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CO_2-CS_2 Geiger Counter

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Experiments on the mechanism and characteristics of a CO_2-CS_2 Geiger counter are reported, and circuits and methods of operation which satisfy the special requirements are described. Counter: The active part is $2\frac{7}{8}$ in. in diameter and 16 in. long, filled to 1 atm, 95% CO_2 and 5% CS_2. The cathode is copper and the anode is 0.005-in.-diam Chromel A. Characteristics: The electrons released by an ionizing particle become attached, probably to CS_2. The self-quenching action of the counter is excellent, no spurious counts are observed in the absence of electronic quenching. However, a dead time of several milliseconds must be imposed electronically because of the long interval during which the negative ions arrive at the anode. Method: An electronic quench is used which is triggered both by the CO_2-CS_2 counter and the anticoincidence ring. This serves to impose the required dead time and also to prevent the firing of the CO_2-CS_2 counter by mesons. Measurements: The plateau was measured to 1900 v above threshold and was found to be level to within 1% from 400 to 1600 v above threshold. The maximum drift time of the negative ions was found to be about 9 msec with a sharp cutoff. Tests with various combinations of gas indicated, but did not prove, that the charge carrier was CS_2. The effects of common contaminants were determined. At voltages over 400 above threshold, 1% O_2 gave no detectable effect; 0.3% SO_2 gave a 2 to 3% reduction in counting rate. Extensive tests of the efficiency were made. Comparisons of the CO_2-CS_2 filling with an argon-ethane filling gave identical results within the experimental error. No basis was found for supposing that there was any failure of the CO_2-CS_2 counter to register counts. Reliability: The counter described has been used for C^{14} dating purposes over a long period of time. There has been no indication that there are variations in counting rate outside those expected on the basis of statistics. The counting rate for "dead" CO_2 in an iron shield 9 in. thick and after anticoincidence is 8 per min.

I. INTRODUCTION

THE CO_2-filled Geiger-Müller counter with CS_2 added as a "quench vapor" was first described by Miller in 1947.[1] More extensive studies of its characteristics were made by Brown and Miller[2] and reported in the same year. In 1949 Mann and Parkinson[3] reported upon a special univibrator quench circuit they had devised for use with the CO_2-CS_2 counter and presented plateau curves obtained with it. In the same year Hawkings, Hunter, and Mann[4] reported upon a study of the absolute efficiency of the CO_2-CS_2 counter which used the quench circuit of Mann and Parkinson. The study on efficiency was made necessary by a series of researches on the half-life of C^{14} which had just been concluded by Hawkings, Hunter, Mann, and Stevens.[5,6] In 1950 Miller, Ballentine, Bernstein, Friedman, Nier, and Evans[7] published on the use of a CO_2-CS_2 Geiger counter in the determination of the half-life of C^{14} and compared the results with those which they obtained with a CO_2 proportional counter.

A CO-CS_2 Geiger counter and its use for a measurement of the absolute counting rate of modern C^{14} were described in 1953 by Mościcki.[8] His counter used a univibrator modulator quench circuit described by Gorgolewski.[9] In 1958 Mościcki published two papers[10,11] on extensive studies (Part I, theoretical; Part II, experimental) of the application of the CO_2-CS_2 Geiger counter to the measurement of C^{14} in natural carbon. His experimental observations on plateau length, pulse delay, multiple pulses, absolute efficiency, and sensitivity to impurities are of particular interest in connection with the present discussion.

In connection with a program of routine C^{14} dating which we have carried on over a period of years[12–16] we have made many experiments on the characteristics of the CO_2-CS_2 counter. Our object was to try to gain a sufficiently clear understanding of the discharge mechanism to enable us to design special circuits or methods of operation which would give optimum performance. In the course of this, we found it necessary to repeat some of the experiments which were reported earlier in the literature and to perform some new ones. Although there still is much to be learned, we believe that we now understand those aspects of the mechanism which dictate the circuit requirements, enabling us to approximate optimum operating conditions. As will be brought out in the pages to follow, we find that when its special circuit requirements are met, this type of counter becomes highly reliable and remarkably insensitive to parameters, such as voltage and gas purity.

The discussion in this paper will be limited to our findings concerning the characteristics of the CO_2-CS_2 counter and to the circuits and methods of operating it in conjunction

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[1] W. W. Miller, Science **105**, 123 (1947).

[2] Sanborn C. Brown and Warren W. Miller, Rev. Sci. Instr. **18**, 496 (1947).

[3] W. B. Mann and G. B. Parkinson, Rev. Sci. Instr. **20**, 41 (1949).

[4] R. S. Hawkings, R. S. Hunter, and W. B. Mann, Can. J. Research **B27**, 555 (1949).

[5] R. S. Hawkings, R. S. Hunter, W. B. Mann, and W. H. Stevens, Phys. Rev. **74**, 696 (1948).

[6] R. S. Hawkings, R. S. Hunter, W. B. Mann, and W. H. Stevens, Can. J. Research **B27**, 545 (1949).

[7] W. W. Miller, R. Ballentine, W. Bernstein, L. Friedman, A. A. Nier, and R. D. Evans, Phys. Rev. **77**, 714 (1950).

[8] W. Mościcki, Acta Phys. Polon. **12**, 238 (1953).

[9] S. Gorgolewski, Acta Phys. Polon. **12**, 152 (1953).

[10] W. Mościcki, Acta Phys. Polon. **17**, 311 (1958).

[11] W. Mościcki, Acta Phys. Polon. **17**, 327 (1958).

[12] H. R. Crane, Science **124**, 664 (1956).

[13] H. R. Crane and James B. Griffin, Science **127**, 1098 (1958).

[14] H. R. Crane and James B. Griffin, Science **128**, 1117 (1958).

[15] H. R. Crane and James B. Griffin, Am. J. Sci. Radiocarbon Suppl. **1**, 173 (1959).

[16] H. R. Crane and James B. Griffin, Am. J. Sci. Radiocarbon Suppl. **2**, 31 (1960).

with an anticoincidence ring, insofar as they differ from standard counter technique. The anticoincidence circuit, power supplies, scalers, etc., which make up the complete C^{14} dating apparatus are not sufficiently novel to warrant a description in this Journal; however, working specifications have been reproduced in Mimeograph form and are available.

II. QUENCHING ACTION OF CS_2

The unusual points of behavior of the CO_2-CS_2 counter can best be treated if we begin by recalling certain pertinent facts which are common to all Geiger discharges. In any gas, whether a quench vapor is present or not, a Geiger discharge is self-terminating. The cumulative ionization which spreads along the length of the wire creates a cylinder of plasma around the wire. Because of their greater mobility, the electrons are quickly collected by the wire, leaving the positive ions as a space-charge sheath. This lowers the electric field near the wire to a value too low to maintain the further production of ionization and the discharge terminates. A new discharge will not start until the cylinder of positive ions has moved out to some distance from the wire, the distance depending upon the overvoltage (volts above threshold) at which the counter is operated.

The function of a quench vapor lies in preventing the discharge from restarting after the sheath has moved away. There are two principal ways in which the discharge may restart: (1) If the gas is transparent in the ultraviolet, the ultraviolet light produced in the discharge may release photoelectrons from the cathode. If the time taken for these electrons to reach the wire allows the sheath to move out far enough, a new discharge can start, and the discharge will repeat indefinitely. This mechanism may be prevented by adding a quench vapor which absorbs ultraviolet light. A well-known example is the addition of alcohol vapor to argon. (2) Free electrons or negative ions may be released at the cathode by the action of the positive ions of the sheath, upon their arrival. This can happen if the ionization potential of the arriving ions is more than twice the work function of the surface. Negative ions may be released if the ionization potential plus the attachment energy of the negative ions is more than twice the work function. These criteria are less stringent than they at first appear to be, because the surface almost always bears oxidized or otherwise altered spots which have lower work functions than that of the pure, clean metal of the cathode. A quench vapor having an ionization potential lower than that of the principal gas will degrade the energy available for the release of electrons or negative ions at the cathode. This degradation takes place through the transfer of charge from the ions of the principal gas to the molecules of the quench vapor, in collisions. Further, if the quench vapor is polyatomic, its ions will, in general, dissipate their energy by dissociating upon contact with the cathode surface instead of by releasing electrons.

A CO_2-CS_2 mixture satisfies the foregoing two criteria, as pointed out by Brown and Miller, and as experiments to be described here will show. Yet why has it been found to be necessary to use an external quench circuit? To answer this, a further property of CO_2 mixtures will have to be discussed.

It was once thought that CO_2 had a strong affinity for electrons, forming negative ions.[2,17] It has since become clear that CO_2 does not attach electrons at all, but that the extreme prevalence of negative ions in CO_2 mixtures is due to another mechanism. Because of its particular molecular energy level structure, CO_2 has a very large cross section for inelastic collisions with free electrons. Consequently, the kinetic energy a free electron gains by falling through the electric field is rapidly dissipated in collisions, so that its "agitation energy" remains at a low value. On the other hand, in a noble gas where the collisions are elastic the electron agitation energy in typical conditions found in a G-M counter may be at least 50 times greater than in CO_2.[18] The cross section for the formation of negative ions of oxygen and several other electronegative molecules increases strongly as the electron kinetic energy decreases.[19] The formation of negative ions in CO_2 is explained by the very great enhancement of the cross section for the formation of negative ions by certain impurities, particularly oxygen, and not by the formation of negative ions by CO_2 itself. It was shown by DeVries[20] that one part in 10^{12} of O_2 in CO_2 in a proportional counter will yield negative ions to such a degree that delayed counts are easily observable. Through extremely good purification techniques, as practiced by those who use the CO_2 proportional counter for C^{14} dating, the formation of negative ions can be kept below the observable level. However, our experiments show that when even a small amount of CS_2 is added to the CO_2 virtually all of the electrons become attached before they reach the anode. Whether the negative ions are derived from the CS_2 itself or from impurities such as O_2 which are inevitably present in the CS_2 is not definitely known, but on the basis of some evidence that will be presented later, the presumption is in favor of the CS_2.

When a particle, say a meson, traverses a CO_2-CS_2 counter, electrons are set free in a range of distances from the wire. They form negative ions and, therefore, have low mobility. The arrival of the negative ions is spread out in time because of the various distances traveled. In our counter, which is 3 in. in diameter and at 1-atm pressure of CO_2, the last ion arrives more than 8 msec after the passage

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[17] H. R. Crane, Phys. Rev. **94**, 1437 (1954).

[18] Tables of the average velocity of electrons in an electric field in various gases, including Ar and CO_2, may be found in Leonard B. Loeb, *Basic Processes of Gaseous Electronics* (University of California Press, Berkeley, California, 1955), pp. 323–328.

[19] A partly theoretical plot of the attachment probability of electrons to O_2 as a function of electron energy may be seen in reference 18, p. 464.

[20] H. DeVries, Appl. Sci. Research (Netherlands) **B5**, 387 (1955).

of the meson. The delay in arrival of the first ion depends upon the path of the meson. By operating the counter in the proportional range, and by connecting the linear amplifier to a fast oscilloscope, we have observed the arrival of the individual negative ions in a time spectrum. In operation as a Geiger counter, the important fact is that the span of time over which the negative ions arrive is greater than the time required for the positive ion sheath to move away from the wire and for the counter to regain sensitivity. Therefore the counter may be fired several times by the negative ions made by a single meson. We have observed that the last discharge in such a group occurs at a delay not greater than the time required for an ion to travel from cathode to anode.

The reason that an external quench circuit has always been found to improve the operation of the counter is now apparent. The quench circuit primarily provides an extended dead time for the recording circuit, so that a group of pulses produced by the same primary particle is recorded as a single count.

To verify the foregoing interpretation, we operated a CO_2-CS_2 counter without any electrical quench circuit, but with a circuit which merely blanked the line to the scaler for approximately 15 msec after each pulse. The mixture was 95% CO_2, 5% CS_2, at a total pressure of 1 atm. The cathode diameter was 3 in. and the anode diameter was 0.005 in. The counter was in an iron shield, so most of the counts were due to mesons. Although the runs were short, they indicated a remarkably long and flat plateau. The threshold was at 5500 v, and there was no indication that the end of the plateau had been reached at 1700 above threshold. The data are given in Table I.

The foregoing tests confirm the idea that an electronically extended dead time is essential for the operation of a CO_2-CS_2 counter, but that an external quench, i.e., a lowering of the voltage after each discharge, is not essential.

III. REDUCTION OF THE NUMBER OF DISCHARGES AS INSURANCE AGAINST SPURIOUS COUNTS

In applications (such as C^{14} dating) where even an occasional spurious count is ruinous, it is a good policy to eliminate as many as possible of the real, but nonessential, counts, for the following reason: We assume that a spurious discharge (one not of radioactive or cosmic-ray origin) can be initiated only by energy which is left over in some form or other from a previous discharge. There are many possible forms: metastable excitations, surface activation, and free radicals are examples. Therefore, each discharge carries with it a certain probablity of causing a subsequent spurious discharge which, in the presence of a good quench vapor, may be extremely slight but never zero. It follows that any reduction in the total number of counts is an advantage.

TABLE I. Plateau for CO_2-CS_2 counter with 15-msec blanking circuit. Cathode 3 in. diam, anode 0.005 in.; 95% CO –5% CS at 1 atm. Starting voltage 5500.

Volts above threshold	Counts/minute
85	190±5
225	210±4
425	211±4
510	209±3
680	206±5
850	210±5
1020	211±3
1190	213±3
1360	218±3
1700	223±3

The reduction in the number of discharges is accomplished through two strategems: (1) We have already pointed out that each meson or beta track gives rise to not one, but to a group of several Geiger discharges. In our circuit, the pulse from the anode of the counter generates a positive rectangular pulse of adjustable length from 10 to 20 msec which is applied to the cathode of the counter. This drops the potential difference between cathode and anode to a value below the threshold, with the result that the discharge which is initiated by the first negative ion to arrive is the only discharge that occurs. This reduces the number of discharges by a factor of 2 to 10. (2) A further reduction is obtained by making a connection which causes the anticoincidence counters to trigger the quench circuit of the CO_2-CS_2 counter. When a meson passes through, the voltage on the CO_2-CS_2 counter is dropped below the threshold before the first negative ion arrives at the anode. There is plenty of time in which to accomplish this. The delay in the firing of the anticoincidence counters is very small (a maximum of about one μsec) while the delay in the CO_2-CS_2 counter ranges up to about 9 msec. The quench is triggered well in advance of the arrival of the first negative ion at the anode in all but the rare cases in which the mesons pass very close to the anode. Thus, in the great majority of cases, the passage of a meson through the CO_2-CS_2 counter does not cause a discharge at all.

Through the two methods just described, the number of discharges per unit of time in the CO_2-CS_2 counter is reduced by a factor of approximately 100.

The circuit used for the above purpose is, in fact, an external quench circuit, since it lowers the voltage after each discharge. The amount of circuitry required to produce a quench is little more than that which would be required to extend the dead time, and we believe the extra insurance provided against spurious counts is well worth the added complexity.

IV. COUNTERS AND SHIELD

The counter used in the experiments described is shown in Fig. 1. The counter shell is made of copper pipe. The metal-to-metal joints are silver soldered, and the metal-to-glass seals are made with high melting point wax. The re-

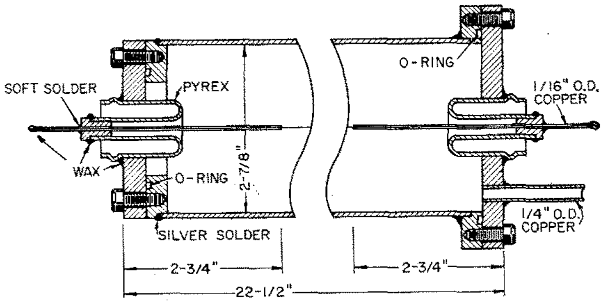

FIG. 1. Cross section of the counter.

movable end plates are sealed with Neoprene O-rings. The wire is 0.005-in.-diam Chromel A (80% Ni, 20% Cr). The ends of the anode wire are sheathed by $\frac{1}{16}$-in.-o.d. copper tubing, a device which is known[21] to be of help in giving a sharp and reproducible limit to the sensitive region of the counter. The anticoincidence ring which surrounds the counter consists of 8 sealed-off counters 2 in. in diameter CO_2-CS_2 and 20 in. in active length, connected in parallel. The whole assembly is shielded by 9 in. of iron.

After the electrical connections have been made, a small metal thimble is slipped over the exposed anode terminal at each end of the counter. This eliminates the possibility of corona, which would give spurious counts.

V. CIRCUITS

A. Amplifier-Quench

An important characteristic of this part of the circuit is the following. When a pulse, from either the CO_2-CS_2 counter or the anticoincidence ring, enters the amplifier, the cathode voltage of the CO_2-CS_2 counter changes to about +700 and remains at that value for a time which we call the quench interval, typically 15 msec. It then recovers to zero abruptly. If a meson passes through the AC ring while a quench interval is in progress, the interval is extended. Sensitivity is restored only after both the CO_2-CS_2 counter and the AC ring have been quiescent for a minimum of 15 msec. Thus ample time is allowed for all the negative ions associated with an AC count to be swept out of the CO_2-CS_2 counter before it is made sensitive, irrespective of the time at which the AC count occurs.

The circuit of the amplifier-quench section is shown in Fig. 2. When 2050 #1 is quiescent its plate is held close to ground potential by grid conduction in 6AC7 #2. When the 2050 fires, its plate potential drops to about −135 v. The plate voltage then recovers along an exponential curve which has its asymptote at +150 v, but it follows this only up to zero, where it is stopped suddenly by the grid conduction in 6AC7 #2. Since the clamping action occurs during the steep part of the rise, the time interval is accurately reproduced from count to count and is insensitive to all circuit parameters except R and C in the plate circuit.

There is a minor qualification to be mentioned in connection with the quench interval. The 2050 cannot be refired until its plate voltage rises above −110. Therefore, after it is fired by a count from the AC ring, there is a dead time of approximately 3 msec, which is the time it takes the plate to recover to −110 v. This means that, if a 15-msec quench interval is used, occasionally the quench interval will actually end 12 msec after the last AC count.

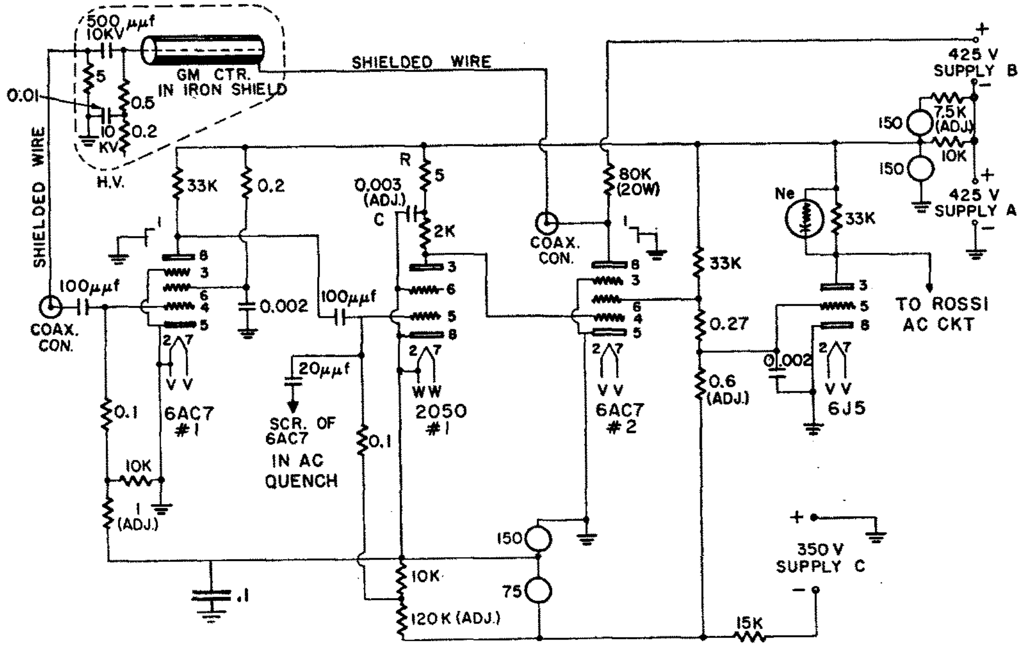

FIG. 2. Schematic diagram of the section of the circuit which contains the amplifier, quench, and pulse inverter. Unless otherwise specified, resistance values are in megohms.

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[21] S. C. Curran and J. D. Craggs, *Counting Tubes; Theory and Application* (Butterworths Scientific Publications, Ltd., London, 1949), p. 118.

The rectangular pulse which appears on the screen of 6AC7 #2 (corresponding to the quench signal) is inverted and delayed about $\frac{1}{2}$ msec by the coupling network to the 6J5. The delay is introduced to be sure that a count in the CO_2-CS_2 counter will never arrive at the anticoincidence circuit ahead of the corresponding count from the AC ring. The inversion of the pulse is required by the anticoincidence circuit, which, in our case, is of the Rossi type. An additional use is made of the 6J5: a neon blinker is driven by its plate signal. The blinker is of convenience in setting the counter voltage.

B. Anticoincidence Quench

The anticoincidence counters are quenched by a univibrator circuit having a quench duration of about 3 msec. A positive, rectangular pulse is taken from this and applied to the Rossi anticoincidence circuit. Also, a positive pulse is fed through a 20-$\mu\mu$f condenser to the grid of 2050 #1 so that the CO_2-CS_2 quench will be triggered by the anticoincidence ring, for the reasons given in Sec. III.

C. Timer

In the system used, the "dead time" may be as high as 15% or even 20%. The variation in dead time over days or months due to changes in the cosmic ray rate, temperature, and aging effects on components, etc., is sufficiently large so that it must be monitored. The problem is bypassed completely in the following way. Sharp pulses are generated at line frequency (60 per second). These are sent through a gate which is open only when the CO_2-CS_2 counter is *not* quenched, and the pulses are counted on a 3-decade scaler. This gives the true integrated sensitive time of the counter for use in calculating the counting rate. Strictly speaking, the result is statistical, since the separation between the test pulses ($16\frac{2}{3}$ msec) is of the same order as the quench interval; however, the number of pulses per day is so enormous that the sampling is highly precise. The ordinary clock, which measures total elapsed time, is not used except for determining, occasionally, the fraction of dead time.

The timer circuit is shown in Fig. 3. One half 6SN7 #1b is overdriven by about 120 v ac from one side of the secondary of the power transformer of power supply A. The plate signal, which is a square wave, is differentiated and used to trigger 2050 #2. The sharp, positive pulses from the cathode of 2050 #2 are inverted in the gate tube, 6AC7 #3, and fed to the 3-decade scaler.

During the quench interval of the CO_2-CS_2 counter, the screen of 6AC7 #3 is negative, preventing the 2050 #2 pulse from reaching the scaler. At all other times the screen of 6AC7 #3 is positive. All signals in the timer circuit are either square waves or sharp pulses, so the result is insensitive to changes in tube characteristics, voltages, or component values.

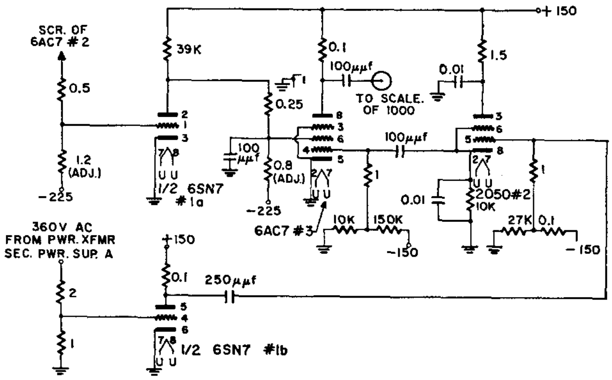

FIG. 3. The timer circuit, which was used to measure the integrated "on" time of the CO_2-CS_2 counter.

VI. OPERATING CHARACTERISTICS

A. Threshold and Plateau

The total and the uncanceled counting rates as a function of voltage above the threshold (which was at 5650 v) are shown in Fig. 4. These were made with a filling of 71 cm Hg of "dead" CO_2 and 3.2 cm Hg of CS_2 (at 23°C). The quench interval was 15 msec. The quench signal was in all cases great enough to drop the voltage below threshold in the quench interval. This necessitated increasing the plate supply voltage of the quench tube to about 2000 v to obtain the upper ends of the curves. The squares, circles, triangles, and crosses denote data taken on different days. The upper curve gives the total count, and the lower curve gives the count after anticoincidence cancellation. The data, with standard deviations, are reproduced in Table II.

Neither of the curves shows a perceptible slope in the central part of the range, which extends from about 500 v above threshold to 1600 v above threshold. The curves show that there is practically no geometric effect; i.e., increase in effective volume of the counter with increasing voltage. As was stated earlier, this effect is reduced through the use of small diameter ($\frac{1}{16}$ in.) sleeves over the ends of the anode wires. The sleeves reduce the distortion of the

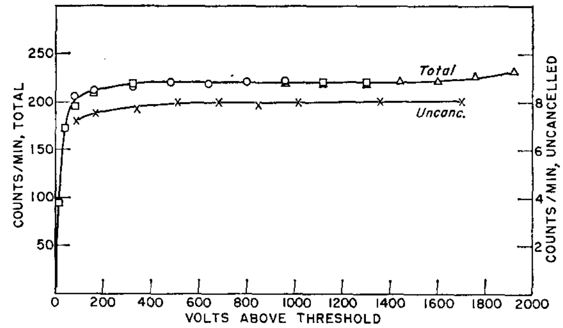

FIG. 4. Plateau curves of the CO_2-CS_2 counter showing the total counting rate and the counting rate with anticoincidence (uncanceled). The uncanceled count curve was obtained in a single run extending over several days. The total count curve was obtained in three separated runs designated by circles, squares, and triangles.

TABLE II. Total and uncancelled counting rates as functions of overvoltage, threshold 5650 v. Filling 71 cm Hg of "dead" CO_2 and 3 cm of CS_2.

Counter volts	Uncanceled counts per min	Counter volts	Total counts per min (±1%)
85	7.20±0.21	15	95,
170	7.56±0.07	40	172,
340	7.70±0.10	80	196, 206
510	8.00±0.06	160	210, 212
680	8.02±0.12	320	217, 219
850	7.90±0.10	480	222
1020	8.02±0.08	640	220
1360	8.07±0.16	800	222
1700	8.06±0.12	960	220, 223
		1120	221, 221
		1300	218, 222
		1440	222
		1600	222
		1760	227
		1920	232

electric field at the ends of the counter. In addition, the fact that the operating range is unusually far (500 v or more) above the threshold, further reduces the insensitive portions of the wire. In fact, we may interpret some of the slope in the earlier part (say 100 to 400 v) as being a geometrical effect, and conclude that at the higher voltages the insensitive portions of the wire are reduced to no more than a couple of diameters of the copper sleeve in length. In our C^{14} dating runs, the voltage is maintained at 500 v above threshold.

B. Time Delay

A quantitative study of the delay in the firing of the counter after the passage of a meson through it was made in the following way. An oscilloscope was connected so that the horizontal sweep was triggered by the discharge in the anticoincidence ring, and so that a vertical pip was produced by the firing of the CO_2-CS_2 counter. (The connection that permits the anticoincidence counters to fire the CO_2-CS_2 quench was removed for this experiment.) Thus the position of the pip on the horizontal axis indicated the time of firing of the CO_2-CS_2 counter with respect to the time of passage of a meson through the AC ring and the CO_2-CS_2 counter. Oscillograms were made, using time exposures. The time spectrum, from the measurement of all the photographs, is shown in Fig. 5.

In addition to the counts which were associated with the mesons that traversed at time-zero on the plot, there was a uniform background, the origin of which is explained as follows. In our counter system there are, in round numbers, 600 mesons per minute which pass through the AC ring and which miss the active volume of the CO_2-CS_2 counter. Only about 200 per minute trigger both the AC's and the CO_2-CS_2 counter. Each of the 600 counts triggers a sweep of the synchroscope. Anywhere in one of these sweeps, an uncorrelated CO_2-CS_2 pip may occur. Such events give a uniform background of pips in the time distribution plot. We calculated the number of these to be expected in the data of Fig. 5 and found it to be in agreement with what appears in the plot, namely about 10 per millisecond interval. Therefore we can say that the distribution of correlated counts goes to zero in the interval between 8 and 9 msec.

A second method was used for finding the maximum delay. The counter was operated in the normal way, with a "dead" CO_2 filling, and uncanceled counts were recorded. The quench interval was gradually shortened with the expectation that when it became shorter than the maximum transit time for the negative ions, the uncanceled count would start to rise. Such a test is highly sensitive, because over 200 mesons pass through the counter per minute, and the late arrival of a single negative ion from these will give an uncanceled count.

After some exploratory runs it was found that the rise in uncanceled counting rate set in suddenly at about 9 msec. Data were then obtained at the four settings listed in Table III with 1% statistical uncertainty.

The results show excellent agreement with the values for maximum transit time found in the former experiment.

C. Evidence That CS_2 or a Product of CS_2 is the Charge Carrier

It is of interest to use the data on delay for the calculation of the mobility of the negative ion in the hope that the result may be of help in determining its species. The conditions of our measurements were anode radius, 0.0064 cm; cathode radius, 3.66 cm; potential difference, 6150 v. These data yield a value for the mobility of $9.4\times10^{-3}/T$ cm^2/v sec, where T is the maximum delay in seconds. The result is approximately 1.04 cm^2/v sec. In attempting to compare this with known mobility data, we were unable to find any information in the literature concerning a CS_2 negative ion, if indeed such an ion exists. Experimental data for the mobility of the negative ions of oxygen in CO_2 are available for approximately the conditions of our CO_2-CS_2 counter.[22]

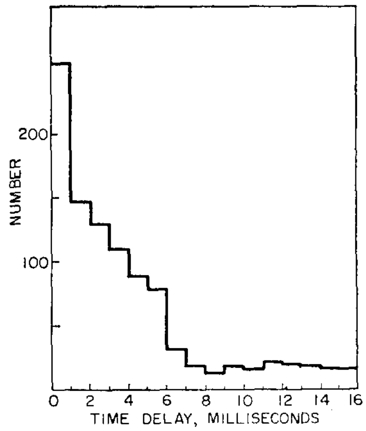

FIG. 5. Histogram showing the delay in the firing of the CO_2-CS_2 counter with respect to the time of firing of the anticoincidence ring.

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[22] E. W. McDaniel and H. R. Crane, Rev. Sci. Instr. 28, 684 (1957).

TABLE III. Uncanceled counting rate as a function of quench duration.

Quench duration in milliseconds	Uncanceled counts per minute
19.3	7.52 ± 0.07
9.7	7.48 ± 0.07
8.3	8.30 ± 0.08
5.1	9.00 ± 0.09

The value is 1.08 cm²/v sec at standard conditions of pressure and temperature. This may be corrected to the pressure and temperature conditions that we used; namely, 73 cm Hg and 26°C, respectively. For the present purpose we shall arbitrarily assume that the pressure was all due to CO_2. The oxygen ions under these conditions should have had a mobility of 1.23 cm²/v sec. This is a higher value than the observed one, 1.04. While this result does not exclude oxygen as the carrier, it does argue for a carrier of lower mobility.

There is a further dissimilarity between our results and the one found for oxygen negative ions in CO_2. In the experiments of McDaniel and Crane[22] the maximum mobility of the oxygen ions was found to be sharply defined. However, ions were also present having mobilities that extended down to about $\frac{1}{2}$ the maximum value. These were assumed to be ions which formed clumps of larger mass. The present results on the uncanceled counting rate vs quench duration show that there is not a detectable number of negative ions (or clusters) with excessively low mobility. In other words, there is no "tail" to the ion mobility spectrum. This is additional evidence against the idea that the carrier in our counter was oxygen.

As a further investigation into the identity of the carrier, the counter was filled with (a) 60 cm Hg of argon, 10 cm ethane, and 2 mm O_2, and (b) 60 cm argon, 10 cm ethane, and 2 mm CS_2. The pulses were observed visually on the synchroscope, with the sweep triggered by the anticoincidence ring. In the case of filling (a), delayed pulses were not seen. In the case of (b), the pulses were delayed in what appeared to be the time spectrum typical of the normal CO_2-CS_2 filling.

In summary, our experimental results lead us to the tentative conclusions that CS_2, or a product of CS_2, is the carrier; that CS_2 has a high cross section for capture of electrons, even in a gas in which the agitation energy is high; and that the resulting negative ion breaks up readily in the field near the wire. A further inference can be drawn from the experiments on impurities. The lack of sensitivity of the counting rate to the presence of electronegative impurities indicates that under the conditions which applied to our counter, the impurities are poor competitors with the CS_2 in terms of cross section for the capture of electrons.

D. Effect of Contaminants

Tests for the effects of air as an impurity in the CO_2-CS_2 counter were made by Mościcki.[11] His data indicated that a partial pressure of 4 mm Hg of air depressed the counting rate by about 13%. He further remarked that the count recovered partially after several days of operation.

We made several tests of the effect of oxygen and of SO_2 as contaminants, these gases being chosen because of their strong electronegative characteristics. The total counting rates are given in Table IV, with the rates for the uncontaminated counter given for comparison. The latter data are the same as those used for the plateau curve (Fig. 4), and are used for comparison because they were taken immediately before the data on the contaminated counter were taken, and under the same conditions.

The data show that at the voltage at which we normally operate the counter (about 500 v above threshold), 1% oxygen contamination has no perceptible effect, but that it does seem to depress the counting rate slightly at voltages near the threshold. 0.3% SO_2 has a more marked depressing effect at low voltages, and it seems to persist, to the extent of 2 or 3 %, up to the highest voltage used. The data indicate that the effects of these two contaminants can be safely neglected, at the small concentrations at which they would occur in a normal filling, and at 500 v above threshold. As an explanation of the effect of SO_2, it seems reasonable to suppose that the negative ion formed from SO_2 has considerably less than a 100% chance of breaking up and releasing the electron in the field near the wire. In our counting system, the fact that an electronegative contaminant attaches electrons is not objectionable in itself, if the ion breaks up near the wire. The time delay caused by attachment has no effect on the counting rate.

An additional indication of the insensitivity of the counter to impurities is furnished by an observation we have made repeatedly: namely, that commercial tank-CO_2 of gas-well origin gives the same counting rate, to within the statistical accuracy, whether it is put into the counter directly from the tank, or first precipitated to $CaCO_3$ and then passed through our standard sample preparation procedure.

TABLE IV. Effect of electronegative contaminants on counter plateau.

Volts above threshold	Counts/min 1% O_2	Counts/min 0.3% SO_2	Counts/min uncontaminated
80	180	140	201
160	208	150	211
320	214	207	218
480	222	not run	222
640	219	214	220
800	224	220	222
960	221	215	222

In connection with the effects of impurities, it is appropriate to mention one which Mościcki found and called a "forming" effect. We, also, have noticed the behavior but have not made measurements upon it. It occasionally appears as a decreased counting rate when counting is started on a sample, particularly if the voltage is set close to the threshold. It may persist for an hour or even several hours. We think the effect can be explained in the same way the initial fog in a cloud chamber is explained. The gas initially contains some condensation centers (molecular clumps and impurities) which attach charges and which have less than 100% chance of breaking up in the field near the wire. The gas is soon cleared of these condensation centers by precipitation upon cathode and anode.

The large effects of air contamination which Mościcki found are not considered to be in conflict with our results. He worked very near the threshold (about 100 v), a point at which our data show that there is, in fact, a marked depression of the counting rate. He also let air into his counter continuously while making the test, so that the "forming" effect may have been important. These facts may well account for his observation of a 15% depression in counting rate.

E. Efficiency

As a direct check on the efficiency of the counter, we present in Table V a set of comparison runs between a CO_2-CS_2 filling and an argon-ethane filling, taken successively and with all conditions identical. The AC counters were turned off.

The difference in the first row is real but not significant, because 170 v above threshold is not on the flat part of the plateau curve. In the last three rows, there is agreement to within the statistical accuracy.

Another set of comparison runs, taken half a year later and with a different counter, is shown in Table VI. The AC counters were turned off.

The synchroscope arrangement described in the section on time delay was used to observe, visually, the pulse in the Ar-ethane filled counter. As far as could be determined by visual inspection, all the pulses were coincident with the AC pulses to within less than 0.1 msec, indicating that, in contrast to the case of the CO_2-CS_2 counter, the electrons were unattached.

TABLE V. Comparative efficiencies of CS_2-CS_2 and argon-ethane fillings.

65.5 cm CO_2, 3 cm CS_2		56.5 cm Ar, 6 cm ethane	
Volts above threshold	Counts per min, total	Volts above threshold	Counts per min, total
170	198±2.0	170	191±2.5
340	204±1.5	340	204±1.6
510	207±1.5	510	208±2.0
680	208±1.5	680	210±1.8

Using the same counter and gas fillings as for the preceding set of data, we ran a comparison in which the scattered radiation from a small radium source was introduced. This was a way of producing low energy secondary electrons in the counter, of energies comparable to those of C^{14} beta rays. The results are shown in Table VII. All the data were taken at 500 v above threshold.

In the above experiment there is a reason for expecting a slightly higher counting rate from the CO_2-CS_2 filling than from the argon-ethane filling, and which should be mentioned, even though the differences are barely outside of the standard deviations. As described earlier, the integrated time during which the anode is at counting voltage is read accurately from the timing scaler. However, since the negative ions produced by a single C^{14} beta ray or by a single meson arrive at the anode over a small spread in time, the effective "on" time of the counter is slightly increased over that given by the timing scaler. (We might call this a penumbra effect.) We have not attempted to measure this correction to the CO_2-CS_2 counter efficiency. However, if we make the reasonable guess of 2 msec for the average time dispersion in the arrival of the negative ions, we can easily calculate the correction to the "on" time of the CO_2-CS_2 counter. It is about 3% greater than that given by the timer scaler in the "with anticoincidence" runs just listed and about 1.5% greater in the "without anticoincidence" runs. The runs with the argon-ethane filling are, of course, not subject to the correction. The differences calculated are consistent with the observations. In Table II and in the plateau curves of Fig. 4, for which the count was about 200/min with the anticoincidence counters off, the correction is only about 0.7%. It should be possible to evaluate the correction just discussed by making a series of runs at various values of quench interval. It would be important to do so, however, only if absolute measurements of the counting rate were to be made, as, for example, in the determination of the half-life of C^{14}.

TABLE VI. A comparison similar to Table V taken on a different counter six months later.

73 cm CO_2, 3 cm CS_2		65 cm Ar, 11 cm ethane	
Volts above threshold	Counts per min, total	Volts above threshold	Counts per min, total
		80	79±3
		160	156±1
		320	214±1.5
480	212±0.7	480	215±1.5
640	212±0.7	640	212±1.5
		720	214±2
800	214±0.7	800	207±1.5
		960	215±1.5

For the sake of completeness we should mention two other effects which can contribute minor differences between the counting rates of the CO_2-CS_2 and the argon-

TABLE VII. Comparison of CO_2-CS_2 and argon-ethane fillings in presence of low energy scattered radiation. All data at overvoltage of 500.

	73 cm CO_2, 3 cm CS_2 Counts/min	65 cm Ar, 11 cm ethane Counts/min
With anticoincidence	251 ± 2	247 ± 3
Without anticoincidence	467 ± 3	453 ± 5

ethane filled counters: (1) The electron density and the atomic number of the gas have effects which are energy dependent. The former is nearly the same in the two cases, but the latter is different by more than a factor of two. (2) In the argon counter, as the electrons are pulled to the anode from far out in the counter, there is appreciable lateral diffusion. This slightly increases the volume from which electrons can reach the sensitive part of the wire. In the CO_2-CS_2 counter, the charge is carried by negative ions which have a much smaller diffusion coefficient, so a smaller effect is to be expected.

A different kind of check on the efficiency of the CO_2-CS_2 counter may be found in the interpretation of the plateau curve (Fig. 4). The fact that the plateau is exceedingly flat indicates that certain field-dependent effects which might cause a loss of counts are essentially absent. These effects are (1) recombination of the electrons or negative ions with politive ions before reaching the anode wire, (2) failure of the negative ions to break up in the strong electric field near the wire to yield free electrons which can make avalanches, and (3) premature extinction of the incipient Geiger discharges as they begin spreading along the wire. All three of these effects would be expected to diminish markedly with increase in voltage. We have seen that this does not happen: that over a range of voltage of more than 20%, the counting efficiency is constant to within about 1%.

In summary, we may remark that we have gone to some lengths to investigate the efficiency of the CO_2-CS_2 counter because we are aware that it has been held in question. In the two instances[4,7] in which the half-life of C^{14} was obtained by means of the CO_2-CS_2 counter, results were found which are in disagreement with the presently accepted value. The situation in this respect was summarized by Miller *et al.*[7] in 1950: These authors proposed that the fault lay in an inherent loss of counts in the CO_2-CS_2 counter amounting to about 15%. In the light of our own checks on the efficiency and in consideration of the nature of the plateau curve of the CO_2-CS_2 counter, we can only conclude that the disagreement must have been due to some other cause.

VII. GENERAL COMMENTS ON OPERATION

In this section, several items of general interest relating to the practical use of the CO_2-CS_2 counter will be reported; the special lore relating to the C^{14} dating procedure will be omitted (The latter is available in another form, as mentioned earlier.)

A. Filling

Because of the insensitivity of the counter to impurities, there is no necessity for evacuating the counter completely. We pump it to about 1 mm Hg with a mechanical pump, and flush with CO_2 (of gas well origin) from a tank. It is then ready to be filled with the sample to be measured. CS_2 vapor from a flask containing the liquid is first introduced to 3 cm Hg pressure, then the CO_2 sample is transferred to the counter by means of a liquid nitrogen trap to a total pressure of 1 atm.

B. Background and Counting Rate

The material of which our counters are made is electrolytic copper, prewar stock. The background counting rate (with anticoincidence) when the counter is filled with "dead" CO_2 is 8/min. A systematic search could very well reveal a source of material having a lower radioactive contamination level.

C. Choice of Pressure and Other Parameters

We have not operated the counter at more than 1 atm pressure. We see no reason why at least 2 atm would not be feasible. Since the negative ion drift time is long and an appreciable fraction of the total measuring time consequently must be sacrificed to deadtime, a reasonable compromise must be made among the several variables. The approximate interrelations among these are as follows: The drift time τ from cathode to anode is $(PR^2/KV)\log_e(R/r)$, where P is the pressure in atmospheres, R and r are the cathode and anode radii in cm, respectively, V is the voltage, and K is the mobility in cm^2/v sec at atmospheric pressure. Empirically, the threshold voltage V_0 of the counter varies approximately as $(rP)^{\frac{1}{2}}$. It is relatively insensitive to R. If we make this substitution and omit the $\log_e(R/r)$ factor, which is varying only slowly, we see that the drift time depends upon the dimensions and pressure approximately as $R^2(P/r)^{\frac{1}{2}}$.

For the C^{14} dating application the important consideration is the mass of gas M contained per unit length of the counter; $M\sim R^2P$. Combining the expressions for τ, M, and V_0, we see that $M\sim TV_0$. If we make various combinations of P, R, and r such as to keep τ constant, we find the simple result that the mass of gas that can be put into the counter (per unit length) is proportional to the threshold voltage. There are, however, two incidental advantages to be gained by going in the direction of smaller R: (1) The cathode area will be decreased, which will bring about a decrease in background counting rate, and (2) the size of the AC counter bundle will be decreased, which will result, through a decrease in meson count, in an increase in the "on" time. (The "on" time is about 85% in the present system.)

In summary, there appears to be no reason for not increasing M by a factor of at least 2. Such a design might be the following: $R=2$ in., $P=4$ atm, and $V_0=11\,000$ v, replacing the present values of $R=3$ in., $P=1$ atm, and $V_0=5500$ v. The factor r would remain the same. M would be up by a factor of 2 and the background from the cathode wall would be down by a factor of $1/\sqrt{2}$. In return for these advantages, it would be necessary to exercise more care in avoiding corona and it would be necessary to use a high pressure filling system.

D. Reliability

Our observations of two CO_2-CS_2 counters operated continuously for many months at a time uncovered no evidence of variations in counting rate outside those expected from the counting statistics. On several occasions, following several months of continuous operation, we found a sudden rise in the background count; for example, an increase of 2–4 counts/min over a 12-hr period. Replacement of the anode wire always cured the trouble, which was probably due to a small point or spot which had developed by a sputtering process. That this can happen is not surprising, in view of the fact that the anode in the CO_2-CS_2 counter is bombarded by negative ions.

We have tried tungsten, platinum, and Chromel A for the wire, and have found the last to be the least subject to this effect.

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Generation of High Power Radio-Frequency Pulses by Means of an Exploding Wire Technique

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A method of generating high power radio-frequency pulses is described which utilizes the "current dwell" period which can occur when a thin wire is exploded by means of a high current. The rf oscillation occurs in a resonant circuit placed in series with the exploding wire. A circuit is described in which is obtained a 6-μsec duration pulse of 2.74-Mc oscillations.

SOME plasma physics experiments require high power radio-frequency pulses having a duration of only a few microseconds. One method of obtaining such pulses using a saturable core reactor has been described by Westendorp and Hurwitz.[1]

Another method of obtaining such pulses involves discharging a capacitor through a triggered spark gap in series with a fine copper wire and a radio-frequency tank circuit. The initial surge of current which passes through the copper wire before it explodes charges the tank circuit. For some microseconds after the wire has exploded, the tank circuit is effectively isolated from the low frequency charging circuit. (This period is referred to as the "current dwell" in exploding wire literature.[2]) During this dwell time, the tank circuit oscillates with a decay time determined only by the losses in the inductive and capacitative sections of the tank circuit. This method of charging the tank circuit removes the necessity for a conventional switch in that circuit and thus eliminates a reduction in the Q of the circuit due to ohmic losses in such a switch.

The circuit diagram is shown in Fig. 1. The component values listed are for a particular experimental test. The low

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[1] W. F. Westendorp and H. Hurwitz, Jr., Rev. Sci. Instr. **31**, 662 (1960).

[2] W. G. Chace and H. K. Moore, *Exploding Wires* (Plenum Press Inc., New York, 1959).