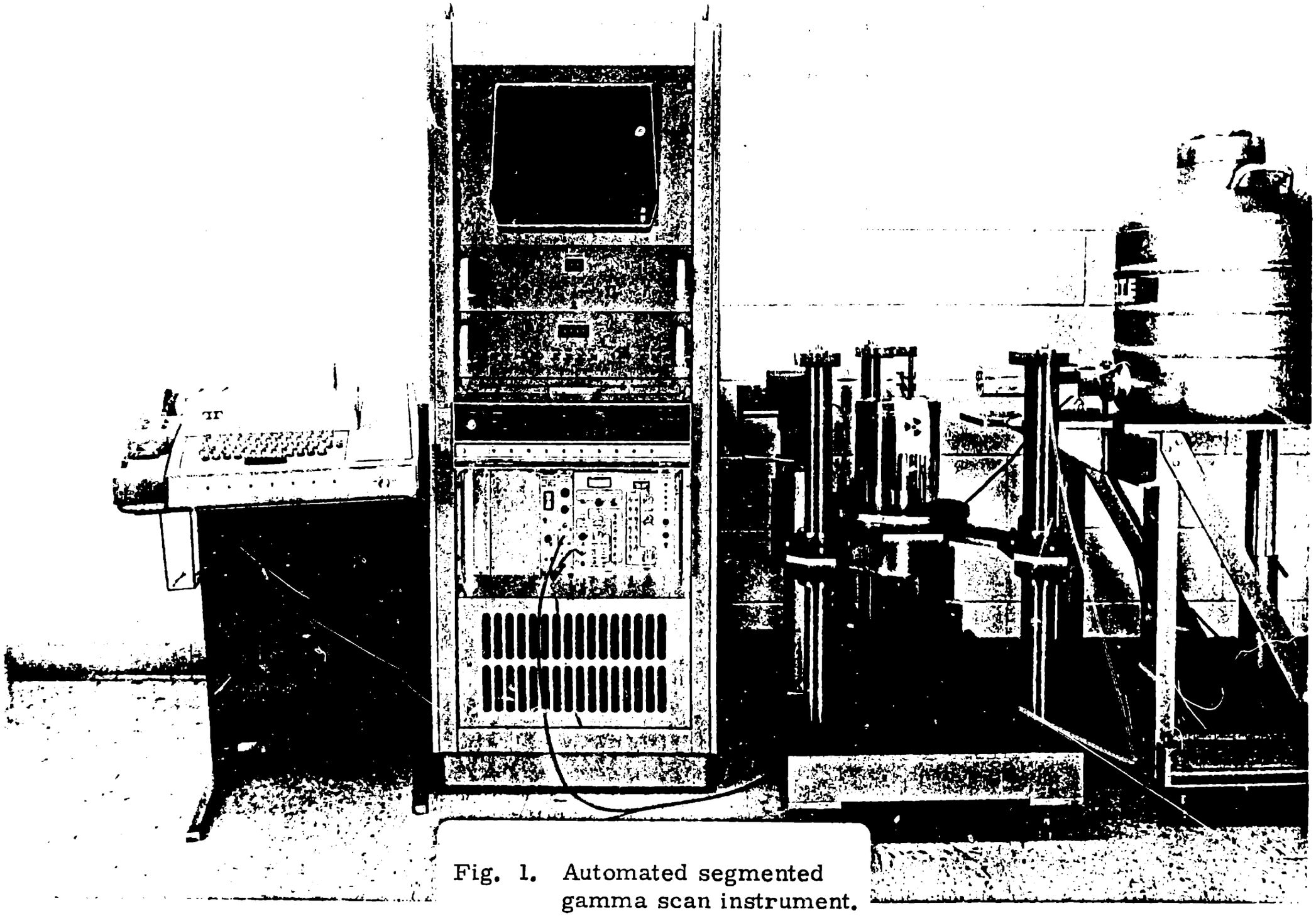


Fig. 1. Automated segmented gamma scan instrument.

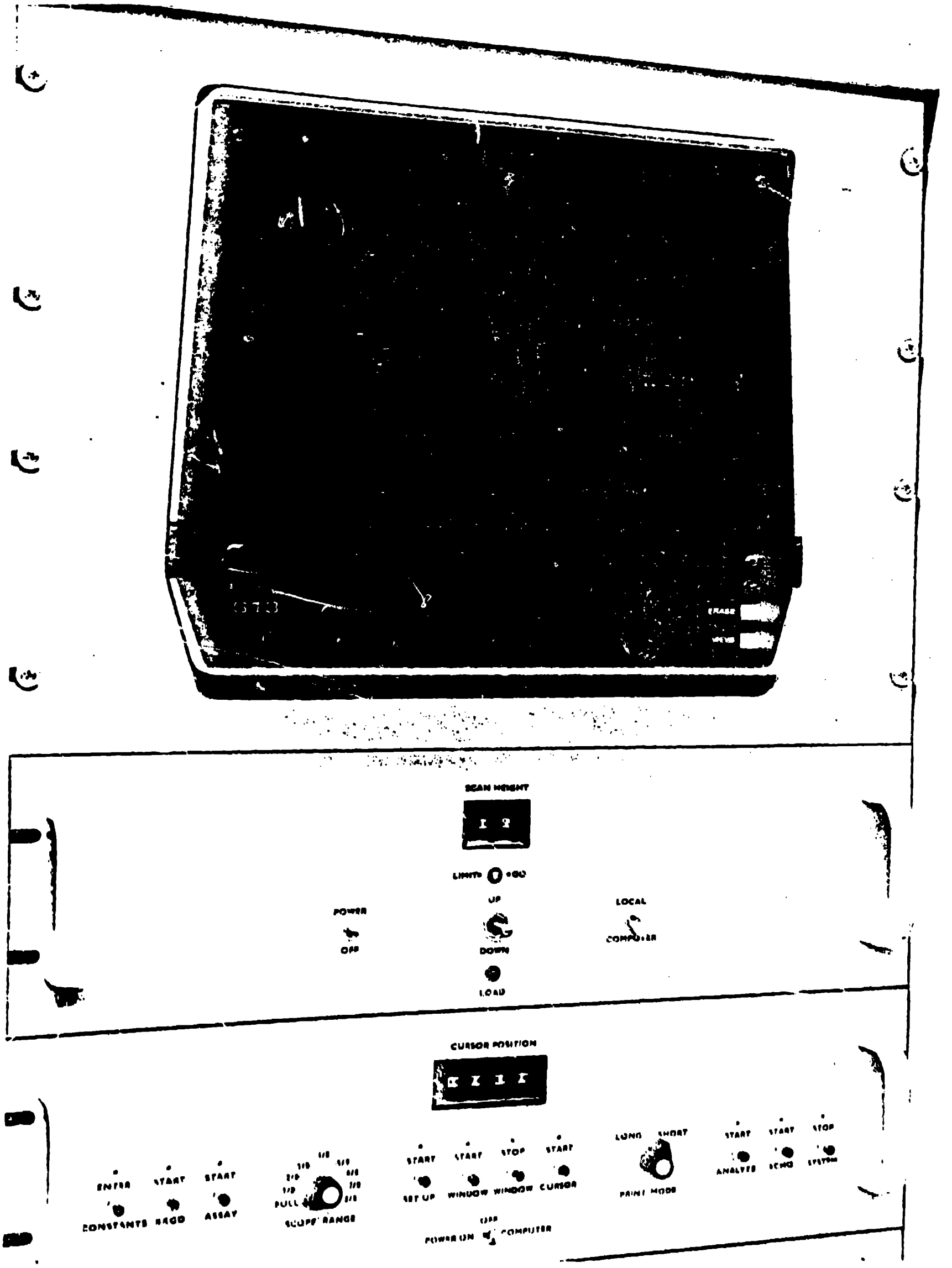


Fig. 2. Control panel.

SAMPLE: ASH STANDARD (ARHCO) #42
ESTIMATED PU = 299 GRAMS, PU 239 = 275.08 GRAMS

RUN ENDED

SEG #	LIVE TIME	TRANSMISSION	ASSAY PEAK	CORR FACTOR
1	+1.0095	+.9855	+24.8	+1.0169
2	+.9690	+.6186	+635.1	+1.2168
3	+.9446	+.1582	+1736.8	+1.9322
4	+.9730	+.1574	+2155.3	+1.9353
5	+.9613	+.1644	+2216.4	+1.9098
6	+.9695	+.1655	+2038.3	+1.9057
7	+.9605	+.1885	+1906.0	+1.8302
8	+.9518	+.2419	+1627.2	+1.6897
9	+.9514	+.4422	+1162.1	+1.3753
10	+.9930	+.7842	+535.5	+1.1120
11	+.9853	+.9393	+74.2	+1.0364
12	+.9956	+.9155	-9.3	+1.0470
13	+.9709	+.7637	-3.4	+1.1235
14	+.9839	+.8558	+20.9	+1.0750

FIRST AND LAST SEGMENTS FOR ASSAY? 1, 12

TOTAL CORRECTED COUNTS = +23898.22

ASSAY = +276.95 GRAMS EST. STD. DEV. = +3.69

Fig. 3. Sample assay printout.

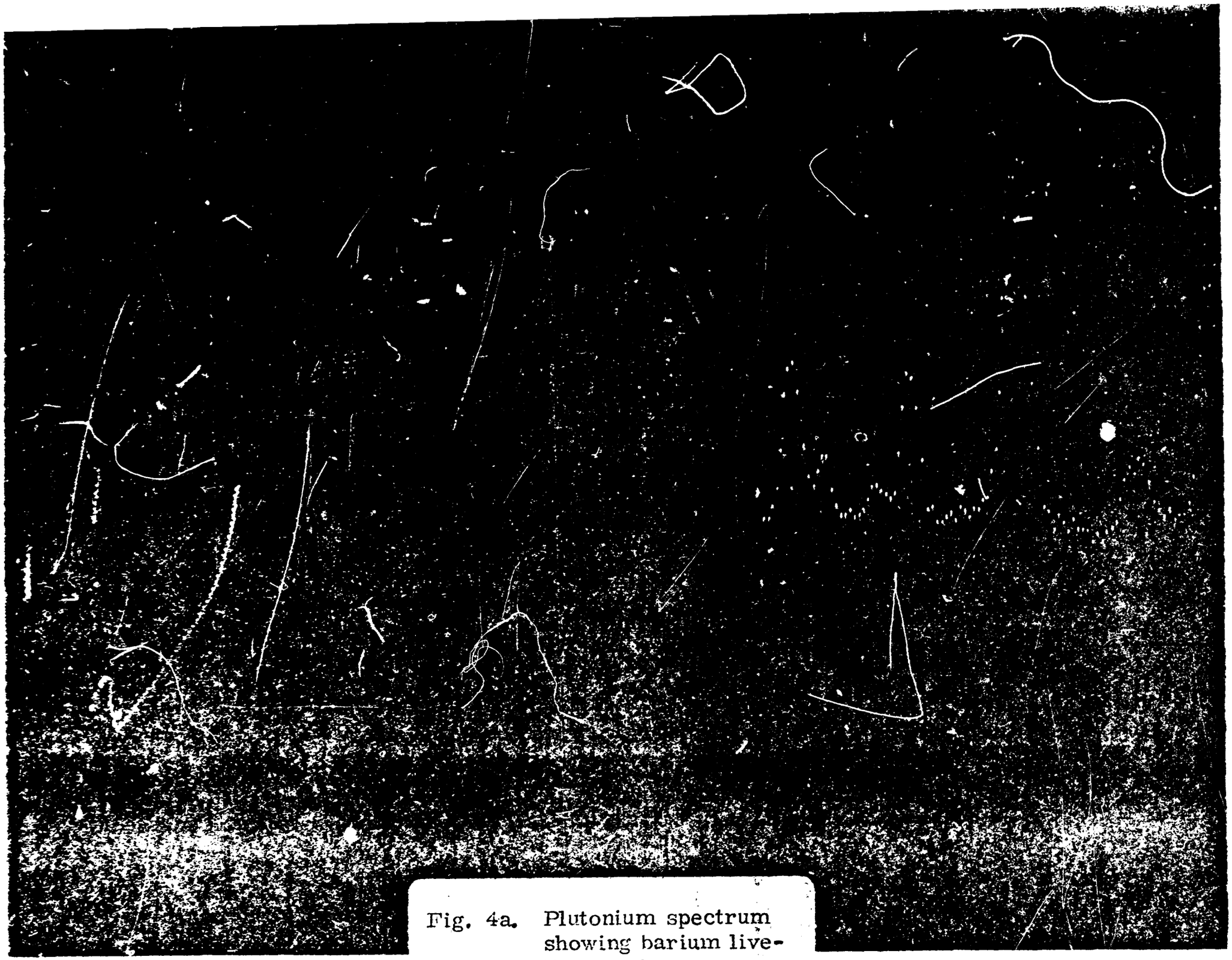


Fig. 4a. Plutonium spectrum showing barium live-time peak.

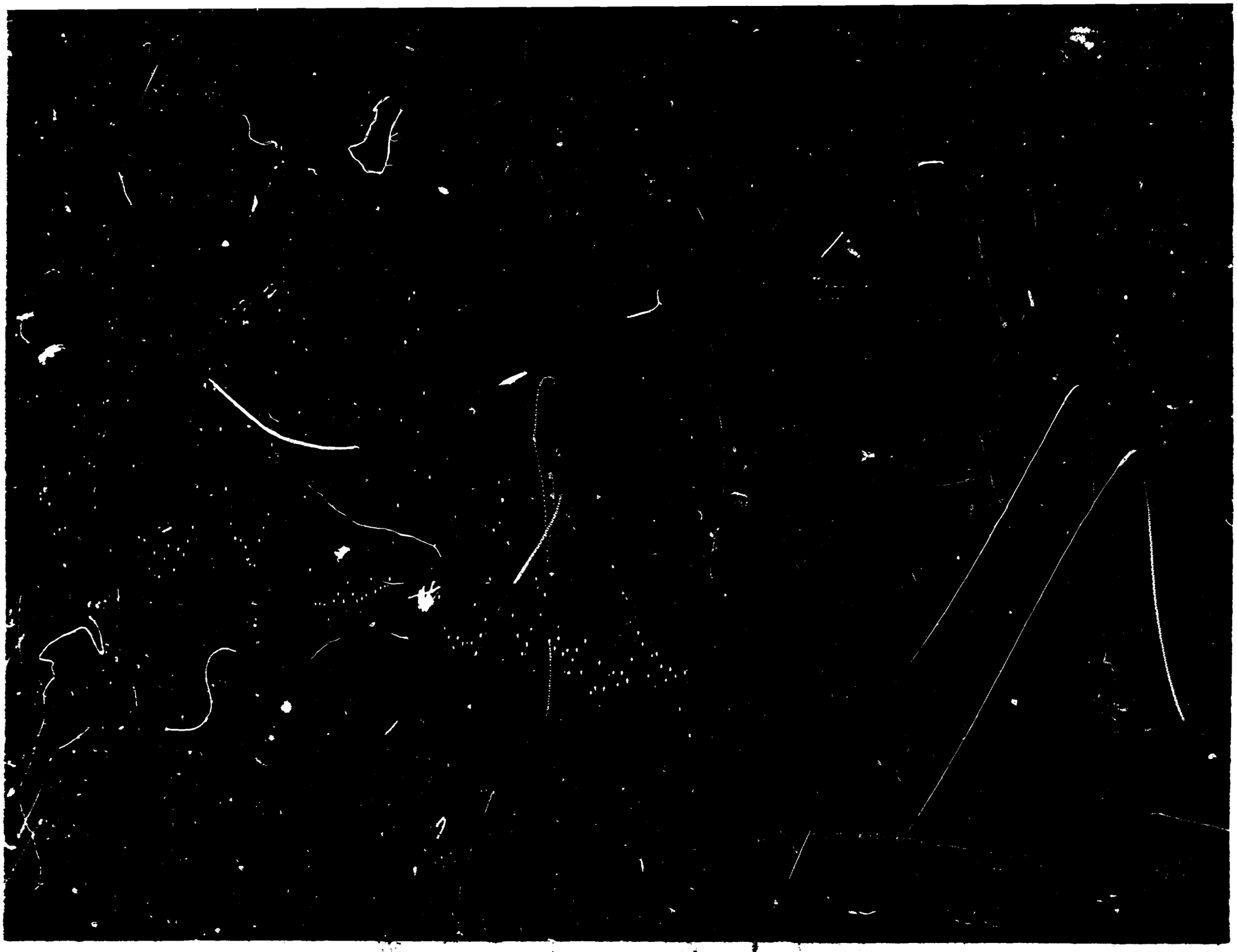


Fig. 4b. Plutonium spectrum showing transmission and Pu peaks.

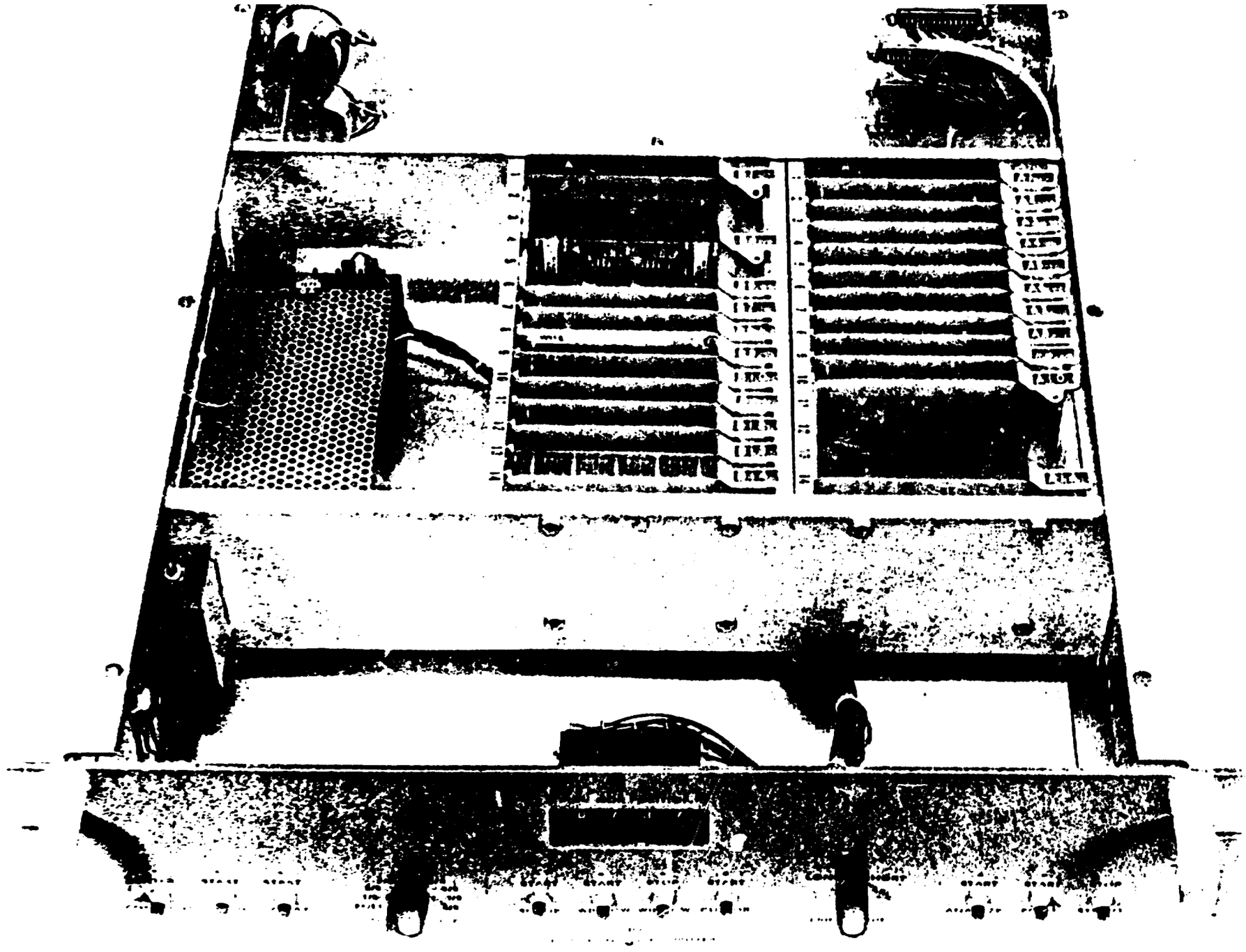


Fig. 5. Main control chassis.

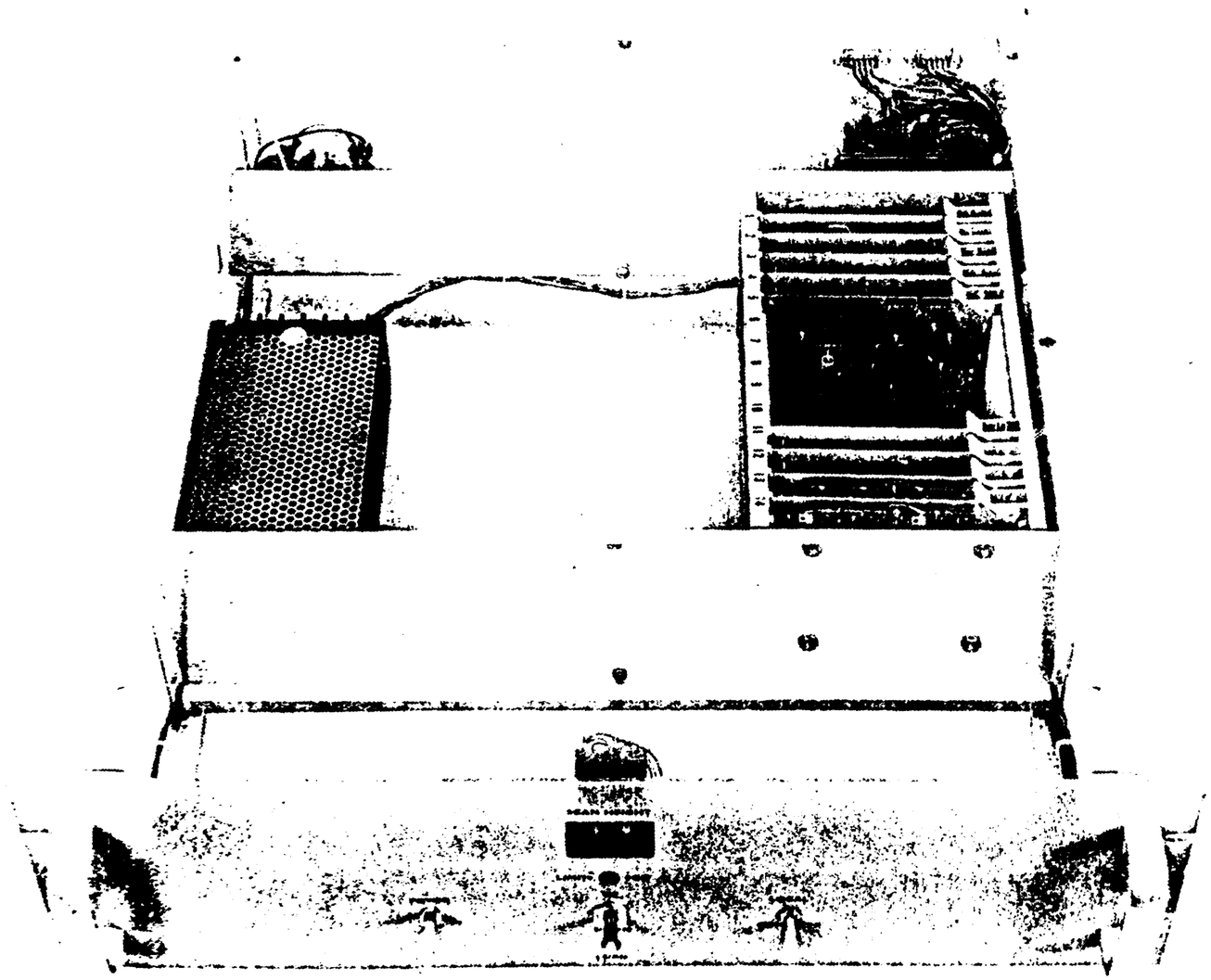


Fig. 6. Motor control chassis.

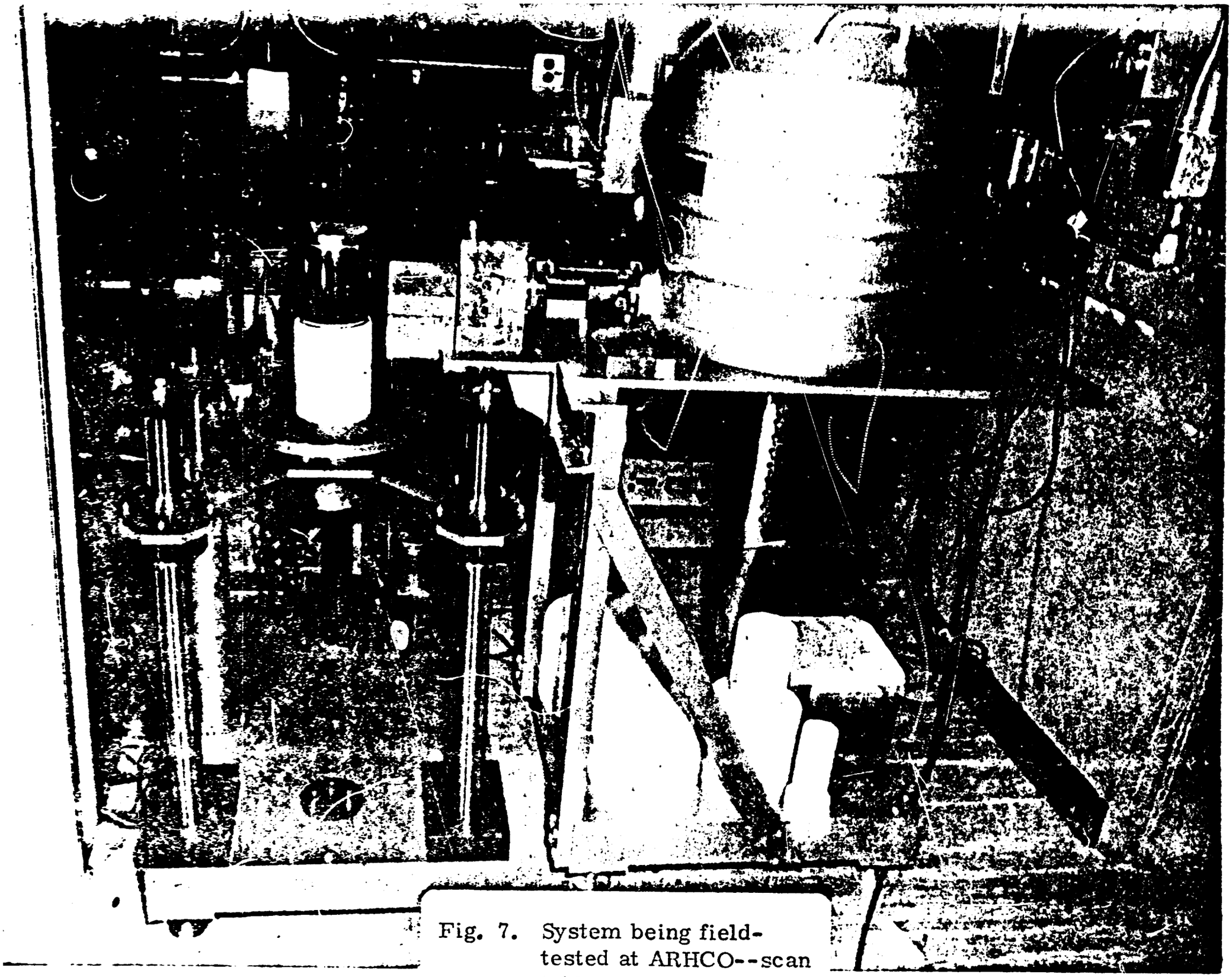


Fig. 7. System being field-tested at ARHCO--scan table.

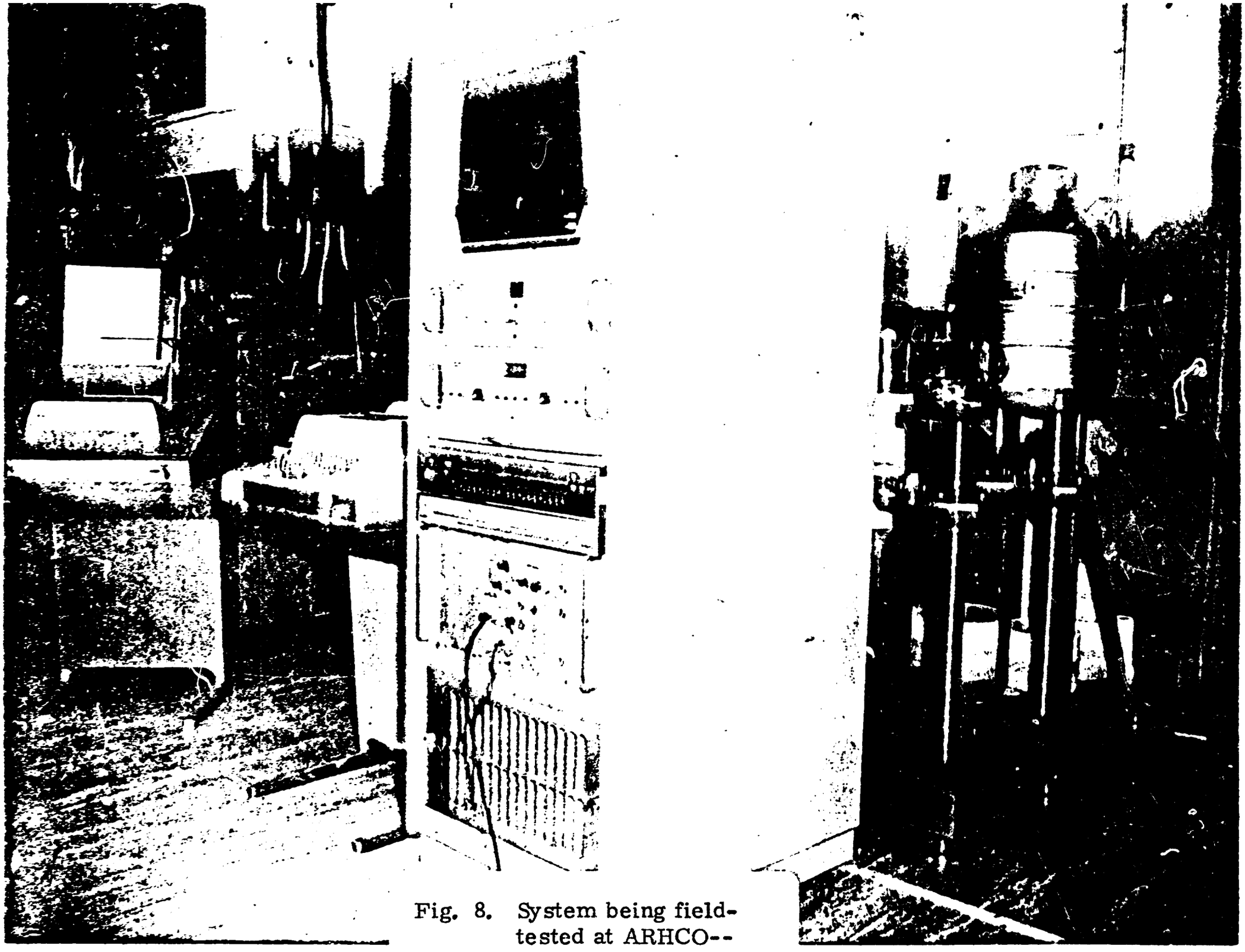
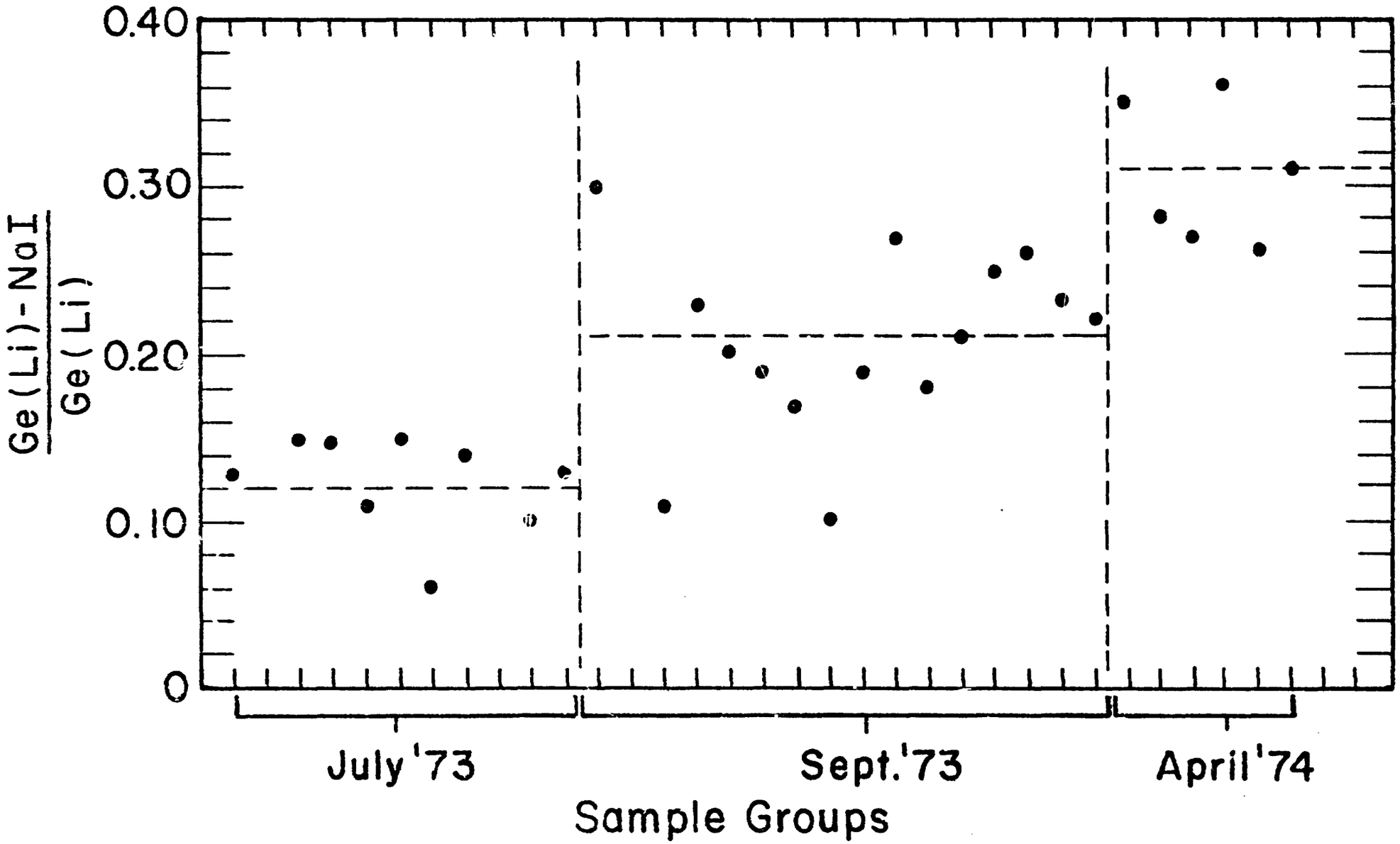


Fig. 8. System being field-tested at ARHCO--



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