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The Half-Life of Radiocarbon (C^{14})

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The half-life of C^{14} has been found to be 5720±47 years by means of the use of mass spectrometrically analyzed $C^{14}O_2$ as a part of the counter gas in brass wall Geiger counters. Some of the problems of absolute β-counting with internal counters have been investigated, particularly the coincidence and end corrections.

I. INTRODUCTION

THE half-life of radiocarbon (C^{14}) has been measured several times.[1–7] The discrepancies in the results and the increasing importance of radiocarbon seemed to call for a further measurement of the half-life. The use of the gas counting technique as a method of accurate, absolute measurement for radioactivities also appeared to be worth investigation. It was further realized that since most of the measurements available at the time this research was begun used solid counting techniques, the use of an entirely different procedure would lend additional weight to any agreement with the earlier results. Finally, it was thought likely that the gas counting procedure was capable of somewhat higher accuracy than the solid counting method.

Previously published results are tabulated in Table I, together with the methods of measuring the number of C^{14} atoms in the sample used and the methods of measuring the radioactivity of the sample.

Since the first three values depend upon more uncertain methods of determining the isotopic composition of the samples, they are not comparable to the later values; but it is clear that the more recent results scatter somewhat more than the estimated errors given by the authors. The agreement, however, is sufficiently good as to suggest that the true value lies near the mean of these values. The error in this mean is difficult to estimate, but it might be permissible to take it from the observed deviations from the mean. The result of such calculations is 5687±100 years. The difficulty in accepting such a mean is the possibility of systematic errors, especially since most of the measurements used solid counting techniques. It is true, however, that the different researches used different detailed procedures and different considerations for such corrections as back scattering, self-absorption, geometrical efficiency, and counter efficiency. It is interesting, therefore, that this mean lies close to the result of the present research, 5720±47 years.

The general method used in this research was the following. A sample of barium carbonate obtained from the Isotopes Division in Oak Ridge was converted to carbon dioxide, the isotopic abundance of C^{14} determined, and an accurate dilution of several thousand fold performed. After mixing, carefully measured portions of the diluted sample were counted. The counting was done with brass-walled counters of different diameters and lengths filled with an argon-alcohol mixture to which the samples of CO_2 were added. The samples varied

TABLE I. Summary of the previously published values for the half-life of radiocarbon.

Half-life (years)	Method for isotopic composition of sample	Method for radioactivity of sample	Reference
10^3-10^5	Estimated bombardment yield	Solid carbonate counting: $CaCO_3$ in screen wall counter	1
26,000 ±13,000	Estimated bombardment yield	Gas counting: CO_2 in counter	2
21,000 ±4000	Estimated bombardment yield	a. Solid sample counting: sample with thin window counter b. Gas counting: CO_2 in counter	3
4700±470	Mass spectrometer (1.71%)	Solid carbonate counting: $BaCO_3$ with thin window counter	4
5300±800	Mass spectrometer (3.23 and 3.35%)	Solid carbonate counting: $BaCO_3$ in low absorption counter	5a
5100±200	Mass spectrometer (3.23 and 3.35%)	Solid carbonate counting: $BaCO_3$ in low absorption counter	5b
6400±200 or 6100 ±200	Mass spectrometer	Gas counting: (Miller technique[a,b])	6
7200±500	Mass spectrometer	Solid carbonate counting: $BaCO_3$ with thin window counter	7

[a] W. W. Miller, Science **105**, 123 (1947).
[b] S. C. Brown and W. W. Miller, Rev. Sci. Inst. **18**, 496 (1947)

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[1] S. Ruben and M. D. Kamen, Phys. Rev. **59**, 349 (1941).

[2] M. D. Kamen, Manhattan Project Literature, A-316.

[3] A. S. Langsdorf, Jr., and R. L. Purbrick, Manhattan Project Literature, CP-G-3272.

[4] Reid, Dunning, Weinhouse, and Grosse, Phys. Rev. **70**, 431 (1946).

[5a] L. D. Norris and M. G. Inghram, Phys. Rev. **70**, 772 (1946).

[5b] L. D. Norris and M. G. Inghram, Phys. Rev. **73**, 350 (1948).

[6] Hawkings, Hunter, Mann, and Stevens, Phys. Rev. **74**, 696 (1948).

[7] L. Yaffe and Jean Grunlund, Phys. Rev. **74**, 696 (1948).

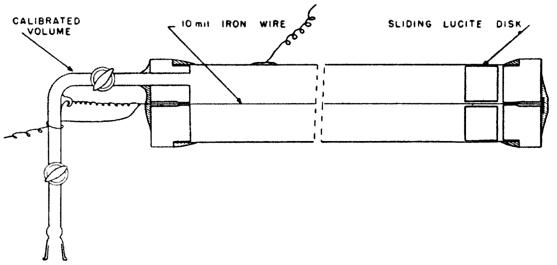

FIG. 1. Counter with Lucite ends.

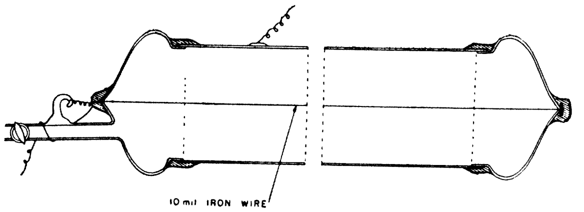

FIG. 2. Counter with glass ends.

from 0.01 to 0.1 cc S.T.P. End corrections were determined by comparing the results per unit volume of active gas from counters of the same diameter but of different lengths; a possible small variation in end correction was observed for counters of different diameters. It was assumed on the basis of previous experience reported in the literature and the basis of the absence of any appreciable variation with radius, that the counters were 100 percent efficient except for the end correction. Several CO_2 master samples were used, varying in composition from 1.24 to 6.14 atomic percent C^{14}. There was no systematic variation attributable to either the master samples or the various methods of introducing the CO_2 into the counters.

II. PREPARATION AND ISOTOPIC ANALYSIS OF THE MASTER SAMPLES; PREPARATION OF THE WORKING SAMPLES

Two methods were used for the evolution of CO_2 from the barium carbonate. The first method utilized the reaction of acid with the carbonate. About 10 mg of the Oak Ridge barium carbonate were placed in the bottom of a tube to which a ground joint had been sealed. The tube was immersed in liquid nitrogen and one or two cubic centimeters of concentrated sulfuric acid pipetted down the side of the tube so that the acid froze before coming into contact with the carbonate. While still immersed in liquid nitrogen the tube was connected to the vacuum line and thoroughly evacuated. The pump was then turned off and the tube allowed to warm to room temperature with consequent evolution of the CO_2. The gas was condensed into a 50-cc bulb to which two sample tubes with break-off seals had been attached. The sample was allowed to stand in the bulb to ensure equilibrium with any CO_2 adsorbed on the glass walls or in the grease on the stopcock. After an hour or two one of the sample tubes was sealed off and analyzed on the mass spectrometer. The dilution was then made and the second tube sealed off to bracket the composition of the gas actually used in the dilution. It was found that the two compositions agreed within the error of the mass spectrometric analysis.

TABLE II. The compositions and dilution factors for the radioactive carbon dioxide samples measured. Column 2 gives the isotopic composition of the master sample and column 3 the factor by which the master was diluted. Column 4 is the ratio of column 3 to column 2.

Designation	Isotopic composition of master (atomic percent C^{14})	Dilution factor	Ratio stable to C^{14} atoms
Ia	1.28±0.01	1592	124370
Ib	1.28±0.01	6410	500780
Ic	1.28±0.01	1670	130470
Aa	3.28±0.03	22750	693600
A	3.28±0.03	7410	225900
IIa	2.55±0.03	3722	145960
IIb	2.55±0.03	4410	172940
Fa	2.55±0.03	803.4	31505
$\alpha 1$	6.144±0.042	6215	10160
$\alpha 2$	6.131±0.038	7521	122670

The second method of evolving CO_2 from barium carbonate, suggested by Dr. W. W. Miller in a private conversation, was the metathesis of the carbonate with lead chloride. The Oak Ridge barium carbonate was mixed with dry lead chloride powder in a mortar and transferred to a tube on the vacuum line. The tube was evacuated and the pumping continued during the initial stages of heating in order to outgas the mixture. The pump was shut off when the temperature reached 100°C. CO_2 was evolved as the temperature was raised, the evolution appearing complete and quantitative at 350°C. In terms of the metathetic reaction

$$BaCO_3+PbCl_2=BaCl_2+PbO+CO_2, \quad (1)$$

a 100 percent excess of $PbCl_2$ was used. A typical result by this procedure was that a sample of barium carbonate labeled 5.9 percent C^{14} by the Oak Ridge Isotopes Division yielded CO_2 of 6.144 ±0.042 percent composition, indicating no appreciable dilution. By contrast, the acid evolution method gave samples varying from 1.24 to 3.28 percent from barium carbonate labeled 3 percent, the variation presumably being due to the introduction of inert CO_2, either in the acid or from the freezing of the acid in air.

The dilution was performed by allowing part of the master sample to expand from its storage bulb through the vacuum line into a McLeod gauge. A 0.3281-cc bulb had been sealed to the capillary of the McLeod, and the active CO_2 was forced into this bulb by raising the mercury. The vacuum line was thoroughly evacuated and the pressure of the CO_2 in the bulb measured. The vacuum line was flushed with inert CO_2, and a pressure of CO_2 allowed to build up, sufficient that, as the mercury was lowered, inert CO_2 was forced through the mercury into the McLeod to a pressure of 40 or 50 cm. The vacuum line was evacuated and the total pressure in the 503.1-cc McLeod volume measured. Care was exercised throughout the operation to keep the mercury level in the McLeod above the side arm to prevent the active sample from diffusing into the line. The gas was allowed to stand for a time to insure diffusive mixing although the bubbling attendant, upon the transfer of the inert CO_2 through the mercury, was sufficiently vigorous as to have assured fairly complete mixing. The sample was then transferred by condensing with liquid nitrogen into a trap sealed to a 500-cc storage bulb with a stopcock.

Each master sample was usually used to make more than one working sample. Table II summarizes the data on the masters and working samples.

The technique used for the isotopic analysis was identical with that described by Norris and Inghram.[5b] It is considered that the accuracy of the measurement is not less than 0.6 percent, the principal error being uncertainty about the relative ionization probabilities of $C^{12}O_2$ *versus* $C^{14}O_2$. The precaution of using molecular flow conditions between the sample tube and the ion chamber eliminates any correction for fractionation of the gas during flow into the mass spectrometer source. The instrument was that used by Norris and Inghram.

III. ABSOLUTE GAS COUNTING

A. Counter Construction and Operation

As mentioned in the Introduction, an argon-alcohol mixture was selected for the counter gas because of previous experience with the mixture and the extensive information as to its high efficiency.[8] The standard pressures employed were 1.2 cm alcohol and 7 cm argon. Two types of counters were used. The principal type, shown in Fig. 1, consisted of a brass cylinder with flat Lucite plugs waxed into the ends. Both De Khotinsky and Apiezon "W" waxes were used. The central electrode, 0.22-mm diameter, was iron wire carefully cleaned with fine emery before being put into the counter. The Lucite plugs were turned on a lathe and were $\frac{3}{4}''$ thick, extending $\frac{3}{8}''$ into the counter. For about half the data, a movable Lucite disk, slightly smaller in diameter than the counter and $\frac{3}{4}''$ thick with a $\frac{1}{8}''$ hole through the center, had been placed in the counter to facilitate mixing the gases. In later measurements this disk was omitted. The second type of counter, used for a few early measurements, was a brass tube with glass ends, as shown in Fig. 2. Its use was discontinued because the end correction was so much larger than that for counters with flat ends.

It was found that the presence of small partial pressures of inert CO_2 in the standard argon-alcohol filling had very little effect on counter operation. Figure 3 shows plateau curves for the addition of various amounts of CO_2. The evidence clearly indicates that pressures of CO_2 up to 7 mm are tolerable; and that the pressure range in this research, namely, 0.01 to 0.5 mm, was completely safe. The precaution was always taken to record the response to an external standard (a piece of an old copper cyclotron-target). It was observed that whereas CO_2 at these pressures had no effect on the observed rate, considerable care had to be taken to guard against certain slightly volatile materials which reduced counter efficiency. Apparently these materials arose in the wax or in the Lucite, and extensive pumping was required for their removal. In the two and a half years spent on this research, this happened only 5 or 6 times in several hundred counter fillings. With the exception of these few runs, the counters always responded properly to the standard regardless of whether or not there was an active filling in the counter.

Memory effects were quite serious during the first weeks, especially with newly built counters. The background after evacuation and refilling with argon-alcohol was several percent higher, following the counting of an active CO_2 sample, than before. The initial background rate was restored by acid washing the counter, but heating to temperatures of 50–60°C for 12 to 14 hours in the presence of inert CO_2 did little good. Treatment with water

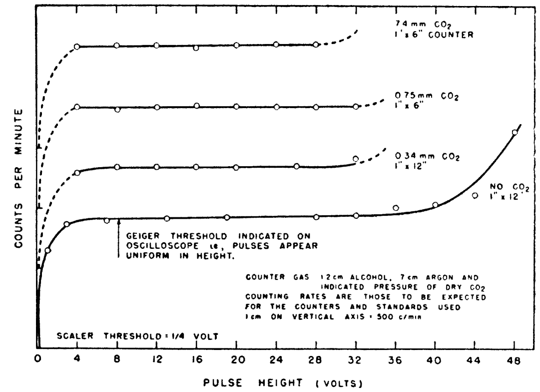

FIG. 3. Effect of CO_2 on counter plateaus. *Note:* Due to scale down, bottom right-hand line should read ". . . 1 cm on vertical axis = 1500 c/min.

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[8] K. Greisen and N. Nereson, Phys. Rev. 62, 316 (1942).

TABLE III. Physical characteristics of the counters.

	Diameter × length (inches)	Volume between ends (cc)	Dead volume (percent of total external to brass cylinder)
A.	1×6	48	34.3
	1×12	106	16.4
	2×12	484	24.5
	2×24	1065	13.1

	Diameter × length (inches)	Volume between ends (cc)	Total volume (cc)	Dead volume (percent)
B.	1×6	75.8	76.5	0.92
	1×12	153.8	155.1	0.79
	1.5×9	263.0	265.5	0.57
	1.5×18	521.0	522.6	0.31
	1.5×27	778.3	780.1	0.23
	2×12	614.0	615.7	0.18
	2×24	1232.0	1233.0	0.09

vapor and CO_2 was fairly effective in reducing the background even at room temperature. The explanation probably is the formation of a chemical compound on the surface of the brass. The preparation of the cylinder before the counter was assembled included washing in dilute nitric acid and rinsing in distilled water. Presumably this left a surface oxide film relatively free of carbonate. Subsequent exposure to CO_2 would encourage the formation of zinc and copper carbonates. These being relatively stable compounds, particularly the zinc carbonate, only very high temperatures, acid washing, or exchange with CO_2 could be expected to effect their removal. Since this notion explained the observations, it was concluded that an efficient pretreatment ought to be exposure of the newly constructed counter to inert CO_2 until the layers were saturated. CO_2 at 1 atmosphere was allowed to stand in the counter for twenty-four hours at room temperature. This pre-treatment proved to be completely effective provided no appreciable amounts of moisture were allowed to enter the counter, and for all practical purposes, the difficulty was solved. This procedure should serve for ion chambers and other instruments constructed of brass, and the notion applies more or less generally to other detection devices constructed of metal and operating with CO_2.

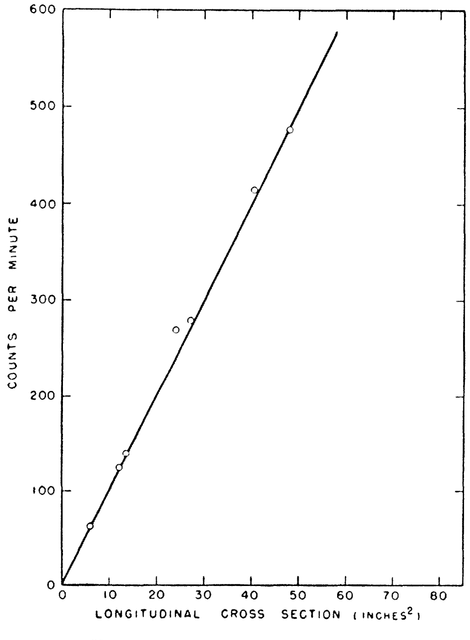

FIG. 4. Counter backgrounds.

The counter was operated on a scale of 64 with a commercial unit sold by Instrument Development Laboratories (Model 161). It was found necessary to modify this unit for the larger counters by the insertion of a 3-megohm variable resistor in the first multivibrator circuit of the set. Prior to this change, somewhat disconcerting behavior was observed with the larger counters, namely, that pulses were doubly recorded when the pulse height exceeded a certain value and that consequently the counting rate doubled about half-way along the plateau. Counters 2″ in diameter and larger invariably showed this doubling, whereas it was never observed with 1″ counters even though these were made as long as twelve inches. The small alteration mentioned, however, was the only one necessary to use the commercial unit with even the largest counters.

The central wire was connected directly to the input lead and the counter wall grounded through a standard cathode-ray oscilloscope. This enabled the operator to determine the true pulse shape and not its first derivative, and proved convenient in setting the operating voltage so that reproducible pulse heights were obtained. It was observed that the middle of the plateau usually corresponded to a given pulse height; this served as a check on the counter behavior. There was also a linear relationship between pulse height and voltage, a change of 2 volts in the pulse height corresponding to a ten-volt change in the operating voltage.

The counters were shielded with $1\frac{1}{2}''$ of lead at the sides, ends and bottom. The top was left unshielded for convenience. Table III gives the dimensions of the counters used, Part A, the "glass counters" and Part B, the "Lucite counters" used throughout most of the work.

It was observed, particularly with the Lucite counters, that the background rate was a linear function of the longitudinal cross-sectional area of the counters. Table IV gives the data for the Lucite counters when shielded as described and lying in a horizontal position.

These data are plotted in Fig. 4. It is clear that linearity is fairly well established and that the slope of the line is in agreement with numerous

TABLE IV. Background *vs.* area for lucite counters.

Counter dimensions (inches)	Background (c/min.)	Ratio of bkgd./area (c/min./in.^{2})
1×6	64	10.7
1×12	125	10.4
1.5×9	137	10.1
1.5×18	285	10.6
1.5×27	415	10.2
2×12	275	11.5
2×24	480	10.0

TABLE V. Resolving times for the lucite counters.

Counter dimensions (inches)	Resolving time τ (microminutes)
1×6	3.86±0.2
1×12	4.20±0.15
1.5×9	5.61±0.5
1.5×18	5.81±0.2
1.5×27	5.50±0.3
2×12	6.93±0.15
2×24	7.04±0.14

reports in the literature on backgrounds under similar conditions. It should be remarked that the addition of 1½″ of lead above the counter reduces the slope approximately 30 percent, so that a rough rule for the background at sea level of a counter shielded in all directions by 1½″ of lead is 7 c/min./in.^{2}

B. Coincidence Correction

The coincidence corrections were determined by the use of carefully paired Cs^{137} sources mounted on thin copper disks. The formulas used were

$$y_1 = x_1 \exp\tau y_1, \tag{2}$$

$$y_2 = x_2 \exp\tau y_2, \tag{3}$$

$$y_1 + y_2 = x_{1,2} \exp\tau(y_1 + y_2), \tag{4}$$

where the y's are the true rates, the x's the observed rates, and τ the resolving time of the counter. The exponential form of the coincidence correction law was used rather than the linear form, although calculations with the linear form showed little difference in the correction obtained. In the case that $x_1 = x_2 = x$, then y_1 will be equal to y_2, (y), and a general simplification results. In fact, since from (2), (3), and (4),

$$y_1 y_2/(y_1 + y_2) = x_1 x_2/x_{1,2},$$

substitution gives

$$y = 2x^2/x_{1,2} \tag{5}$$

which, when replaced in Eq. (2) or (3), gives a useful expression for τ.

$$\tau = (x_{1,2}/2x^2)\ln(2x/x_{1,2}). \tag{6}$$

In using this method it must be realized that the background is measured as a part of both x_1 and x_2, and that it enters only once in the measurement of $x_{1,2}$. This correction has been made throughout this work.

The values obtained for τ for the Lucite counters are listed in Table V.

The apparent conclusion to be drawn from these data is that the value of τ is a function of counter diameter alone. This appears reasonable on all bases previously considered.[9]

The values of τ tabulated above agree within experimental error with those obtained by means

TABLE VI. Half-life data for glass counters.

Sample	Vol. $C^{14}O_2$ used (μ cc S.T.P.)	Specific activity (c/min./μ cc)
A. 2″×24″ counter (net vol.=1065 cc between cap ends, total vol.=1225 cc)		
Ic	1.57	5150±600
Ia	0.508	5570±250
Ia	0.671	5620±250
Ia	1.163	4880±250
A	1.463	4540±400
B. 2″×12″ counter (net vol.=484 cc, total vol.=649 cc)		
Aa	0.870	4200±150
Ic	1.39	4680±300
Ic	0.189	5380±250
Ib	0.183	5560±250
Ia	0.446	5280±250
	Average	4960±150
C. 1″×12″ counter (net vol.=106 cc, total vol.=127 cc)		
Ic	1.28	4840±250
Ic	0.427	4520±250
Ib	0.180	4900±300
	Average	4710±200
D. 1″×6″ counter (net vol.=48 cc, total vol.=73 cc)		
Ib	0.165	3480±250
Ib	0.393	3970±250
Ic	0.278	3850±200
Ic	0.485	3740±200
Ib	0.138	4360±250
	Average	3860±150
E. Calculated half-life		
1. 2″ counters		
Geom. end correction:	24″ $\frac{1225}{1065}\times 5120$	$=5900\pm150$
	12″	$=6650\pm200$
Formula end correction:	$\frac{1225\cdot 5120-649\cdot 4960}{1225-649}$	$=5330\pm600$
Half-life		
Geom. end correction:	24″	6010 years±150
	12″	5320 years±200
	Formula	6600 years±600
Average (geom.)		5665±300
2. 1″ Counters		
Half-life		
Geom. end correction:	12″	6230±300
	6″	6030±300
Formula end correction:		6030±900
Average (geom.)		6130±300

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[9] S. A. Korff, *Electron and Nuclear Counters* (D. Van Nostrand Company, Inc., 1946), Chapter 4.

TABLE VII. Half-life data for Lucite counters, 1″×6″ counter.

Sample	Vol. of $C^{14}O_2$ (μ cc S.T.P.)	Specific activity (c/min./μ cc S.T.P.)	Date
IIb	0.4259	5631±50	12/12/46
IIa	0.5592	5575±50	12/13/46
IIb*†	0.4153	5730±285	2/19/48
IIa*†	0.5456	5768±60	3/19/48
IIb*†	0.3914	5778±100	6/30/48
IIb‡†	0.2567	5700±60	7/22/48
IIb‡†	0.1422	5905±90	7/27/48
(IIa‡†§	0.2540	5504±60	8/4/48)
(IIa‡†§	0.3162	5298±110	8/5/48)
(IIa‡†§	0.2989	5597±60	8/9/48)
(IIa‡†§	0.2049	5778±110	8/10/48)

Average (corrected for dead volume) 5780±36

1″×12″ counter

IIa	0.7952	5838±50	12/4/46
IIb	0.7320	5881±50	12/4/46
IIb	0.5156	5737±50	12/12/46
IIa	0.5935	5580±50	12/12/46
IIb*†	0.3741	5888±230	2/19/48
IIa*†	0.4934	6196±60	3/19/48
IIb*†	0.3561	6017±60	6/30/48
IIb‡†	0.3134	5910±60	7/21/48
IIb‡†	0.4035	5988±60	7/22/48
IIb‡†	0.2234	5918±110	7/28/48
(IIa‡†§	0.3981	5683±85	8/4/48)
(IIa‡†§	0.3376	5694±60	8/5/48)
(IIa‡†§	0.4730	5792±90	8/9/48)
(IIa‡†§	0.4158	5688±110	8/10/48)

Average (corrected for dead volume) 5941±50

1½″×9″ counter

IIb*	0.7126	6170±50	1/23/48
IIb*†	0.7228	5993±300	2/19/48
IIb*†	0.7730	5910±150	3/9/48
IIa*†	0.9109	6143±300	3/23/48
IIb*†	0.9131	5911±300	6/25/48
IIb*†	0.7221	5980±110	6/30/48
IIb‡†	0.4588	5832±60	7/22/48
IIb‡†	0.1684	5553±220	7/29/48
(IIa‡†§	0.7281	5732±120	8/5/48)
(IIa‡†§	0.6128	5551±190	8/10/48)
α1‡†	0.3163	6045±100	10/27/48
α1‡†	0.1912	6060±100	11/1/48
α2‡†	0.5673	5961±60	11/8/48
α2‡†	0.6250	5763±100	11/11/48
α2‡†	0.5247	5808±60	11/12/48

Average (corrected for dead volume) 5975±45

1½″×18″ counter

IIb*†	0.5150	6335±600	3/9/48
IIa*†	0.6069	6144±110	3/23/48
IIb*†	0.4391	6154±90	5/7/48
IIa*†	0.4820	6133±60	5/13/48
IIb*†	0.9829	5561±160	6/25/48
Fa*†	0.9362	5739±150	5/27/48
IIb‡†	0.8269	6133±90	7/21/48
IIb‡†	0.8699	6169±120	7/23/48
IIb‡†	0.2964	6366±190	7/29/48
(IIa‡†§	0.5825	5843±60	8/5/48)
(IIa‡†§	0.9132	5866±150	8/10/48)
α1‡†	0.6053	6073±60	10/21/48
α1‡†	0.5960	5904±110	10/22/48
α1‡†	0.6259	5992±110	10/27/48
α2‡†	0.7372	6118±120	11/24/48

Average (corrected for dead volume) 6082±59

1½″×27″ counter

IIb*†	0.4352	6326±120	3/9/48
IIa*†	0.5143	6125±60	3/23/48
IIb*†	0.3720	6071±120	5/7/48
IIa*†	0.4080	6289±250	5/13/48
Fa*†	0.5286	5802±60	5/27/48
IIb*†	0.5549	6103±120	6/25/48
IIb‡†	0.7608	6103±120	7/23/48
IIb‡†	0.4430	6022±120	7/29/48
α1‡†	0.5654	6078±120	11/1/48
α2‡†	1.121	6188±120	11/8/48
α2‡†	0.7540	6008±60	11/11/48
α2‡†	1.032	6165±120	11/24/48

Average (corrected for dead volume) 6121±38

2″×12″ counter

IIa	0.4500	5760±50	12/3/46
IIa	0.9500	5736±50	12/4/46
IIb	0.6740	5501±50	12/4/46
IIa	0.3600	5627±60	5/29/47
IIb	0.3440	6118±50	5/30/47
IIa	0.2575	5887±500	6/3/47
IIa	0.1930	5815±100	6/3/47
IIa*	0.3560	6130±45	6/5/47
IIa*	0.7190	5920±40	6/5/47
IIa*	0.4150	6185±50	6/11/47
IIa*	2.4800	6125±100	6/11/47
IIa*	0.6380	6175±100	6/11/47
IIb*†	0.7599	5921±150	2/26/48
IIa*†	0.7864	6231±180	3/2/48
Fa*†	4.2610	5894±500	3/1/48
IIa*†	1.0004	6275±400	3/19/48
IIb*†	1.2446	5888±120	5/3/48
IIa*†	0.7004	6180±300	6/11/48
Fa*†	1.0600	5770±150	6/26/48
IIb*†	1.2380	5977±250	6/24/48
IIb*†	1.1600	6015±180	6/25/48
IIb‡†	0.5191	6013±80	7/22/48
(IIa‡†§	0.7506	5922±120	8/4/48)
(IIa‡†§	0.8856	5865±200	8/9/48)
α1*†	1.2699	6008±240	10/7/48
α2*†	1.0526	6218±190	10/13/48
α1‡†	0.7096	6002±40	10/18/48
α1‡†	0.6276	5998±60	10/20/48

Average (corrected for dead volume) 5987±38

2″×24″ counter

IIa	0.8329	6244±50	12/6/46
IIb	0.7946	6245±80	12/6/46
IIb	0.8170	5944±60	12/6/46
IIb	0.2650	6166±120	12/6/46
IIb	0.1780	6121±120	By Pumping
IIb	0.1140	6772±500	12/11/46
IIa	0.3230	6143±90	5/22/47
IIb*	1.0070	6134±50	1/23/48
IIb*†	1.0770	6256±370	2/26/48
Fa*†	6.0390	6224±600	3/1/48
IIa*†	1.1146	6442±130	3/2/48
IIa*†	0.8295	6386±190	3/19/48
IIb*†	1.1087	6163±120	5/3/48
IIa*†	0.6274	6320±90	5/11/48
Fa*†	0.9499	5927±100	5/26/48
IIb*†	0.8396	5886±90	6/24/48
IIb*†	0.7864	6045±60	6/25/48
IIa*†	0.4223	5984±60	6/29/48
IIb‡†	0.5841	5986±60	7/21/48
IIb‡†	0.7220	5980±150	7/28/48
IIb‡†	1.0163	5938±120	7/26/48
α1*†	0.8698	6027±120	10/7/48
α2*†	0.7218	6191±120	10/13/48
α1‡†	0.6290	6090±60	10/18/48
α1‡†	0.7765	6060±60	11/19/48

Average (corrected for dead volume) 6126±30

* New std. vol. introduction technique.
† Pot. in IDL.
‡ Premixing technique.
§ Doubtful premix dilution value, omitted in calculating half-life.

of a single sweep cathode-ray oscilloscope whereon the resolving time was observed visually. They agree also with values obtained by following the decay of 54-minute In through several half-lives, as well as with values reported in the literature for counters with argon-alcohol fillings.

The register appeared to record faithfully for rates up to 40,000 c/min. It was assumed, therefore, that its characteristics did not affect the resolving time determinations in which the maximum rates did not exceed 30,000 c/min., nor did they affect the coincidence loss during the half-life measurements, since most of the measurements were made at rates well below 10,000 c/min. Furthermore, because the rates were less than 10,000 c/min., the coincidence corrections were never more than a few percent.

C. Sample Introduction

The problem of introducing the CO_2 samples into the counters was one of the most serious encountered. The method first used involved measuring the pressure of CO_2 in a 0.982-cc bulb and condensing this CO_2 with liquid nitrogen into a glass U-trap between the counter and the vacuum line. The stopcock between the trap and the line was closed and the CO_2 allowed to sublime into the counter. The residual active CO_2 was swept into the counter in the course of introducing the alcohol and argon. Since incomplete transfer was suspected, a second method was devised. A second stopcock was sealed to the counter filling tube (see Fig. 1), and the volume between stopcocks calibrated. With the stopcock towards the vacuum line open and the stopcock adjacent to the counter closed, active CO_2 was introduced into the vacuum line and the pressure and temperature measured. The calibrated volume was closed off from the line and the stopcock to the counter opened. The line was evacuated, and the alcohol and argon were let into the counter. This method seemed to be quite satisfactory except for the possibility that grease from the stopcock might change the volume by ½ to 1 percent. A third procedure consisted of introducing a known amount of CO_2 into a liter bulb, adding approximately a liter of argon and measuring the total pressure. Since the volume of the bulb was known, it was then possible to calculate the CO_2/argon ratio. After standing several hours to insure diffusive mixing, the gas was ready for filling counters. The counters, the volumes of which were known, were first filled with alcohol at its vapor pressure at 0°C; the CO_2-argon mixture was added and the pressure of the alcohol plus the mixture measured. When necessary, additional argon was used to bring the total pressure to 8.2 cm. The data from all three methods agreed well on the average, although the scatter seemed to be greatest in the first and least in the third.

TABLE VIII. End correction calculations.

Sample	Counter	Specific activity (corrected for dead volume)		Calc. end correction (percent)
		A. Premix data		
IIa	1″×6″	5599±72		6.2±3.2
	1″×12″	5772±52		3.0±1.6
	1½″×9″	5719±172		5.2±5.4
	1½″×18″	5867±117		2.5±2.6
IIb	1″×6″	5816±70		6.1±3.2
	1″×12″	5992±60		2.9±1.5
	1½″×9″	5862±115	(1½×18)	11.6±6.9
			(1½×27)	5.4±5.1
	1½″×18″	6196±99	(1½×9)	5.4±3.2
			(1½×27)	−5.9±7.6
	1½″×27″	6075±120	(1½×9)	1.8±1.7
			(1½×18)	−4.0±5.2
	2″×12″	5941±100		2.1±4.0
	2″×24″	6004±90		1.0±2.0
α1	1½″×9″	6108±92	(18)	−2.2±3.8
			(27)	−0.39±3.4
	1½″×18″	6042±66	(9)	−1.1±1.9
			(27)	2.5±7.0
	1½″×27″	6092±120	(9)	−0.13±1.1
			(18)	1.6±4.5
	2″×12″	6011±100		2.1±3.9
	2″×24″	6075±60		1.1±2.0
α2	1½″×9″	5914±53	(18)	7.5±4.4
			(27)	4.2±2.3
	1½″×18″	6137±122	(9)	3.6±2.1
			(27)	−2.8±7.0
	1½″×27″	6080±73	(9)	1.4±0.8
			(18)	−1.9±4.6

Counter	Average specific activity (corrected for dead volume)		Calc. end correction (percent)
	B. Standard volume data		
1″×6″	5749±36		8.5±2.7
1″×12″	5793±60		4.1±1.3
1½″×9″	6050±42	(18)	2.3±3.0
		(27)	3.7±1.2
1½″×18″	6120±88	(9)	1.1±1.5
		(27)	3.8±4.5
1½″×27″	6199±46	(9)	1.2±0.4
		(18)	2.5±3.0
2″×12″	6068±34		3.0±1.4
2″×24″	6158±30		1.5±0.7
	C. Total half-life data		
1″×6″	5780±35		6.9±2.1
1″×12″	5978±50		3.3±1.0
1½″×9″	5975±45	(18)	5.0±1.7
		(27)	4.3±1.3
1½″×18″	6124±47	(9)	2.4±0.8
		(27)	1.2±2.8
1½″×27″	6148±29	(9)	1.4±0.4
		(18)	0.8±1.8
2″×12″	6006±34		3.9±1.25
2″×24″	6126±30		1.9±0.6

D. End Correction

The problem of determining the inefficiency of the counters, due to inhomogeneous and weakening fields near the counter ends, was handled by comparing the observed rates per unit volume of radioactive CO_2 for counters of the same diameter but

TABLE IX. End corrections.

Counter diameter	$D/L=1/6$	$D/L=1/12$	$D/L=1/18$
A. Premix data			
1″	6.2±1.5	3.0±0.8	——
1½″	4.2±1.5	2.2±1.3	1.2±0.5
2″	2.1±2.0	1.1±1.0	——
B. Std. vol. data			
1″	8.5±2.7	4.1±1.3	——
1½″	3.2±1.5	2.0±2.0	1.4±0.5
2″	3.0±1.4	1.5±0.7	——
C. Total data			
1″	6.9±2.1	3.3±1.0	——
1½″	4.5±1.5	2.3±1.0	1.3±0.7
2″	3.9±1.3	1.9±0.6	
Average (independent of diameter)	4.7±0.6	2.3±0.5	1.3±0.4
K	0.269	0.270	0.231
Average (by diameter)			
1″	7.2±2.0	3.5±1.0	——
1½″	4.0±1.2	2.3±1.0	1.3±0.4
2″	3.0±1.2	1.5±0.6	——

of different lengths. If the observed rates are called R and r for counters of length L and l, respectively, and the count rate per unit volume for 100 percent efficiency is called ρ, then

$$\rho(L-l)=RL-rl, \tag{7}$$

or

$$(\rho-R)/R=(l/(L-l))(1-r/R) \tag{8}$$

for the end correction for the longer counter, and

$$(\rho-r)/r=L/(L-l)((R-r)/r) \tag{9}$$

for the end correction for the shorter counter. The accurate determination of the end corrections was attempted only for the Lucite counters. For the glass counters, the formulas were used, and the results were in agreement with the assumption that the active volume ended at the beginning of the glass inserts. The corrections, however, were so large that the use of these counters was discontinued.

The data for the Lucite counters seem to indicate that the end correction is primarily dependent upon the ratio of length to diameter rather than upon the actual size. There is, however, some evidence for a slight variation in end correction from diameter to diameter, and the corrections were applied in both ways. Since the difference in the final results was small, the two were averaged. It is believed that the error in the end correction was the largest single error in the determination of the half-life. Since the magnitude of the end correction is small, the maximum correction being 7 percent, this error is less than 1 percent in its effect on the final result.

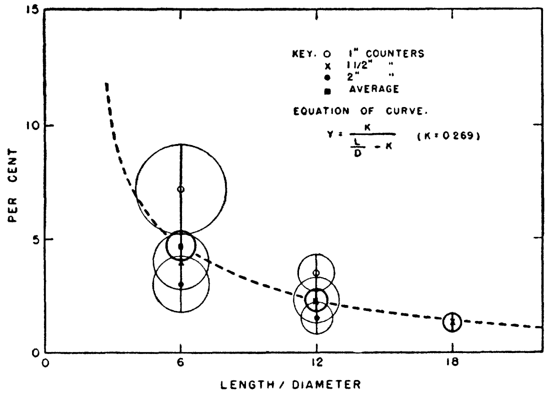

FIG. 5. End corrections.

It is quite conceivable that a short-range radiation such as tritium or certain K capture gases will show a different end correction than C^{14}. It is

TABLE X. Summary of half-life data by sample and counter; independence of master sample.

Counter	Sample	No. runs	Average specific activity (corrected for dead volume)	Corrected specific activity *a	*b
1″×6″	IIa	1	5821	6094	6240
	IIb	4	5831	6105	6250
1″×12″	IIa	3	5917	6053	6124
	IIb	7	5953	6090	6161
1½″×9″	IIa	1	6178	6468	6425
	IIb	7	5941	6220	6179
	α1	2	6108	6395	6352
	α2	3	5898	6175	6134
1½″×18″	IIa	2	6157	6299	6299
	IIb	6	6138	6279	6279
	Fa	1	5757	5889	5889
	α1	3	6009	6147	6147
	α2	1	6137	6278	6278
1½″×27″	IIa	2	6221	6314	6302
	IIb	5	6139	6231	6219
	Fa	1	5815	5902	5890
	α1	1	6092	6183	6171
	α2	3	6134	6226	6214
2″×12″	IIa	13	6015	6298	6195
	IIb	7	5930	6209	6108
	Fa	2	5842	6116	6017
	α1	3	6016	6299	6196
	α2	1	6229	6522	6416
2″×24″	IIa	6	6259	6403	6353
	IIb	12	6077	6217	6168
	Fa	2	6081	6221	6172
	α1	3	6064	6203	6155
	α2	1	6196	6338	6289
Average, all counters	IIa	28		6276±37	6278±24
	IIb	48		6193±17	6195±15
	Fa	6		6032±49	5992±42
	α1	12		6245±28	6204±24
	α2	9		6308±37	6266±30
Average				6213±30	6187±31
Half-life				5700±28	5724±29

*a Average end corrections, independent of diameter.
*b Average end corrections, by diameters.

Table XI. Summary of half-life data by counters (total averages, disregarding sample differences).

Diam./ length	Counter	Average specific activity	Corrected specific activity *a	Corrected specific activity *b
1/6	1″×6″	5780±25	6052	6196
	1½″×9″	5975±31	6256	6214
	2″×12″	5976±30	6257	6155
1/12	1″×12″	5941±35	6078	6149
	1½″×18″	6082±41	6222	6222
	2″×24″	6126±21	6267	6218
1/18	1½″×27″	6121±26	6213	6201
	Average (103 runs)		6192±22	6193±10
	Half-life		5719±20	5719±10

*a Average end corrections, independent of diameter.
*b Average end corrections, by diameters.

intended to measure this effect to determine any difference.

Note added in proof.—Measurements with A^{37} (34.1d; K capture) which give 2.8 kev pulses of Auger electrons have given no evidence of dependence of the correction on diameter and the corrections found agree within a small error with those reported here for C^{14} (dotted curve in Fig. 5). These results will be published later elsewhere.

IV. EXPERIMENTAL DATA AND RESULTS

Table VI presents the data obtained in the fall of 1946 with the glass counters described earlier. Part E of the table summarizes the half-life calculations from these data. A geometrical end correction was used rather than the formula end correction discussed in the previous section, because the errors in the individual rates were too large for the differences to be significant.

Table VII is a similar summary of the data from the Lucite counters. Specific activities are calculated from the observed rates after correcting for coincidence losses and background. The dead volume correction for each counter is made for the average specific activity. The data from sample IIa premixing are omitted in the average because of an error in the preparation of the CO_2-argon mixture.

Table VIII contains data used in calculating end corrections, Part A, data from the premix method of introducing CO_2; Part B, data from the calibrated volume method, and Part C, total averages. The premix data for sample IIa are included in the end correction calculations because an error in the dilution does not affect the end correction.

Table IX is a summary of the calculated end corrections.

The average end corrections are plotted as a function of length to diameter ratio in Fig. 5, and smooth curve drawn for the end correction independent of any diameter effect. The theoretical equation for the curve is given with the value for the constant K derived from the data. This equation is derived easily on the assumption that the correction depends only on the ratio of length to diameter.

Table XII. Summary of half-life values; precision and best value.

A. Individual values	
1. Glass counters	
1″	6130±300 years
2″	5665±300 years
Average	5898±200 years
2. Lucite counters	
a. Average by samples	5700±42 years
b. Average by samples (diameter dependent end correction)	5724±57 years
c. Total average (independent of sample)	5719±40 years
Average	5712±46 years
B. Best value (average by inverse square of errors)	5720±47 years (mean solar)

The average specific activity obtained for each working sample with each counter is listed in Table X. The corrected specific activity is calculated for each value with (1) end corrections as a function of length/diameter only and (2) end corrections varying by diameter.

Table XI is a summary of the average specific activities independent of sample. End corrections are made in two ways, as above.

Table XII summarizes all the results, together with the standard deviation of the mean. The final result, 5720±47 mean solar years, is obtained by weighting individual results according to the inverse square of their errors.

V. DISCUSSION

The published values for the half-life of radiocarbon (see Table I) exhibit a scatter somewhat beyond the errors estimated by the authors, and apparently random. The mean of the last 5 values in the table, when weighted according to the errors quoted by the authors in each case, is 5687±100 years. It is difficult to know what significance the standard deviation ±100 years has in this calculation. The agreement between the result of the present research, 5720±47, and the mean 5687 ±100, should not be overstressed since the validity of such an average is uncertain. It is interesting, however, that some accord does exist, and may add to confidence in a value close to this point.

The precision quoted for the present result (47 years) is the standard deviation of the mean; statistical deviations of the observed specific activities, the errors in the coincidence and end corrections, the error in the spectrometric analyses, and dilution errors were considered in computing this error. The "safe error" may be considered to be about twice this, or, in round numbers, ±100 years.