

LIQUID SCINTILLATION COUNTING  
RECENT APPLICATIONS AND DEVELOPMENT  
VOLUME II. SAMPLE PREPARATION AND APPLICATIONS

TEFLON VIALS FOR LOW-LEVEL C-14  
LIQUID SCINTILLATION COUNTING

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*This manuscript presents a partial re-examination of results from a paper published by Calf and Polach on the use of Teflon vials for liquid scintillation of carbon-14 samples. Five Teflon vials similar to "Design A" of Calf and Polach, except for an increased vial capacity (from 5 to 6 ml), were fabricated; their  $^{14}\text{C}$  counting performance was compared with low  $^{40}\text{K}$  glass vials. Use of the Teflon vials resulted in an increase in carbon-14 counting efficiency of from 5-to-10 percent using a Packard 3255<sup>1</sup> liquid-scintillation counter optimized to yield a maximum figure of merit  $E^2/B$ . However, no reduction in the background counting rate was observed for a 3-ml benzene sample added to 1-ml of scintillation solution, as opposed to the significant reduction observed by Calf and Polach. Tests on Teflon vials show that the loss of hydrocarbon-scintillation mixture through the vial is less than 0.5 percent by weight per week.*

*The background count rate at 5,000 feet above sea level measured in this laboratory is 22 percent higher than the rate at sea level, using a similar instrument and operational conditions. The increased background is caused by cosmic ray produced muons and nucleons. Shielding experiments and an analysis of the background components are presented.*

<sup>1</sup>*The use of brand name in this paper is for identification purposes only and does not imply endorsement by the U.S. Geological Survey.*

*The increase in counting efficiency, plus the higher detection sensitivity obtainable by increasing the volume ratio of sample benzene to toluene scintillation solution from 3:1 to 5:1, extends the carbon-14 age limit determinable by liquid scintillation counting from the present 37,000 years to 42,000 years.*

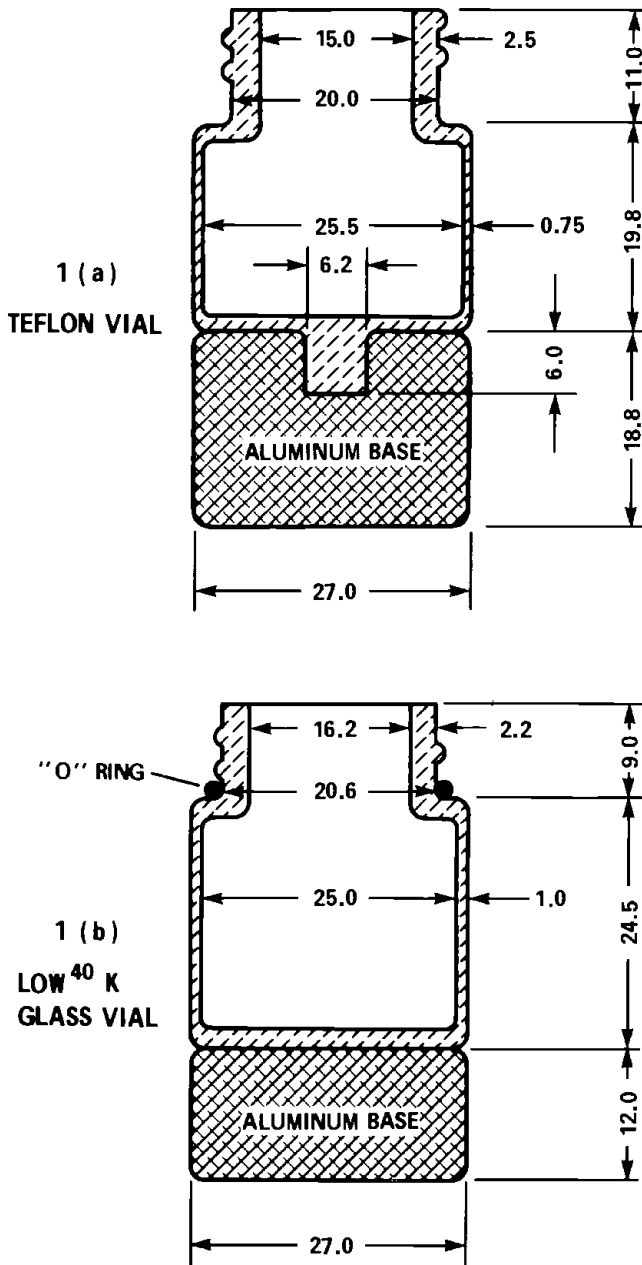
## I. INTRODUCTION

The liquid-scintillation counting method for carbon-14 age dating is used by many radiocarbon dating laboratories throughout the world. Maximum energy of beta particles of carbon-14 decay is only 154 KeV, so penetrating power of these particles is weak. Furthermore, carbon-14 activity measured in most samples is low in specific activity and a high-counting efficiency is required. Therefore, the sample containing carbon-14 should be converted to a compound compatible with liquid-scintillation counting.

Benzene serves well in this capacity as a scintillation solvent and is used by many dating laboratories. The chemistry of benzene synthesis involves quantitative conversion of the sample specimens to carbon dioxide (De Vries, 1955; Rafter, 1954); followed by conversion to acetylene (Barker, 1953; Noakes and others, 1965); and finally, catalytic trimerization to benzene (Noakes and others, 1963; Polach and Stipp, 1967). Impurities in the product benzene, such as acetone, will act as a quenching agent in reducing counting efficiency (Noakes and others, 1965). Fraser and others (1974) concluded that impurities are present in the acetylene generated from the lithium carbide hydrolysis step and should be removed.

In addition to the high scintillation yield required from the scintillation liquid, counting efficiency variations will result from the use of different types of counting vials. Calf and Polach (1974) introduced the use of Teflon vials; they improved slightly on carbon-14 detection efficiency compared with low  $^{40}\text{K}$  glass vials, and also decreased the background counting rate.

Aside from the requirement for higher-counting efficiency, background count rate of the instrument should also be kept as low as possible by: (1) use of construction materials free of radioactive contamination; (2) shielding the detector with a sufficiently thick absorber; and (3) use of an anticoincidence



SCALE: 1.5 MILLIMETERS = 1 MILLIMETER

FIGURE 1. Counting vials.

guard-ring detector. Noakes and others (1973) described a new kind of low-level liquid scintillation counter using an annular NaI(Tl) crystal scintillator anticoincidence shield, which reduced the counter background by 82 percent. The high cost of annular crystal guards restricts their use to applications where the advantages justify the expense. Hartley and Church (1974) also described a low-level liquid scintillation counter constructed without annular anticoincidence guards which reduced background crosstalk between the two photomultipliers by optical coupling and electronic means.

The U.S. Geological Survey's Denver Radiocarbon Laboratory has been routinely dating ground-water samples using a liquid-scintillation counting method. This paper discusses increasing the efficiency and lowering the backgrounds of counting systems with long-term stability. The first section will re-examine and compare the performance of low  $^{40}\text{K}$  glass vials currently in use in the Denver Laboratory with that of the Teflon vial used by Galf and Polach. Sources of the background and the use of extensive lead shielding to identify the contribution of cosmic-ray components are reported.

## II. EXPERIMENTAL METHODS

### *A. Sample Vials*

Various containers for liquid-scintillation solutions have been described in detail by Painter (1974). Polyethylene vials, though lower in background count-rate, swell appreciably, as toluene, xylene, and dioxane (scintillation cocktail added to benzene) permeate the vial wall. Nylon vials also have low backgrounds and high-counting efficiency, but are very susceptible to photoluminescence, and the decay time is considerably longer than polyethylene or glass vials. Their interaction with hydroxylic solvents is another disadvantage. Therefore, only low  $^{40}\text{K}$  glass or Teflon vials are used in benzene liquid-scintillation counting. Five Teflon vials were manufactured from DuPont Polytetrafluoro Ethylene (PTFE)<sup>1</sup> rod (Fig. 1a) similar to design "A" of Galf and Polach (1974), with an increased volume capacity of 6 ml. This design is also similar to our existing low  $^{40}\text{K}$  glass vials (Fig. 1b). The width of both vials (Teflon and glass) are the same as a standard 20-ml glass vial. The height of the aluminum base is chosen so the center of the Teflon and glass containers will be placed at the center of the photomultiplier

tube axis. The glass vial is masked with black electrical tape above the level reached by the top of the scintillation liquid, to minimize the background of low  $^{40}\text{K}$  glass. Vial caps are molded from urea-formaldehyde with polyethylene disk liners.

### *B. Sample Preparation*

A counting solution is prepared by mixing sample-benzene with 1 ml of Amersham's Phase Combining Solvent (PCS), a proprietary liquid scintillation mixture containing scintillants (2, 5-Diphenyloxazole). For background counting, reagent benzene (dead in terms of  $^{14}\text{C}$ ) is used. The modern reference standard used is NBS oxalic acid converted to benzene.

In the past, a modern standard was freshly prepared each time from oxalic acid and corrected for isotopic fractionation by determining the  $^{13}\text{C}/^{12}\text{C}$  ratio of benzene. This correction is necessary, because it was reported by Polach (1972), that benzene synthesized from oxalic acid was not satisfactorily reproducible for reasons of isotopic fractionation. Furthermore, the modern standard, after mixing with the scintillator solution and transferring to the vial cannot be reused, due to long-term evaporation through the vial cap resulting in "apparent" loss of sample solution and activity. Because of frequent use and tedious benzene synthesis, a secondary modern standard is currently in use. A standard stock solution of 250 ml is prepared by adding 1 ml of  $^{14}\text{C}$  labeled toluene (13,900 disintegration per minute, dpm) to a 249 ml of PCS to give an activity of 55.6 dpm per milliliter. A secondary modern standard is then made up each time by mixing 1 ml of this stock solution with the required amount of reagent benzene. This secondary standard is calibrated against the oxalic-acid standard (corrected for isotopic fractionation) twice each year, and whenever a new batch of stock solution is made. The use of a secondary standard not only has an advantage of reproducible results, but also avoids the frequent time-consuming and costly synthesis of benzene from oxalic acid, when samples are to be counted.

The teflon-vial memory effect is studied by counting the background count rate both immediately before and after secondary standard counting. Vials are rinsed twice with reagent benzene and vacuum heated to  $60^{\circ}\text{C}$  for four hours, after counting the standard reference. The leakage rate of the

counting solution through a Teflon vial is determined by placing the weighed vial containing the counting solution in a liquid-scintillation counter at 8°C temperature, storing for one week, and reweighing to the nearest milligram.

### *C. Liquid Scintillation Spectrometer*

Packard model 3255 Tricarb and Nuclear Chicago Mark I liquid scintillation spectrometers (LSS)<sup>1</sup> were used for the experiments.

1. Packard 3255 liquid scintillation spectrometer was bought in late 1976. The detector system consists of two bialkali photomultiplier tubes (PMT) operated in summed-coincidence principle. A lead shield (2" thick X 8" X 8") directly above the counting vial located in the sample changer compartment is provided, in addition to a regular 2" lead shield around the counting chamber. An optional adjustable discriminator module (3051) was used to set the operating conditions. The setting of the lower discriminator was 80; the setting of the upper discriminator was 900. High voltage to the photomultiplier tubes is decreased while increasing the amplifier gain, to maintain counting efficiency of 70 percent.

2. Nuclear Chicago Mark I liquid scintillation spectrometer. The spectrometer, bought in 1966, has a higher background than the Packard instrument. The detector system is also surrounded by a 2" lead shield around the counting chamber; however, no extra 2" lead shield above the counting vials is provided. Operating principles and conditions are essentially the same as in the Packard instrument. Since the Packard instrument has a lower background, it is used routinely for tritium and carbon-14 sample counting. The Nuclear Chicago Mark I is utilized for background-shielding experiments.

### *D. Background Shielding*

Lead bricks used for shielding experiments were fabricated from lead wool used by the U.S. Navy during World War II. The radioactive impurities of lead wool were prechecked before fabrication with Ge(Li) gamma detector to assure their low level. The laboratory is housed in a single story, concrete-block building in Arvada, near Denver, at an elevation of approximately 5,000 feet above sea level.

The influences of extra lead shielding around a Nuclear Chicago Mark I Spectrometer (NCMS) in various configurations were investigated:

1. Four-inch shielding along one side (right) of the spectrometer close to the detector system;
2. Item 1 plus 6 lead bricks arranged in 4" thickness covering a space 12 X 8 inches square, directly above the vial;
3. Item 1 plus 8 lead bricks arranged to provide a 4" thickness, covering a space 16 X 8 inches square directly above the vial;
4. Item 1 plus a layer of 2" lead covering the entire top of NCMS;
5. Item 1 plus two layers of 2" lead covering the entire top of the NCMS.

These experiments were made to determine the contribution of cosmic-ray components to the background. Because of the large size of the NCMS and the limited number of lead bricks (approximately 200 pieces), no further shielding experiments were conducted.

### III. RESULTS AND DISCUSSION

#### *A. Carbon-14 Counting Efficiency, Figure of Merit and Factor F*

Comparisons of  $^{14}\text{C}$  counting efficiency for low  $^{40}\text{K}$  glass and Teflon vials are given in Table 1. One point concerning factor F in the table must be made clear. The figure of merit,  $E^2/B$ , as commonly used by manufacturers of spectrometers is inappropriate as a merit expression for low-count rates, because background is volume-dependent, and the counting efficiency factor, E, in  $(E^2/B)$  is not related to the sample volume used. Felber (1962) introduced a factor,  $\text{No}/B^2$ , where No = Modern reference standard; 0.95 oxalic acid in cpm; and B = background, cpm. This is particularly suitable for the evaluation of carbon-14 age-related calculations and is used by many dating laboratories. Calf and Polach (1974) called this factor, F, ( $F = \text{No}/B^2$ ), and gave it the name of "Radio-carbon Dating System Figure of Merit." In this factor, background B and No count rates are both volume-dependent. Calf and Polach (1974) also derived the equation for maximum-measurable carbon-14 age corresponding to the lowest detectable sample count rate based on three sigma criteria as follows:

$$\text{Max. Age} = 8033 \ln \left( \frac{t}{18} \right)^{\frac{1}{2}} + 8033 \ln \left[ \text{No}/B^{\frac{1}{2}} \right],$$

where the first term is time-dependent, and the second term is F-dependent. By increasing the counting time and the factor F, the obtainable age is increased.

The instrument used for routine  $^{14}\text{C}$  counting is a Packard Model 3255. It was set up in mid-1977, and operated with  $^{14}\text{C}/^{14}\text{CQ}$  preset counting conditions for all samples (including biological samples). In June 1978, we chose to optimize counting conditions using the adjustable discriminator module set up for maximum  $E^2/B$ , specifically for carbon-14 samples, with a sample size of 3 ml benzene plus 1 ml of the phased combining solvent (PCS) scintillator solution. The background count rate using low  $^{40}\text{K}$  glass vials was reduced significantly from 13.05 and 12.77 to 8.84 count per minute (cpm) while counting efficiency decreased only slightly. In February 1979, Teflon vials were substituted for low  $^{40}\text{K}$  glass vials. Background count rate did not show any significant reduction, while counting efficiency increased by 5 percent. Further experiments with increased volume of benzene (4 ml and 5 ml) in both types of vials indicate nearly 10 percent increase in counting efficiency for Teflon vials, and comparable counting efficiency for glass. For background

Table I. Comparison of  $^{14}\text{C}$  Counting Efficiency  $E^2/B$ , F of 6 ml Low  $^{40}\text{K}$  glass and Teflon Vials

Period	Low $^{40}\text{K}$ glass					Teflon vial					Remarks	
	Benzene (ml)/PCS (ml)	Bkgd CPM	0.95 ox <sup>a</sup> CPM	Percent E <sup>b</sup>	F <sup>c</sup>	Bkgd <sup>d</sup> CPM	0.95 ox <sup>d</sup> CPM	Percent E	F	$E^2/B$		
Aug. '77- Dec. '77	3/1	13.32	21.30	59.70	6.84	287	No Teflon vial was used during these periods				Counter using $^{14}\text{C}/^{14}\text{CQ}$ preset counting conditions	
Jan. '78- May '78	3/1	12.77	22.09	61.91	6.18	300	No Teflon vial was used during these periods					
June '78- June '79	3/1	8.84	20.81	59.33	7.00	385	8.66	22.49	63.03	7.69	464	Counter set up for max. $E^2/B$
June '78- June '79	4/1	10.70	26.73	59.61	8.32	346	8.76	31.11	70.80	10.62	673	Counter set up for max. $E^2/B$
June '78- June '79	5/1	11.30	33.64	61.56	8.88	336	8.80	38.48	70.63	12.87	587	Counter set up for max. $E^2/B$

<sup>a</sup> 0.95 ox = 95 percent of NBS oxalic acid standard count rate corrected to the year 1950 and  $\delta^{13}\text{C} = 18$  o/oo PDB.

<sup>b</sup> Percent E = Using the data that 95 percent of NBS oxalic acid is equivalent to  $13.53 \pm 0.07$  dpm per gram C.

<sup>c</sup> F = Radiocarbon system figure of merit  $F = \frac{0.95 \text{ ox}}{(\text{Bkgd})^{1/2}}$

<sup>d</sup> Values quoted for Bkgd and ox standards are average of five Teflon vials.

count rate, Teflon vials do not show any obvious increases, yet low  $^{40}\text{K}$  glass shows an increase of 1.2 cpm for each additional milliliter of benzene added. The reasons for this observation will be given later; note here that sample size is a significant factor in determining background count rate between the low  $^{40}\text{K}$  glass and Teflon vials. Low  $^{40}\text{K}$  glass vials have lower backgrounds when less than 3 ml of benzene are used; Teflon vials are better for volumes of 3 ml or larger.

Comparison of background count rate in the Denver laboratory with the other USGS laboratory in Reston, Virginia, using the same background sample and instruments operated under the same conditions, reveals that extra background caused by the increased high-altitude cosmic radiations (5,000 ft above sea level) is approximately 2 cpm (for 3-ml benzene and 1-ml PCS in Teflon vial), which is about 22 percent based on our background count rate of 8.56 cpm.

Both types of vials show approximately the same figure of merit,  $E^2/B$ , for a sample volume of 4-ml to 5-ml benzene. However, they reflect a significant increase in the radio-carbon system figure of merit,  $F$ . This is because the Teflon vial does not increase in background with sample volume increase. The advantage in using the Teflon vial with a large sample size is very promising. If 4,000 minutes (min) is a reasonable counting time, substitution of  $t$  and  $F$  values into the maximum age equation yields 37,000 years for 3-ml-to-1-ml of benzene-PCS solution in the low  $^{40}\text{K}$  glass, and 42,000 years for 5-ml-to-1-ml of benzene-PCS solution in the Teflon vial; this results in an extension of 5,000 years in maximum-age limit. However, larger benzene volumes require additional tens of gallons of ground-water samples for processing, which will increase the manpower and work-time for old age samples. Therefore, 5 ml of benzene was chosen as a maximum sample size for ground water carbon-14 dating at present. For other geological samples, where larger samples can be obtained and processed without difficulties, significant increase in age limit is anticipated by using even larger benzene sample sizes and Teflon vials. Addition of dry scintillant directly into the sample benzene may be preferred to keep the total volume small. Also, increasing the concentration of dry scintillant in the PCS may be needed to attain the same counting efficiency.

*B. Permeability and Memory Effect of Teflon Vial*

The loss of sample through polyethylene vial is well-known. Rapkin and Gibbs (1963) reported that toluene leakage rate through polyethylene wall was approximately .7 percent per day. The loss in mass not only decreases the sample count rate, but also may diffuse into other counting vials in the sample-changer compartment, resulting in contaminations.

Five Teflon vials were tested for their resistance to leakage. Table II shows the loss of sample, stored in the refrigerated LSS, at a temperature of 8°C for a period of one week. The loss rate is 0.2 percent per week. Since no series of samples, standards, and backgrounds will be counted for more than one week, no further test was performed for a longer period.

The glass vial also shows a comparable sample loss rate to the Teflon vial with the loss probably occurring along threads or through the urea-formaldehyde screw-cap lined with a polyethylene disk. Caps with cork and aluminum foil liners may provide better seals. However, metal foil liners are more likely to contain radioactive impurities, and were not used. Memory effects are insignificant for all Teflon vials. The magnitude of this effect was measured by determining the background count rate before and after counting of

Table II. Loss of Counting Solution from Teflon Vials and Low <sup>40</sup>K Glass Stored at 8°C.

Vial no.	Vial type	Counting Solution <sup>a</sup>			
		Initial wt. (g)	Wt. after one week (g)	Wt. Loss (g)	Percent loss
1	Teflon	3.5656	3.5588	0.0068	0.19
2	Teflon	3.5621	3.5595	.0026	.07
3	Teflon	3.5612	3.5593	.0019	.05
4	Teflon	3.5623	3.5551	.0072	.20
5	Teflon	3.5682	3.5656	.0026	.07
6	Glass	3.5634	3.5541	.0093	.26

<sup>a</sup> 3 ml benzene added to 1 ml of phase combining solvent (PCS).

Table III. Memory Effect of Teflon Vials

Vial no.	Background (cpm) on Feb. 17, 1979	Secondary std. (cpm) on Feb. 23, 1979	Background after std. <sup>a</sup> (cpm) on Feb. 27, 1979
1	8.44 ± 0.10	47.60 ± 0.25	8.51 ± 0.10
2	8.20 ± 0.10	47.64 ± 0.25	8.07 ± 0.10
3	8.55 ± 0.10	47.44 ± 0.25	8.47 ± 0.10
4	8.65 ± 0.10	47.56 ± 0.25	8.42 ± 0.10
5	8.74 ± 0.10	48.26 ± 0.10	8.64 ± 0.10

<sup>a</sup> Teflon vials are washed twice with reagent benzene followed by vacuum heated to 60°C for 4 hours.

secondary modern standards. The data are given in Table III and are self-evident. It is worth noting that the secondary, modern standard made from hot spikes gives a very good reproducible count rate, while the background count rate changes slightly from vial to vial. This is because it is very difficult to make a thin, uniform side wall for all Teflon vials.

### C. Sources of Background

The major components of background in a LSS are as follows:

1. Electronic noise from photomultiplier tubes;
2. Light pulses associated with the operation of photomultiplier tubes;
3. Radioactive contamination in the materials, lead shielding, and surroundings;
4. Cosmic-ray induced radiations.

Component 1 can be minimized to below detection by: a short resolving time, a discriminator, and operating in a coincidence mode. Component 2 can be reduced by operating the photomultiplier tubes at a lower voltage. Glass has a tendency to electroluminescence and arcing on the envelope of the photomultiplier tubes, when high voltage is applied to the pins of the tube, especially when humidity is high;

therefore, instruments should be operated in a low-humidity environment. Component 3 can be kept to a minimum by selecting materials free of radioactive impurities. Component 4 is usually the major problem. Though it can be reduced to some extent by lead shielding, the most effective approach requires a deep underground laboratory with massive lead shielding around the LSS.

To find out how the background can be reduced, an analysis has been made of the contributions from different sources of radiations to background. The counting rates were determined twice: first with the empty chamber and second, with a blackened vial inserted into the chamber. By doing this, the photomultipliers cannot detect photons from a vial. The  $^{14}\text{C}$  channel counting rates are shown in Table IV. In addition, counting rates of empty sample vials and sample scintillation solutions in varying amounts are also shown for both low  $^{40}\text{K}$  glass and Teflon vials. The following information can be extracted from the table:

1. The empty-chamber counting rate of 12.00 cpm indicates an optical crosstalk between the two photomultipliers, which apparently constitutes the major part of the  $^{14}\text{C}$  channel background.

Table IV. Sources of Background Counts in  
Packard Model 3255 LSS

Counting conditions	Low $^{40}\text{K}$ glass $^{14}\text{C}$ channel (CPM)	Teflon vial $^{14}\text{C}$ channel (CPM)
5 ml background benzene + 1 ml PSC counting solution	11.30 $\pm$ 0.20	8.80 $\pm$ 0.20
4 ml background benzene + 1 ml PCS counting solution	10.10 $\pm$ 0.20	8.75 $\pm$ 0.20
3 ml background benzene + 1 ml PCS counting solution	8.84 $\pm$ 0.19	8.56 $\pm$ 0.17
Empty sample vial	6.78 $\pm$ 0.15	8.38 $\pm$ 0.18
Black vial	2.20 $\pm$ 0.10	2.20 $\pm$ 0.10
Empty chamber	12.00 $\pm$ 0.20	12.00 $\pm$ 0.20

2. The background count rate decreasing from empty chamber to empty vial means that the vials are acting as an obstacle between two phototubes, either blocking the light transmission or reflecting away the light originating from one phototube to another.

3. If the background contribution is from cosmic events interacting with the liquid counting solution, the background count-rate should decrease with decreased sample volume. The fact that the observed count rates of Teflon vials do not depend on, or slightly depend on, background-sample volume, as seen from Table 1, indicates the background caused by cosmic interactions with sample liquid is insignificant. In the case of low  $^{40}\text{K}$  glass, the observed dependence is attributed to reduced optical coupling. When sample volume in the glass vial is reduced, the ability of light to be transmitted through the vial is reduced, because a second and third glass-air interface is created which will scatter the light away, resulting in a decreased coincidence count. Contribution from cosmic events interacting with the glass wall of the vial are also possible. However, this source is likely small, in view of the fact that the counting rate of an empty low  $^{40}\text{K}$  glass vial is significantly lower than an empty Teflon vial (This is only a conjecture).

4. Also, the significantly lower count rate of an empty low  $^{40}\text{K}$  glass vial compared with an empty Teflon vial suggests that a Teflon vial has a very low light absorption, and, when filled with counting solution, a Teflon vial has higher light-scattering properties than a glass vial.

5. As discussed in component 1, the dominant background in the LSS is caused by optical crosstalk. It was established that the effect of cosmic events interacting with the liquid scintillating solution is negligible. Noise pulses from photomultiplier tubes are usually cut down to less than 1 cpm by electronic means (Packard Instrument's manual, 1977). Radioactive impurities in carefully-selected materials and surroundings should not contribute more than 1 or 2 cpm. Therefore, the major contribution to optical crosstalk is caused by cosmic-ray interaction with the two photomultiplier tubes; further support to this argument will be given below in background shielding.

#### *D. Background Shielding*

Primary cosmic rays interact with the upper atmosphere to produce mesons (mostly  $\pi$  mesons) and nucleons. Many  $\mu$  mesons (or muons), produced as decay products of  $\pi$ -mesons during their passage through the air, constitute most of the cosmic radiation found in the lower atmosphere (Friedlander and others, 1964).

The nucleons consist mainly of photons, electrons, and positrons, and compose about 10 percent of the cosmic radiation at sea level. A fraction of muons and nucleons are absorbed while traversing the atmosphere. Variations in absorber thickness (air mass) are proportional to variations in barometric pressure. The absorption length for cosmic-ray muons at sea level in the atmosphere is  $4,000 \text{ g/cm}^2$  (Cocconi, 1951), and for cosmic nucleons,  $120 \text{ g/cm}^2$  (Hayakawa, 1969). Thus, it is clear that muons have significantly higher penetrating power than nucleons, about 33 times in the air. The probability of absorption rapidly increases with the atomic number,  $Z$ . In terms of lead shielding, these muons can penetrate a meter or more, while nucleons can be stopped in several inches of lead.

As the intensity of cosmic radiation increases with increasing altitude, we expect cosmic-ray nucleon components to increase more rapidly than muons. The Denver-area laboratory is situated at an altitude about 5,000 feet above sea level. Therefore, extra lead shielding around the detector system of LSS should effectively cut down the fast-rising component of cosmic-ray nucleons; this is true, as seen in Table V. A layer of 2-inch lead covering the whole top and 4 inches along one side (near the detector system) reduces the  $^{14}\text{C}$  channel background of Nuclear Chicago Mark I from 10.10 cpm to 7.99 cpm. An additional 2 inches of lead on the top gives a small further reduction to 7.24 cpm. This fact, with others mentioned in section C, support the argument that the major background is caused by the interaction of cosmic rays with the photomultiplier tubes, resulting in crosstalk between the two tubes. The small background reduction observed with an additional layer of shielding is due to high penetration of the cosmic-ray muon component, which requires massive lead shielding to remove.

TABLE V. Influences of Lead Shielding on Counter Background using Nuclear Chicago Mark I LSS

<i>Shielding conditions</i>	<i>Background of 3 ml reagent benzene + 1 ml PCS in Teflon vial in CPM</i>
(1) Without extra shielding	$10.10 \pm 0.10$
(2) 4" lead shielding along one side of LSS near detector system	$8.58 \pm 0.09$
(3) 4" lead shielding along one side of LSS near detector system + 4" lead brick 12" X 8" sq. in. centered over the vial.	$7.93 \pm 0.08$
(4) 4" lead shielding along one side of LSS near detector system + 4" lead brick 16" X 8" sq. in. centered over the vial.	$7.83 \pm 0.08$
(5) 4" lead shielding along one side of LSS near detector system + a layer of 2" lead covering whole top of LSS	$7.99 \pm 0.08$
(6) 4" lead shielding along one side of LSS near detector system + 2 layers of 2" lead covering whole top of LSS	$7.24 \pm 0.04$

Radiocarbon dating laboratories at the University of Washington, Seattle (Stuiver and others, 1976) and the Physics Institute at the University of Berne, Switzerland (Oeschger and others, 1976), have constructed underground laboratories more than 30 feet below the earth's surface. These laboratories were able to reduce the muon flux by a factor of 10 or more, and also the secondary neutron and gamma radiations induced by muons. As a consequence, a significant background reduction can be achieved by locating the LSS in an underground laboratory.

#### IV. CONCLUSION

A significant increase in F can be obtained by using a Teflon vial with a benzene sample size larger than 3 ml. For less than a 3-ml sample, both glass and Teflon vials are comparable.

Background observed in an LSS results mainly from optical crosstalk caused by interactions of cosmic events with photomultiplier tubes. The nucleon component of cosmic radiation is significantly higher at high altitude than at sea level; it can be shielded by an extra few inches of lead. However, the meson component cannot be reduced to any significant amount unless the LSS is operated with extensive lead shielding in the underground laboratory. Use of annular anticoincidence guard counters with an LSS operated in the underground laboratory possibly would be the best solution for obtaining the lowest background for radiocarbon dating purposes.

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