

## MODEL DYS LOW-LEVEL LIQUID SCINTILLATION COUNTER

Jiang Han-ying, Lu Shao-wan, Fu Shi-mi, Zhang Wen-xin,  
Zhang Ting-kui, Ye Yuan-zhen, Li Mei-fen, Fu Pei-yun,  
Wang Shu-xian, Peng Cheng-hang and Jiang Pei-dong,  
(Institute of Biophysics, Academia Sinica, Beijing, China)

### ABSTRACTS

Two models of a low-level liquid scintillation counter, the DYS-1 and DYS-2, were designed and manufactured in our laboratory. The DYS-1 is more sensitive than the DYS-2. For 5 ml  $^{14}\text{C}$  benzene samples, an efficiency of 80% and a background of 0.54 cpm are obtained from DYS-1; its figure of merit,  $(E^2/B)$ , exceeds 10,000. The detection limit for tritium in water (50 ml water and 50 ml Instagel scintillator) is approximately 1 Bq/l water ( $t = 30$  min,  $P = 68.3\%$ ). The sensitivity of DYS-2 is better than conventional liquid scintillation counters; for 5 ml  $^{14}\text{C}$  benzene samples, the efficiency of the DYS-2 is above 70% and its  $E^2/B$  varies from 1700 to 2500 depending on the construction of material of the counting vial. When a 10 ml sample of water is mixed with 10 ml of Instagel, a detection limit better than 4 Bq/l water ( $t = 30$  min,  $P = 68.3\%$ ) can be attained.

The DYS-1 is a single sample manual liquid scintillation counter which can be used to measure samples in various sizes between 2-100 ml with optimum geometry. The DYS-2 provides an automatic sample-changing system with a capacity of 10 vials and can be used to measure 5-20 ml samples under optimum conditions. Both instruments permit preset counting time and print raw data automatically.

### INTRODUCTION

During the last two decades, tremendous efforts have been made in many laboratories to reduce the background of liquid scintillation counters and augment the volume of samples designed to improve its sensitivity.

Carbon dating has become a widely used method for the geologist and archaeologist, etc. The method of measurement in  $^{14}\text{C}$ -dating has

been altered and the liquid scintillation counter is gradually superseding counting with a gas detector in most laboratories. In general, about 5 ml of benzene is synthesized from a sample resulting in a good figure of merit.

Tritium in the environmental water has become the objective of measurement since 1950. The measurement of  $^3\text{H}$  concentration in the earth surface water, underground water, rain, snow and ice, etc. provides important information on hydrology, meteorology, and geo-chemistry, etc. A wide range of tritium values from less than one to a few hundreds Becquerels should be measured. Large volume sample LS counters have been developed for the direct measurement of tritium in the environmental waters. Some samples which have a very low specific activity must be enriched before each measurement is taken. Sometimes, the amount of sample is inadequate after enrichment for large volume sampling, and some small-sized sample vials may still be needed. Two models of low level liquid scintillation counters were built to meet the various requirements.

#### INSTRUMENTATION

The main detector uses two EMI 9635QB, two-inch diameter "Quantacon" photomultipliers. It has a high quantum efficiency due to a bialkali photocathode (potassium-cesium-antimony) and a low background noise, less than 10,000 cpm at 25°C. A pair of EMI 9635QB in the DYS-1 counter was selected from over ten tubes. The EMI 9635QB used in the DYS-2 counters were not selected.

A sample is placed between two axially opposed photomultiplier tubes and a 20 ml sample chamber (see Fig. 1) is used as a the basic sample chamber for both the DYS-1 and DYS-2. The copper chamber has a teflon inner lining which can provide good reflection. When using some small spacers or regulating the position of the elevating rod; 2 ml, 5 ml, 10 ml and 20 ml samples can be measured under optimum conditions. The axis of two photomultipliers and central horizontal plane of sample are coincident and a good geometry can be obtained.

In the DYS-1 counter, a 50 ml or 100 ml sample chamber can be mounted. Both were made of copper and coated with MgO or lined with teflon along the inner surface.

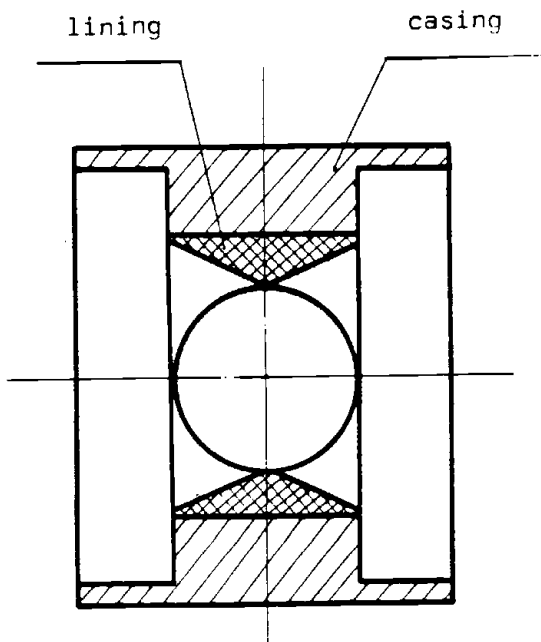


Figure 1. The schematic diagram of 20 ml sample chamber.

All of the vials are made of quartz. Vials for the 2 - 50 ml samples are cylindrical and the 100 ml sample size is a drum vial. Their sizes and shapes are shown in Fig. 2. Vials made of other materials, such as teflon, low-potassium glass, may be used in these counters only if their size and shape are adapted to the chamber.

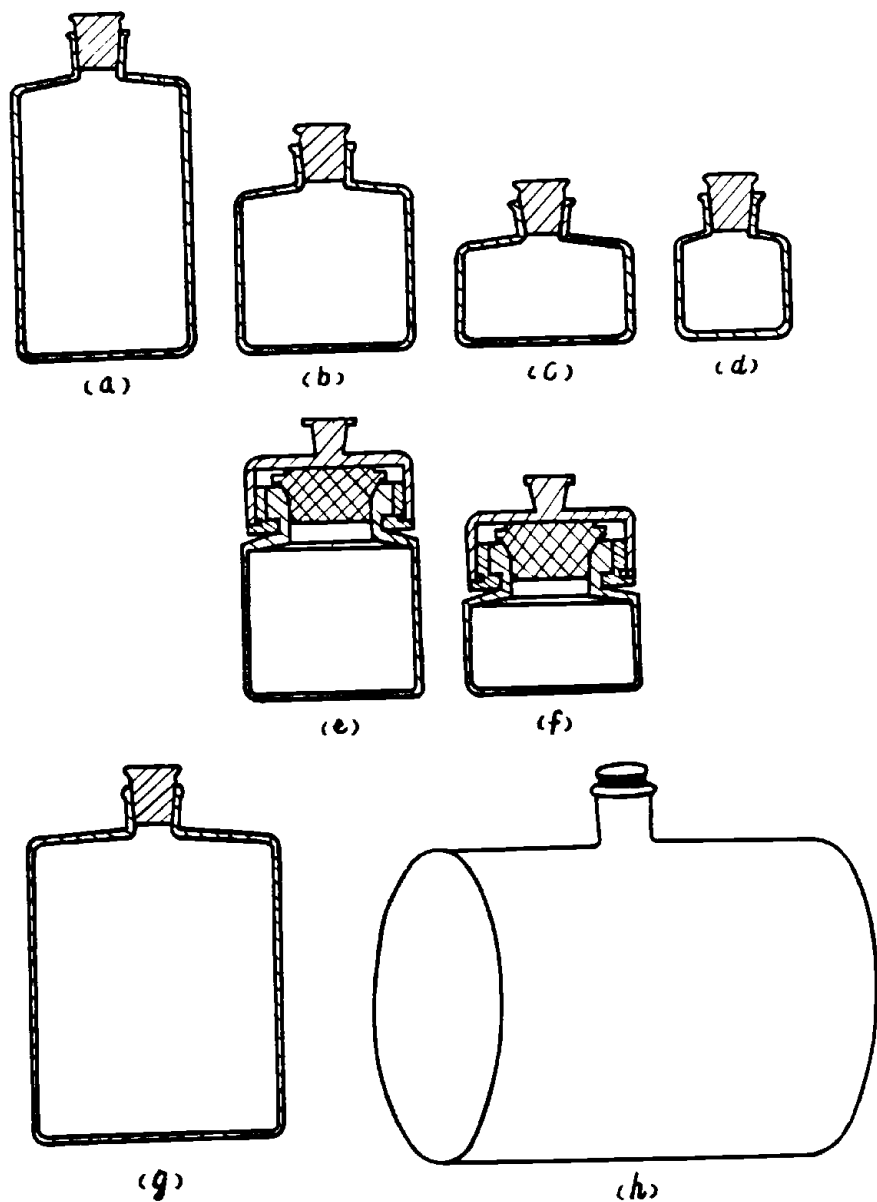


Figure 2. The schematic diagram of sample vials. (a)(b)(c)(d)(g) - 20 ml, 10 ml, 5 ml, 2 ml, 50 ml: cylindrical quartz vials. (e)(f) - 10 ml, 5 ml; teflon vials. (h) - 100 ml: drum vial.

Figure 3 shows a block diagram of DYS-1 and DYS-2. Most of the electronics were made up of standard NIM bin components, of which some were obtained from the Beijing Nuclear Instrument Factory (BNIF). Some parts and elements were modified according to the requirements of low level measurement. A few units, such as the quasi-logarithmic amplifier, controller, etc., were fabricated in our own laboratory, in form of NIM bin components. The AC line filter, stabilized AC power supplier, driving and control circuit of reversible motor were mounted in two small boxes.

The emitter-follower preamp follows each of the photomultipliers (PMT). One output signal can be obtained from the preamp, except in the main detector of the DYS-1 where two output signals are obtained.

The amplitude of output pulses from the preamps of the main detector are proportional to the number of photoelectrons emitted from the photocathode of each PMT; they are amplified by two quasi-logarithmic amplifiers (QLA). When the amplitude of the input pulses ranges from a few mV to 10V, the output of the QLA ranges approximately from 0.3V to 5V with a pulse width of  $\sim 0.5 \mu\text{s}$ .

Two timing single channel analyzers (T-SCA) receive the output pulses from the two QLAs, respectively. The T-SCA provides differential amplitude analysis, i.e. energy analysis of all input pulses and gives timing output pulses.

DYS-2: A coincidence-anti-coincidence (CAC) circuit receives the output pulses from the two T-SCAs through its input terminals I and II, respectively. If the two input pulses arrive at the CAC within a fixed specified interval, usually about 50 ns, the output I of the CAC will provide a pulse to the scaler. A printer can automatically print out the raw-data at each end of the preset counting time as selected by a switch in the scaler. The machine can also be operated under a programme in which an automatic sample change from the circular, ten position sample train is followed by counting and print-out.

DYS-1: The first output pulses of the preamps of the main detector are fed to a dual summing quasi-logarithmic amplifier (DSQLA) where they are added together and logarithmically amplified.

The output pulses of the six preamps from the six photomultiplier tubes of the anti-coincidence detector are sent to a linear mixer (LM)

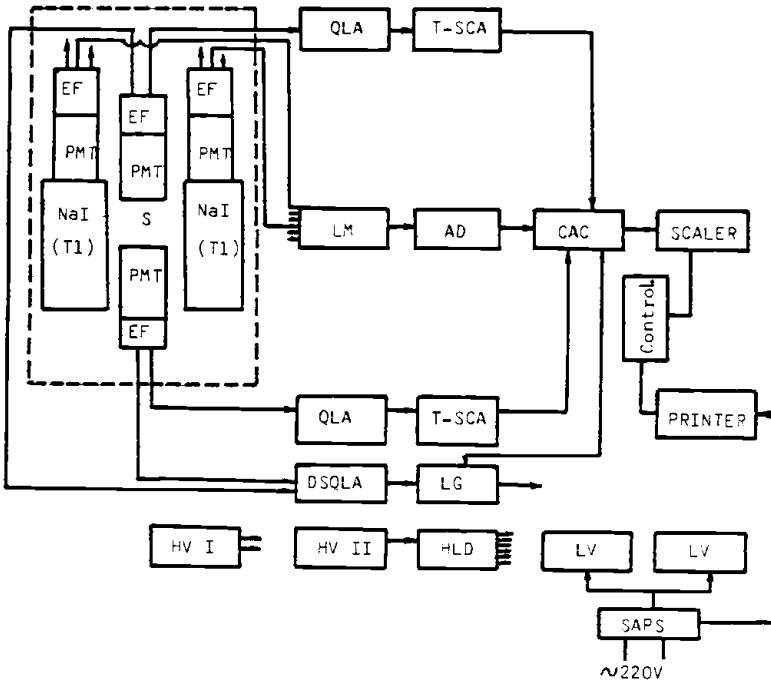


Figure 3a. Model DYS-1.

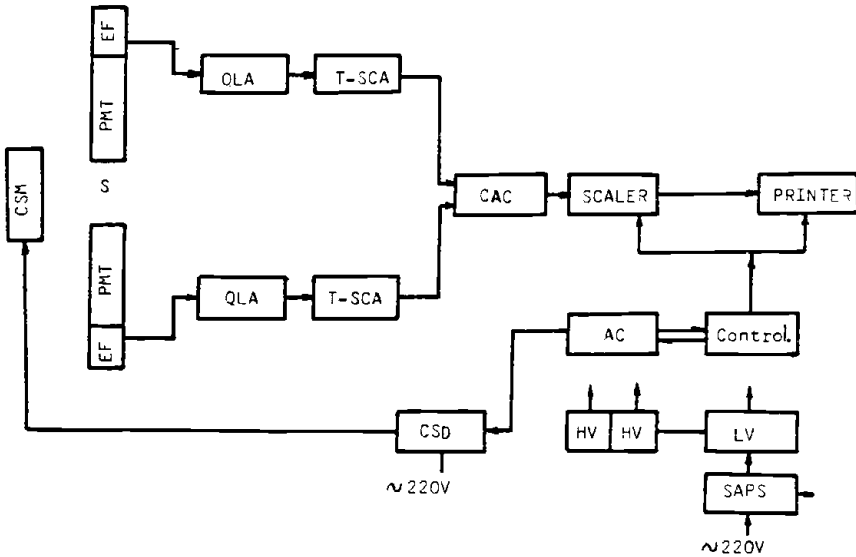


Figure 3b. Block diagram of DYS-2. QLA: quasi-logarithmic amplifier. T-SCA: timing single channel analyzer. CAC: coincidence-anti-coincidence. AC: automatical changing sample. CSD: changing sample driving. SAPS: stabilized AC power supplier. CSM: changing sample machinery. EF: emitter-follower.

and added together. The output of LM is then examined by an amplifier-discriminator (AD). If it exceeds a certain threshold, the AD sends a pulse to the CAC as the third input pulse. Therefore, there are three input lines connected with the CAC, two from the main detector, one from the anti-coincidence detector whose function will be discussed in detail in the section about the anti-coincidence detector. If the three input pulses appear simultaneously, it means the input pulse comes from external radiation and no output pulse occurs. If both of T-SCAs give simultaneously output pulses within approximately 50 ns, but no pulse at the output of AD during the same period, pulse is generated at output II of CAC. This is the case when a flash is caused by a particle emitted from the sample itself in scintillator. The scaler counts the pulses from output II of the CAC and the printer prints the raw-data at the end of each preset time under program control. Regardless of the presence of the AD, as long as there are two output pulses from the two T-SCA within the specified interval, an output pulse from output I of the CAC can still be obtained.

The output of the DSQLA and the output I of the CAC can be directed to a linear-gate circuit (LG), the output pulse of the LG can be sent to a T-SCA or a multi-channel analyzer to analyze the energy spectrum or to correct for quenching.

The effect of a cross-talk discrimination circuit on the background of carbon-14 has been tested. The pulse analysis logic used has the property that the output pulses from two PMTs are analysed firstly according to their amplitude, i.e. their energy analog and then for coincidence within a specified interval. This decreases the background of the  $^{14}\text{C}$  channel measurably due to reduction of the background caused by cross-talk between the opposing PMTs in the sample chamber. A signal caused by cross-talk is not derived from photons produced by the sample, it is derived from photons produced in either PMT. When photons are produced in one PMT, the signal created in that PMT is larger than the signal produced in the other due to the reduction of photons resulting from the distance between the two PMTs. Laney<sup>1</sup> designed a cross-talk discriminator circuit based on this principle. With this discriminator circuit, the figure of merit for  $^{14}\text{C}$  is increased from 340 to 610. With the pulse

analysis logic described here, in the DYS-2, the figure of merit for 5 ml  $^{14}\text{C}$  benzene sample can be increased from 940 to 1580. In the DYS-1 where an anti-coincidence detector is introduced, the figure of merit for a 5 ml  $^{14}\text{C}$  benzene sample can be increased from 9600 to 11800. The background due to cross-talk of the  $^{14}\text{C}$  channel for the DYS-2 can be decreased from 8.15 cpm to 4.43 cpm. The  $^{14}\text{C}$  channel cross-talk background in the DYS-1 can be decreased from 0.24 cpm to 0.13 cpm. The reduction in the DYS-2 is greater than that observed for the DYS-1 which is better shielded against cosmic rays and environmental gammas. This indicates the cross-talk background comes mainly from cosmic rays and external radiation.

#### SAMPLE AND BACKGROUND STANDARD

1. Standardized solutions of  $^3\text{H}$ ,  $^{14}\text{C}$ -n-hexadecane and  $^3\text{H}_2\text{O}$  (Amersham Co.) were used. The accuracy of  $^3\text{H}$ -activity was  $\pm 3\%$  and that of  $^{14}\text{C}$ -activity was  $\pm 2\%$ . The tritium-free water was obtained from Tianjin.

2. Scintillation solutions: Toluene (The First Chemical Agent Factory, Shanghai). Benzene (The Chemical Industry, Factory of Beijing). Scintillator PPO (England), POPOP and B-PBD (The Chemical Agent Factory of Shanghai). Instagel scintillator (Packard Co., USA). Triton X-100 (ROTH; Rohm + Haas). The water used to dilute the standardized  $^3\text{H}_2\text{O}$  was distilled water.

Toluene scintillation solution was prepared as 7g PPO + 0.35 g POPOP per liter of toluene; Triton X-100 scintillator was prepared from a mixture of toluene scintillation solution and Triton X-100; the mixing ratio was 2:1. The benzene scintillation solution was prepared from 5 g PPO + 0.5 g POPOP per liter of benzene or 6 g B-PBD per liter of benzene.

3. Standard samples: Standard samples of 5 ml, 10 ml and 20 ml of  $^3\text{H}$  and  $^{14}\text{C}$  were made of the standardized solutions of  $^3\text{H}$ ,  $^{14}\text{C}$ -n-hexadecane and 5 g PPO + 0.5 g POPOP per liter of benzene. A standard sample of 5 ml  $^{14}\text{C}$  used in the long-term stability measurement and a

standard sample of 2 ml  $^{14}\text{C}$  were made from the standardized solution of  $^{14}\text{C}$  and 6 g B-PBD per liter of benzene. Standard samples used for measuring  $^3\text{H}_2\text{O}$  detection limit in 10 ml, 20 ml, 50 ml and 100 ml volume were made from standardized  $^3\text{H}_2\text{O}$  and Instagel with a mixing ratio of 1:1. A standard sample used for long-term stability measurement of the  $^3\text{H}_2\text{O}$  sample was made from the standardized  $^3\text{H}_2\text{O}$  and Triton X-100 scintillation solution, with a mixing ratio of 2:3. Standard background samples were made by adding the  $^3\text{H}$ -free water instead of  $^3\text{H}_2\text{O}$  and used as the "blank" samples.

All of the standard samples were held in quartz vials. Ground quartz stoppers for the vials were glued in place with 914 adhesive agent. The standard samples can be used for a few years, free from any leakage. All of the standard samples were equilibrated with air.

#### THE DYS-1 LOW LEVEL LIQUID SCINTILLATION COUNTER

1. Shielding and sample-changing: The DYS-1 was placed on the ground floor of a four-story laboratory building. The main detector consists of a sample chamber connected with two PMTs horizontally opposed. The sample chamber is surrounded by an annular NaI(Tl) crystal which functions as the anti-coincidence detector. These are coupled to the sample-changing machinery and framed in a 1100 x 400 x 350 mm steel chamber with a black interior. The steel chamber is placed inside a 1300 x 600 x 550 mm lead box built from 20 x 10 x 5 cm lead bricks. There is 150 x 100 mm hole at the top of the lead box which is covered with a 255 x 205 x 100 mm rolling lead lid. When a sample is being changed, the lead lid is manually moved and a reversible motor moves the sample chamber out along the middle hollow of the anti-coincidence crystal. After a sample is changed manually, the reversible motor is started again, and returns the sample chamber back to its counting position. Finally, the lead lid should be pulled back to cover the hole. This instrument is illustrated in Fig. 4.

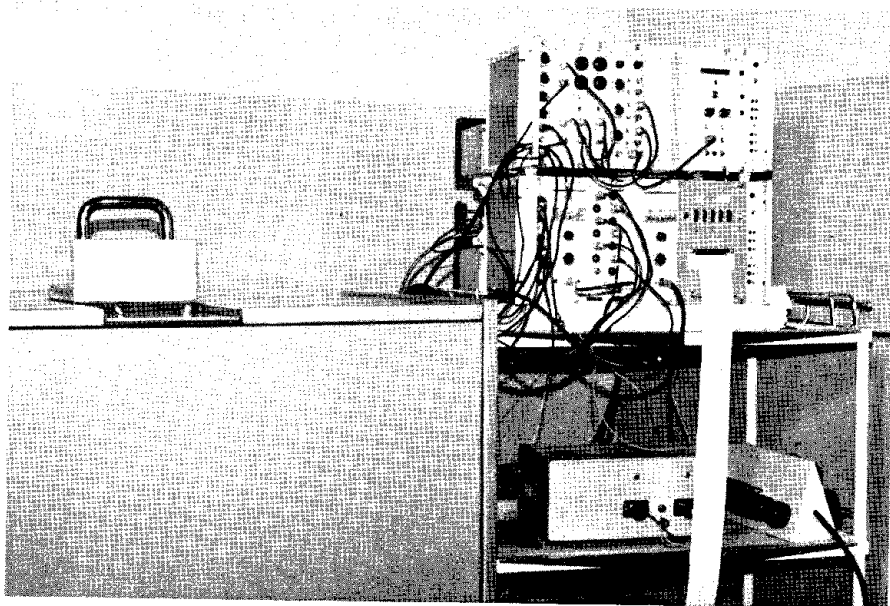


Figure 4. Model DYS-1 low-level LS counter.

2. The anti-coincidence detector consists of an annular (NaI(Tl) crystal (200 mm O.D., 150 mm in length and 80 mm I.D.) and six photomultipliers which are used to collect the photons from the crystal. The NaI(Tl) crystal was not constructed from potassium-free materials. There are six windows, glass windows (7.5 mm thick) on it in order to accommodate the six PMTs (GDB-52).

When cosmic or external hard radiation simultaneously pass through the NaI(Tl) and sample, the signal obtained from both the main detector and anti-coincidence detector is removed by the electronics. Since the detection efficiency of NaI(Tl) for hard radiation is very high, the background which comes from external radiation can be largely removed. This principle has been previously applied in low level counters by J.E. Noakes et al.<sup>2</sup> who was the first to apply a NaI(Tl) crystal as an anti-coincidence detector in a liquid scintillation counter.

Table 1. The effect of an anti-coincidence detector on the background of a 20 ml sample in the DYS-1.

	empty chamber (cpm)	full spectrum (cpm)	<sup>3</sup> H channel 56% Eff (cpm)	<sup>14</sup> C channel 77% Eff (cpm)	<sup>3</sup> H <sub>2</sub> O channel 23% Eff (cpm)
NaI(Tl) off	17.7±0.14	31.6±0.2	6.09±0.06	6.11±0.08	3.49±0.06
NaI(Tl) on	1.43±0.03	6.31±0.11	2.17±0.04	1.73±0.05	1.21±0.03
reduction %	92	80	64	72	65

The effect of the anti-coincidence detector is illustrated in Table 1. The background of the empty sample chamber of the whole energy channel was reduced from  $17.7 \pm 0.4$  cpm to  $1.43 \pm 0.3$  cpm; about 92%, due to the use of the NaI(Tl) crystal. The background of the empty sample chamber results mainly from external radiation which produces Cerenkov light in PMTs and other transparent materials within the main detector. The sample background in the whole energy channel was reduced from  $31.6 \pm 0.2$  cpm to  $6.31 \pm 0.11$  cpm; about 80%, due to the use of the NaI(Tl) crystal. This revealed that a portion of the sample background was caused by residual radioactivity inside the lead box.

3. The experimental results of the DYS-1 are shown in Tables 2 and 3, the efficiency of 20 ml tritiated benzene sample is 56% with a background of 2.17 cpm, correspondingly the figure of merit is greater than 1400; when the detection efficiency of a carbon-14 benzene sample of 5 ml is 80%, the figure of merit is over 10,000.

The measured results of tritium in water are shown in Table 3. In order to compare low level liquid scintillation counters, Moghissi et al.<sup>3</sup>, proposed the Y value, the detection limit, provided the counting time is t and P = 68.3%.

Table 2. The measured results of 5 ml, 10 ml and 20 ml samples in the DYS-1.

	efficiency (E)	background (B)	E <sup>2</sup> /B
20 ml	<sup>3</sup> H channel 56%	2.17±0.04	1450
	<sup>14</sup> C channel 77%	1.73±0.05	3450
10 ml	<sup>3</sup> H channel 58%	1.79±0.03	1890
	<sup>14</sup> C channel 77%	0.91±0.03	6550
5 ml	<sup>3</sup> H channel 58%	1.35±0.04	2500
	<sup>14</sup> C channel 80%	0.54±0.02	11800

Table 3. The measured results of <sup>3</sup>H<sub>2</sub>O sample

sample volume	water volume	efficiency E	background B(cpm)	detection limit (Tritium Units TU)				(EM) <sup>2</sup> /B
				30	100	500	1000 min	
10 ml	5 ml	28%	0.99±0.03	18	10	4.5	3.2	19500
20 ml	10 ml	23%	1.21±0.03	12	6.7	3.0	2.1	43700
50 ml	25 ml	18%	3.54±0.06	10.5	5.8	2.6	1.8	59800
100 ml	50 ml	17%	6.63±0.08	7.9	4.3	1.9	1.4	106000

$$Y = \frac{\sqrt{B}}{2.22EM\sqrt{t}} \quad nCi/l = \frac{\sqrt{B}}{0.00714EM\sqrt{t}} \quad TU = \frac{16.7\sqrt{B}}{EM\sqrt{t}} \quad Bq/l$$

Here, we define B, background in cpm; E, efficiency in cpm/dpm; M, the volume of water within the sample in ml. As it is shown in Table 3, Y value of DYS-1 with the sample of 20 ml is approximately 1.2 Bq/l (t =

100 min). In general, the tritium in most of the water of earth's surface can be directly measured with 20 ml sample. With a sample of 100 ml (50 ml water and 50 ml Instagel) under optimum conditions, an efficiency of 17% with a background 6.63 cpm can be obtained. Thus, the best detection limit is 0.2 Bq/l (1000 min).

We selected several water samples provided by the Institute of Geological-Mechanics for a comparison of the results of the measurement between the direct counting method and the pre-enrichment method is shown in Table 4. The results of the two methods are identical within the errors of determination. The sample for direct counting contained an 8 ml water sample with 12 ml Triton X-100 scintillator; the sample for the enrichment method contained 4 ml of enriched water and 6 ml Triton X-100 scintillation solution. Triton X-100 scintillator used here was made from ROTH Triton X-100 which has higher background than the Rohm + Haas Triton X-100, as shown in Table 5. If a 100 ml sample with Rohm + Haas Triton X-100 scintillator or Instagel was employed, the sensitivity would be even higher and more accurate.

Table 4. The comparison of results between direct counting and counting after enrichment.

sample	Xi 3	Xi 4	Yang 4	Yang 7	Tai 5
direct (TU)	7±7	20±7	36±6	42±7	113±6
indirect (TU)	6.4±.3	18.7±.6	36.5±.7	59.4±.8	117±1

Table 5. The comparison between two kinds of Triton X-100

Trademark	Roth	Rohm + Haas
background (cpm)	1.33±0.05	0.99±0.03

4. The long term stability of the detection of efficiency and background was checked during the period from September, 1981 to April, 1982. The time required to measure the efficiency for each counting period was longer than 10 min, the stability deviation was minimal and the changes were attributed to the instrument. The time required to register the background at each measurement was longer than 500 min. An average value and a statistical standard deviation at each measurement was calculated. The results indicate that the measured standard deviation was 0.9% for  $^3\text{H}$  efficiency of a 20 ml sample; 0.4% for a 5 ml  $^{14}\text{C}$  sample and 1.8% for a 10 ml  $^3\text{H}_2\text{O}$  sample. The background was very stable. The experimental standard deviation did not exceed the statistical standard deviation.

MODEL DYS-2 LOW LEVEL LIQUID SCINTILLATION COUNTER

The main detector is enclosed in 5 - 7 cm of lead. The part near the sample chamber is shielded with 7 cm of lead; the other areas with 5 cm of lead. There is a 2 cm lead brick over the sample changer entrance. The lead shielding weighs about 150 kg.

Table 6. The performance of DYS-2

sample*	vial	efficiency E %	background B(cpm)	$E^2/B$	Y (TU) (t=30', P=68.3%)
5 ml $^{14}\text{C}$	quartz	73.4-75.8	3.01-3.28	1700-1850	-
benzene sample	LK** teflon	73.8-76.4 75.6-80.5	2.66-3.13 2.41-2.87	1750-2200 2250-2500	- -
10 ml $^3\text{H}_2\text{O}$ sample	quartz	20.1-23.7*	2.14-3.26*	-	36-41
20 ml $^3\text{H}_2\text{O}$ sample	quartz	19	5.0	-	30

\* The values are a range of values obtained from four sets of data

\*\* Low-potassium glass

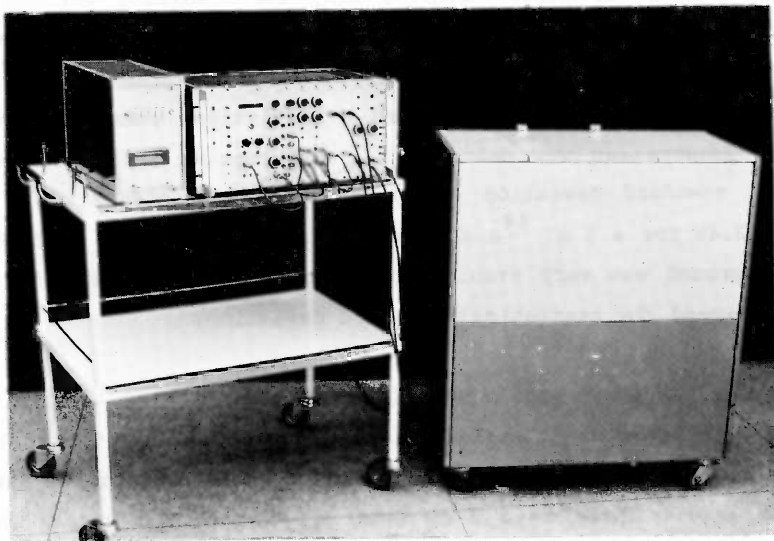


Figure 5. Model DYS-2 low-level LS counter.

The main body of the counter consists of four parts: machinery frame, sample changer, detector and sample elevator. The frame was made of zinc plated square-steel tubing and mounted on four small wheels. A ten position rotating sample changer allows samples to be counted, one-by-one, in sequence. When a sample arrives at the appropriate place, the elevator can transport the sample down into the sample chamber for measurement (Fig 5). The elevator and sample support can be adjusted to measure 5 - 20 ml samples in optimum geometry. The experimental results for the DYS-2 are shown in Table 6. These data were measured from four sets of samples. Vials with low potassium glass yield an efficiency for a 5 ml  $^{14}\text{C}$  benzene sample of over 70% with the figure of merit of 1700, the best being 2200. With teflon vials, the figure of merit can be as large as 2500. With a 10 ml sample, 5 ml water and 5 ml Instagel, the Y value is 5 Bq/l; with a 20 ml sample, 10 ml water and 10 ml Instagel, the Y value is 4 Bq/l ( $t = 30$  min). Water samples of several Bq/l can be measured

directly. With the sample changing automatically, long term stability of a 5 ml  $^{14}\text{C}$  sample and background sample during a one week period was observed. The experimental standard deviation did not exceed the statistical standard deviation.

#### REFERENCES

1. B.H., Laney, "Electronic Rejection of Optical Crosstalk in a Twin Phototube Scintillation Counter" in "Organic Scintillator and Liquid Scintillation Counting", D.L. Horrocks and C.T. Peng (eds.), Academic Press, New York, 991, 1971.
2. J.E. Noakes, M.P. Neary and J.D. Spaulding, "Tritium Measurements with a New Liquid Scintillation Counter", Nuclear Instruments and methods, Vol. 109, 177, 1973.
3. A.A. Moghissi, H.L. Kelley, J.E. Regnier and M.W. Carter, "Low-level Counting by Liquid Scintillation Counting - I. Tritium Measurement in Homogeneous Systems", International Journal Appl. Radiat. Isotop., Vol. 20, 145, 1969.