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A LOW LEVEL RADON COUNTING SYSTEM

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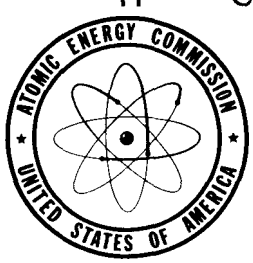

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ABSTRACT

A stable, convenient system for low level counting of radon is described. The system permits a precision of ±1% with an efficiency of about 80%. The lower limit of measurement is the amount of radon in equilibrium with about 10^{-14} grams of radium. A vibrating reed electrometer is used to amplify slow pulses obtained from an ionization chamber operated at 300 volts. The cumulative pulse count is recorded by a mechanical register following amplification of the output of the electrometer. Special care in construction and selection of the ion chamber materials permits an alpha background as low as 15 cph for a 4-liter volume. Chemical pre-treatment of the gases is unnecessary for most samples.

INSTRUMENTATION

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INTRODUCTION

Many experimental arrangements have been proposed for counting radon gas. The earliest method[1] involved integration of ion current produced by radon and its solid decay products by means of an electrometer. This method was satisfactory for high radon concentrations but for low level work it does not have the required electronic stability and displays large fluctuations in background. This latter effect was reduced by using (Evans[2] and Urry[3]) two ion chambers back to back to compensate for such fluctuations. For the long time periods required for measuring low level samples the apparatus provided for automatic voltage calibrations and photographic recording of the trace. This integration method is cumbersome and is not as sensitive or as stable as some of the later techniques.

At this stage in the development of radon measurement, two different methods were followed. One involved the use of the ion chamber as a proportional counter at high voltage (~2000 volts), thus requiring a high gain, fast amplifier. This system permits high

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1. E. Rutherford, Radioactivity, 2nd Ed., Cambridge University Press, London (1905).

2. R. D. Evans, Rev. Sci. Instr. 8, 99-102 (1935).

3. W. D. Urry, J. Chem. Phys. 4, 40-48 (1930).

level as well as low level counting. It was proposed first by Curtiss[4] and most recently improved by Hudgens, et al[5] so that the 10^{-14} grams Ra range could be reached. The other method was to employ the ion chamber as a low voltage ion-collection device and with large amplification to detect pulses rather than to integrate the ion current. Two detecting systems have been employed: (a) earlier the output from an FP-54 electrometer tube was recorded photographically from a Leeds and Northrup Type R galvanometer in a balanced bridge circuit[6]; (b) more recently, a vibrating reed electrometer has been utilized with a Brown pen recorder[7].

Both high voltage and low voltage chambers have given satisfactory performance in low level radon counting. The high voltage method requires rather elaborate chemical clean-up of the gases to be used. This type of counter is particularly sensitive to water and acid vapors, carbon dioxide and oxygen. The long-range stability required in the high voltage method places severe restrictions on the electronic design and construction of the high voltage supply and the accompanying high gain, fast amplifier. The main advantage of this

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4. L. F. Curtiss and F. J. Davis, J. Research, Nat'l. Bur. Standards 31, 181-196 (1943).

5. Hudgens, Benzing, Cali, Meyer and Nelson, Nucleonics 9, 14-19 (1951).

6. G. D. Finney and R. D. Evans, Phys. Rev. 48, 503-511 (1935).

7. P. M. Hurley, Dep't. of Geology and Geophysics, M. I. T. (private communication).

system appears to be the range of activity which it can handle. It was designed by Hudgens, et al.[5] for use with radon concentrations ranging from those of radioactive ores down to ordinary rocks. For this purpose it is admirably suited. For the lowest levels of radon concentration in naturally occurring substances the inherent difficulties in this system appear greater than in the low voltage method.

The low voltage method as described above has one serious handicap. The thousands of alpha pulses per run (if the counting error is to approach 1%) must be counted visually. This is a demanding task if many determinations of high precision are to be made. On the other hand, for special studies where the size or shape of peak can be used to differentiate various isotopes or to determine the location of radiation[8], the pen record is valuable.

The system described below is based on the low voltage ionization chamber - vibrating reed electrometer arrangement. It not only combines the features of electronic stability and simplicity of chemical pre-treatment but, most important, includes an electronic circuit for totalizing the slow pulses which are passed by the electrometer. The system has given satisfactory performance for over a year with an efficiency of about 80%, and a very low background.

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8. P. M. Hurley, Bull. Geol. Soc. Am. 61, 1-8 (1950).

APPARATUS

Ionization Chamber

A detailed drawing of the ionization chamber is shown in Figure 1. The 4 liter chamber is constructed of carefully selected stainless steel to obtain the lowest possible background. The dimensions are not critical. The use of multiplace "o" ring seals and metal stopcock and ball joint makes the chamber very rugged and easily disassembled for cleaning. The size of the insulators prevents appreciable surface leakage under the normal operating conditions.

General Circuitry

The basic components of the detecting and counting system are shown in the block diagram of Figure 2. The detecting arrangement is quite conventional, consisting of a large grid resistor R in series with the ionization chamber and supply voltage V. The ionization current I gives rise to a pulse of magnitude IR, which is then amplified for recording purposes. When the center probe is connected to the negative side of V, the reed delivers a negative-going pulse.

The choice of R is determined primarily by the time constant R'C, where R' (composed primarily of R) is the effective resistance to ground, and C consists of the distributed capacitance of the input grid and collecting-electrode system. R'C should be small enough so that with the maximum average rate of arrival of pulses there is small

probability of one following another within the time R'C; on the other hand, R'C should be large enough to permit the full voltage pulse to be developed. C is fixed by the geometry of the detecting circuit, and with the system described the optimum value of R was found to be 2×10^{10} to 3×10^{10} ohms. However, over an even wider range R was not found to be critical, and values ranging from $R=10^{10}$ to 4×10^{10} ohms were found usable. Resistors manufactured by the Victoreen Instrument Company of Cleveland, Ohio, were employed. These can be conveniently mounted directly in the input sleeve of the electrometer-head, or in a special adapter provided specifically for the Victoreen-type resistor.

The supply voltage V was not found to be critical, and was somewhat arbitrarily taken at 300 volts. Voltages as low as 90 volts were found to give comparable results. New commercial B-batteries gave good service for periods as long as twelve months and although the system is very sensitive to small operational variations in the battery voltage, it was not found necessary to attempt improvement of the stability of the B-battery supply. With aging, erratic fluctuations of battery voltage eventually set in, quite spontaneously, and the necessary battery change is indicated by the appearance of large amplitude "noise" in the electrometer output.

The model 30 vibrating reed electrometer manufactured by the Applied Physics Corp. of Pasadena, California, is suitable for amplification of pulses of this kind and gives dependable operation.

The chief advantages of the instrument are its high sensitivity and freedom from zero-drift over long periods of time. The reed head is placed on top of the ionization chamber which is suspended in a wood frame set on a foam rubber mat. The mat effectively damps out mechanical vibration to which the reed is highly sensitive. The usual precaution of grounding the reed is observed, but no other measures are necessary to provide quiet, stable operation. With the parameters of the detecting system as described above, the pulses (as seen by the reed) range in amplitude up to 2 mv, which permits consistent operation of the reed on the 10 mv scale. The output of the reed may be used to drive a recording milliameter, or a voltage signal may be taken off the output resistance loop and introduced into an electronic counting system. For reasons already mentioned, the latter alternative is much to be preferred and the development of a satisfactory electronic counter constituted the major problem of the program.

Counter Circuit

The overall function of the counter amplifier circuit is to activate a register drive circuit for each pulse delivered from the reed. Therefore, the circuit must (1) respond to all pulses over their characteristic range of rise time, (2) resolve all pulses delivered by the detector-reed system, and (3) by incorporation of an electronic discrimination level reject pulses whose amplitude is less than a specified

minimum. This level of discrimination should, moreover, be maintained at a constant value within narrow limits, for many weeks.

The basic circuit in operation at present is shown in Fig. 3. Tubes V_1 and V_2 constitute a feedback amplifier of high stability and low noise level. The output of this amplifier is then used to trip V_3, a mono-stable, cathode-coupled multivibrator, which in turn fires the register-drive tube V_4, and consequently the register (Veeder-Root series 1248). Filament and d. c. supply voltages are obtained from a commercial regulated power supply (a modified model 28-111, Lambda Elec. Corp. is readily adaptable for this purpose). In earlier models a scaling circuit was incorporated prior to the register drive stage, but this feature was subsequently dispensed with, inasmuch as the resolution time of the Veeder-Root register was demonstrated to be adequate for count rates up to 1000 cph.

The pulses delivered by the reed to the counting system range in magnitude up to . 34 volts, with an average close to . 15 volts. The rise time of these pulses, as measured on a C. R. O., ranges from about . 01 to . 5 sec. For low-level counting, a resolution time of . 1 sec. is adequate. The amplifier itself has sufficient band width to respond to all pulses, so that the pulse response of the system is determined by the characteristics of the multivibrator. The input noise level is more than 20 db below the noise of the reed. Although more gain was incorporated in the amplifier, a gain of 200 was found

to give optimum performance. The coupling between the two sections of V_2 acts as a low pass filter, designed primarily to remove a small 450 cycle signal coming from the reed.

The output of the amplifier is capacitatively coupled to the input grid of the mono-stable, cathode-coupled multivibrator. The level of activation of the miltivibrator is determined by the bias of the input grid (the input side is non-conducting when the tube is in the quiescent condition). The discriminator potentiometer permits a maximum grid bias of about 10 volts below cathode. The discriminator rejects noise from the reed output which ranges in amplitude up to about 30 millivolts maximum, when the reed is operated on the 10 mv scale. It will be noted that some real pulses will also be rejected inasmuch as the pulse height (dependent upon the path-length of the α-particle in the ionization chamber) ranges over a continuum of values from zero to the maximum value. However, the distribution is concentrated toward the higher amplitude portion of the spectrum, and pulses giving rise to signals less than 30 mv on the reed output are a small fraction of the total.

A battery supplied discrimination potential level is open to some objection because this does not insure constant potential difference between grid and cathode, due to the possibility of cathode drift. Although cathode drift was assumed negligible, attempts to in-

corporate dry cells were not successful, inasmuch as the decrease of terminal voltage of the cells changed the discrimination level within a short time, in spite of the use of large bleeder resistors. The use of wet cells was not explored because of other objectional features.

The stability of discriminator potential obtained by electronic means is limited by the regulation of the d. c. source. An alternative to rigid regulation of the d. c. source consists of selecting that set of operating conditions which will minimize the effect of small variations in the potential level of discrimination. The latter course was followed with the result that as currently operated, the system will tolerate a variation of about 1. 3% in the 150 volt supply of the circuit. After initial aging of all electronic components, the minimum period of stable operation (i. e., in which no measurable change in counting efficiency was detected) for three counters is about one week, and occasionally a unit may run as long as one month (continuous operation) before a change in counting efficiency necessitates adjustment of the discriminator potentiometer setting. This change is thought to be primarily due to drift in the discriminator grid bias. To further the long period stability of the counter, long-life tubes and high-stability resistors were employed as indicated in the schematic (Fig. 3). The 110 volt a. c. operating potential for both the reed and power supply was supplied from a constant voltage (Sola) transformer.

The parameter controlling the pulse response of the counter is the RC time constant of the multivibrator (R18 and C9 in Fig. 2). The optimum value was found to be .75 sec., which permits resolution of all pulses delivered by the reed. With this value, the counter does not respond to pulses of rise time longer than .5 sec. which, however, are of infrequent occurrence.

The register drive circuit is similar to those used in standard electronic counters. The necessary grid bias is obtained from a voltage doubler circuit operated off the filament supply.

CALIBRATION

Calibration of the radon counting system was accomplished by the use of standard radium solutions supplied by the National Bureau of Standards. Two calibration flasks were prepared, each containing 1×10^{-12} grams of radium. Prior to use, the flasks and distilled water used were checked for radium contamination. The flasks were sealed and stored for at least thirty days to allow equilibrium between radium and radon to be established. The radon was then transferred into the ionization chamber by flushing with nitrogen, and the pressure of the chamber was brought to one atmosphere. An interval of 3 to 4 hours must be allowed before counting to permit the daughter products of radon to achieve equilibrium, thereby simplifying the calculation[5].

Although there are actually two daughter α emitters which are in equilibrium with the radon, both of these are solids and have been found to condense on the inner wall of the ionization chamber[9]. Under these circumstances each of the daughters has a counting efficiency of 50% and the total activity after equilibrium has been achieved, is two times the activity of the radon. The efficiency as used here is defined as the percentage of the expected activity from the standard radium solution which is recorded by the detection device. This factor was measured at the time the apparatus was built and then was checked occasionally in the succeeding months. The reproducibility of this constant between the two flasks used, in several different ionization chambers and through a period of almost 2 years, has remained constant (within statistics) as seen in Table 1 below.

Table #1 Absolute Efficiency of Radon System

Flask	Ion Chamber	Date	Efficiency
C	7	Aug. 1951	77.0±2.5%
A	1	Aug. 1951	80.0±2.0%
A	5	Sept. 1951	77.0±2.0%
C	1	Oct. 1951	77.8±1.8%
A	3	Oct. 1951	76.3±1.8%
C	5	Apr. 1952	79.3±1.4%
		Average Efficiency	77.9±1.4%

9. R.B. Graveson, Health Physics Division, New York Operations Office. A.E.C. (private communication)

By the use of this efficiency factor, several rock specimens of known radium content, supplied by the Bureau of Standards, were then measured as a further check of the absolute accuracy of the system. These rocks were melted in a graphite resistance fusion furnace modified after that described by Evans and Goodman[10]. The resulting gas was dried and counted in the ionization chambers. Results are tabulated in Table 2.

Table #2 Accuracy of Radon System

Sample	Bureau of Stds. Value g Ra x 10^{-12} / g	Measured Value g Ra x 10^{-12} / g
Graniteville Granite	3.3±0.1	3.27±0.10
		3.36±0.09
		3.35±0.09
		3.10±0.09
		3.22±0.07
	Av.	3.26±0.05
Carthage L.S.	0.15±0.03	0.145±0.03

From Tables 1 and 2 it can be seen that both the accuracy and precision of the present radon measurement technique as a means of determining radium content of solids or liquids is entirely satisfactory to about 3%.

Although necessary to establish the absolute counting efficiency of the radon system, the method outlined above is rather time-consuming and cumbersome. Each flask must be allowed to accumulate radon for at least thirty days. Special precautions must be taken in making the run to insure that all radon is removed from the solution and that no contaminating material is included. Because it is desirable to make daily checks on the operating level of the counting apparatus, a secondary calibration of the relative efficiency of the counters was devised, utilizing standards prepared at this laboratory. The secondary standard is prepared by mounting a very small piece of uranium-containing mineral (in this case zircon) inside an ionization chamber in such a way so as to shield the detector from all particles emitted by the mineral, yet allowing the radon released to diffuse freely into the volume. This was accomplished by use of a small stainless steel cup ($\sim$1 cm radius) mounted open-end down on the base of the chamber. Several small holes were drilled in the top of the inverted cup and the zircon speck mounted on the under side. With this arrangement all of the α's from the solid were prevented from causing ionization in the chamber proper, while the escaping radon was quite free to fill it. After having mounted the radon source, the ionization chamber was sealed, evacuated and filled to 1 atm. pressure with dry, radon-free N_2. This ionization chamber was then put on a counting unit and the activity was measured daily and plotted as a function of

time. The resulting curve indicated that the activity was due entirely to radon build-up and after approximately a month leveled off to a constant value of steady state equilibrium. This standard chamber was then counted several times before and after efficiency determinations made with the Bureau of Standards radium solutions. In this way the secondary standard of known activity was prepared. This secondary standard has proven extremely useful as a day-to-day check of the sensitivity level of the counters, which, when having drifted from the level of 78% counting efficiency, can be brought easily to this level by adjustment of the discriminator level. At the present time two such secondary standards are in operation, one with an activity of about 900 cph and the other about 500 cph.

BACKGROUND

The background of the six ionization chambers now in operation varies between 14 and 30 counts per hour and has remained constant in most cases for long periods of time. The origin of this residual activity has been found to be entirely from the internal metal parts of the chamber. The nitrogen used as filling gas is stored for at least 30 days to allow any appreciable radon to decay. Runs made with evacuated chambers have shown that spurious counting of electronic origin contributes less than 0.1 cph.

Optimum background is obtainable only by careful selection of the stainless steel used to construct the ionization chambers. This is accomplished by pre-counting the steel "pots" before any machining is done. After the parts are constructed, a washing in aqua regia, thorough rinsing in triple-distilled water followed by careful drying, has been found to aid materially in lowering the background due to surface contamination. Once assembled and vacuum sealed, the background rate of a chamber has been found to remain completely constant.

CONCLUSION

A system has been devised for the radiometric determination of small quantities of radon. The method combines the desirable features of electronic stability and chemical simplicity. The system can be applied to the measurement of radium in naturally occurring solids, liquids and gases. The same electronic system can be effectively used for low level alpha counting of solids (thick or thin source) with a parallel plate ionization chamber.

ACKNOWLEDGMENTS

The authors wish to acknowledge the considerable aid of many colleagues in the development of this procedure. The ionization chamber was modified after models in use by J. E. Hudgens, New

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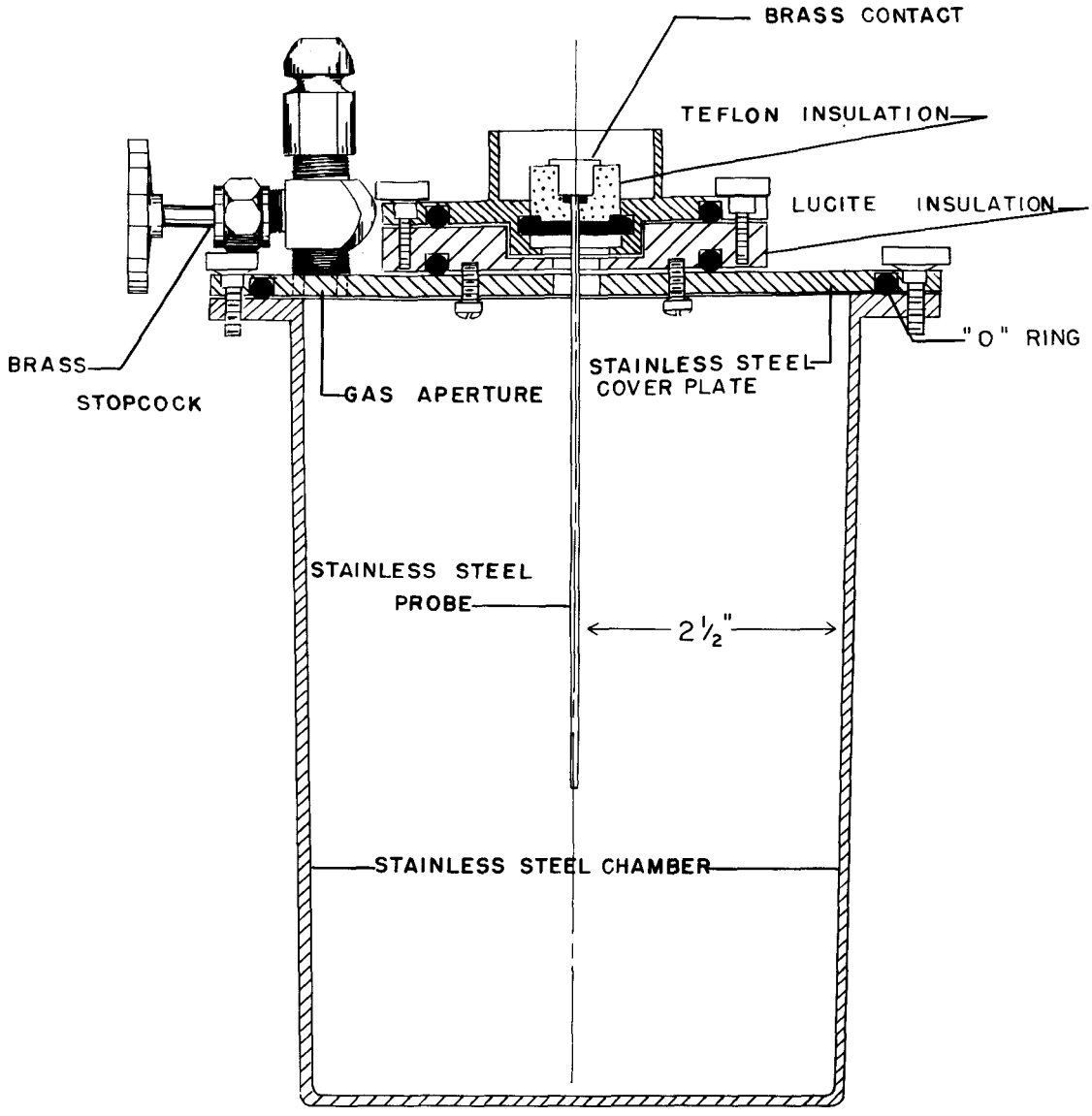

Fig. 1—Ionization chamber.

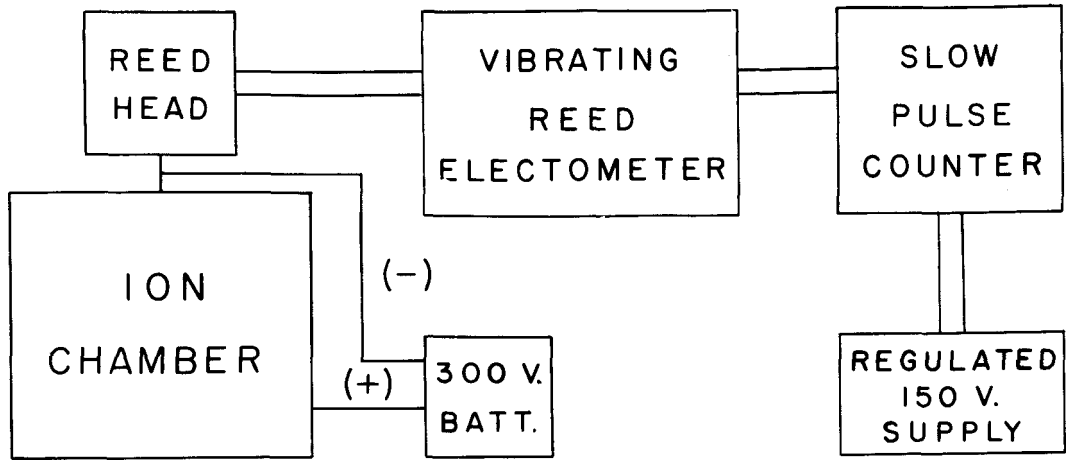

Fig. 2—Block diagram of detecting and counting circuit.

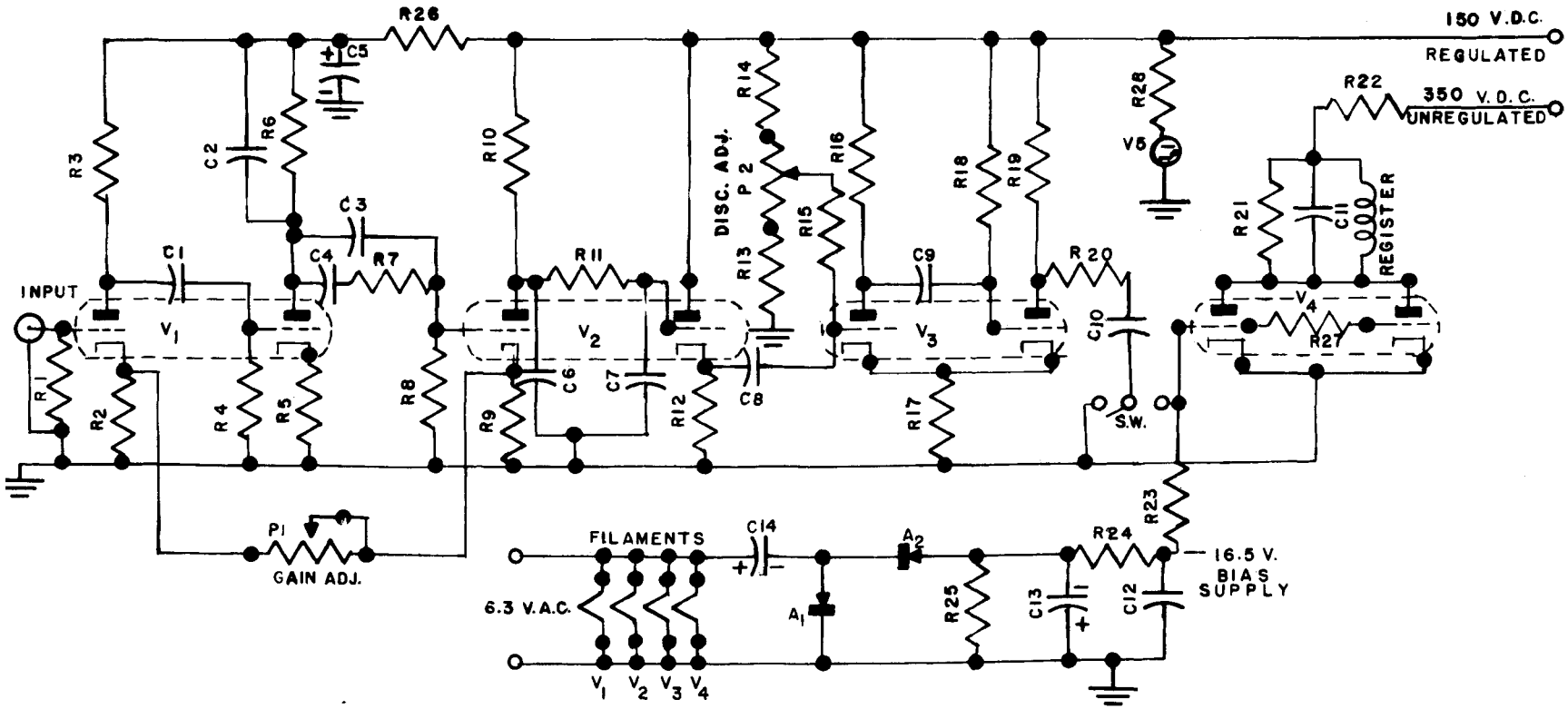

Fig. 3—Schematic for counter circuit.

LEGEND FOR FIGURE 3

R1	150 K, 1/2 watt	R18*	1.5 Meg.	C6	.25 MF, 400V
R2*	3300 W	R19	12 K, 1 watt	C7	.25 MF, 400V
R3	270 K, 1 watt	R20	200 K, 1 watt	C8	.25 MF, 400V
R4	680 K, 1/2 watt	R21	220 K, 1 watt	C9	.5 MF, 600V
R5*	3300 W	R22	2000 W, 5 watt	C10	.1 MF, 600V
R6	270 K, 1 watt	R23	1 Meg, 1/2 watt	C11	.02 MF, 400V
R7	6.8 Meg. 1/2 watt	R24	1 Meg, 1/2 watt	C12	.1 MF, 600V
R8	680 K, 1/2 watt	R25	10 K, 1 watt	C13	50 MF, 25V
R9*	470 W	R26	8000 W, 5 watt	C14	50 MF, 25V
R10	47 K, 1 watt	R27	330 W, 1/2 watt		
R11	100 K, 1/2 watt	R28	510 K, 1 watt	P1	10,000 W WW, 4 watts
R12	47 K, 1 watt			P2	10,000 W General Radio 371A
R13*	33 K	C1	.1 MF, 600V		
R14*	100 K	C2	.01 MF, 400V		
				A1-A2	Federal 404D3552
R15*	270 K	C3	.1 MF, 600V		
				V1	5691
R16	15 K, 1 watt	C4	1.0 MF, 600V		
				V2	5692
R17*	10 K	C5	40 MF, 450 V		
				V3	5692
				V4	6SN7
SW	SPDT toggle switch				
				V5	NE-51 Neon Lamp

* Aerovox carbofilm, CP 1/2, 5% tolerance on all other resistors.