A NEW LIQUID SCINTILLATION COUNTER FOR MEASUREMENT OF TRACE AMOUNTS OF ^{3}H and ^{14}C

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Abstract

A newly designed liquid scintillation counter has been built specifically for the measurement of low specific activity samples of ^{3}H and ^{14}C. The instrument's electronics measure fast-pulse time intervals rather than performing pulse height analysis. Massive graded shielding and a background guard shield are utilized to minimize external radiation for low background counting. Figure of merit (E^2/B) of the counter is an order of magnitude superior to present commercial liquid counters. The external background guard shield was evaluated in the absence of a massive graded lead shield. The use of such a background guard shield in commercial liquid scintillation systems is considered.

Introduction

In an earlier paper (1) we described a new kind of low-level liquid scintillation counter that was designed, built, and tested in our laboratory. The importance of tritium measurements were stressed and performance data for the counter were given there in terms of ^{3}H.

It is certainly true that the quantative detection of other β emitting nuclides in low specific activity is of interest. The counter previously mentioned is not limited to the analysis of ^{3}H, but rather can be used to detect and quantify other β emitters.

^{14}C is a nuclide which is widly used and counted by both the physical (2,3,4) and life scientist. More specifically, carbon dating has become a widely used tool of the geologist, archaeologist, etc. By converting the carbonaceous sample to benzene (5) and counting in a liquid scintillation counter of the conventional type, age measurements can be estimated from the counts collected during the counting interval, t, with some precision . However, with the instrument described here and the above sample, either its' age could be estimated on the basis of a shorter counting interval, t' and the same precision or its' age determined with greater precision where the count is taken during the same interval t; and, in principle, older samples can be analyzed with the new system. This advantage is based on the larger sized sample that can be counted with a lower background. Moreover, such an advantage can be applied to other experimental systems.

Modern, commercially available, liquid scintillation counters employ pulse height discrimination (where pulse height is a much convolved energy function) as a means of utilizing the coincidence counting technique. In the low-level liquid scintillation counter described here, we employ time discrimination to implement the coincidence counting technique. The use of an annular NaI(Tl) crystal scintillator shield which served to reduce the counter's background by 82% was another difference between the low-level liquid scintillation counter and a commercial liquid scintillation counter. The high cost of the crystal annulus precludes its' general use. Therefore, a lower cost scintillating plastic annulus was tested with the low-level liquid scintillation counter. The results were compared with those obtained under the same circumstances, but with the NaI(Tl) crystal in place in an effort to determine the efficacy of such an approach to low level counting.

Experimental Procedure

A) COUNTER: Conventional liquid scintillation counters use the coincidence counting technique which places the sample between two photomultiplier tubes which share a common axis. For a data pulse to be generated at the counter's scaler, two pulses per beta event must arrive at the coincidence gate within a specified time interval, usually 20 nsec. Differential counting is accomplished by the use of pulse height discriminators which select or reject data pulses on the basis of their beta energy analog, pulse height. Background radiation is excluded through the use of lead shielding. No method is employed to discriminate between external and internal background noise.

The counter reported in this paper differs considerably from commercially available liquid scintillation counters. Particularly in the way in which raw data pulses are tested and selected and/or rejected; as well as, the manner in which shielding is employed. Fig. 1 shows the instruments' control panel and shielded counting chamber. A detailed discussion of these features follows.

B) ELECTRONICS: Fig. 2 shows a block diagram of the counter. With the exception of the photomultiplier tubes, all of the electronics were made up from state of the art ORTEC NIM bin components. A pair of RCA 4501/V4, 2" diameter, head-on "Quantacon" photomultiplier tubes (PMT) were obtained from the Industrial Tube Division of RCA Inc. as well as, the four RCA 6199 photomultipliers used in conjunction with both the crystal and plastic shield. The 4501/V4 tubes were selected because of their high quantum efficiency (31% at 385±50 nm), high gain (secondary emission ratio of 50 for the first dynode with 1000 V between the cathode and the first dynode) and low dark current (less than 10,000 c/m at 22°C). The high quantum efficiency is achieved by using a bialkali photocathode (potassium-cesium-antimony) and high gain by using a secondary emitting surface of galliumphosphide.

Electrical pulses, whose area is proportional to the number of photons sensed at the photocathode of the two photomultiplier tubes, are amplified by two preamplifiers (PA), one for each PMT. Only pulse shaping due to variation in the fall-time of the PA is done by the PA.

The output of the two PA's is directed to two delay line amplifiers (DLA) which provide shaping for all output pulses and expand the pulse amplitude by a selected gain factor of from 3 to 1000. Since no two PMTs can be expected to exhibit exactly equal gain for a given number of photons, equal sensitivity in each DLA is effected with the DLA gain adjustments. While both unipolar and bipolar output pulses are generated by the DLA, the bipolar output pulse was selected in our case since it is double delay-line shaped and thus provides a precision measure of time at the baseline crossover which is independent of pulse amplitude. The fastest integration time was selected to give the best pulse shapes. The height of an output pulse can range from 0 to 10 V and is dependent only on the area of the input pulse.

The output pulses of both DLAs are collected at a dual sum and invert amplifier (DSI). The DSI serves only to collect and make available for displaying and/or recording all of the raw data pulses produced by the two PMTs. This includes not only pulses dependent on beta

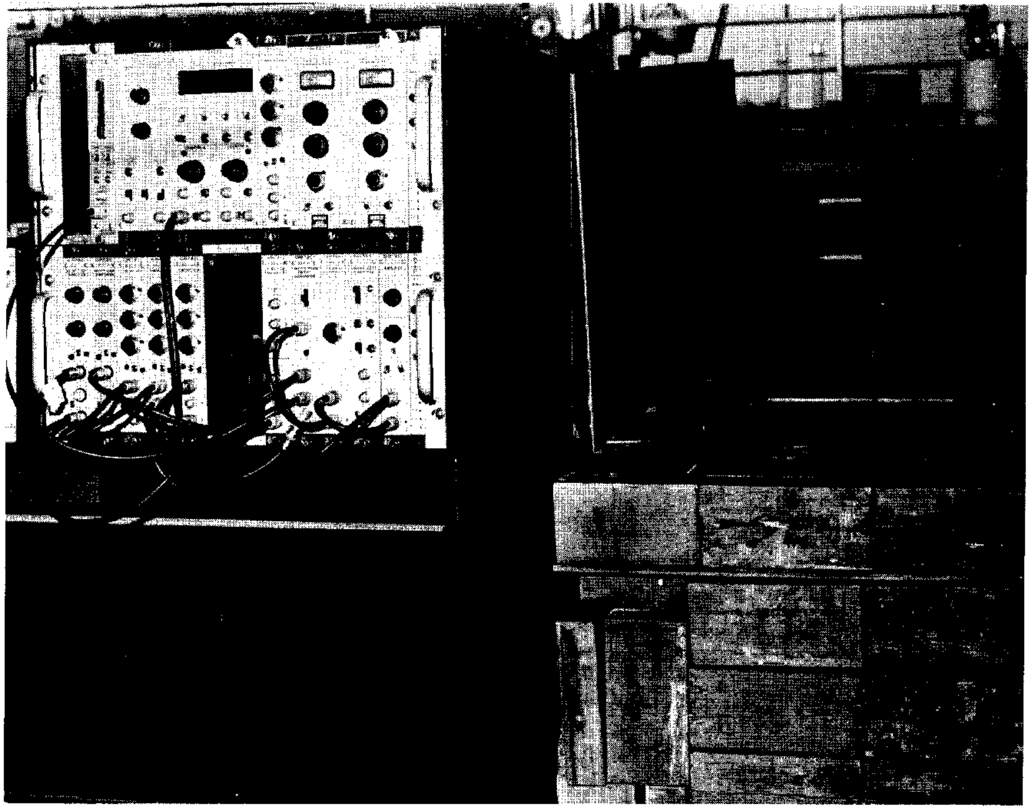

Figure 1. Over-all view of the low-level liquid scintillation counter - includes massive graded lead shield and electronics.

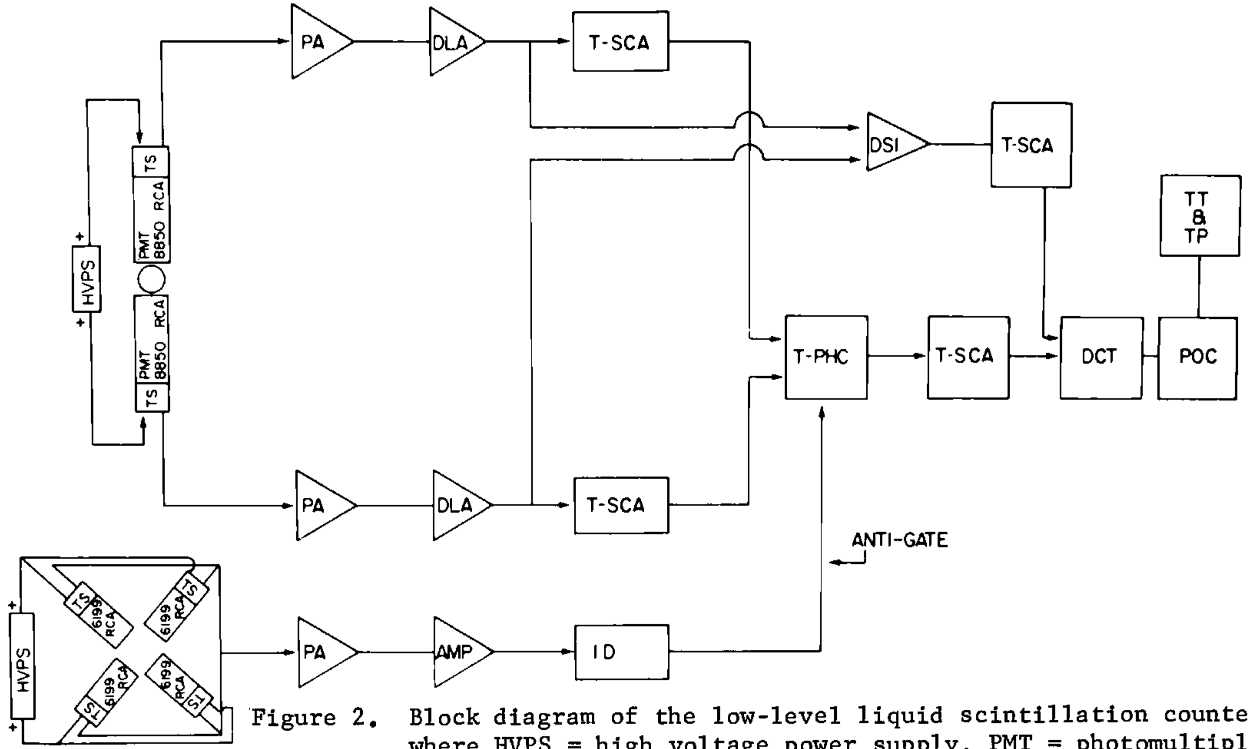

Figure 2. Block diagram of the low-level liquid scintillation counter where HVPS = high voltage power supply, PMT = photomultiplier tube, PA = pre-amplifier, T-SCA = timing single channel analyser, DSI = dual sum and invert, AMP = amplifier, ID = integral discriminator, T-PHC = time to pulse height converter, DCT = dual counter and timer, POC = printout control and TT&TP = teletype and tapepunch.

decay, but also the dark current or singles from each PMT.

The output pulse from each DLA also becomes the input pulse for a timing single channel analyzer (T-SCA). The T-SCA first tests the pulse for amplitude. The amplitude of this well-shaped bipolar input pulse is proportional to the number of photons that give rise to it at the PMT. Pulses which satisfy the criteria of the discriminators and thus occupy the pulse height interval delta E above E, become data pulses while the other pulses are rejected. The setting of the lower discriminator, E may be varied from 100 mV to 10 V and the setting of the interval above E or delta E may be varied from) to 10 V. The baseline crossover point of the data pulses initiates both a +5 V (nominal) square wave whose rise time is less than 20 nsec. and whose width is nominally 500 nsec. and a -0.6 V fast logic pulse whose rise time is less than 5 nsec. The fast logic pulse is selected since only a fast trigger is needed whose width is less than 20 nsec. A delay of 1.1 microsec to 100 nsec. can be introduced, as required, between the baseline crossover point of the data pulse and the leading edge of the fast logic pulse. It is worth noting that regardless of the input pulse amplitude, the fast logic output pulses of the T-SCAs are all of equal amplitude and differ from one another only in time. Note also that the fast logic output pulses from the T-SCA are initiated not only by scintillations but also by PMT dark current or singles. The contribution to the output of the T-SCAs by the PMT singles is eliminated by the coincidence requirement imposed by the next electronic operation.

The time to pulse height converter (T-PHC) receives three inputs, two of which are the output pulses of the two T-SCAs and one anti-gate input pulse from either the crystal or the plastic shield within which both the sample and the PMTs are placed. The latter signal's source will be dealt with in detail in the section of this paper concerned with background shielding. Briefly, however, this signal is used to eliminate a large fraction of the external radiation which gets through the lead graded shield. The delay setting of the two T-SCAs, which provide two of the inputs for the T-PHC is such that if two pulses leave the DLAs simultaneously, the resulting T-SCAs, and usually more due to differences in intrinsic electronic delay along the two paths followed by the data pulses. So that the output pulse of a given T-SCA will always occur prior to that of the other T-SCA, sufficient delay is introduced into the latter. For the experiment described here a minimum difference of 35 nsec. is used, the reasons for which will be discussed below.

The T-PHC, under the influence of the T-SCA's input pulses, generates a bipolar output pulse whose amplitude is proportional to the time difference between the two fast logic input pulses, (note that two such fast logic pulses are produced per beta event). Of the two fast logic pulses, the one from the T-SCA with the least delay always arrives at the T-PHC first and serves as a start pulse for the timing gate; the other fast logic pulse serves as it's stop pulse. If the time interval separating the start and the stop pulses is less than the selected gate range, an output pulse is produced. If, however, the stop pulse arrives after a time interval which is longer than the selected gate range, no output data pulse is generated. Moreover, if an anticoincidence pulse arrives at the T-PHC within the selected gate range along with a start and stop pulse, the generation of a data pulse is disabled. The logic being that if all three input pulses exist within the gate range, then an external event has given rise to them and is thus determined to be false data or background. For our purposes the 50 nsec. range was selected so that the probability of selecting false data would be minimized. Other gate ranges that are switch selectable included: 250 and 500 nsec. The selected gate range defines the range of the pulse height distribution of the T-PHC output pulses. Recall that the amplitude of the output pulses of the T-PHC is proportional to the time interval separating the start and stop fast logic pulses. The output pulse of the T-PHC is bipolar and has a constant pulse shape which is independent of it's amplitude. These pulses provide the input for yet another T-SCA whose task is to impose time discrimination on them and generate a square wave output pulse whose amplitude is +5 V (nominal) and 500 nsec. wide. For this T-SCA the E or lower discriminator is properly termed the T (for time) discriminator and the delta E termed delta T.

One scaler of a dual counter and time (DCT) receives the output pulses of the T-SCA discussed above. The other scaler of the DCT may receive as an input either the output of the DSI or that of it's timer. In this study the timer output was selected as this scaler's input so that count rates could be measured. Besides creating the time base with which any or all of the data are scaled, the DCT can be set up to display the data in either scaler. By feeding the output of the DCT to a print-out control POC, shown in Fig. 2 , the contents of both scalers can be permanently preserved on either a punched paper tape or a teletype printout or both.

C) NaI(Tl) CRYSTAL: A Bicron Inc., Mod. 7.5 HWS annular crystal, whose dimentions are 8" O.D., 5" in length, and 3½" I.D. was used in conjunction with four RCA 6199 PMTs and associated electronics as a background shield and is shown in Fig. 2 & 3 . With the sample and the two RCA 4501/V4 nested within the annulus of the crystal, all but a small fraction of the background radiation reaching the crystal and sample is detected, giving rise to an anticoincidence pulse at the T-PHC. Such a pulse identifies the two fast logic pulses from the preceeding T-SCA's as false data and aborts a data pulse produced from them.

As before, the PA provides amplification, but virtually no shaping. The amplifier (Amp) is used to provide further amplification and Gaussian shaping of the unipolar output pulse. The integral discriminator (ID) requires an imput pulse of at least a 50 nsec. width and up to +10 V amplitude. Such input pulses are provided by the Amp when the proper gain of X4 is selected. The output of the ID is composed of those pulses initiated by Amp pulses whose amplitude exceeds or equals the discriminator setting. The output signals are 500 ns wide square pulse of +5 amplitude. It is necessary to introduce considerable delay in the anticoin pulse so that its time of arrival at the T-PHC corresponds to within 50 nsec. of that of the associated start and stop fast logic input pulses.

The effect of the NaI(Tl) crystal, an anticoincidence guard, was measured with a 25 ml background sample in a quartz vial of high optical quality. When the crystal was disabled, a count rate of 10.40±0.09 c/m was observed in an optimized window, but with the crystal in operation, the background dropped to a count rate of 2.10±0.02 c/m. The difference in the two count rates shows a 81.5% reduction in background when the NaI(Tl) crystal shield is used.

D) PLASTIC SHIELD: The plastic (NE-102) background shield was obtained from Bicron Inc. It was an annulus

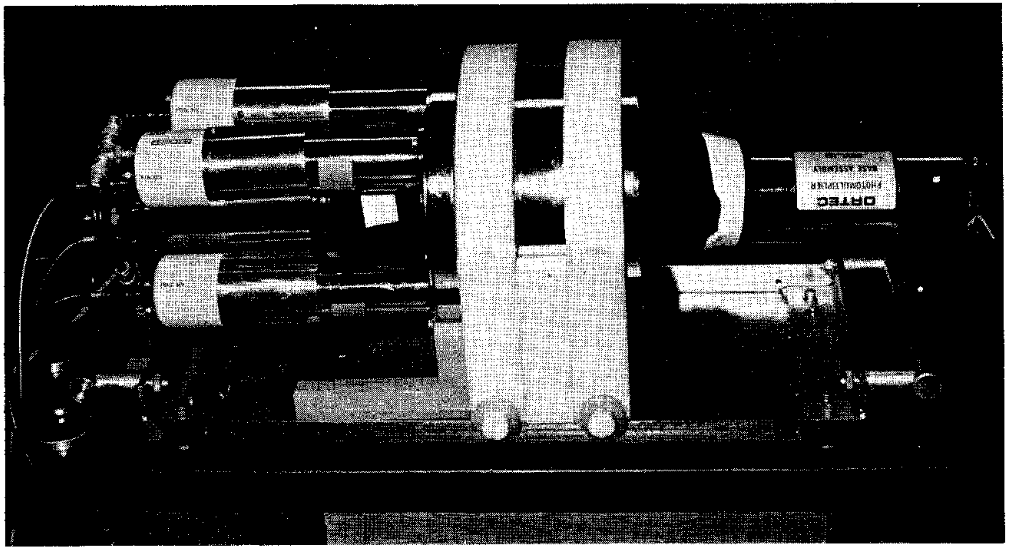

Figure 3. Background guard shield exposed with sample and PMT's mounted. (This particular one is the NaI(Tl) crystal.

whose dimensions are 5" in length, 9.5" O.D. and 3.5" I.D. Fig. 4 shows the above shield alone and Fig. 2 shows it diagramatically as a part of the system. Mechanically and electronically its' performance is exactly analogous to that described for the NaI(Tl) crystal shield.

Results and Discussion

It was our objective with this study to compare the performance of the NaI(Tl) crystal shield with that of the plastic (NE-102) shield, where, in both cases, no graded lead shield was present.

The composition of the scintillation solution used for all measurements was: 6.25 g PPO (2, 5-diphenyloxazole) and 0.4 g POPOP (1, 4-di-2-(5-phenyl-oxazole) benzene) thoroughly mixed into 1000 ml of benzene. In all cases the final concentration of the fluors in the sample was adjusted to that given above so that constant scintillator efficiency was insured. All chemicals used were of scintillation grade and obtained commercially.

Each background sample was composed of the scintillator described above and was measured into the vials with class "A" pipets. One hundred minutes was selected as the counting interval, and each of the background count values reported is the average of the counts collected during many counting intervals. The error associated with each background was computed to a 2σ confidence level and is based on the sum of all of the counts collected during each of the counting intervals of a given measurement.

The electronics previously described were routinely calibrated. A precision pulser operating at either 60 or 220 Hz and producing 1 nsec. rise up to +10 V tail pulse was used in conjunction with a fast oscilloscope. The test BNC connector on each PA was used to accept the pulser's input and was subsequently treated, as any data pulse would be, by the system's electronics. Pulse shapes and timing were measured with the scope.

Initially both long and short term stability was studied. Long term stability is illustrated by Fig. 5 . Short term stability was checked with an external γ source (^{137}Cs). In both measurements Chi Square was obeyed.

When the system, as previously described, was used to study ^{3}H, ^{14}C, and background samples, the results shown in Table I were obtained. It is clear that an E^2/B maximum is exhibited at 64.7% relative efficiency. It

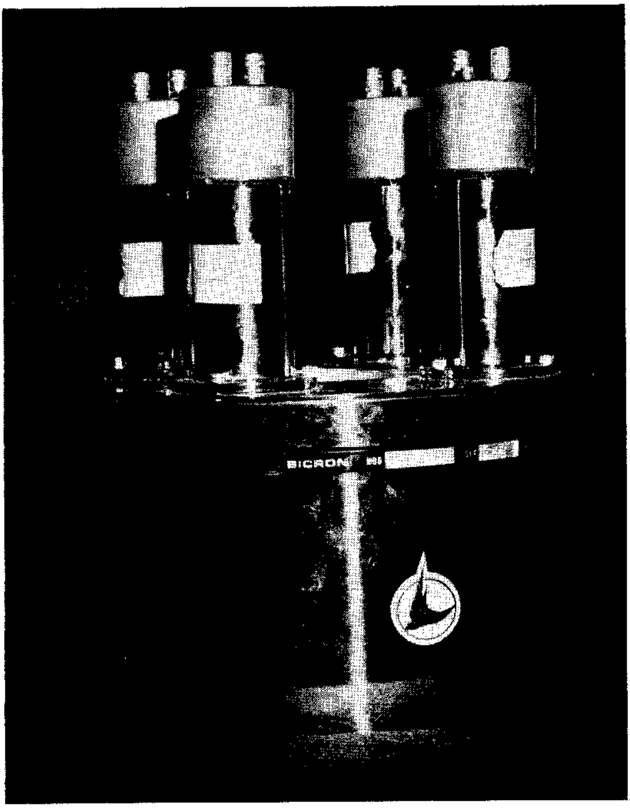

Figure 4. The plastic background guard shield exposed and without the PMT in place.

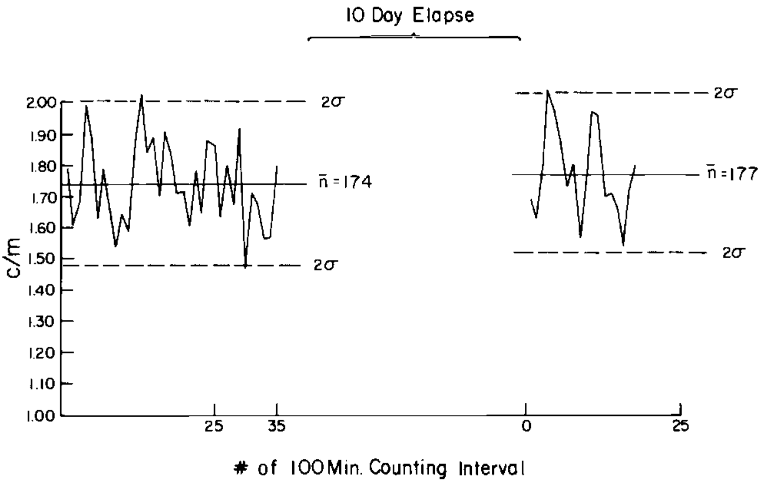

Figure 5. Long term system stability test count data.

TABLE I

Performance data of NaI(Tl) crystal background guard shield when inside the massive graded lead shield. Both ^{14}C and ^{3}H where considered.

Window (%)$^{(a)}$	Bkgd in $^{c}/_{m}$ $^{(b)}$	Eff in %$^{(c)}$	E^2/B $^{(c)}$	E^2/B per ml$^{(c)}$
100.0	4.20±0.5	96.0 (64)	2194 (975)	100 (44)
95.0	3.55±0.41	91.2 (61)	2332 (1041)	106 (47)
88.8	2.11±0.40	85.2 (57)	3440 (1540)	156 (70)
75.9	1.22±0.26	72.9 (48)	4356 (1889)	198 (86)
64.7	0.79±0.24	62.1 (41)	4882 (2170)	222 (98)
55.3	0.71±0.24	53.1 (29)	3971 (1764)	180 (80)
Typical commercial counter (d)	37 (29)^{c}	94 (59)	237 (118)	15 (7)

a) relative % efficiency.

b) all errors expressed in $^{c}/_{m}$ at 2σ confidence level.

c) numbers in parenthesis are for ^{3}H; those without parentheses are for ^{14}C.

d) Nuclear Chicago Mark II bench-top LS counter without the crosstalk level feature active.

should be born in mind that these results were obtained with the use of both the graded and NaI(Tl) crystal shield.

As shown in Fig. 3 , the background guard shield was outside of the graded lead shield, but placed on a bench-top, physically near to the electronics. The sample and PMT's were nested within the annulus, such that they all had a common axis. The PMTs were light-sealed by means of a felt tape of our own construction. Whether the NaI(Tl) crystal or the plastic served as the background guard shield, the above experimental configuration was used. The data shown in Table II illustrates the effectiveness of the background guard shield.

It is clear from an examination of Table II that the NaI(Tl) is a very good passive filter for background radiation and the plastic is not (passive is meant to denote a state of electronic inactivity). Since the average "Z" of the NaI(Tl) crystal is much higher than the plastic these results are reasonable. When the background guard shield is made active the NaI(Tl) again bests the plastic guard shield, by 30% (relative). In view of the high efficiency of the NaI(Tl) crystal, its' superior performance is expected.

Type	Mode	Bkgd in $^{C}/_{M}$ (a)	$^{E^2}/_{B}$ for ^3H (b)	$^{E^2}/_{B}$ for ^{14}C (b)
NaI(Tl)	on	17 ± 0.24	99	227
	off	58 ± 0.48	29	66
Plastic	on	70 ± 0.75	24	55
NE102	off	192 ± 0.88	9	20

(a) errors expressed to 2σ confidence interval.
(b) based on best E^2/B window as shown in Table I.

Table II Performance data of background guard shields in the absence of the massive graded lead shield.

From these results it can be seen that the counting system which employs the NaI(Tl) crystal and no lead gives results comparable with those of typical low cost commercial liquid scintillation counters. The performance of the plastic guard without lead was not as good. With an optimized background guard shield which makes use of a new scintillation material, Bicroguard^{TM}, counter performance equivalent to that of a commercial counter is to be expected. It is worth noting that such performance is obtained without massive graded lead shielding, and thus, such an arrangement may have future commercial application.

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TM Bicroguard, Bicron Inc.