An Automatic Counter for Age Determination by the C^{14} Method

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SINCE 1949, when the first successful measurements of the ages of relics by the C^{14} method were announced by Libby, Anderson, and Arnold (*1*), several laboratories have taken up such studies. All the installations, with the exception of the one to be described here and one other (*2*), have followed quite closely the original design described by Libby (*3*). During the past three years we have developed an apparatus that differs from older ones in some important ways in regard to mechanical construction and circuitry, and we have introduced some modifications into the techniques of measurement. In particular, the apparatus is completely automatic in operation. Since the general ideas of age measurement have been presented in detail in other papers (*4, 5*), the discussion in this article will be restricted to the physics of low background counting and the description of our own apparatus and technique. The topics discussed, which constitute the essential requirements for extremely low background counting, are the following: (1) reduction of cosmic ray background by means of anticoincidence counters; (2) reduction of local soft radiation background by means of a shield; (3) avoidance of the use of metals that contain traces of radioactivity in the construction of both the shield and the counter; (4) frequent alternation between the unknown specimen and known comparison standards to reduce the effect of slow drifts in sensitivity and background; (5) stability of all circuits and components over long periods; (6) reduction of spurious counts, such as those caused by negative ion formation and cathode activation in the counter; and (7) cleanliness and reproducibility in the technique of handling and mounting the samples.

Mechanical Details of the Counter

An essential feature of the counter is that the envelope is long enough to provide "parking space" for three sample cylinders. These are moved from the outside by means of a motor so that, one at a time, they form the cathode around the counter wire. There is a separate clock and count register for each sample cylinder, and these accumulate the total time and counts for each sample separately. A programming system, consisting of a clock and relays, actuates the changing of the samples, switching of the clocks, registers, etc., so that long runs are made without attention.

As shown in Fig. 1, the counter envelope consists of a 3¾″ I D seamless steel tube 61″ long (inside), sealed at the end with flat plates having O-ring gaskets. The three sample cylinders are iron, 9″ long, 3¼″ I D and 3/32″ wall thickness, and have 3/16″ lips at the ends. They are mounted, with 3″ spaces between them, on a piece of ¼″ O D, .03″ wall brass tubing, 33⅝″ long. Six ⅝″ × ⅝″ × 1/16″ fins are silver-soldered to the side of the brass tube, and these serve as mounting lugs for the cylinders. Each cylinder has a tapped hole near each end and is attached to its mounting lugs by screws. The ¼″ brass tube serves as a sheath for a 3/16″ steel lead screw, having a double thread, 13 revolutions/inch, and 27¼″ long, including 1″ at the end which is not threaded. A ½″ long steel nut is silver-soldered to the end of the sheath tube. Graphite with oil is used to lubricate the screw. When the lead screw is turned, the assembly of three sample cylinders moves along so that any cylinder can be brought into the counting position. The assembly of sample cylinders rests on the bottom of the envelope, leaving the 5/16″ space above, which is occupied by the screw and its sheath. No guides are used to keep the assembly in position; gravity seems to serve well enough, provided care is taken to make the lead screw and sheath straight, so that little torque is exerted on the assembly.

The method used for transmitting the rotary drive for the lead screw into the envelope avoids having any rotating shaft or sliding surface form a part of the vacuum seal. It has required no attention in more than a year of steady operation. The principal part is a crank, which is merely a curved piece of shaft (Fig. 1). Both ends of the crank turn in ball bearings. The cup over the outer end of the crank moves in a cone but does not rotate. The cup is driven by a slotted cam or wheel, into which the boss on the cup fits loosely. The piece of rubber tubing connecting the two ball-bearing cups is supported by an internal spring wire helix, so that it will not collapse under vacuum. Both the inner ball-bearing cup and the support for the wheel bearing are silver-soldered to the end plate of the counter envelope. To permit checking on the operation of the drive system, a flashlight bulb is mounted on the inner face of the end plate, with an electrical connection to the outside, and a 1″ glass window is provided.

The lead screw mechanism is driven by a geared

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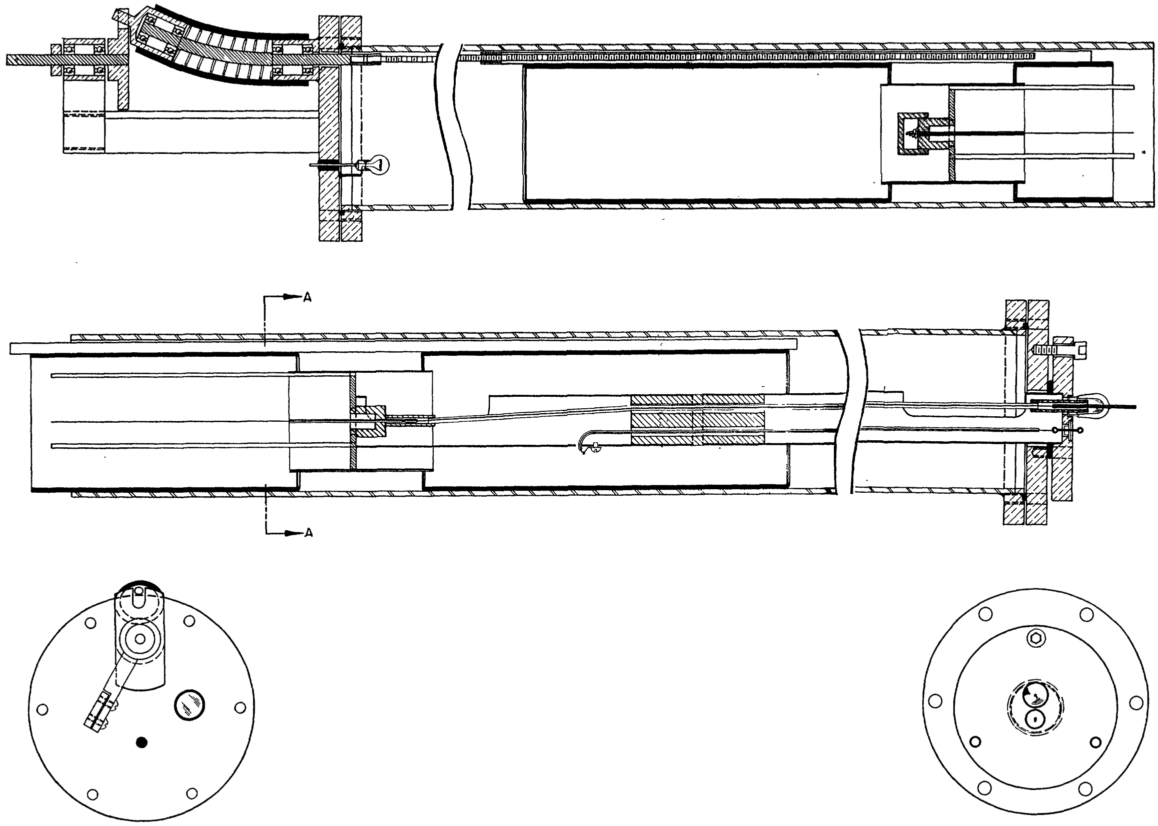

FIG. 1.

reversible motor at 2 rps. A second lead screw, of the same pitch and length, but of larger diameter, is located outside the counter and shield, and is driven by the same motor. The latter carries a nut that travels in unison with the sample cylinders. Adjustable micro-switches are placed so that one is depressed by the nut when the sample assembly reaches each of its three counting positions. These cause the drive system to stop at the correct place after each change.

The anode assembly is cantilevered from the opposite end of the envelope by means of a 1¼″, 1/32″ wall steel tube. Its center line is 5/32″ below the center line of the envelope. The tube is partly cut away at each end to provide access to the electrical connections and is broken electrically near the middle by means of a Lucite plug, in order that the anode supporting structure (including the screen, if used) may be maintained at a potential different from ground. The end plate of the envelope is double, providing for alignment by means of three screws, as shown in Fig. 1.

A circular plate, 2½″ in diameter, is silver-soldered to the end of the 1¼″ tube, and another plate of the same dimensions is supported 11½″ (inside measurement) beyond this by means of three 3/32″ hard steel rods. The plates have slots cut in their edges at intervals of 3/16″. When a screen wall is used, it is made by lacing .01″ copper wire back and forth in these slots. The high voltage insulators, at the centers of the plates, are made of polystyrene. The anode wire is .007″ tungsten, which is fastened at the inner end and pulled by a spiral steel spring at the other end. Pieces of 1/16″ O D copper tubing cover the tungsten wire 1½″ outward from each plate, limiting the sensitive length of wire to 8½″. Pieces of .01″ sheet steel rolled into cylinders 3½″ long and 2½″ in diameter are placed around the end plates and are held in place by friction. Their edges are even with the ends of the copper sleeves on the tungsten wire. These cylinders, with the copper sleeves, limit precisely the length over which the counter is sensitive. They also make it virtually impossible for beta rays to arrive, even after scattering, from a neighboring sample cylinder. All metal parts of the anode assembly are steel, and all joints are made with silver solder. The high voltage connection, which runs inside the 1¼″ steel tube from a Stupakoff lead-in bushing in the end plate of the envelope to the copper sleeve that holds the counter wire, is rubber-insulated. Glass sleeves are slipped over the joints at the ends. After assembly the tungsten wire is cleaned by flaming to white heat with a Bunsen burner.

Shield. The shield is built of slabs of 2″ thick iron "boiler plate." Counting from the center of the shield toward either end, the thickness is 8″ for a distance of 15″, 6″ for the next 7″, and 4″ for the final 16″. The ends are closed with 2″ thick movable slabs. When the shield was first built, about 1 ton of lead, in 100-lb pigs, was stacked on top of the middle section. Later comparisons of counting rate with and without the lead have shown that it reduces the background by not more than .2 cpm.

Circuits

Carbon counter. The voltage of the carbon counter is regulated according to pulse size by means of a circuit previously described (*6*). This maintains the operating voltage at very nearly a constant point relative to the threshold, regardless of shifts in threshold that may occur as a result of aging of the gas in the counter. This has been found to be of value in making long C^{14} runs (48 hr or more) unattended, and in starting within 1 or 2 hours after filling the counter. In a counter containing a large area of metal and rubber, besides the freshly prepared carbon samples, some gradual change in the composition of the gas and hence in the threshold is to be expected. The time constant of the regulator is such that in normal operation the voltage on the counter rises at the rate of about 1 v/sec. When the voltage gets to a high enough value that a count pulse is able to fire the thyratron, the voltage is reduced by 2 v. Thus the counter voltage has a superimposed sawtooth component of about 2 v amplitude and 2 sec period. To indicate the degree of regulation achieved, Table 1 gives the actual voltage maintained on the counter for various values of high voltage supplied to the regulating circuit.

Although the above experiment was done in reverse, so to speak, it can be concluded from the figures that, if the supply voltage were set at 1400 v, the threshold could move about 200 v in either direction and carry the operating point of the counter along with it. The actual threshold, in the above case, was 1125 v. The number of volts above the threshold at which the circuit stabilizes (75 in the above case) is determined by the thyratron bias. The circuit diagram for the unit containing the amplifier, regulator, and univibrator is shown in Fig. 2.

TABLE 1

Supply voltage	Counter voltage
1120	1120
1190	1175
1230	1195
1445	1200
1520	1205
1625	1207
1660	1215
1695	1225
1730	1230
1805	1245

The counter pulses are fed, from the output of the amplifier stage, into a univibrator that has a recovery time of 15 msec. The output of the latter is then differentiated so as to give pulses of about 1 msec duration. These go to the anticoincidence mixer. The artificial "dead time" of 15 msec is introduced to reduce the background of spurious counts. Spurious counts associated with the ion transit time in the counter follow counts within a few hundred microseconds, and those associated with cathode activation (*7, 8*) have a probability that decreases rapidly after each count. The first of these kinds of spurious counts is practically eliminated, and the second is greatly reduced, by the dead time, at the sacrifice of only 3% of the real counts.

External counters, anticoincidence mixer, scaler, and register circuits. There is nothing unique in our anticoincidence mixer, scaler, and register circuits—one standard method would have served as well as another. Therefore, only some brief comments about the system in general are necessary. The external counters are commercial ones, 1″ × 20″, copper wall "self-quenching." Fifteen of them are grouped around the carbon counter envelope and connected in parallel. A univibrator quench is used with these for two reasons: it provides uniform, large pulses to be fed into the mixer, and it allows the counters to be continued in service after their quench vapor has been used up.

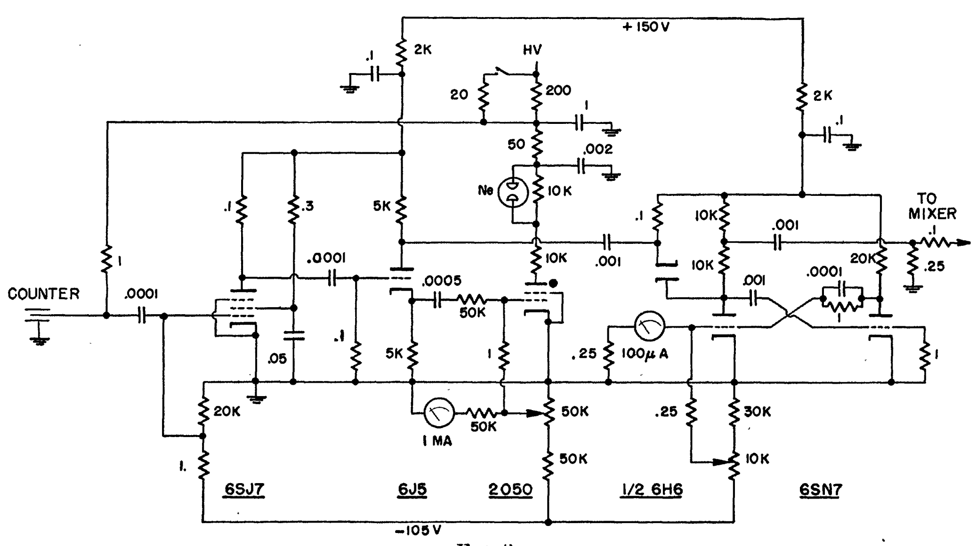

Fig. 2.

Advantage is taken of the fact that the counting rate is very small, and pulses of generous width are used throughout the system—about ½ or 1 msec. In the mixer the pulses coming from the external counters are inverted and added, in a simple resistance network, to those coming from the carbon counter. Since the pulses from both counters have been made uniform by univibrators, this manner of mixing leaves nothing to be desired. The use of a nonlinear mixing scheme, such as the Rossi circuit, would add nothing, since infinite nonlinearity, so to speak, has already been introduced by the univibrators. The anticoincidence pulses thus formed are sent through a scale of four, the output of which goes to the univibrator which actuates the registers. The scale of four is used only because the particular registers used are slow, having a revolving time of ¼ sec.

Power supplies. A dual, electronically regulated high voltage supply (0–2000 v), a negative bias supply (−90 v) and several 6.3-v heater supplies are combined on a single chassis. These circuits are standard and need no description.

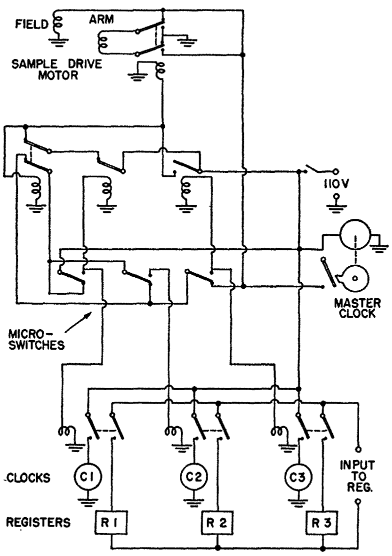

FIG. 3.

Programming system. A complete circuit diagram for the system, which shifts the sample cylinders, switches the clocks, registers, etc., is shown in Fig. 3. A master clock turns a cam that closes a microswitch momentarily to initiate each change of the sample cylinders. The only precaution necessary is that the master contact must remain closed until the nut on the external lead screw has moved off its microswitch, and must be open again before the nut gets to the next position. Provision is made in our cam system to allow unequal times to be spent in the measurements of the three samples. This is necessary in order to obtain the best statistical accuracy for a given total expenditure of time, when the counting rates of the three samples are not the same. If, for example, the unknown is near contemporary, most of the time will be spent on the unknown and the contemporary, and little on the "dead" sample. The remainder of the circuit is believed to be self-explanatory. The only items not shown are two emergency stop microswitches, one at each end of the external lead screw, which cut off the power in case the system should fail to stop at either end position.

A hold-down relay is inserted in the 110-v power line to the entire apparatus so that it will remain off after a temporary power failure. This precaution is essential because the various voltages and filament temperatures do not come up to full value together, thereby causing spurious counts.

OPERATION AND CHARACTERISTICS OF THE COUNTER

Counting rates. In normal operation the counting rates are: external counters, 800 cpm; carbon counter with dead carbon, 125 cpm total and 8.4 uncancelled cpm; carbon counter with contemporary carbon, 16.5 uncancelled cpm. The effective area of carbon is 583 cm^2. The number of cpm/cm^2 above background for contemporary carbon is .0138. When corrected for the 3% dead time of the univibrator blanking circuit, this becomes .0142 cpm/cm^2. This value is somewhat lower than the value .0165 indicated by the data given in a recent paper by the Chicago group (*4*). Although we cannot be certain as to the reason for this difference, it is possible that the elimination of some spurious counts by the blanking univibrator, combined with the absence of a high "drag voltage" on the screen may account for it.

Plateau. The plateau of the carbon counter is surprisingly long. A recent measurement gave an operable range from 1200 to 1600 v with a slope of 1%/100 v.

Screen wall. The screen, or grid, has been assumed to be an important element in the C^{14} counter, and we have carried out some extensive experiments as to its behavior. The counter has been operated over many months, both with and without a screen wall. When operated at a positive potential (up to 150 v) with respect to the cylinder, the screen has been found to have no measurable effect on either the counting rate or the consistency of the rate. In view of the fact that the carbon layer is such a good conductor[2] that its surface potential is that of the metal cylinder, this result is to be expected. The reasons are as follows: When the screen is not present, the potential of the space at the screen radius is easily calculated to be about 50 v positive with respect to the cylinder. If, therefore, the screen is inserted and maintained at +50 v, it will not modify the potential distribution in the counter. Both the outward-drifting positive ions and the inward-drifting secondary electrons will pass through the screen as if it were not there, except that a fraction of each, equal to the geometrical opacity (5%), will be collected by it. (The 5% loss will not be expected to reduce the counting rate by 5%, because each beta ray makes many ion pairs, and it will not often happen that all the secondary electrons from a given

[2] A measurement gave 30 ohms for the resistance across the layer when contact was made to 1 cm^2 of the surface.

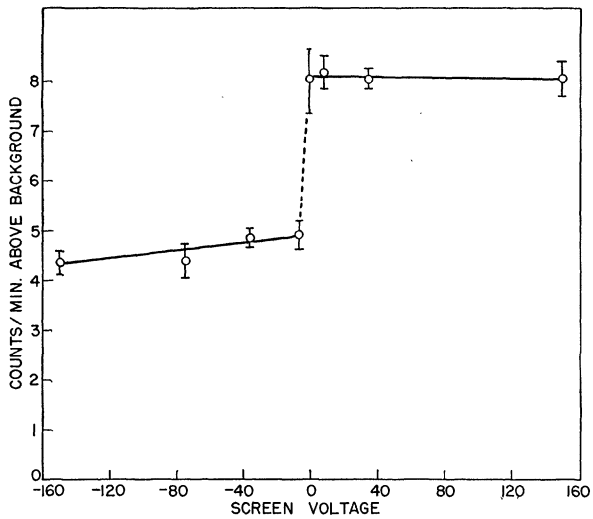

FIG. 4.

beta ray will be collected by the screen). Operated at this potential, then, the screen does not form the real cathode, except to the extent of 5%. If, in the geometrical case described, the screen is given a potential higher than 50 v, it will collect somewhat more than 5% of the positive ions and less than 5% of the secondary electrons, and, if lower than 50 v, the reverse will be true.

If a negative potential of 8 v is applied to the screen, secondary electrons formed in the space between the screen and the cylinder will just be repelled back to the cylinder. By analogy with a triode, this is the cutoff voltage; "μ", computed from the geometry, is about 130. Therefore, when the screen is at 8 or more volts negative, the effective size of the counter is that of the screen. Only those beta rays that produce ion pairs inside the screen will give counts, and the screen acts as a real cathode.

The experimental curve in Fig. 4 shows the counting rate as a function of screen potential, taken under usual operating conditions with a contemporary carbon sample. It is consistent with the arguments presented above.

When operated at a negative potential, a screen forms the real cathode and protects against all the possible bad properties of the carbon layer—poor conductivity, photoelectric sensitivity, spontaneous electron emission, emission of electrons by positive ion collection, roughness, eccentricity, variation in radius, etc. The price paid for this is the reduction in counting rate indicated in Fig. 4. When the screen is operated at a positive potential, the field strength around the anode wire is maintained constant against all geometrical variations in the carbon layer, and protection against the effects of surface charge caused by poor conductivity of the layer is obtained. There is, however, no protection against the three types of electron emission mentioned above. Since our own sample layers are smooth and of uniform diameter, and since the carbon seems to be a good conductor and a well-behaved cathode surface, we have not found any advantage in using a negative screen or, in fact, any screen at all.

Thickness of sample vs. counting rate. A comfortable excess of carbon above that necessary for a layer of saturation depth is not always available, and in some cases it is necessary to work with less than saturation depth. Since the transmission curve for C^{14} beta rays is already well known, an experimental check in this case becomes mainly a check on the uniformity with which the carbon layer is spread in the cathode cylinder. The results of a series of measurements with various quantities of carbon are shown in Fig. 5. The descending curve is the transmission of C^{14} beta rays in aluminum (*7*), which should be very nearly correct for a carbon layer, and the ascending curve is its integral, or the expected thick-layer counting rate. The experimental points show good agreement and indicate that our carbon-spreading technique (*5*) gives a uniform layer, even for a few mg/cm². Visual inspection of the layer confirms this conclusion.

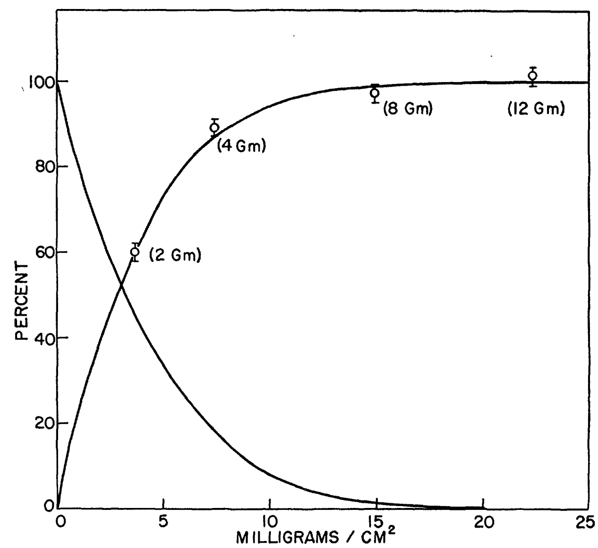

FIG. 5.

Effect of atomic number of cathode on background rate. In comparing the counting rate of a bare cylinder with a cylinder coated with a sample, the difference in efficiencies of the surfaces as "radiators" for conversion of the soft local radiation into secondary electrons must be recognized. As an experiment, the background radiation was increased by about a factor of 10 by placing a wrist watch, having a luminous dial, inside the shield. The mixture of gamma radiation from the watch (radium and its products) is probably not far different from that given by the local radioactive contamination. The increase in counting rate for an iron cylinder was about 3% more than that for a carbon-lined cylinder, and the increase for a tin-lined cylinder was 30% greater than for the carbon-lined cylinder. This shows that, in the same radiation environment, different backgrounds will have to be applied to samples of different atomic numbers.

Reduction of Background Count

Selection of materials for shield and counter.[3] Of the materials having reasonably high density and atomic number, for shielding, lead and iron are the two cheapest and most readily available materials. Lead is rejected because of its radioactive contamination. Lead occurs frequently in association with radioactive ores, and these are not entirely eliminated in the refining process. In addition, there is the long-lived (22 years) radioactive isotope of lead, which cannot be removed, and which occurs in varying concentration depending upon the source of the lead. Its gamma ray is .47 Mev energy. We have found lead to vary in radioactivity but have not found any with low enough background to make it useful as a shield. We have obtained approximately the same result (within 1 cpm) with a 3″ lead shield having a lining of 2″ of copper (bus bar stock) plus ½″ of iron, as with the all-iron shield described.

The envelope of the counter and all the inside metal parts are made of iron, steel, or copper except the anode wire, which is tungsten (not thoriated). No soft solder is used. "Free-machining" brass contains up to 3% lead, so it is not used. Libby has reported satisfactory experience with lead-free brass, but similar material which we were able to obtain was found to be contaminated.

Iron and copper have been used for the sample cylinders with equally good results.[4] A set of stainless steel cylinders was made and found to give a background from 2 to 3 cpm higher than that observed with the iron or copper cylinders. Iron cylinders have an advantage over copper in that they can be cleaned conveniently by etching in dilute HCl. Oxidation is prevented by wiping a thin film of oil on them after etching. It has been found from experience that three cylinders, even when cut from the same piece of stock, cannot be assumed to have the same background counting rates. For this reason, the two permanent comparison samples (contemporary and dead) are calibrated against contemporary and dead carbon placed in the position of the unknown sample. The two permanent samples are, therefore, secondary standards. The same sample is found to give up to .3 cpm difference in different cylinders.

Argon (9 cm) and ethane (1 cm) are used as filling for the counter. So far no trouble has been experienced with commercial argon, although in this Atomic Age no noble gas should be considered to be above suspicion. One of the most abundant of the fission products is Kr^{85} (9.4 years), and undoubtedly there is now much of it in the atmosphere.

Airborne radioactivity. The shield is not airtight. Although no experimental data are available to prove the point, we believe it would have been worth while to have constructed the shield in such a way as to prevent air from drifting through it. It is well known that outdoor air carries radon and thoron in such concentrations as to give, when their short-lived products are included, the order of 1 disintegration/min/l. The concentration of these emanations can be much greater inside a closed building with masonry walls or floors, because of the high uranium and thorium content of some sands. Airborne radioactivity of other kinds is also a possibility in any large research laboratory. The air in a sealed shield would soon become inactive.

The precaution of protecting the carbon samples and the cylinders from unnecessary exposure to air is also worth while. Thorium B (10.6 hr) will be deposited electrostatically on an exposed sample or cylinder and, with its products, which are all of shorter half-life, will give a background count. Radon is not so harmful, because all the half-lives in the chain following it are either very long or very short compared to a day, and they are therefore not expected to contribute measurably to contamination.[5] The atomic bomb tests in Nevada constitute a further hazard. Twice within the past year it has been necessary to discontinue the C^{14} measurements temporarily because of airborne fission products.

[3] Results of the comparison of a large number of common materials for radioactive contamination are given in a paper by J. A. Bearden (*Rev. Sci. Instruments,* **4**, 271 [1933]). A good recent survey of the occurrence of radioactivity in our surroundings can be found in "Disposal of Radioactive Wastes," by E. C. Pitzer, a report, it is understood, that will be published by the AEC Technical Information Service, Oak Ridge, Tenn.

[4] Our confidence in iron has been shaken somewhat by the recent newspaper account of the discovery, by the Jones & Laughlin Steel Co., of a commercially significant concentration of uranium in one of its iron ore deposits in the Upper Peninsula of Michigan.

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[5] A more detailed discussion, with results of some experiments, on α-contamination of carbon samples is given in "Carbon 14: Measurement of Low Level Radioactivity," by David L. Douglas, California Institute of Technology, Oct. 1951 (a progress report on an AEC project).