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Informal Report

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Nuclear-Radiation Measurement Facilities at the Radiochemistry Laboratory at LASL

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

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NUCLEAR-RADIATION MEASUREMENT FACILITIES AT THE RADIOCHEMISTRY LABORATORY AT LASL

by

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ABSTRACT

A brief description is given of the radiation counting facilities and different types of systems used by the CNC-11 radiochemistry group at the Los Alamos Scientific Laboratory.

I. INTRODUCTION

The radiochemistry laboratory was placed in operation in 1948 to measure fission-product nuclides from the Sandstone series of that year. Since that time, it has evolved as the needs of Group CNC-11 have changed and expanded. Today, any nuclear radiation except the neutrino can be measured in a reproducible and quantitative fashion. The use of Geiger counters was abandoned in late 1948 because of their unpredictable life and poor reproducibility.

II. ALPHA COUNTING

The quantitative measurement of alpha radioactivity (^{239}Pu) was originally done using parallel-plate ionization chambers in which N_2 was the working gas. When Group CMB-7 made gas proportional counters reliable in 1949, we put in service a design useful for alpha counting. However, this model gave poor operational experience in the presence of high-beta samples and low-level samples from ion exchange columns. Thus, we returned to using ion chambers in 1976.

Our alpha counters have been stable to within 1% from installation in 1949 to the present. Now we are operating five units, three of which can also measure spontaneous fission decay while counting alpha activity. The working gas presently used is propane because this gas is conveniently available.

Absolute calibration of these counters has been done by three methods. Originally the geometry was determined by calibrating a ^{210}Po solution with a microcalorimeter and then counting an aliquot in the alpha counters. This gave a geometry factor of 51.65% of $4\pi \pm 1\%$. In 1965, a calibration using ^{241}Am that was standardized by 4π alpha, gamma counting gave a geometry value of $51.75\% \pm 0.75\%$. In 1970, a calibration using ^{241}Am that was standardized in a low-geometry counter gave a value of $51.72\% \pm 1\%$.

These counting systems have never been automated for data acquisition because the amount of counting done has never been large.

III. BETA COUNTING

The original beta counters were Geiger counters (Eck & Krebs* glass tubes). Operation was erratic and of limited life. In 1949, they were replaced with

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*Eck & Krebs Scientific Laboratory Glass Apparatus Inc., Long Island City, NY 11101.

methane-flow proportional counters designed and built at the Los Alamos Scientific Laboratory by Group CMB-7.* Radioactive standards of ^{238}U and its daughter products were made to follow counter performance. The counters were calibrated absolutely to determine fissions in ^{235}U by comparison fission counting and chemical separation of ^{99}Mo for beta counting. Stability of these counters has been very good [less than 1% drift in counting rate on one unit (Fig. 1) from 1950 to the present]. Deadtime corrections have always been included in computations of data from these counters. In 1966, the working gas was changed from methane to propane because the operational voltage plateaus were better (see Sec. XII. B).

In 1957, the workload for beta counting had increased to a level where automation was necessary. A wheel system was designed for entering the input data previously being entered manually (Fig. 2). When completed in 1962, the system had nine automatic changers in operation. Data recording is done on an IBM 526 Summary Punch** (Fig. 3).

IV. GAMMA COUNTING

The serious use of NaI(Tl) detectors for gamma-ray measurement began about 1950. However, counting rate stability was a problem. Only after transistorized pulse-height analyzers were developed was reliable gamma counting possible.

V. POSITRON COUNTING

The problem of measuring radioactive nuclides that are neutron deficient in the presence of beta-emitting fission products, such as the pair ^{89}Zr- ^{95}Zr, was solved by using a trochoidal analyzer, which uses a shaped magnetic field to separate positrons from negatrons (Fig. 4). Two such units, now in operation, have an absolute efficiency of 10% of 4π and a separation factor between positrons and negatrons of 10^6.

VI. LOW-BACKGROUND BETA COUNTING

The cosmic-ray flux, which causes background in beta counters, is twice the rate at 7200 ft (altitude) that it is at sea level. As a result, measuring low-activity samples requires some type of active shielding. We have developed a system (Fig. 5) that gives a geometry equivalent to our other beta-counting systems and has a background a factor of 100 lower than that of other counters (0.2 counts/min).

VII. SPECTROSCOPY

The advent of solid-state circuitry for multichannel analyzers (MCAs) and analog electronics fabrication made spectroscopy usable on a routine basis.

VIII. ALPHA SPECTROSCOPY

The performance of the Frisch-grid ion chamber for the measurement of both energy and intensity of alpha-particle sources still cannot be matched. The geometry of this detector is high (50%), its stability is good, and the background is less than that for any other particle detector (0.001 counts/min per channel) (Fig. 6).

Where the ratio of two or more alpha-emitting species of different energies is required, the surface-barrier silicon detector is adequate. Both the ion chamber and silicon detector are in use here. Energy resolution is about the same for both types. All alpha-spectroscopy systems can measure the spontaneous fission rate of a sample while performing alpha spectroscopy.

IX. GAMMA SPECTROSCOPY

A. NaI(Tl)

These detectors, mostly the well type, are used to measure a radioactive nuclide or nuclides after extensive chemistry on a sample eliminates all but one chemical species or after additional work with a

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*One of these models was later erroneously called the Sugarman counter.

**International Business Machines, Armonk, NY 01504.

mass separator provides a radioactive sample containing only one radioactive nuclide (for example ^{173}Lu). They are used with an MCA and readout system connected to a midicomputer for data analysis (Figs. 7–9). The high efficiency and long-term stability and reliability of NaI(Tl) detectors lend themselves very well to problems of gamma-ray measurement.

B. Ge(Li)

The high-energy resolution of these devices makes them useful where little or no chemistry has been done on a sample. They are also useful where more than one nuclide must be measured in a mixture of isotopes. A special application of these detectors is the anitcoincident-shielded well counter where high efficiency, low background, and Compton suppression all contribute to increased sensitivity of the detector system (Figs. 10 and 11).

X. LOW-LEVEL GAS PROPORTIONAL COUNTING

The use of 2-ℓ gas counters for measuring low-energy beta or x-ray emissions, such as ^{3}H,^{14}C, or ^{37}Ar, is a very specialized form of radiation measurement. Elaborate active and passive shielding is required to achieve state-of-the-art results. We have three such systems in operation with the required gas chemistry separation systems (Fig. 12).

XI. AUTOMATION

A. Automation of Gamma Spectroscopy

At present, seven computer-controlled sample changing systems are in operation with Ge(Li) detectors. These units can be programmed to measure 24 samples each, at selected times with selected efficiencies, and do the required data reduction in the associated computers (Fig. 13 and 14). Automation of the well NaI system has begun; so far, one unit is fully automated (Fig. 15). All data collection from these systems is done by the pulse-height-analyzer readout (PHARO) system.

B. Computer Utility

The automated data-collection system for alpha and gamma is served by a combination of computers. The system used until recently was a PDP-9 computer* with data transfer for plotting purposes to a PDP-11/05 that ran two Calcomp plotters.** A back-up PDP-9 was also available to keep the system running 24 hours a day, 7 days a week.

Just recently, the PDP-9 has been transferred to back-up status, and the data acquisition and control system is now based on a PDP-11/60 computer. A transfer switch still allows use of either PDP-9 if necessary.

C. Software and Data Reduction

Our automatic readout system (PHARO) allows the computer to read the data stored in a particular analyzer's memory and to read an accompanying series of digits from a panel of thumbwheel switches that identify the sample being counted. The computer stores the data in a file on a secondary storage medium and places this job in a queue of jobs to be processed.

The processing of a spectrum consists of checking the data for possible readout errors, checking the identification to determine which processing options are to be invoked, and then coordinating the execution of these processing options. The options include a channel-by-channel listing, a plot, and magnetic tape storage. A particular user may prefer to give some special treatment to certain NaI spectra. Standards are examined very carefully. Certain parameters are measured, depending on the type of standard, and compared against historical averages. A quality assurance number is printed out for the operator. Ge(Li) spectra may be analyzed by our in-house gamma-ray spectrometry program called RAYGUN. It produces a list of peaks along with their net area. Then, by searching one of several small libraries, the program identifies a nuclide and estimates how much of it is in the sample. These estimates are then converted to disintegrations per

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*Digital Equipment Corp., Maynard, MA 01754.

**California Computer Products Inc., Anaheim, CA 92801.

minute (DPM) at T_0 for the identified nuclide based on known decay-scheme parameters and a predetermined efficiency curve for the particular detector and sample configuration. Table I is an output listing from RAYGUN.

XII. QUALITY CONTROL OF ALL SYSTEMS

A. Introduction

The procedures that follow are designed to control the quality of the data obtained in alpha and beta counting and in gamma and alpha spectroscopy. They insure that the various detectors behave consistently from day to day.

B. Alpha Counting

Uncovered ^{239}Pu standards (plutonium electroplated on platinum disks) and background are counted daily. If abnormal fluctuations in the count rate of the standard are observed, a "plateau" is run, or other special diagnostic procedures are applied.

TABLE I

SAMPLE ANALYSIS FROM RAYGUN

```
SHOT=8317 JUG=60 MASS=113 CTR=76 RCT= 93.851 ZT= 86.492(A0) CT= 93.838(6P) CL=  20.0 CO=60            SN=526

ANALYTICAL RESULTS USING [1,3]ELIB3.DAT

 NUCLIDE           DPM AT     PEAK  PEAK   DPM AT     ATCMS AT
         ENERGY ZERO TIME   FRAC  ERR  COUNT TIME ZERO TIME  ERROR


 1 NA 24 • • • • 7.043E+07 • • • • • • 2.024E+04  9.143E+10  11.9

         1369.  7.043E+07  1.00  11.8
 2 FE 59 • • • • 8.639E+04 • • • • • • 7.706E+04  8.002E+09   6.2

          141.  1.014E+05  0.15  14.2
         1099.  8.698E+04  1.00   8.2
         1292.  8.562E+04  1.00   9.4

 3 MO 99 • • • • 2.979E+04 • • • • • • 4.722E+03  1.712E+08  19.0

          141.  2.979E+04  0.87  14.2
THE FOLLOWING IDENTIFICATION MAY BE AMBIGUOUS

 4 SM153 • • • • 1.938E+06 • • • • • • 1.412E+05  7.832E+09   3.8

          108,  4.629E+07  1.00   5.2
          104.  1.938E+06  1.00   3.7

 5 EU152 • • • • 1.862E+04 • • • • • • 1.061E+04  1.071E+11  18.3

          121,  1.297E+04  1.00  18.8
          412.  2.806E+05  1.00  12.6
          344,  3.829E+03  UPPER LIMIT VALUE ONLY
          779.  5.826E+03  UPPER LIMIT VALUE ONLY
THE FOLLOWING IDENTIFICATION MAY BE AMBIGUOUS

 6 GD153 • • • • 9.894E+04 • • • • • • 9.687E+04  4.973E+10   3.8

          100,  4.450E+04  1.00   5.2
          104.  9.894E+04  1.00   3.7

 9 W 187 • • • • 3.396E+06 • • • • • • 2.033E+04  7.025E+09  11.8

          134.  3.906E+06  1.00  23.0
          479.  3.324E+06  1.00  13.7
          618.  2.114E+06  UPPER LIMIT VALUE ONLY

10 NP239 • • • • 1.768E+06 • • • • • • 2.021E+05  8.629E+09   2.6

          106.  1.671E+06  1.00   3.3
          210,  1.727E+06  1.00  11.8
          228,  1.974E+06  1.00   5.2
          277.  1.682E+06  1.00   5.1
          320.  1.550E+07  1.00   5.5
          334.  1.356E+06  0.93  23.7
```

The plateau, which is determined once a week, is a plot of counting rates versus voltage applied to the central wire anode of the gas detector. From this measurement, we can determine whether overall gain (gas and electronic) has changed. The voltage is increased in 100-V steps and a 5-min count is taken at each step. We look for changes in the shape of the curve. If the plot shows abnormalities (experience permits the ready recognition of abnormalities), proper maintenance steps are taken.

Once a month, a special ^{239}Pu standard is counted on every alpha counter. The decay rate of this standard is known absolutely, and if the counters are operating normally, they must give the correct counting rate. Any counter that does not give the correct count is repaired.

NOTE: In our laboratory, a mode of operation has been chosen that accepts standard deviation limits of ±1.5 as normal statistical variation. If the fluctuations in count rate exceed these limits, a second count is taken; normal limits for this determination are ±3.0 standard deviations.

C. Beta Counting

Daily counts of a ^{234}Pa (U_3O_8 source) standard and background are taken. Except for the fact that no monthly absolute standard count is made, what applies to alpha counting applies here.

D. Gamma Spectroscopy

MCAs with two types of detectors, NaI and Ge(Li), are used for gamma spectroscopy.

1. NaI Detectors. To determine whether the analyzers are operating consistently, ^{137}Cs standards and background are counted daily. The electronic gain of the system (energy span) is adjusted to place the photopeak of the standard in a specific channel. If, on any day, the electronic gain settings for the standard are different from those of the previous day, then the system is examined to determine the reason for the anomalous behavior.

Each day, total counts under the photopeak of the standard are determined. If the system is operating consistently, the number of counts should be approximately the same from day to day. The same statistical considerations are applied as for alpha and beta counting.

2. Ge(Li) Detectors. Standard and background counts are made daily. For these detectors, the standard is ^{152}Eu. The electronic gain of the system is adjusted and checked in the same manner as for NaI detectors.

Two options are available for the absolute calibration of the detectors for intensities versus energies of gamma rays. One makes use of a source of mixed radionuclides obtained from National Bureau of Standards. This source provides absoluely known gamma-ray intensities for calibration purposes. The second option uses an intensity-calibrated ^{152}Eu standard (also supplied by the National Bureau of Standards). The second option is preferable because the single nuclide decays with the same long half-life (the standards can be used for many years) whereas some of the nuclides in the mixed standard have short half-lives (new standards must be obtained yearly).

Deviations in the positions, intensities, and full width at half maximum (energy resolution) are monitored for abnormal fluctuations.

E. Alpha Spectroscopy

Two types of spectrometers with MCAs are used for alpha spectroscopy: Frisch grid and silicon diode. Background counts and the alpha spectrum of a mixed ^{238}Pu and ^{239}Pu standard are taken daily. The important quality control parameters in alpha spectroscopy are (1) the full width at half maximum of the alpha-particle full-energy peak, (2) the ratio of the full-energy peak integrals of the ^{239}Pu and the ^{238}Pu, and (3) the absolute intensity of the ^{239}Pu integral. These parameters are monitored daily for abnormal statistical fluctuations, which indicate that maintenance is necessary.

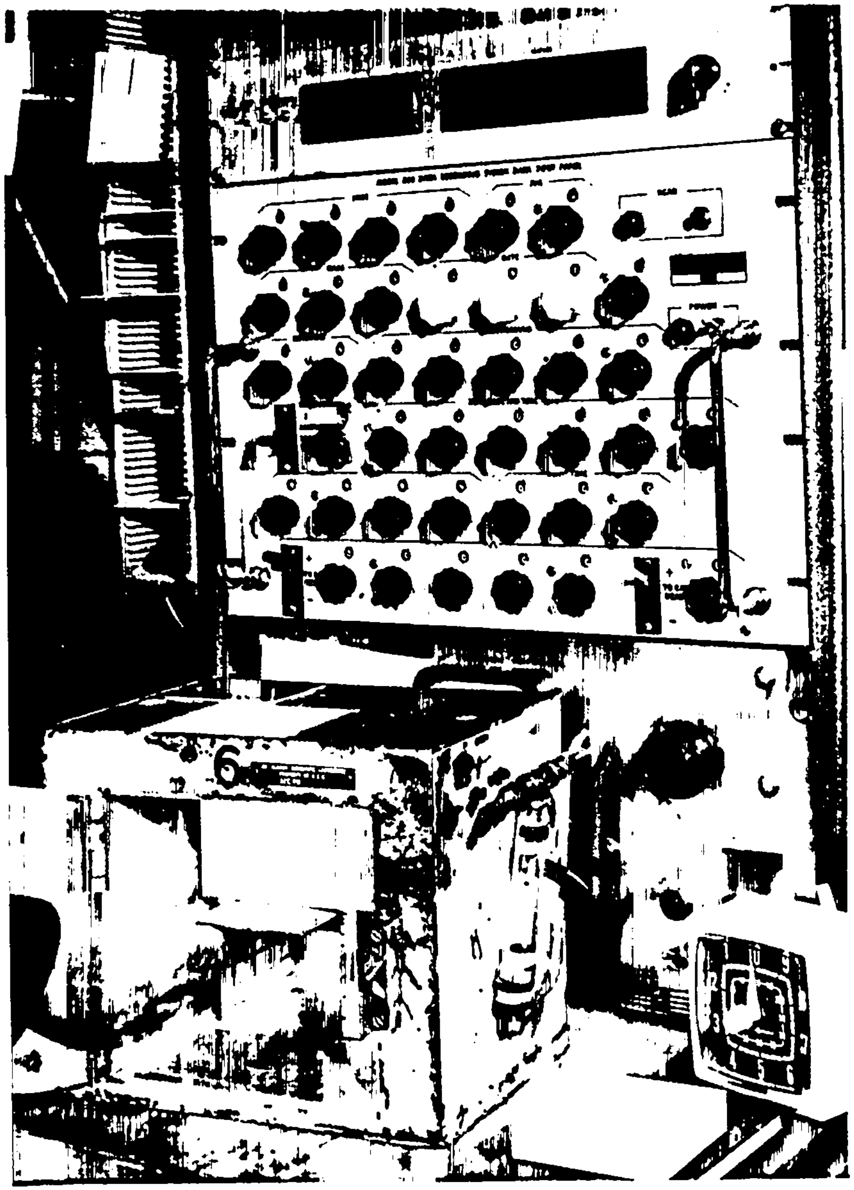

Fig. 1.
Beta counter and readout.

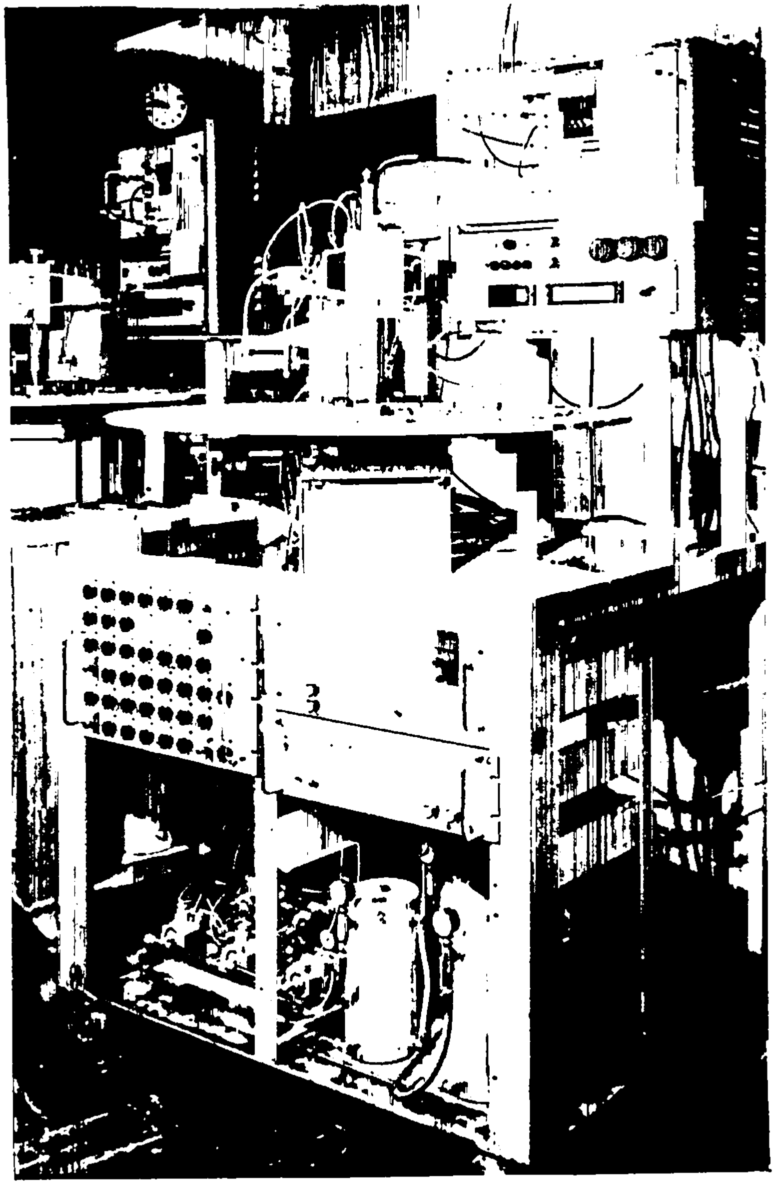

Fig. 2a.
Automatic beta counter.

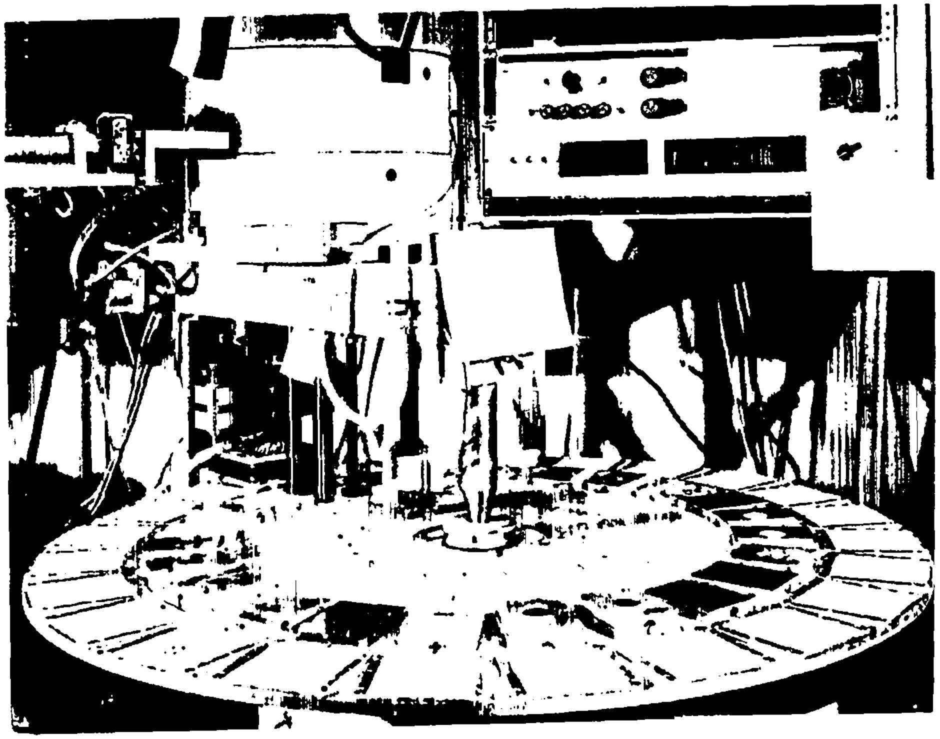

Fig. 2b.
Wheel carrying samples on the beta counter.

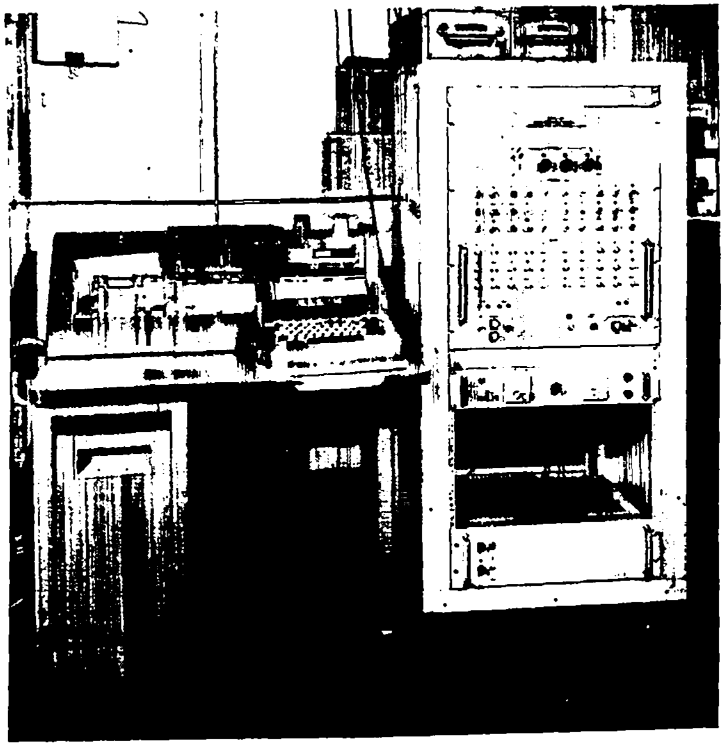

Fig. 3.
Card punch and multiplixer for the automatic beta-counting system.

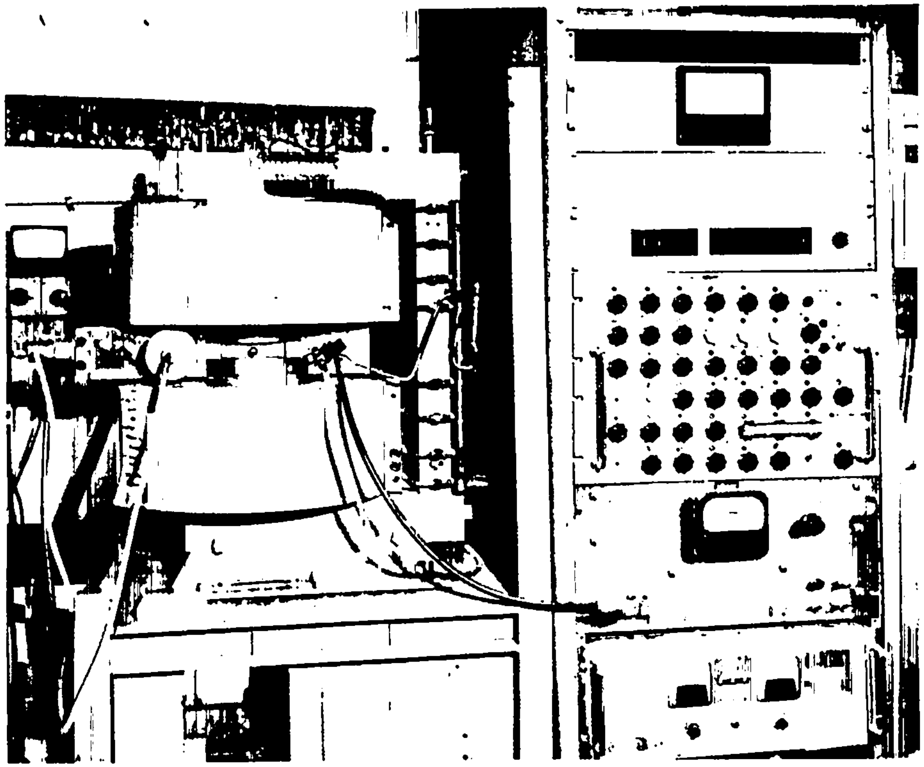

Fig. 4.
Trochoidal analyzer and readout.

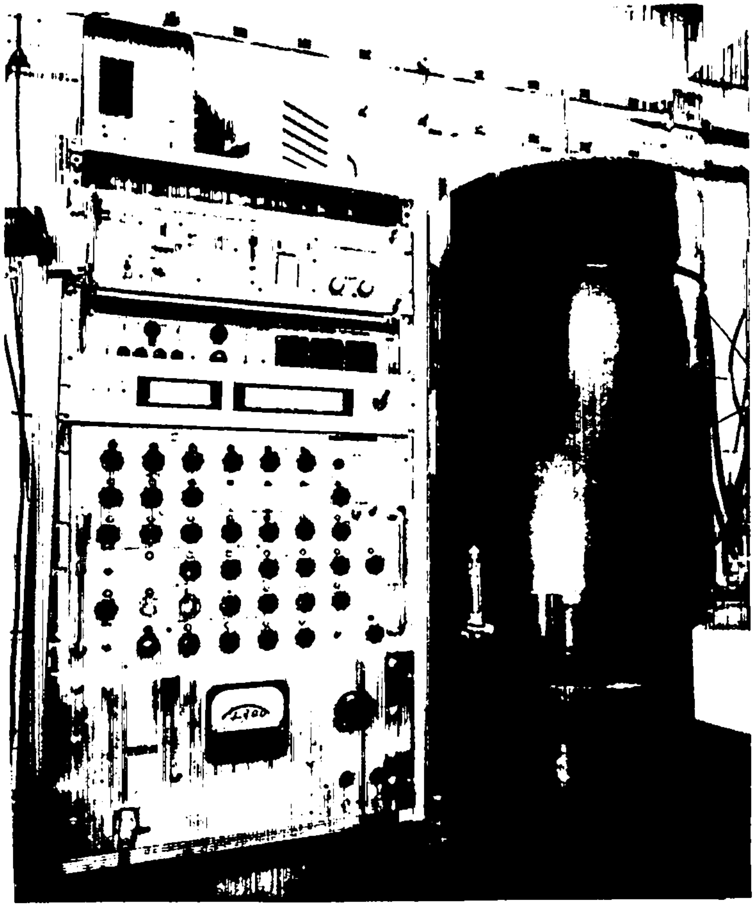

Fig. 5a.
Ultralow-level beta counter and readout.

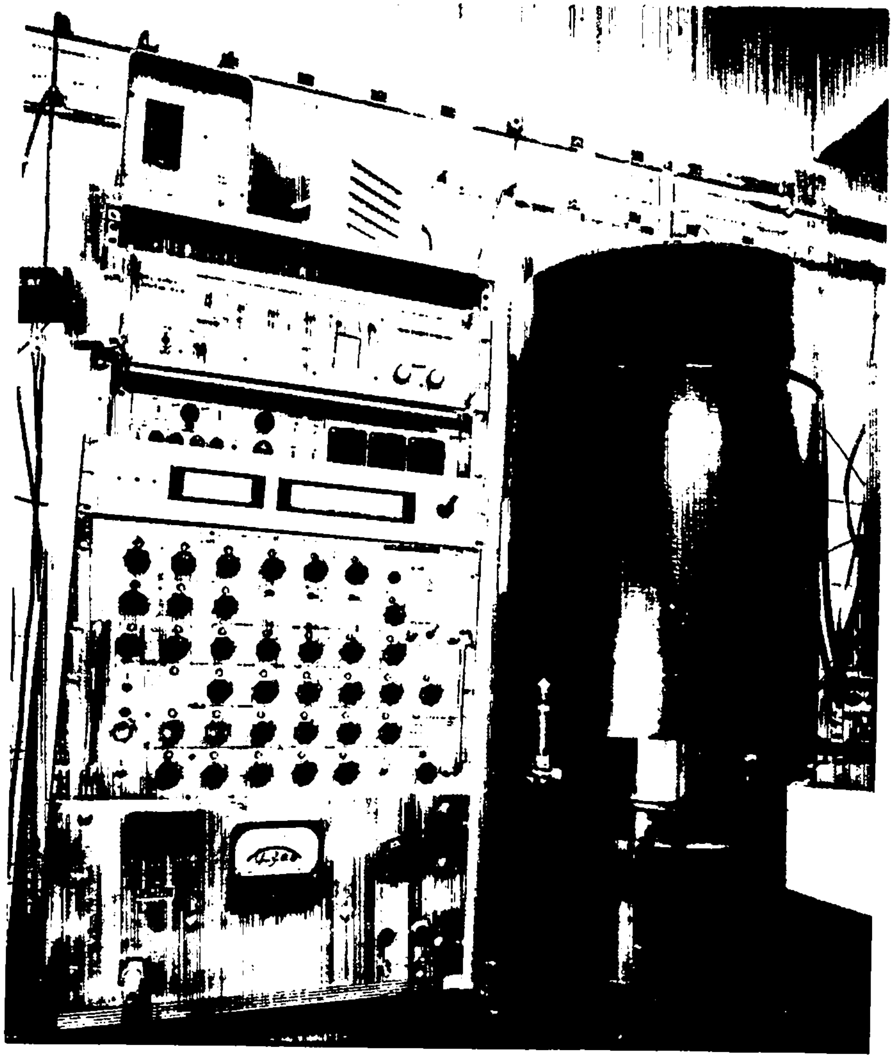

Fig. 5b.
Ultralow background counter, anticoincidence shield in place.

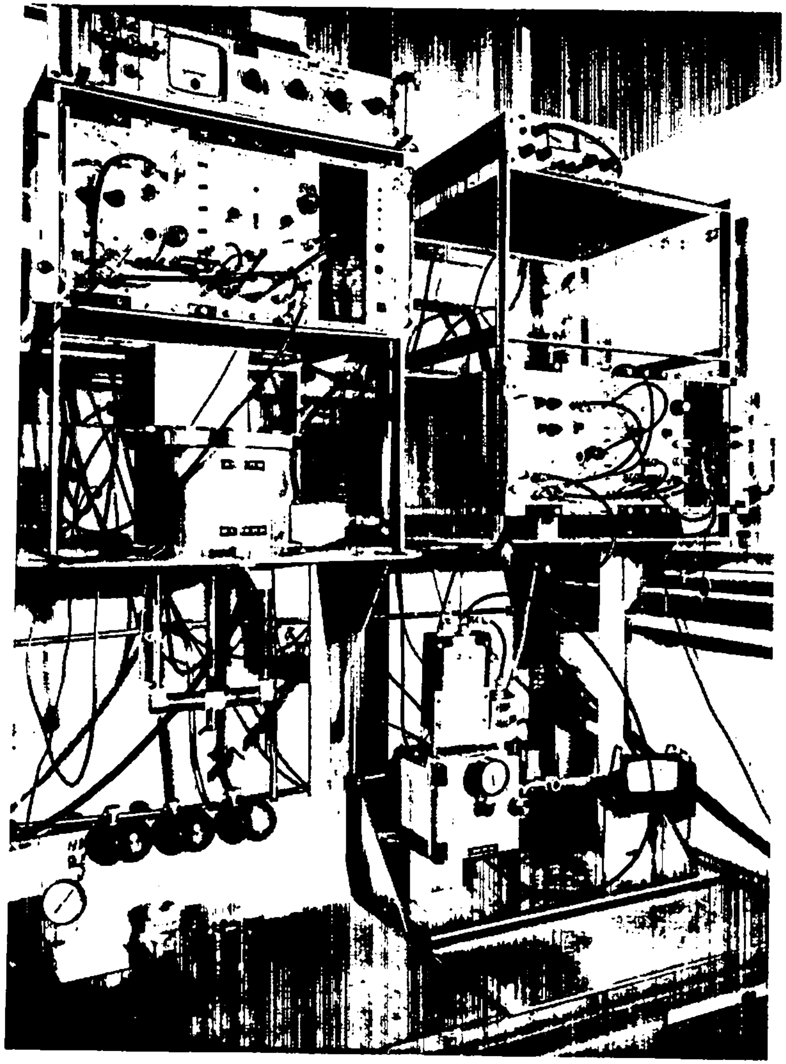

Fig. 6.
Frisch-grid and surface-barrier alpha spectrometers.

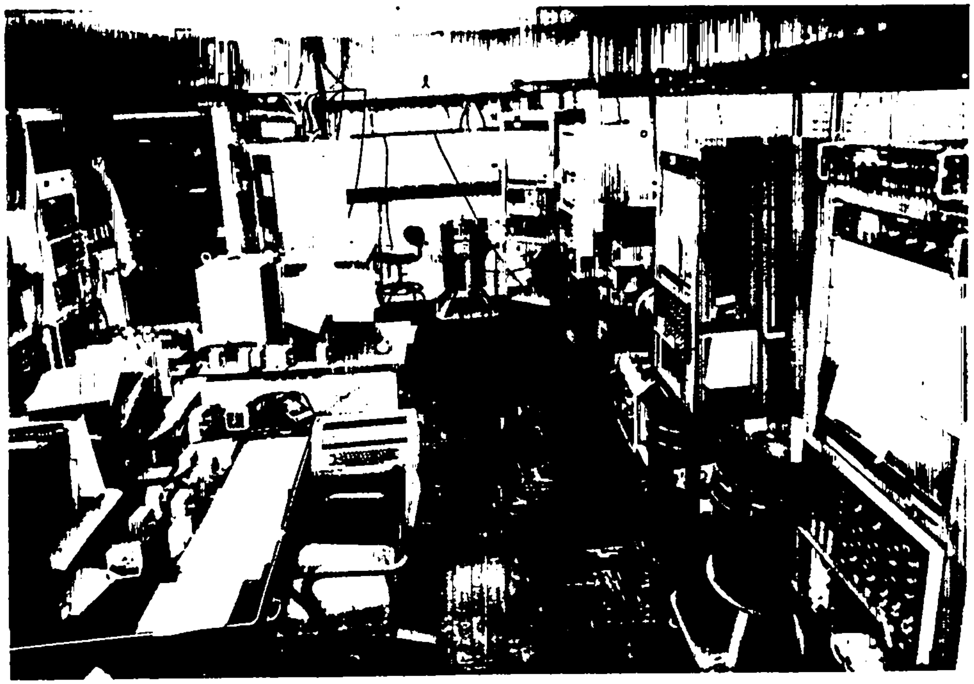

Fig. 7.
NaI gamma-spectroscopy laboratory.

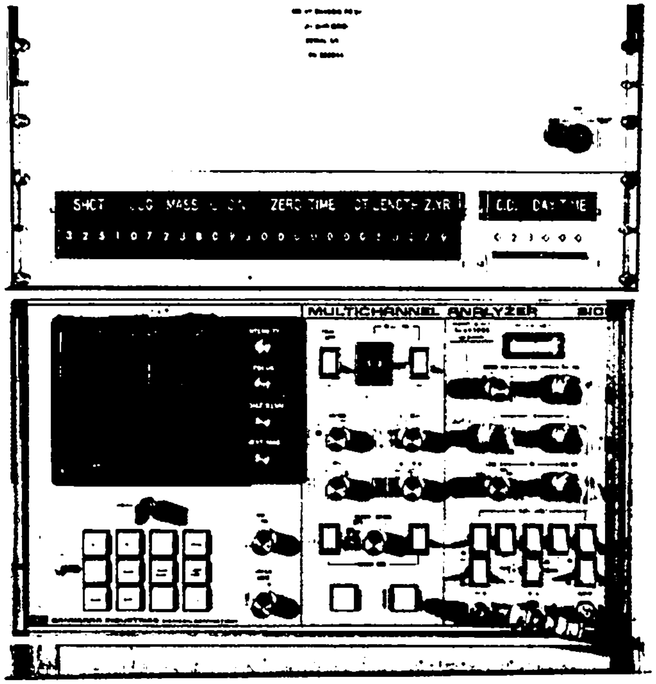

Fig. 8.
Pulse-height analyzer and readout (PHARO) system for spectroscopy.

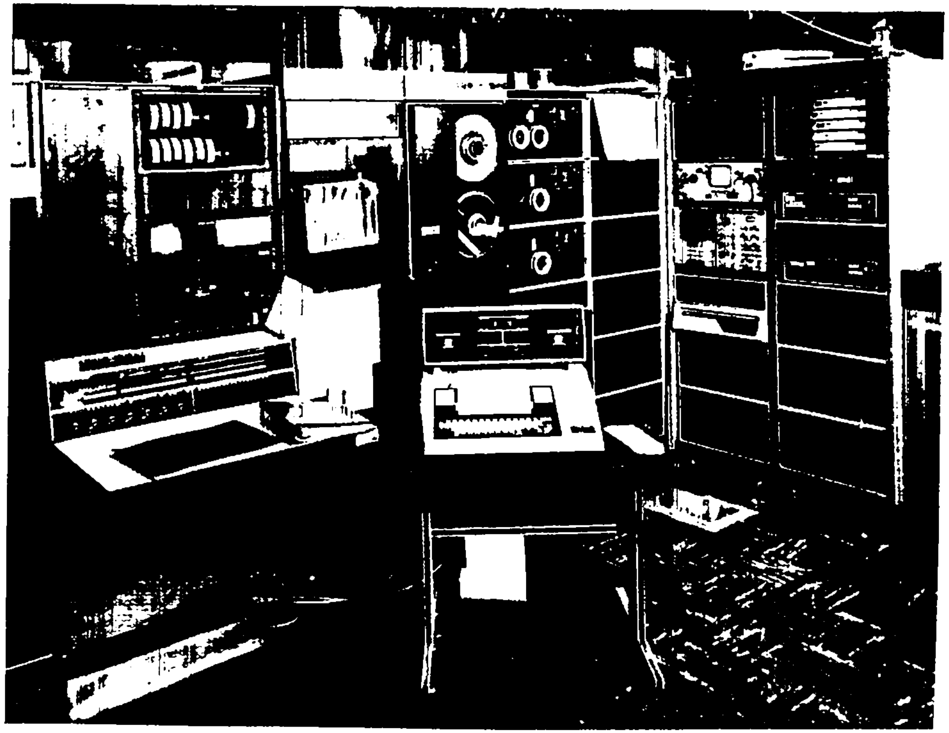

Fig. 9.
Computer for data acquisition and processing.

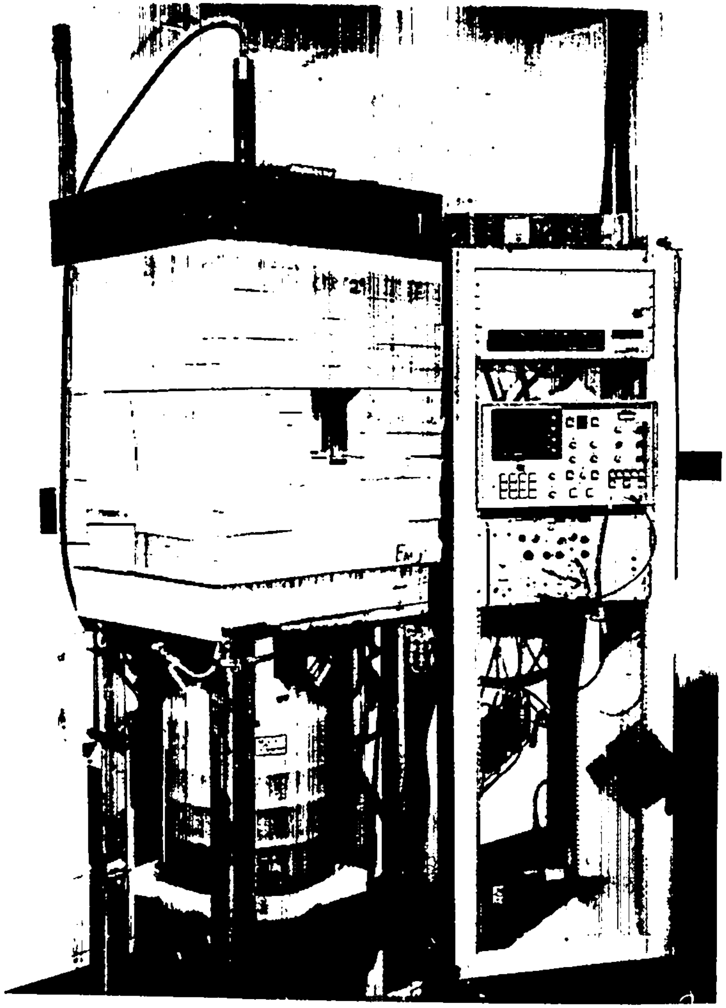

Fig. 10a.
Ge(Li) well detector with anticoincidence shield.

Fig. 10b.
Close-up of the anticoincidence shield for the Ge(Li) well detector.

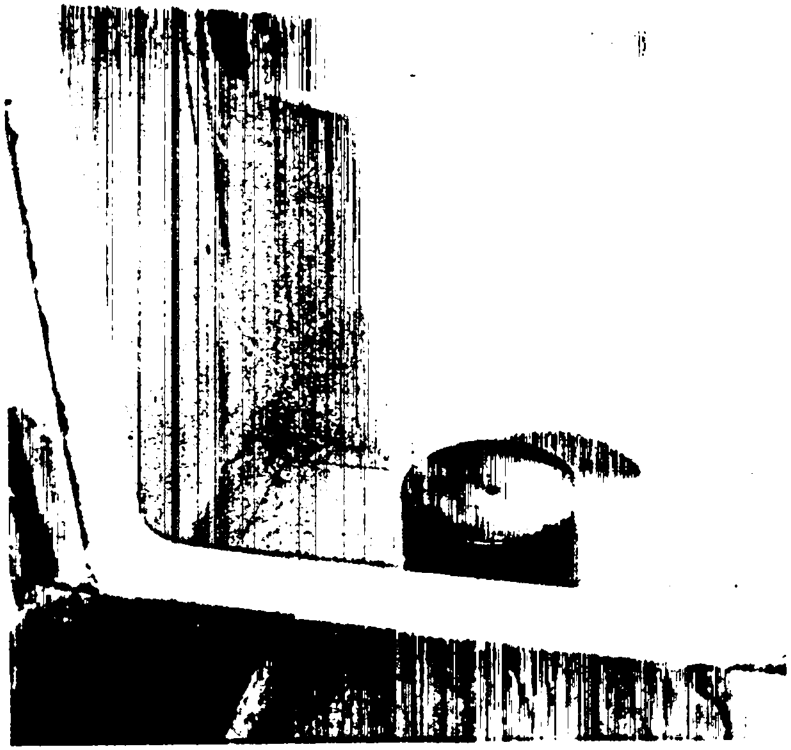

Fig. 11.
Anticoincidence shield of the well detector in the raised position.

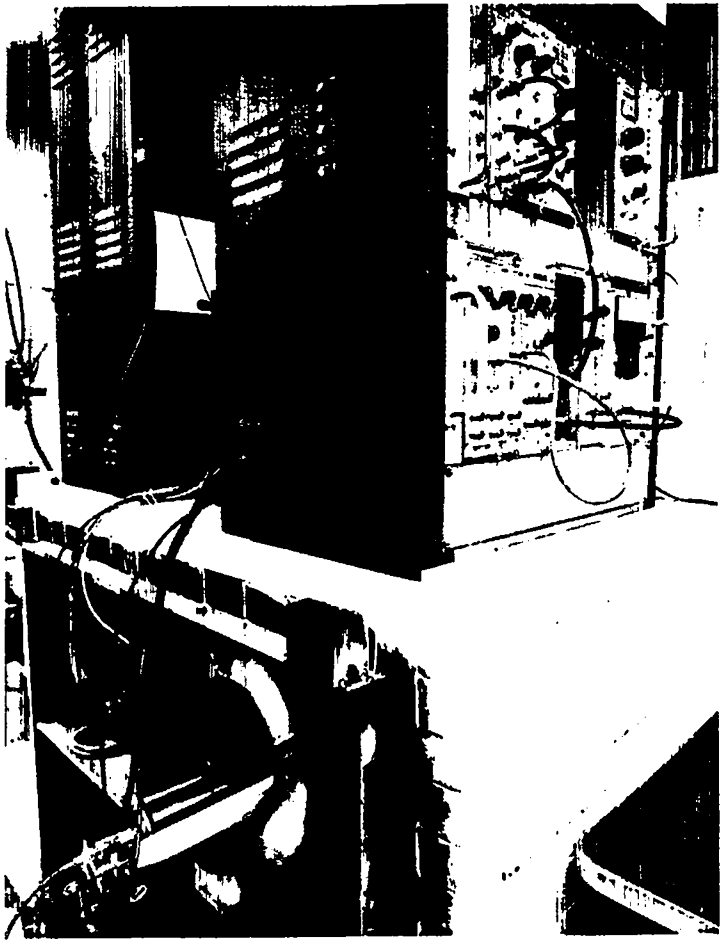

Fig. 12a.
Low-level ^{37}Ar and ^{3}H proportional counting system.

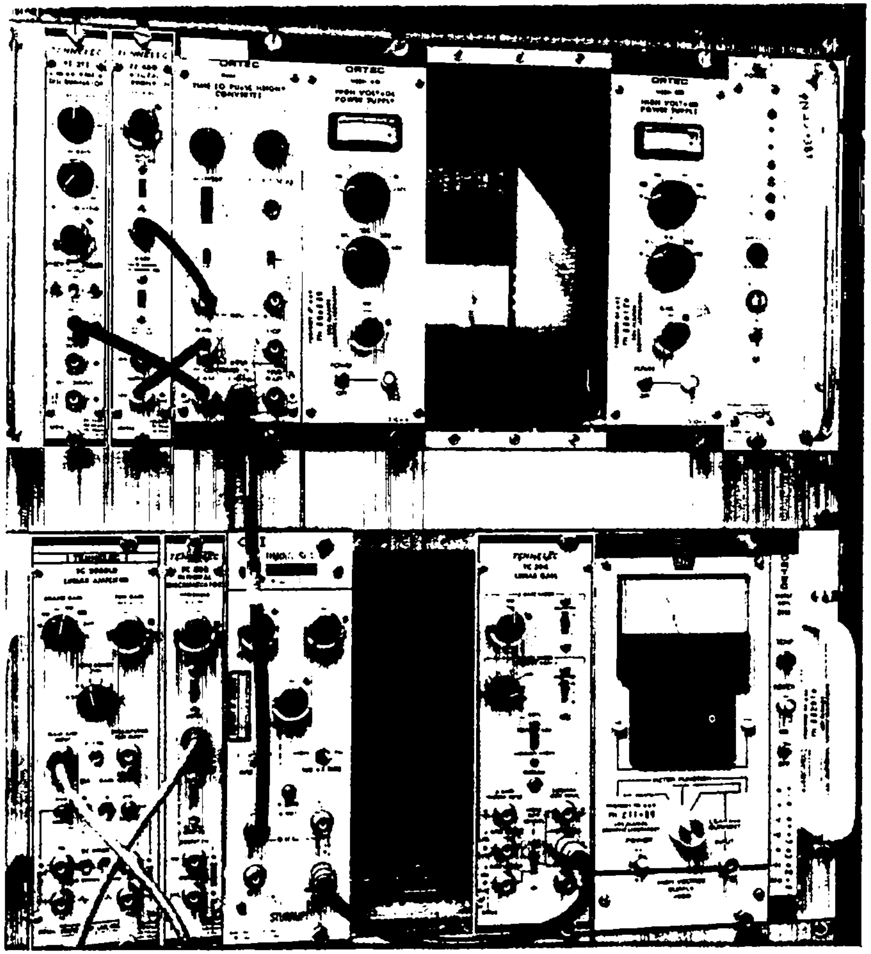

Fig. 12b.
Electronics of the low-level ^{37}Ar and ^{3}H counting system.

Fig. 13.
Automatic sample changers for Ge(Li) gamma spectroscopy.

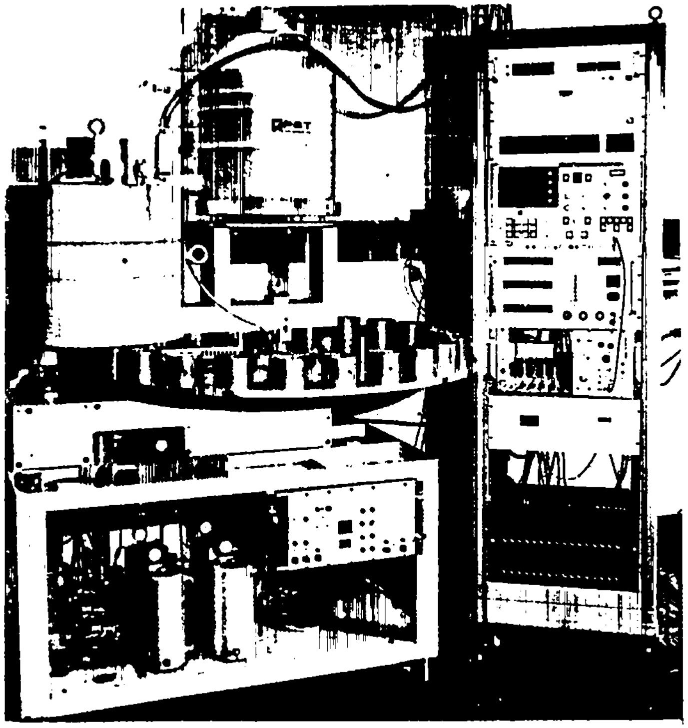

Fig. 14.
Automatic sample changer (wheel type) for Ge(Li) gamma spectroscopy.

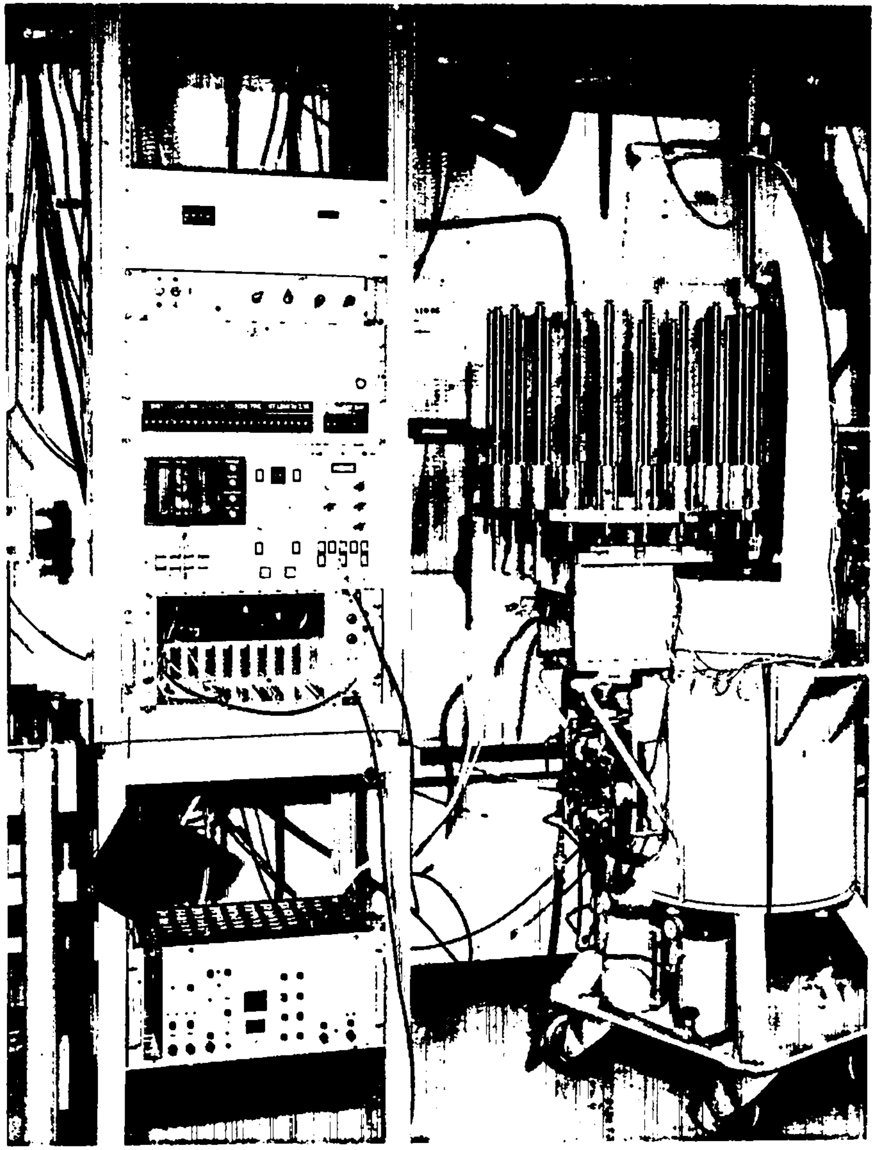

Fig. 15.
Automatic sample changer of the NaI well counter for gamma spectroscopy.

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