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AN AUTOMATIC MASS SPECTROMETER FOR RELATIVE MEASUREMENTS OF URANIUM ISOTOPES

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ABSTRACT

A mass spectrometer has been developed for making automatic relative measurements of uranium isotopes. This report describes the major features which enable the analyst to operate several spectrometers of this type concurrently. The instrument is programmed to make a determination in 13.5 minutes, and a digital ratiometer and printer in the measuring system provide printed data output. The cost of making ratio-of-ratios determinations has been reduced over 40 percent, the variance of a determination has been lowered significantly (by a factor of 2.5 for the measurements usually made), and the cost of maintaining the automatic spectrometers is 30 percent less.

AN AUTOMATIC MASS SPECTROMETER FOR RELATIVE MEASUREMENTS OF URANIUM ISOTOPES

INTRODUCTION

Automation of mass spectrometers in the ORGDP laboratory has resulted in more efficient use of both instrument and analyst time.

Previously, an analyst introduced samples into the spectrometer by manually operating several valves, adjusted a leak to control the ion intensity, continually changed the accelerating voltage to hold the instrument "on peak", manually set decade resistors to obtain isotopic ratio readings when samples were changed, and copied the decade and recorder readings. There was considerable chance for error in making these adjustments and interpreting the recordings.

The major objectives of this project were to increase analyst and instrument production to meet the demands for more analyses, improve precision, and reduce the cost of analyses.

SUMMARY AND CONCLUSIONS

Instrumentation was developed and incorporated in a General Electric, Nier-type (1), six-inch radius isotope mass spectrometer to make its operation completely automatic. With this system an analyst is required only to attach and purify samples, collect printed data, remove the samples, and determine that the instrument is in proper operating condition. Automatic sequencing equipment controls the sample inlet system, adjusts the leak, and activates equipment which prints out the mole ratios for the two samples being compared.

Analyst effort was reduced by stabilizing the supplies to maintain the instrument "on peak" and by installing air-operated manifold valves and a servo-controlled leak. A digital voltmeter with printer output compares the signals from the dc. amplifiers and relieves the analyst of the tedious job of making the measurement and copying data.

Maintenance of the instruments was reduced by incorporating simple circuitry, quality components, and a minimum number of batteries.

The automation and rebuilding of twelve spectrometers has reduced the total analytical costs by more than 40 percent. Spectrometer maintenance has been reduced by 30 percent.

COMPONENTS

One of the most important requirements of an automatic mass spectrometer is that the ion beams be measured only when centered in the collector slit or "on peak." A previous comparison of "peak sweeping" and "on peak" methods of measuring an ion beam showed no significant difference between their precisions; however, the data can be accumulated more quickly by the "on peak" method since no time is lost by repetitive scanning. Therefore the ion accelerating voltage supply and magnet current regulator were designed for maximum stability.

The program control, manifold, and leak control system were designed for maximum flexibility since they would be used on spectrometers measuring a wide range of isotopic concentrations and possibly requiring different time schedules and intensity control levels.

High Voltage Supply and Magnet Current Regulator

The accelerating supply and magnet regulator both control the beam position. To determine minimum stability requirements for "on peak" operation, a vacuum tube voltmeter was installed in the accelerating voltage lead to the source. By measuring accurately the minimum voltage change in either direction required to reduce the maximum peak height perceptibly, the stability requirements of the high voltage supply were determined. The maximum allowable instability was ± 50 millivolts at 1600 volts, or one part in 32,000. At this level of regulation the beam would remain centered in the slit with no observable change in the ratio, assuming no change in the magnet current. By applying similar criteria to the magnet supply and optimizing the regulating characteristics of the magnet and high voltage supplies, it can be shown that the instrument will remain on peak only so long as simultaneous deviations of these supplies are each less than one part in 100,000.

The high voltage supply in figure 1 contains a three-stage regulator utilizing a type 6SL7 tube as a comparison amplifier followed by a type 2C53 voltage amplifier driving another 2C53 as the series regulator (2). A 48 volt error signal is obtained from a 3.5 megohm total load resistance divider which is tapped to provide 48 volts when the output of the supply is 2000 volts. The comparison amplifier compares the error signal to a 48 volt reference battery. The divider is also tapped to provide 2.8 volts which is used as a test voltage to determine the stability of the supply (3).

A resistance divider with a resistance ratio change of less than one part in 100,000 per degree centigrade provides a proportionate sample of the output. The temperature effect is further reduced by placing the divider in a water-cooled oil bath. The 48 volt reference battery is also water-cooled to reduce the effect of temperature changes and increase the battery life. The long-term drift of the supply which normally cannot be corrected by the electronic regulator is reduced to an insignificant factor by the use of a servo amplifier as a drift compensator. The servo compares the error signal to the reference and makes the necessary corrections by adjusting the reference grid of the 6SL7 tube.

The magnet current regulator has the same type of comparison amplifier that is utilized in the high voltage supply and uses an identical servo-system for drift correction. The 48 volt error signal is obtained from a 192 ohm sampling resistor in series with the magnet coils. The resistor is made of manganin wire wrapped around a cylinder which is water-cooled to minimize changes in the error signal. It is also tapped at 10 ohms from ground to provide a 2.8 volt signal for checking stability. The comparison amplifier is referenced to the same water-cooled battery that is used for the high voltage supply. In this manner partial compensation for reference battery drift is obtained. If the instrument could be operated at the point on the magnet saturation curve at which this compensation is a maximum, it would remain on peak with magnet current changes as large as one part in 10,000 (however, this point occurs at a few hundred volts less than normal accelerating voltage). To obtain both a low magnet current and a high accelerating potential, the magnet pole pieces were shaped to concentrate the field.

The high voltage and magnet supplies can usually operate for more than 8 hours with total noise and drift of less than one part in 90,000. With supplies of this stability a spectrometer can remain "on peak" for 8 hours without need for manual adjustment, and "on peak" periods as long as 34 hours have been observed.

Program Control

This unit shown in figure 2 provides the timing of each step necessary for precise measurements, furnishes power for the operation of the manifold, and controls the sequence of the determination. The control is comprised primarily of an Eagle Signal Corporation Multiflex timer, two Automatic Electric stepping switches (models 44 and 45) and six relays. The timer has seven contacts which can be set individually for any time up to five minutes and the time cycle can be changed by replacing two gears. One contact times the pumpout, another the time a sample is in the ion source, and a third delays the operation of the automatic leak. Others activate the printing cycle and provide power for operation of the program control unit.

The model 45 stepping switch is a 25-step, six-level unit which controls the operation of the manifold, and furnishes power to position indicator lights and holding power for the timer.

One determination requires twelve steps, making it convenient to use one revolution of the stepping switch for two determinations thereby increasing the life of the switch.

The model 44 stepping switch is an 11-step, three-level switch which is used to control the printing cycle of the program. After sample admission the timer delays printout for one minute for the intensity and observed ion ratio to stabilize. The stepping coil is in parallel with the entry solenoid of the printer and, when a print pulse is received, the stepper is advanced. After ten prints the stepper "homes" through the eleventh contact and advances the main program to the pumpout position.

The manifold control, although on a separate panel, is an integral part of the program control. Switches for manual operation of individual manifold valves are provided so samples can be changed at the end of a determination. An automatic-manual selector switch disables these switches when in the automatic position thus eliminating the possibility of accidental misoperation of the manifold during a determination.

Sample Inlet System

The arrangement of the manifold is shown in figure 3. Hoke 1384 diaphragm-type valves have been adapted to air operation. Air pressure to the diaphragm-type air motors is controlled by Skinner Electric solenoid valves which are energized by the program control. To avoid leak fractionation differences, one leak is used. This design necessitates evacuating part of the high pressure side of the system between introductions of the samples and maintaining an evacuated "buffer" volume between the two samples to prevent mixing if a valve leaks. The manifold is mounted inside the spectrometer frame with only the sample connectors protruding. It is compact and easily replaced for cleaning or changing the concentration range of the instrument.

Leak Control

Gas flow from the inlet system to the ion source is controlled by an air-operated leak valve with a spring-loaded, air-opened motor. The leak, developed by UCNC at Paducah, is a bellows-sealed, plastic-seated valve with a 0.030 inch port.

The leak control (figure 4) is comprised of a servo amplifier, a 20 rpm. motor and a Moore "Nullmatic" air regulator. A 2:1 gear reduction from the motor to the regulator makes its maximum driving speed 10 rpm. Further reduction of the speed is obtained by adjusting a potentiometer installed across the quadrature windings of the motor.

The signal from the high-current amplifier is attenuated and compared to a reference battery by the servo-amplifier. The level of the ion beam intensity can be adjusted with a potentiometer which changes the amount of attentuation. The speed control can be used to match the response time of the leak to the system, to allow the use of maximum gain with little "hunting".

To avoid initial intensity surges when sample is admitted to the spectrometer the leak operation is delayed about 6 seconds by the program control, but remains active for the remainder of the "sample-in" period. In the event of excessive pressure rise in the analyzer during a determination, the leak is disabled by a pressure-sensing relay in the vacuum gage panel. This prevents the leak from opening excessively and possibly "flooding" the source with sample. If the leak is disabled in this manner a buzzer warns the analyst.

Emission Regulator

The emission regulator was revised as shown in figure 5 to provide better regulation. Two type VR75 tubes in the original circuit were replaced by one VR150; another stage was added to the regulating circuit for increased gain and a type 6AS7 tube was used in the output.

Pressure Gages

The pressure gage for the source region is a Penning type (5) which is equipped with a meter relay in the pressure-indicating circuit. In case of a slight pressure increase the meter relay contacts close, de-energizing the leak and setting off an alarm buzzer. The current of the electronic pump on the analyzer tube is used as an indication of that pressure. The manifold pressure gage is a conventional Pirani type gage.

Measuring System

The measuring system is designed around single-stage preamplifiers and identical dc. amplifiers for both high and low-current channels. Each preamplifier uses a type 5886 electrometer tube mounted in a magnetic shield, and both are inclosed in an evacuated housing mounted directly on the collector envelope. At the isotopic level of natural uranium, a 2×10^{10} ohm input resistor in the high-current preamplifier results in a high-current amplifier output of 10 volts; the corresponding low-current input resistor is 10^{11} ohms.

The necessity of manually adjusting decade resistors to obtain ratio readings was eliminated by the use of a Non-Linear Systems Model 550 five-place digital ratiometer. The ratiometer is a stepping switch type with a sensitivity of 100 microvolts when 10 volts is used as a reference. Since a ratiometer has no standard reference voltage, the 10 volt signal from the high-current amplifier output is the reference signal. When the ratio of the outputs of the two amplifiers remains balanced within ± 100 microvolts for about two seconds, a print control circuit in the ratiometer transmits an impulse to a Model 1910 Clary Printer which prints the ratio function. When ten ratios are accumulated they are totaled and the program advances to the next step. Thus, chart reading and averaging errors are eliminated.

When the sample is removed from the spectrometer the ratiometer loses its reference voltage and would normally continue to seek a null balance, causing excessive wear on the stepping switches. To prevent this, a contact in the program control timer de-energizes the print control system and "locks" the ratiometer except during the printing cycle.

Analyzer System

The analyzer tube rack is constructed of 3/8" thick aluminum sheet with one inch brass rods for spacers to provide rigid and easy mounting of components.

The analyzer is equipped with differential pumping using an 80 liter per second mercury diffusion pump for the source region and an eight liter per second electronic pump for the tube, with the source exit slit acting as the differential pressure slit. The high speed pump on the source combined with differential pumping reduces the effect of sample impurities on the ratio (4).

The collector envelope is made of stainless steel and is designed so the preamplifier housing mounts directly on it and forms the bottom flange. The high and low-current collector leads are very short; in fact, the low current collector is mounted directly on a Stupakoff seal feeding into the preamplifier, thereby making it possible to obtain a very low noise level. The preamplifier housing is easily removed for ready access to the collector. Slit adjustments are made by means of a bellows and retaining cap. A unique system of ball joints and screws provides precise and rigid positioning of the source magnet.

PROCEDURE

The programming of a determination is shown in figure 6. When the ratio of sample A to sample B*is to be determined the analyst sets the automatic-manual switch of the manifold control to the "manual" position, attaches samples, evacuates the connectors, pumps the samples to remove impurities, and determines that sample will enter the spectrometer with no pressure rise. The automatic-manual switch is then returned to "automatic" and the sequence is started by pushing the "start" button. Two isotope ratio readings of sample B bracketed with three readings for sample A constitute the normal amount of data collected for a single determination. The first step in the program is used as a conditioning step for sample B so that the readings for A and B will be affected equally by each other and a valid memory correction can be applied. No data are taken from this entry. Sample B is then pumped out for 30 seconds and sample A admitted. After a six-second delay, the leak starts controlling and, after a one-minute print delay to stabilize the pressure in the source region, the ratiometer is "unlocked" and the printing cycle is started. After ten prints are accumulated, sample A is pumped out, the printer totals, and the program continues for four more sample entries. The observed isotopic ratio of the two samples is then determined according to the formula in figure 6. A typical determination is shown in figure 7.

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*Sample B must be a known standard if percentage composition of sample A is to be determined.

PERFORMANCE

Twelve automatic spectrometers have been assembled as shown in figure 8 and used for about four years. On the type of measurements made most frequently, the ratio-of-ratios variance per determination has been reduced from approximately 25×10^{-8} on the manual instruments to about 10×10^{-8} on the automatic type. Prior to the automation program an analyst usually made less than 15 determinations per shift; he can now easily attend three or four spectrometers, average more than 40 determinations per shift, and may occasionally run 80.

The improved pumping system, leak control, and automatic programming which reduced misoperation have increased ion source life by about 40 percent which, along with the improved circuitry, has resulted in a 30 percent decrease in maintenance costs.

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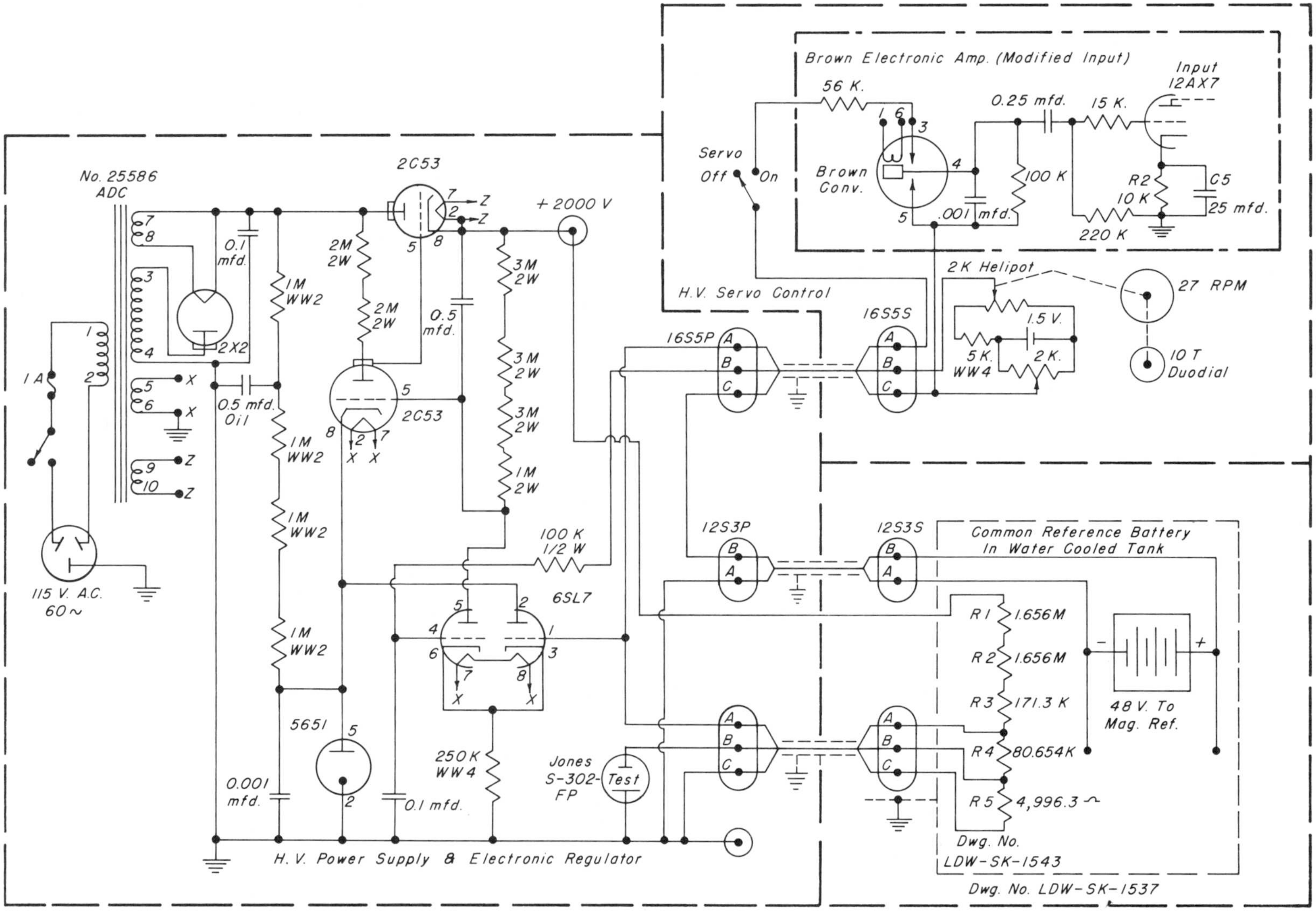


Figure 1

HIGH VOLTAGE POWER SUPPLY & SERVO

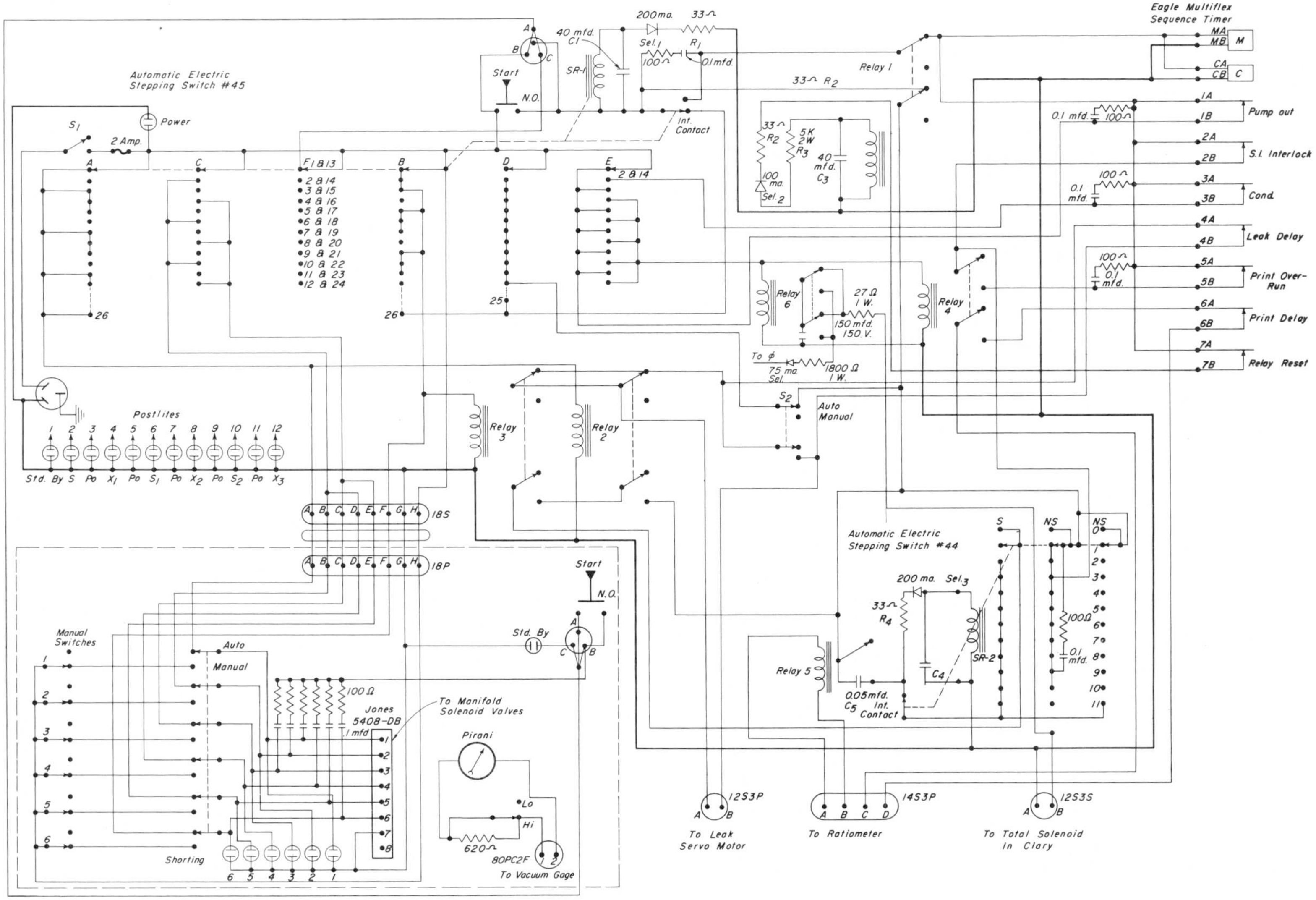
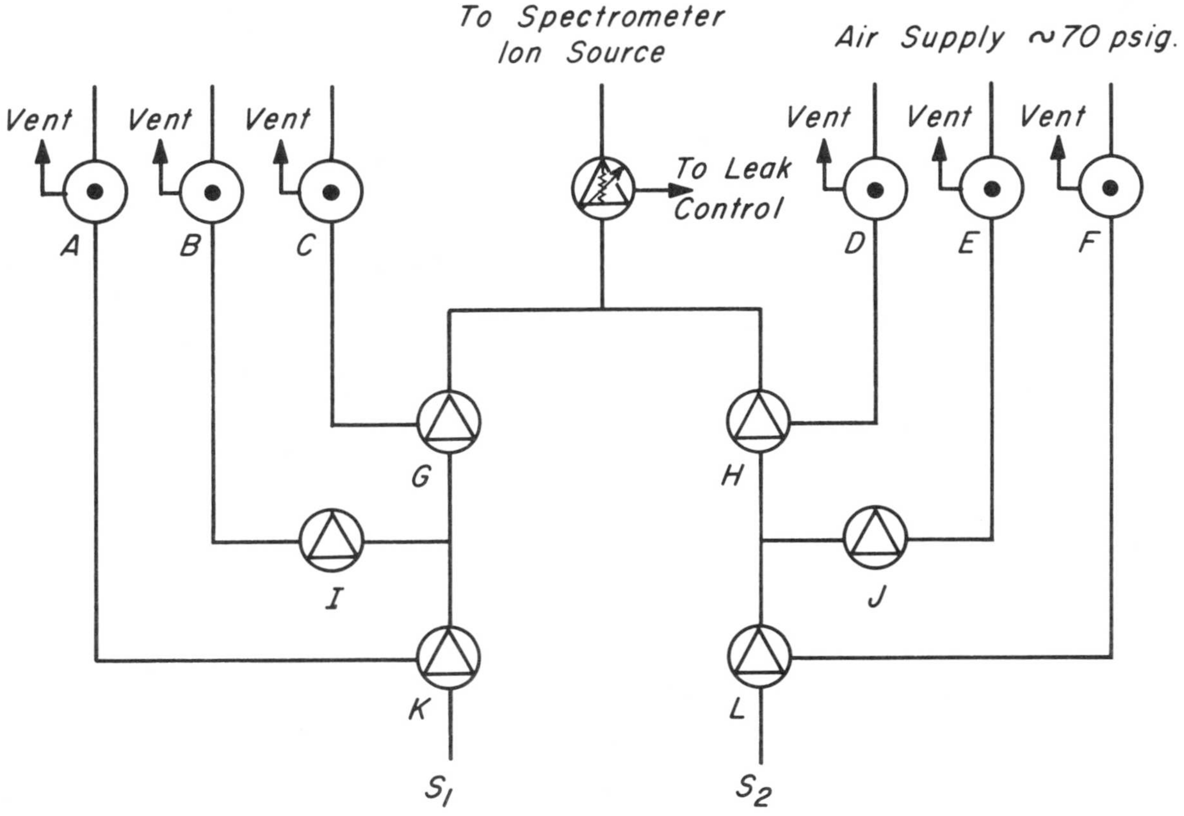


Figure 2

PROGRAM CONTROL AND AUXILIARY MANIFOLD CONTROL



LEGEND:

A, B, C, D, E, F - Skinner Electric Valves

G, H, I, J, K, L - Special Pneumatic Valves

Figure 3

SAMPLE INLET SYSTEM

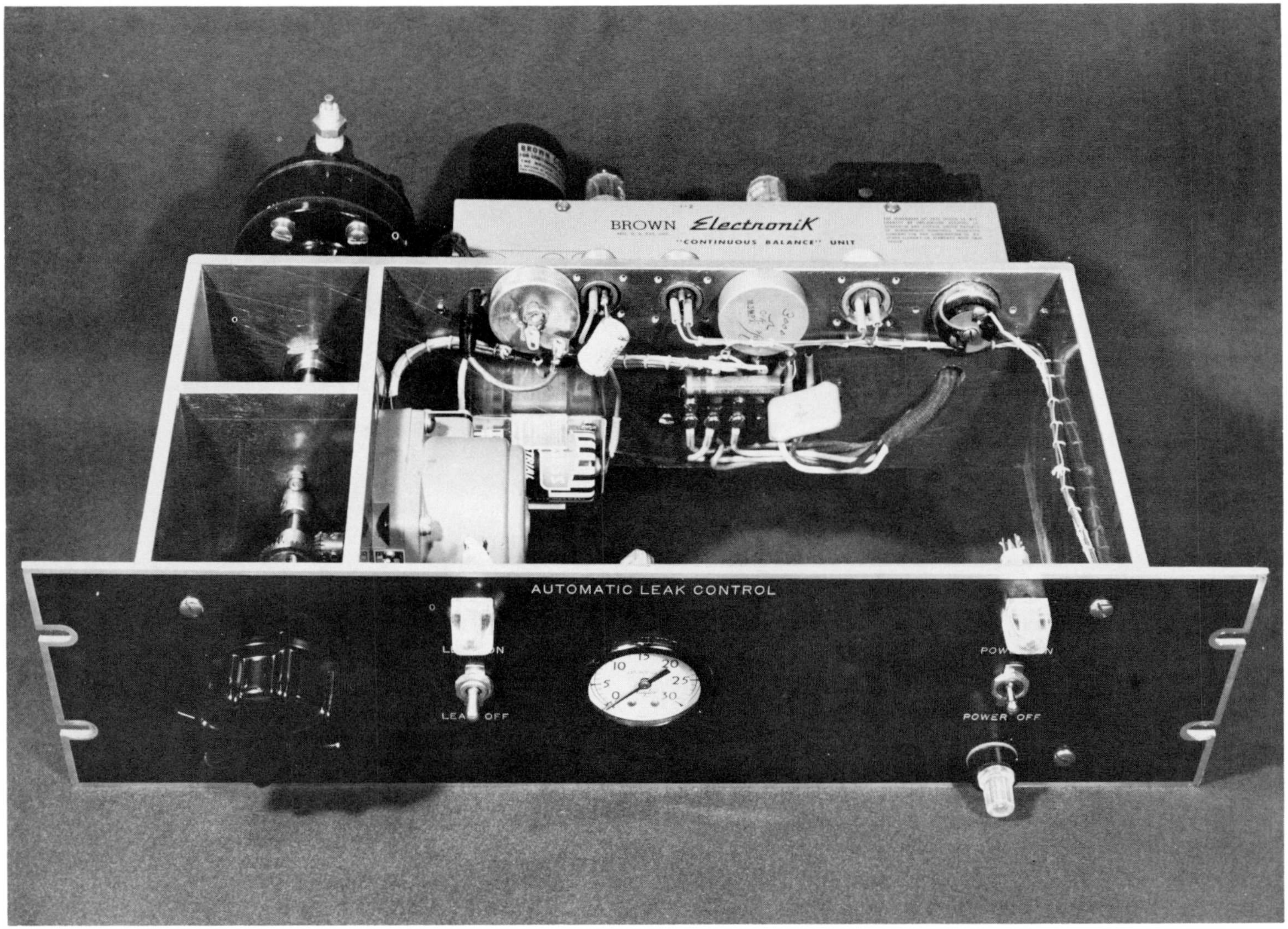
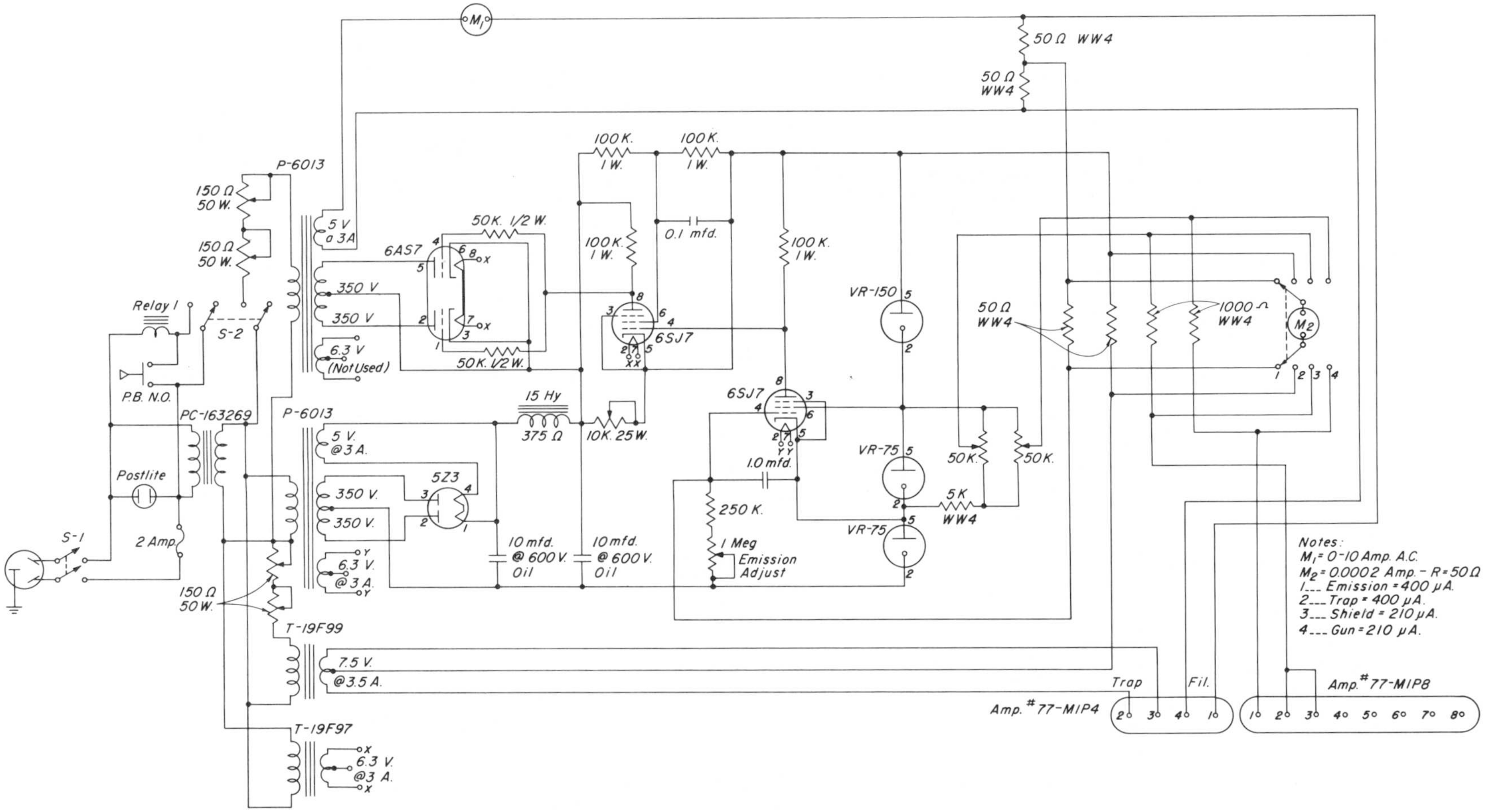


Figure 4

AIR-OPERATED LEAK SYSTEM



Notes:
M_1 = 0-10 Amp. A.C.
M_2 = 0.0002 Amp. - R = 50 Ω
1___ Emission = 400 µA.
2___ Trap = 400 µA.
3___ Shield = 210 µA.
4___ Gun = 210 µA.

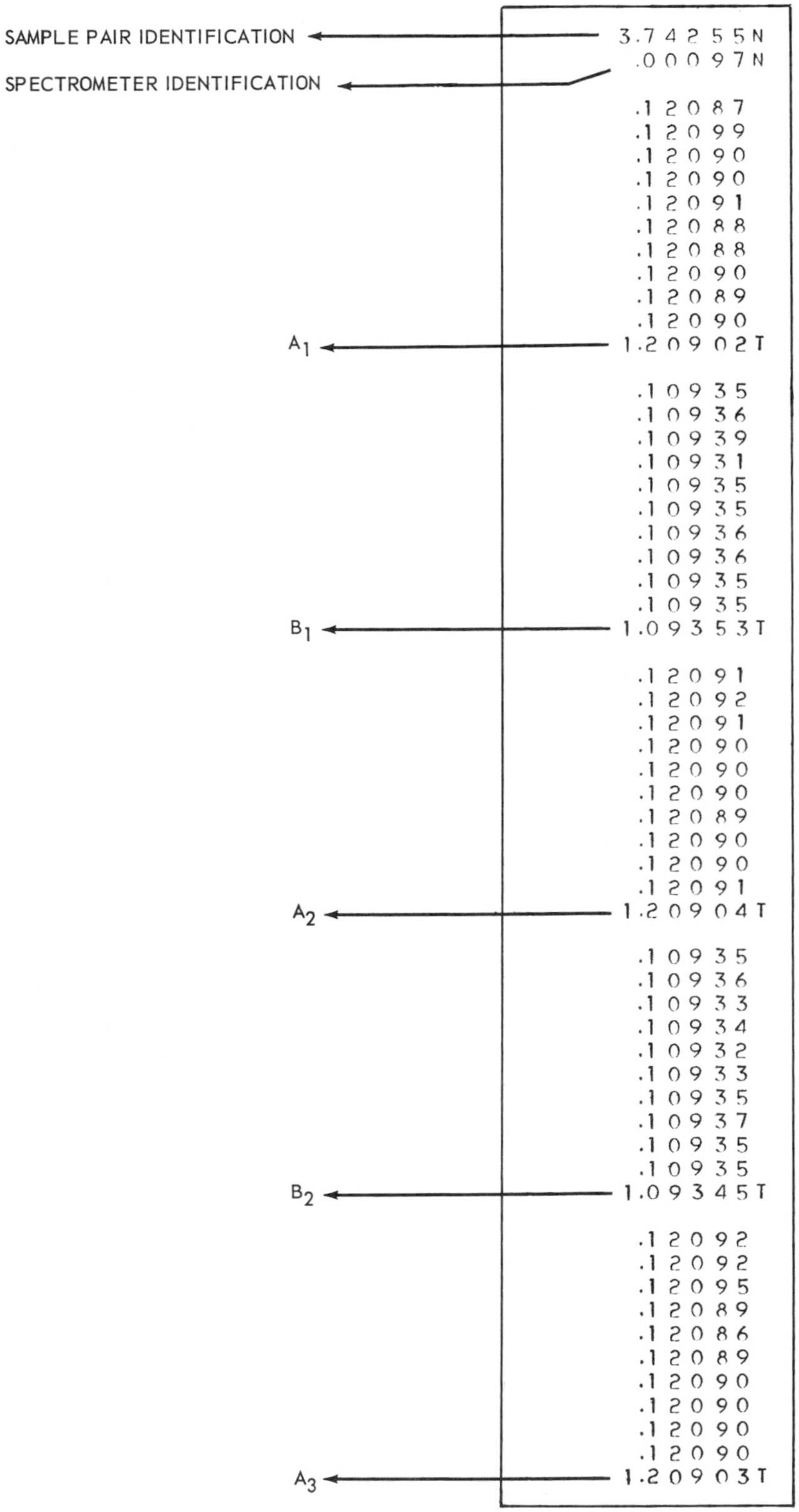
Figure 5
EMISSION REGULATOR

		MINUTES
MANUAL	Attach Samples	
	Evacuate Connectors	
	Pump Samples	
	Start Sequence	
AUTOMATIC	Sample B (No data taken in this step)	1.0
	Pump Out	0.5
	Sample A_1	2.0
	Pump Out	0.5
	Sample B_1	2.0
	Pump Out	0.5
	Sample A_2	2.0
	Pump Out	0.5
	Sample B_2	2.0
	Pump Out	0.5
	Sample A_3	2.0
	Standby	
	Total	13.5

$$\frac{\dfrac{A_1 + A_2 + A_3}{3}}{\dfrac{B_1 + B_2}{2}} = R_o = \frac{\left(\dfrac{U^{235}}{1 - U^{235}}\right)_A}{\left(\dfrac{U^{235}}{1 - U^{235}}\right)_B}$$

Figure 6

STEPS IN DETERMINATION OF RATIO A/B



```
 3.7 4 2 5 5 N
  .0 0 0 9 7 N

  .1 2 0 8 7
  .1 2 0 9 9
  .1 2 0 9 0
  .1 2 0 9 0
  .1 2 0 9 1
  .1 2 0 8 8
  .1 2 0 8 8
  .1 2 0 9 0
  .1 2 0 8 9
  .1 2 0 9 0
 1.2 0 9 0 2 T

  .1 0 9 3 5
  .1 0 9 3 6
  .1 0 9 3 9
  .1 0 9 3 1
  .1 0 9 3 5
  .1 0 9 3 5
  .1 0 9 3 6
  .1 0 9 3 6
  .1 0 9 3 5
  .1 0 9 3 5
 1.0 9 3 5 3 T

  .1 2 0 9 1
  .1 2 0 9 2
  .1 2 0 9 1
  .1 2 0 9 0
  .1 2 0 9 0
  .1 2 0 9 0
  .1 2 0 8 9
  .1 2 0 9 0
  .1 2 0 9 0
  .1 2 0 9 1
 1.2 0 9 0 4 T

  .1 0 9 3 5
  .1 0 9 3 6
  .1 0 9 3 3
  .1 0 9 3 4
  .1 0 9 3 2
  .1 0 9 3 3
  .1 0 9 3 5
  .1 0 9 3 7
  .1 0 9 3 5
  .1 0 9 3 5
 1.0 9 3 4 5 T

  .1 2 0 9 2
  .1 2 0 9 2
  .1 2 0 9 5
  .1 2 0 8 9
  .1 2 0 8 6
  .1 2 0 8 9
  .1 2 0 9 0
  .1 2 0 9 0
  .1 2 0 9 0
  .1 2 0 9 0
 1.2 0 9 0 3 T
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Figure 7

PRINTED TAPE - ONE DETERMINATION

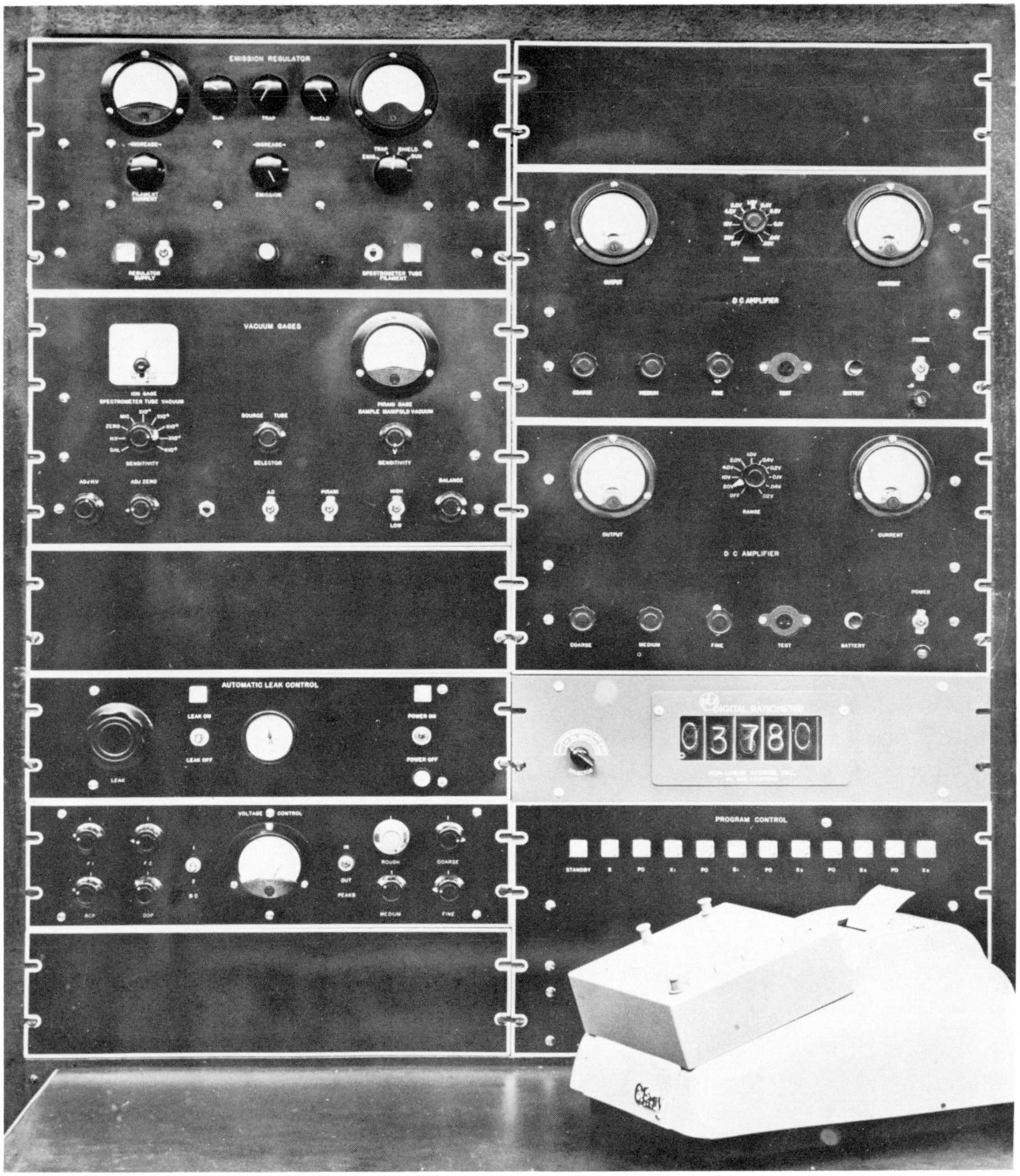

Figure 8

AUTOMATIC, RELATIVE SPECTROMETER