

DETECTION OF WEAK BETA EMITTERS BY LIQUID SCINTILLATION COUNTING TECHNIQS

I. INVESTIGATIONS ON THE OPTIMAL DETECTING CONDITIONS OF SOFT BETA EMITTERS.

By

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I. Introduction

Marked substances are often employed in order to clear up the mechanism of chemical reactions. In the organic, and above all in biochemistry — the most frequently used radioactive isotopes, the ^{14}C , the ^3H and the ^{35}S , are weak beta emitters their energy being in the order of 0.1, 0.01 MeV, respectively, and therefore the applications of special counting technics are needed. The most often used methods are those of the ionization chamber, of the proportionate counter the special GM. tube and the liquid scintillation counting technics.

In the course of our work some problems concerning the liquid scintillation counting technics were investigated with instruments available at the Institute of Physical-Chemistry of the Technical University, Budapest and in the Isotope Institute of the National Commission for Atomic Energy.

In the case of liquid scintillation counting the active sample to be measured is directly solved in the scintillator and this is the source of numerous theoretical and practical problems. In the liquid scintillator systems consisting of some solvent, mostly toluene and scintillators (phosphors), the radioactive radiation primarily excites the solvent molecules. This excitation energy moves by various mechanisms to the phosphor which converts a portion of it into light.

Some substances even the increasing concentration of a fluorescent solute hinders the energy transfer and thus the light output decreases.

This fact causes a serious problem, because often the active sample itself or its impurities show such an effect. The mentioned phenomenon is called quenching and the substances are called quenching agents.

A light-tight container surrounds the vial containing the scintillator and the active sample and couples it to the photomultiplier, which performs the transformation and primary amplification of the light impulses. The pulses obtained at this output are further amplified and registered in a scaler.

The other critical part of the detecting process is the above-mentioned photomultiplier, because at its output we obtain impulses, when in the vial

containing the scintillator radioactive substances are not present, because of the high gain required by the low pulse-height. The counts thus observed are the so-called background, consisting of two parts;

a) the photomultiplier background, i.e. the noise of the light-tight closed tube,

b) the background originating in the scintillation medium superposed on the former.

In low-level counting scintillation technics point "a" is one of the main drawbacks, but there are several possibilities to decrease this effect:

- 1) the use of quiet tubes e.g. type EMI "S",
- 2) lowering the temperature to reduce thermionic emission,
- 3) the use of special electronic instruments;

I. coincidence or anticoincidence circuits,

II. pulse amplitude or pulse shape discriminators.

From this short introduction which points out only some of the problems it can be seen that this counting technique gives rise to severe requirements concerning the instruments as well as the selection of the counting parameters, so first of all the examination and comparison of the available instruments were needed.

The aims were: testing the stability of the instruments, photomultiplier qualification, settling the optimal operating conditions, examination of the correctness which may be obtained by determinating the absolute activity of ^{14}C by the extrapolation method. The applied equipments were a scintillation counter of the type SK-2 manufactured in Poland after an EKCO licence and a scaler of the LL-1 Mod. B type, a single-channel counter with a lower level discriminator between 5-50 V. The photomultiplier is of the EMI 9514 S type.

To compare with the data of a Packard Tri-Carb spectrometer model 3003 were investigated, too. This apparatus is a coincidence counter, the multipliers are of type RCA. It is equipped with a triple-channel analyser and with a combination of a lower and upper level discriminator.

2. The high voltage and temperature dependence of the dark current

The stability examinations of the equipments were carried out according to the method published by KESZTHELYI [9]. The following disparity was investigated with minimum 20 scaling

$$\frac{S^2 - \bar{n}}{\bar{n}} < 3 \sqrt{\frac{2}{p-1}} \quad (1)$$

where $S^2 =$ the relative variance

$\bar{n} =$ the average of the scaled impulses,

$p =$ the number of the one minute long scalings.

These series of scalings were repeated several times and the following "k" value was built up:

$$k = \frac{S^2 - n}{\bar{n} \sqrt{\frac{2}{p-1}}} \quad (2)$$

After KESZTHELYI if the $k < 2$, then the equipment can be considered to be good.

After our investigation with the Packard apparatus $k < 0.5$ (of 5 series), but with the Polish apparatus $k_{\max} = 2.6$ (of 3 series). This indicates that the apparatus can be used only if moderate accuracy is required.

As the H. V. adjust is a potentiometer, the high voltage dependence of the counts was measured at various discriminator bias (10, 20, 30 V) using a ^{137}Cs source and a scintillation crystal. The obtained counting rate was plotted against the high voltage.

The deviation of the counts at a given set up is smaller than the standard deviation (in this case: 0.7%). On the other side the obtained averages deviate without the limit of error around the curves. This shows that the applied H. V. setting is not exact, which is in a good agreement with the above mentioned fact i.e. this apparatus can be used only in case of moderate accuracy.

In the literature numerous methods can be found to qualify the photomultiplier [2, 7, 9, 14, 16], but either they do not give a complete picture or the apparatus has to be partly dismantled, or such electronic equipments are required, which cannot be found in every radiochemical laboratory. To avoid these problems we attempted to qualify the photomultiplier by the investigation of its dark noise.

After readjusting the detector for liquid scintillation counting, the high voltage, discriminator level and amplifier gain dependence of the dark noise was examined. At first the tests were carried out at room temperature, because with this equipment we wanted to measure only ^{14}C and ^{35}S , and after the catalogues the EMI "S" tubes are quiet so they can be used also at room temperature. The results can be seen in Fig. 1. Each of the obtained curves could be divided into two linear sections. In the first part the dark noise is max. 100 cpm., and its direction tangent ~ 0 , whereas in the second section the dark noise is greatly influenced by the high voltage. Therefore it is advisable to choose the high voltage setting within the first range.

An increased high voltage has to be used in case of decreased amplifier gain or higher discriminator level. During counting series it was found that

the operating time and the liability of the temperature of the surroundings greatly influenced the reproducibility, and therefore the deviations of the counts were often greater than 10%. Besides it seemed unreasonable that the direction tangents of the last measured series (gain: 250 ×) decreased to $\frac{1}{3}$ of the former ones. For these reasons it became necessary to thermostat the photomultiplier. Because of the small temperature difference we decreased

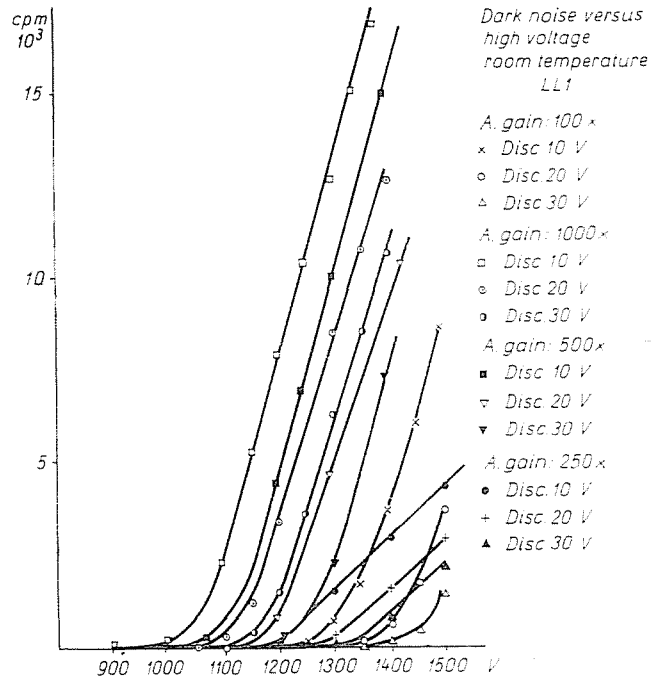


Fig. 1

the heating output of the thermostat by using a toroid transformer to 150–300 W. Thus it was possible to keep the temperature (the minimum value of which was 14 °C) with an accuracy of ± 0.02 °C by a tapwater temperature of 12–13 °C.

Using the thermostat the dark noise was measured in the function of the high voltage, the amplifier gain and the discriminator level at a temperature of 14, 17, 20 and 25 °C (Fig. 2, 3). The shapes of the curves are the same as that of the above mentioned ones, and it became obvious that the direction tangent of the second section is independent of the amplifier gain and of the discriminator voltage at a given temperature, since the adequate values are equal except one within a 5% error-limit. (Table 1)

The first sections are common at all measured temperatures and the origins of the here measured counts are uncertain. (The possible causes can

Table 1
Direction tangents: cpm./V

Temperature °C	Disk. 10 V		Disk. 30 V Gain: 500
	Gain: 500	Gain: 100	
25	250	250	210
20	120	116	122
17	88	92	88
14	74	74	70

be: fluorescence of the glass envelope of the photomultiplier, Cherenkov effect, probably optical or ionic feedback.)

Above a threshold value determined by the scaling parameters the thermionic emission of the photocathode will considerably exceed the precedent effect, i.e. the second section is reached, which is, according to our data, directly proportional to the high voltage and its direction tangent is a function of the temperature. Therefore, this section is characterized by the temperature dependence of the thermionic emission of the photocathode and the high

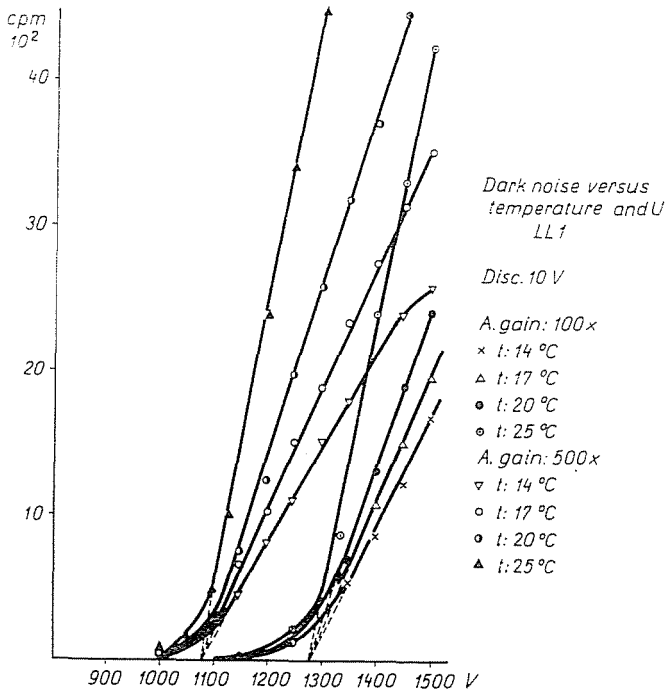


Fig. 2

voltage dependence of the counting rate, which is, according to HARTMANN [7], equal to the high voltage dependence of the average current amplification.

Thus with the determination of these curves the two principal characteristics of the photomultiplier can be calculated.

The temperature dependence of the thermionic emission is described by the Richardson—Dusham equation [15] according to which

$$i_T = A \cdot T^2 \cdot e^{-\frac{W_k}{kT}} \quad (3)$$

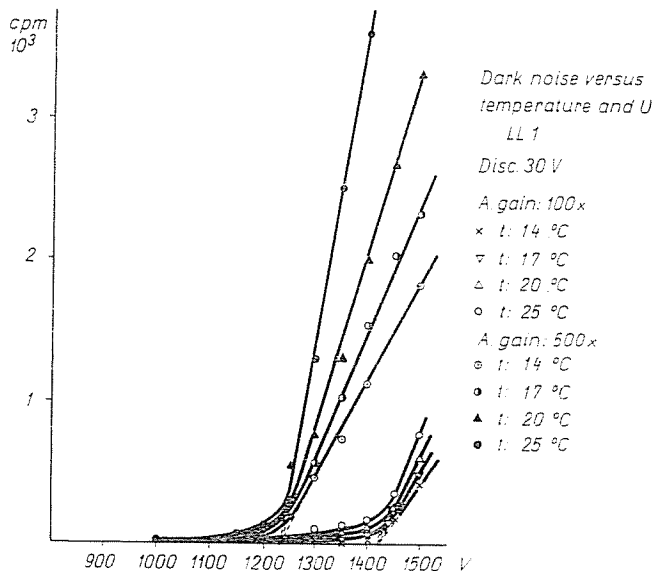


Fig. 3

where: i_T = the total of the emitted electrons,

T = the temperature in K°,

W_k = work function,

k = Boltzmann constant,

A = a constant.

The relation of direction tangents measured at a temperature of 25 °C and at 14 °C did not correspond to the value calculated from (3) when the work function 1–1.3 eV of the alloy (CsSb) was used. (Theoretical value: 4.8–7.3, calculated value: 3.4.) This fact points to the presence of other influencing factors, and at the same time it can be seen too, that the data established in literature according to which the dark noise decreases to its half value at 10–15 °C temperature decrease around room temperature is a rather far-fetched approximation.

It was also investigated in what conditions the Richardson—Dusham equation results are identical data with the above stated experimental ones.

According to our calculations in the case of 1 eV work function substituting $t = 72^\circ\text{C}$ and 61°C , respectively, the relation of the direction tangents is 3.29, in the case of 1.3 eV work function at $t = 119^\circ\text{C}$ and 108°C the relation of the direction tangents is 3.34. Within the limit of error these values are already equal to the experimental data, but such a great temperature difference ($47^\circ\text{C} - 94^\circ\text{C}$) is most unlikely between the photocathode and the cooling agent.

These results indicate, that in this case the use of the Richardson—Dusham formula relating to the thermionic emission is not adequate but the field-emission must be taken into consideration too, as the positive field strength arising on the surface of the cathode furthers the emission of electrons from the cathode material.

At low field strength the increase of the thermionic current due to the effect of the electric field is described by the Schottky formula [15] after which

$$i_{TE} = i_T \cdot e^{\frac{0.44 E}{T}} \quad (4)$$

where: $E =$ the field strength in V/m.

Of course, that formula can be applied only at homogeneous fields, therefore at the cathode-first dynode space it can be only approximately used. At high field strength on account of the tunnel effect the measured currents are by several orders of magnitude higher as the so calculated ones. At planes the thus observed currents can be calculated with the Fowler—Nordheim formula [15].

$$i_h = \frac{6 \cdot 2 \cdot 10^{-6}}{W_B} \left[\frac{W_E}{W_B - W_F} \right]^{1/2} \cdot E^2 \cdot e^{-\frac{6 \cdot 8 \cdot 10^7 (W_B - W_F)^{3/2}}{E}} \quad (5)$$

where:

$W_F \approx W_{F_0}$ the maximal zero energy (eV),

$W_B = W_{F_0} + W_k$ (eV)

and if E is substituted into V/m we obtain i_h in the dimension A/m^2 .

As it is obvious from equation (5) that the cold emission will be significant only above the field strength of 10^7 V/m, we at first considered only the Schottky correction, effective at low field strength. Thus the deviation of the calculated and experimental values decreased only slightly, as the absolute value of the correction factor in our case is ~ 1.15 , and by the calculation of the ratio of the counting rates only the $e^{1/T}$ factor remains, which is in this case ~ 1 .

According to BAY [2, 3] and SIMONYI [15] the above observed deviation can be solely explained by the presence of an additive cold emission current since formula (5) is valid only for homogeneous surfaces and for currents macroscopically measurable. The microscopic unevennesses of the photocathode surface on account of the edges and peaks can also give rise locally to such a field strength increase, that in their surroundings cold emission may occur. On the output of the photomultiplier the impulses produced by those single electrons are also detected. On the basis of this theory we completed the Richardson—Dusham formula with an empirical correction factor.

We intend to stress that the numerical value and shape of this correction is only correct for the given photomultiplier and counting parameters, as we did not take into consideration the field strength dependence of the cold emission, since in the investigated interval its variation was max. 50%.

First it was attempted to define the approximate value of the A constant and W_k , as for the last we could not find accurate data. The value of A must also be determined experimentally, though theoretically it may be calculated, but the data obtained in practice are always smaller by some orders of magnitude.

According to our calculations:

$A \sim 0.1 A/\text{cm}^2 (K^\circ)^2 = 5.3 \cdot 10^{17} \text{ electron} \cdot \text{cm}^{-2} \text{ sec}^{-1} (K^\circ)^{-2}$
while on the other side:

$$A_{\text{theoretical}} = 1.2 \cdot 10^2 A/\text{cm}^2 (K^\circ)^2 = 7.5 \cdot 10^{20} \text{ electron} \cdot \text{cm}^{-2} \text{ sec}^{-1} (K^\circ)^{-2}$$

when

$$W_k = 1.3 \text{ eV}.$$

The motive for higher W_k is that the applied photomultiplier is a special quiet tube which contains a cathode material of lower efficiency i.e. its working function is higher.

Thus the resultant equation is as follows:

$$i = 5.3 \cdot 10^{17} T^2 e^{-\frac{1.5 \cdot 10^4}{T}} + B \quad (6)$$

where the dimension of i : electron/cm² sec.

Calculated from the direction tangents scaled at a temperature of 298 K^o and 287 K^o $B = 1$ electron/cm² sec, and the thermionic emission is 2—5 electron/cm² sec. On the basis of these data taking into consideration that the cathode surface is $\sim 15 \text{ cm}^2$, the measurable counting rate is $3 \cdot 10^3 - 10^4$ cpm.

With the second table we wish to prove the necessity of the correction factor. In the first column the temperature values can be found, in the second

the determined direction tangents, in the third the theoretical ratios and in the fourth the ratios of the direction tangents calculated with the correction factor.

Table 2

Temperature K°	Ratios of direction tangents		
	det. value	value calculated with (3)	value calculated with (6)
$\frac{298}{287}$	3.4	7.2	3.41
$\frac{293}{290}$	1.34	1.78	1.41
$\frac{293}{287}$	1.65	3.1	1.78
$\frac{298}{290}$	2.66	4.23	2.65

We want to stress repeatedly that for an exact discussion of this phenomenon series of measurements are needed in a wide voltage and temperature range. At the same time for the comparison and qualification of the photomultipliers it is sufficient to determine the direction tangents i.e. to characterize the temperature dependence of the dark noise, and this can be carried out without the knowledge of the exact mathematical formula.

As the second section is linear it can be stated besides the above mentioned, that the average current amplification factor of the photomultiplier is directly proportional with the high voltage i.e. the (7) formula

$$M = k \cdot V^z \quad (7)$$

is modified as follows:

$$M = k(V - V')$$

where V' is the intersection on the abscissa axis, V is the high voltage, z and k are constants, and $a = 1$ by the given EMI 9514 S photomultiplier.

Fig. 4 (dark noise versus temperature) shows that though the counting rate increases exponentially with the temperature, operating with a 30 V discriminator level and with 1200 V high voltage the dark noise increase, within this temperature range can be neglected. Therefore it is advisable to decrease it with a high level discrimination as in this case a slight temperature deviation does not influence the precision of the scaled data, and thermostation is not required. So tap water can be used for cooling if its temperature is within 14–25 °C.

On the basis of these discussions it can be stated, that for the qualification of the photomultiplier valuable data can be obtained by determining the characteristic curves (perhaps in a more extensive temperature range) shown in Figs 2, 3 and simultaneously from the same data on the basis of Fig. 4 the optimal temperature interval, the discriminator level and the claim for temperature stability can be stated.

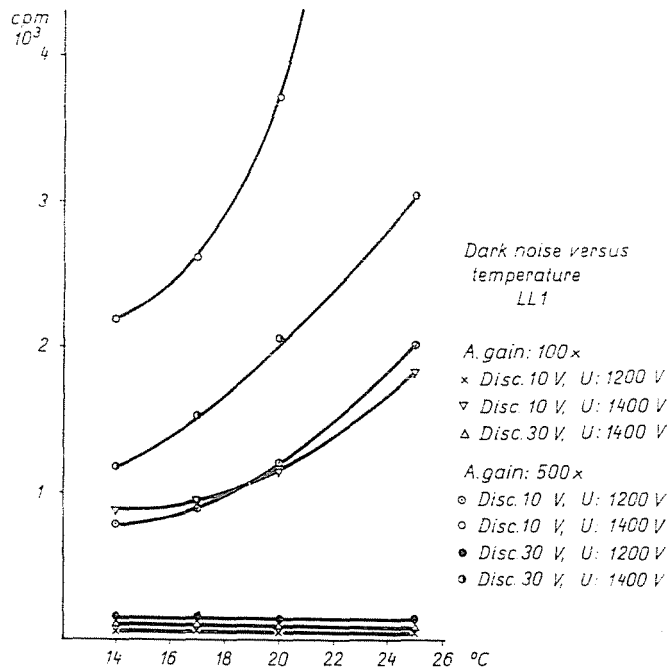


Fig. 4

3. The optimal counting parameters, the determination of the operating point

Several methods can be found in literature to establish the optimal operating conditions [1, 6, 8, 10, 11, 14].

As in the case of liquid scintillation counting, on the curves obtained plotting the counting rate against the high voltage, there cannot be found a well defined plateau, it is necessary to draw up beside the $R_S = f(U)$ also the $R_B = f(U)$ curves (where R_S : counting rate, R_B : background, U : high voltage). The characteristic knee of the R_B curve determines the maximal high voltage to be used.

Often different figures of merit are counted from these scaled data and plotted against the high voltage. Most frequently maximum of the following

curve is chosen as the criterion of the operating point:

$$\frac{(R_S - R_B)^2}{R_B} = f(U) \quad (8)$$

but the following curve can be used too:

$$\text{signal/noise} = f(U)$$

The application of the curve: square of the efficiency divided by the background has been reported, too. The balance point operation — after PACKARD — is also a widespread method [13]. Operating with a differential discriminator of high channel width, the obtained counting rate is plotted against the high voltage and the maximum of this curve is chosen as an operating point. The reasons for this selection are: the maximal sensitivity and a low background at a given discriminator setting. The resultant counting rate deviation of some factors influencing the counting in the sample, in the optical equipment, in the electronics are the lowest there.

The aim of the investigations were: determination of the optimal counting parameters, comparison of the figures of merit found in literature and to lay down their application limits.

We also examined the following curve:

$$\eta = f(U) \quad \eta = \text{efficiency}$$

because with its knowledge the minimal measurable activity (A_{\min}) [5] can be easily determined.

$$A_{\min} = \frac{5.49 \cdot 10^4}{\eta \cdot v} \sqrt{R_B} \mu\text{C/ml} \quad (9)$$

where: v = the volume of the sample in ml.

With the LL-1 equipment the above discussed curves were determined at the temperature of 14 °C with amplifier gain: 100 \times and 500 \times , discriminator level: 10 V, 30 V, with 10 ml "Gamma" scintillator solution (0.4% p-terphenyl, 0.05% POPOP in toluene) and with 0.1 ml standard (^{14}C) toluene (specific activity: $4.7696 \cdot 10^5$ dpm/ml \pm 1.5%).

On the basis of the scaling results (Figs 5, 6, 7, 8, 9) it was found more suitable to use the signal/noise = $f(U)$ and the $\eta = f(U)$ curves simultaneously for the determination of the optimal operating conditions, because

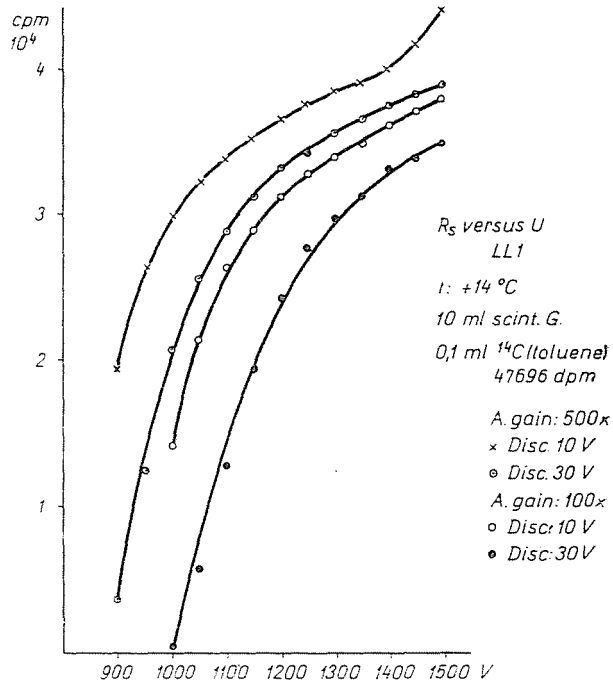


Fig. 5

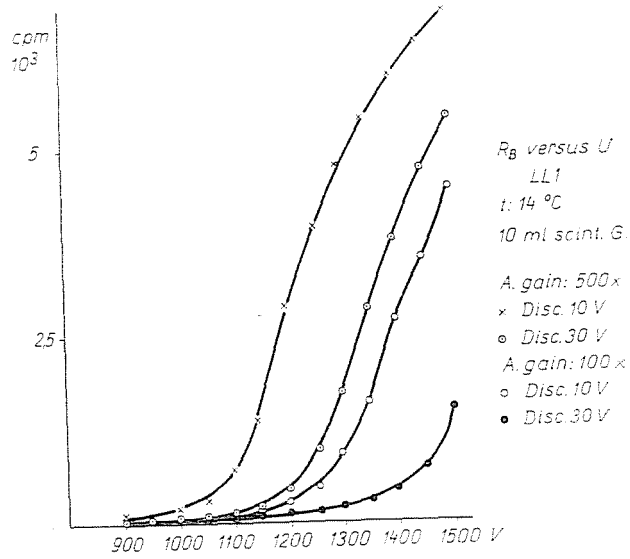


Fig. 6

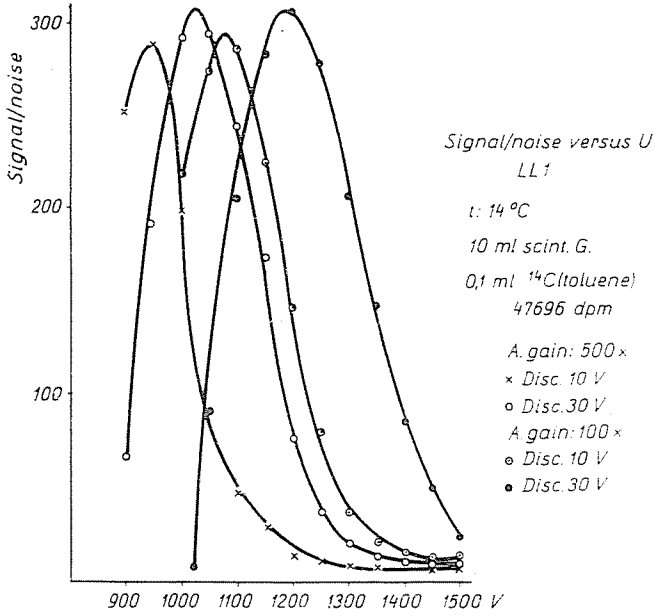


Fig. 7

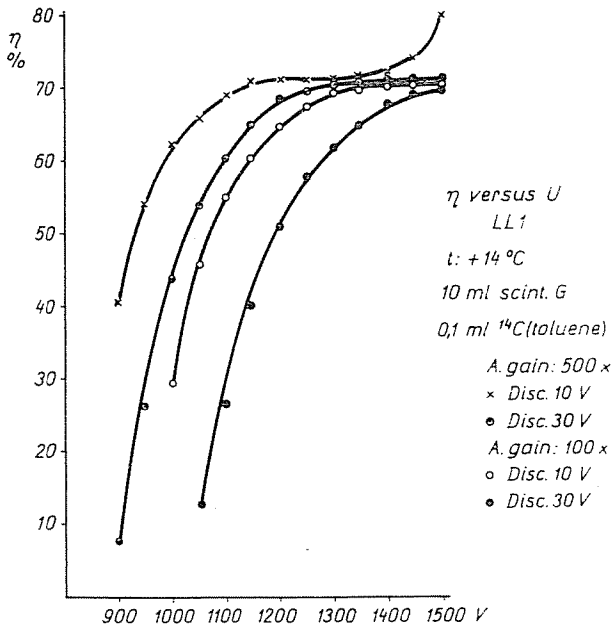


Fig. 8

a) following our approximate calculations if the signal/noise > 100 (but not only around the maximum), it is difficult or impossible to evaluate the $\frac{(R_S - R_B)^2}{R_B} = f(U)$ curve,

b) the above-mentioned two curves are more descriptive and it is more simple to determine on the basis of these curves the smallest measurable activity at a required signal/noise level,

c) at the same time besides the formerly discussed operating point (where the optimum of the signal/noise ratio is the determining factor) a so-called detecting operating point can be determined, where the maximum of the net counting rate i.e. the maximal efficiency is the criterium.

On the basis of these considerations it was found that an amplifier gain: $500 \times$ and a discriminator level of 30 V is the optimal, in spite of the parameters not being better than measured at amplifier gain: $100 \times$ and discriminator level: 30 V. The shape of the maximum of the signal/noise $= f(U)$ and of the

$$\lg \frac{(R_S - R_B)^2}{R_B} = f(U)$$

curves is there even more plain (according to our data the logarithmic scale is more convenient as the shape of the dilating curves become that of a parabola) but with 200 V increased high voltage is needed, which is unfavourable for the stability and the duration of the multiplier tube [7]. Therefore, in spite of the minimal advantages this settling is not favourable.

With amplifier gain: $500 \times$ and discriminator level: 30 V the five characteristic curves are represented in Fig. 10, and thus the obtained high voltage optimum was 1050–1100 V. In this case $\eta = 54\text{--}60\%$, the signal/noise ratio is 300–240 for ^{14}C . The smallest activity to be measured at a signal/noise ratio: 10 is 0.6 $\text{m}\mu\text{C}$.

For comparison similar scalings were also performed with the Packard Tri-Carb apparatus. There was no possibility to modify the high voltage at this equipment, and for this reason the characteristic curves were taken up as a function of the amplifier gain and discriminator level, as after the above-mentioned curves and the data found in literature, the alteration of the amplifier gain and discriminator level could be compensated by the alteration of the high voltage i.e. alteration of the photomultiplier's current amplification.

For scintillator 0.4% PPO and 0.03% dimetil-POPOP was used in (Reanal) toluene. The PPO and the dimetil-POPOP was produced by the Packard firm. The scintillating solutions were stored in brown fluid flasks and they were dosed with brown automatic buret or with a pipetting equipment. No nitrogen bubbling was applied.

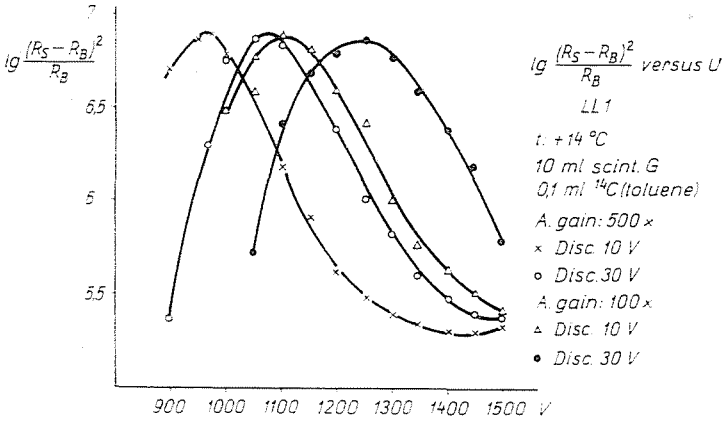


Fig. 9

The following for scaling prepared, sealed standards of the Packard firm were used:

^{14}C : 51900 dpm \pm 1% (toluene)

^{36}Cl : 44800 dpm \pm 1% (Cl-benzene)

As internal standards we partly used the products of the Packard firm and partly of the N. C. A. E. Isotope Institute.

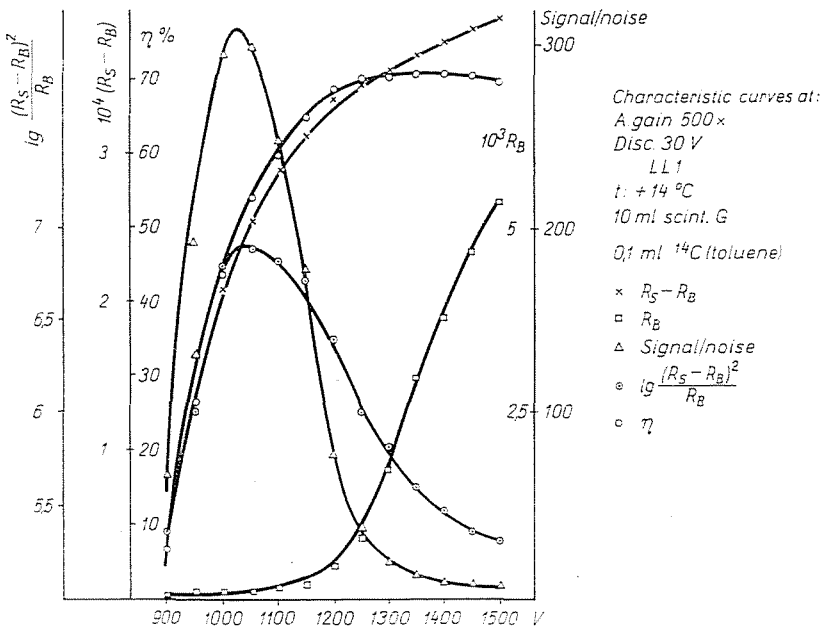


Fig. 10

According to our investigations the p.a. toluene produced by the Reanal firm can be used as scintillation solvent though at the scaling of ^{14}C it causes $\sim 10\%$ decrease in efficiency, and its purity fluctuates, and so difficulties arise especially at the scaling of ^3H , as depending of the quality of the toluene efficiencies between 24–40% were measured. Producing a larger quantity of scintillator solution this failure could be eliminated within a limited time, that is the p.a. toluene could be used with good results without any further purification.

On Fig. 11 the counting rates of the ^{14}C and ^3H samples (0.1 ml standard, 10 ml scintillator) are plotted against the amplifier gain at various discriminator bias.

At discriminator bias $\infty - 50$ the obtained curves are of an integral type, the others are of differential type though at ^{14}C on account of its higher energy this cannot be well perceived.

The monotone decreasing curves of Figs 12, 13, and 14 point to the fact that the balance point determination on the basis of Fig. 11 can also be used in that case when, the maximum of the signal/noise ratio and of the

$$\lg \frac{(R_S - R_B)^2}{R_B}$$

curves (Figs 12, 13, 14) lies outside the efficiency limits, or the curves cannot be estimated on account of their deformed shape. This phenomenon occurs particularly at scaling radiations of small energy, as here the noise cannot be considerably decreased by discrimination. Thus for the scaling of ^3H at the optimal setting the efficiency is 39% at amplifier gain of 40%. The large background and the unfavourable signal/noise ratio is caused by the considerable ^3H contamination of the vials.

It is possible, on account of the difference in the energy of the beta particles of ^3H and ^{14}C to simultaneously assay both of these isotopes with a double-channel apparatus supplied with the combination of a lower and upper level discriminator. In the possession of the above curves we can fix a middle-level discriminator with the aid of which the mixture of ^3H and ^{14}C can be counted.

For example in our case with a 500 middle-level discriminator at an amplifier gain of 60% the lower channel contains only 6% of carbon-14 and the upper channel only 15% of tritium. According to Figs 15 and 16 the efficiency is

$$\begin{aligned} &\text{at } ^{14}\text{C}: 67.5\% \\ &\text{at } ^3\text{H}: 35\% \end{aligned}$$

while the maximal efficiency is 79% and 40%.

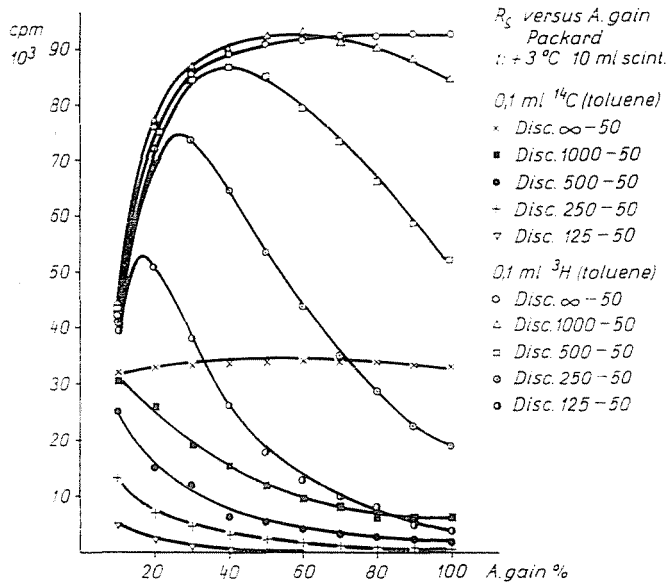


Fig. 11

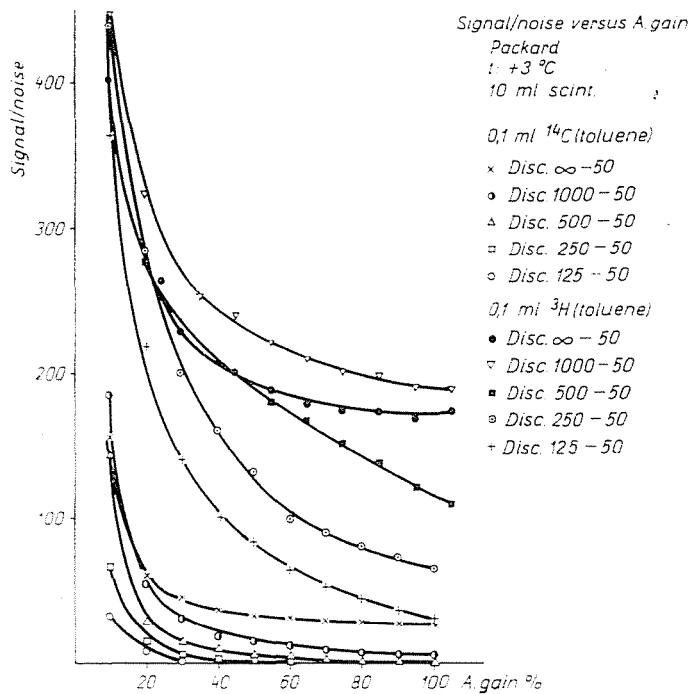


Fig. 12

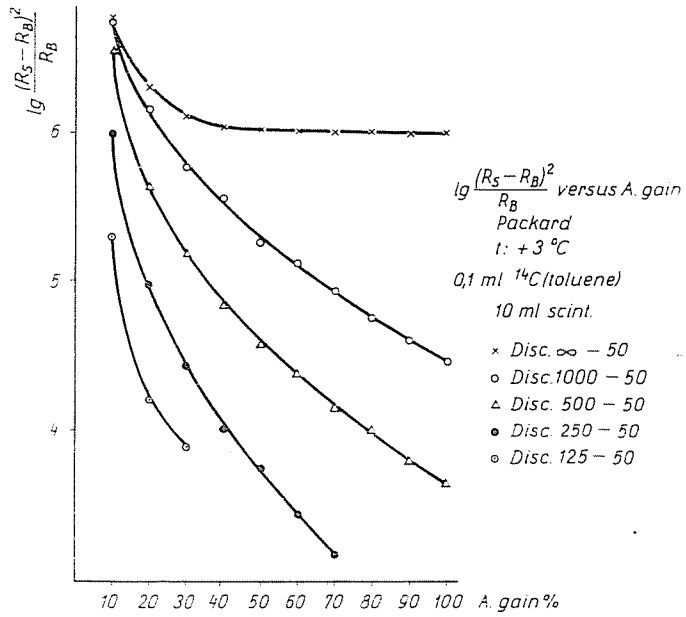


Fig. 13

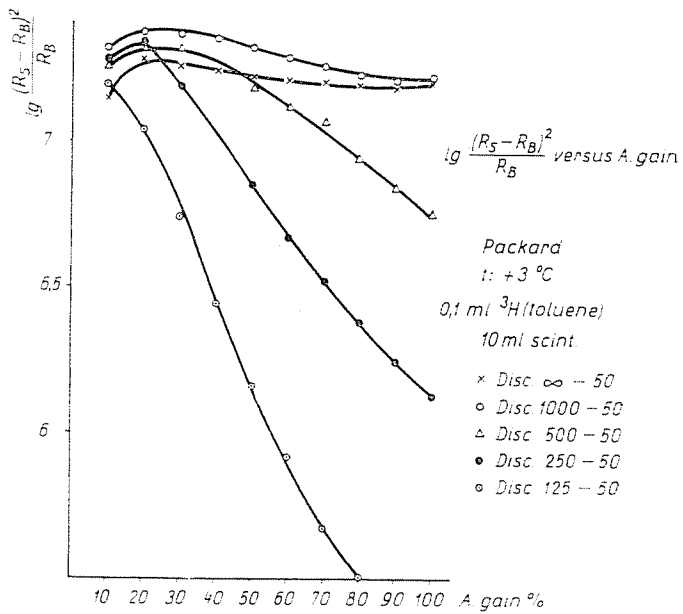


Fig. 14

It is also advantageous to know the efficiency ratio of ^{14}C and the ^3H to be able to select the applied discriminator settings.

It can be seen from Fig. 17 that the application of 60% amplifier gain was correct, because in the first channel:

$$\eta_{^3\text{H}} = 2.5 \eta_{^{14}\text{C}}$$

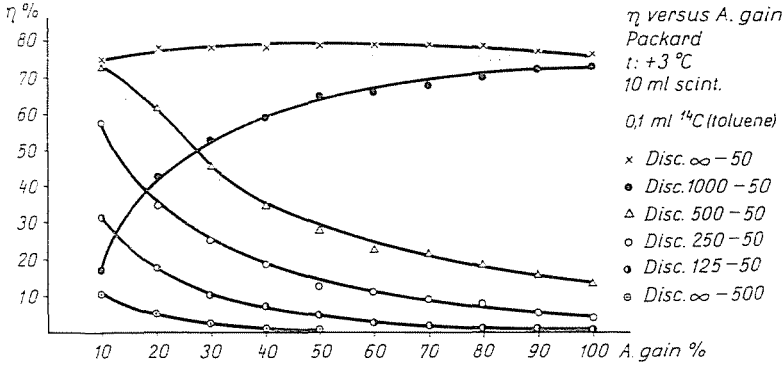


Fig. 15

and in the second channel:

$$\eta_{^{14}\text{C}} = 12 \eta_{^3\text{H}}$$

The efficiency ratio increases with increasing amplifier gain by tritium counting, but rapidly decreases by the counting of carbon-14. If the decrease

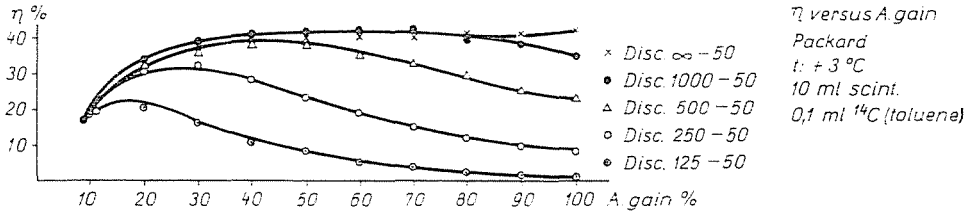


Fig. 16

of the tritium impulses is wanted in the upper channel the middle-level discriminator must be set between 500–1000.

4. Absolute activity determination with extrapolation method

On account of the quenching phenomena, to have a true knowledge of activity it is not sufficient to determine the counting rate of the active sample, but the efficiency has to be determined, too. The efficiency is mostly determined with the internal standard method; the active sample is "spiked"

with a small quantity of a solution of known activity and the increment of counts thus observed is attributed to the added standard. But in some cases, for example: in lack of standard, sealed sample, etc. other methods must be used. It is characteristic for the scintillation counting technics that the obtained pulse heights are proportional with the energy of the particles, and so their energy spectrum can be determined.

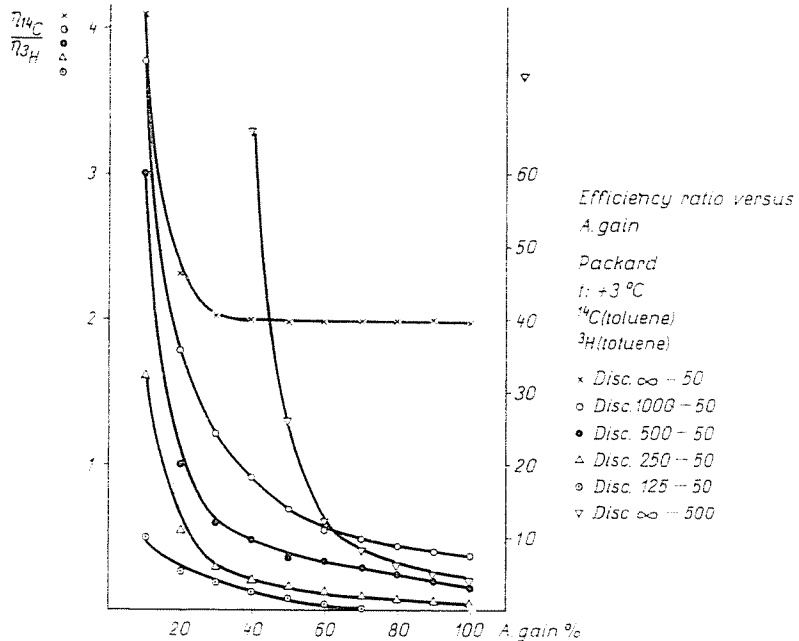


Fig. 17

According to WANGERMANN [17] with the graphical extrapolation of the higher energy range of the integral spectrum to the zero energy, at liquid scintillators, the total activity can be obtained if:

- a) the shape of the spectrum is linear, for example, ^{14}C , ^{198}Au ,
- b) the dark noise of the equipment is minimal (that can be ascertained by cooling or with coincidence circuit),
- c) the integration is carried out with an integral discriminator,
- d) and the 4π geometry is ascertained.

Graphical extrapolation is needed because of the lower threshold of the discriminators and of their inaccuracy at small energies.

In practice depending from the isotopes only 80–90% of the true activity can be obtained by this method [4]. The maximal accuracy is 2–3% but in the case of strong quenching or non-linear spectrum (for example ^{35}S) the error of the method is greater.

The aim of these investigations was to fix the obtainable exactness using this method for the determination of carbon-14, that is, what measure of correction is needed as the data found in literature are not complete and their accuracy is under $\pm 10\%$.

The first investigations were carried out with Packard ^{35}Cl and ^{14}C standards. Various amplifier gains were applied to clear up their influence on the curves i.e. on the results, as well as to make a comparison between the data obtained with carbon-14 and with an isotope of harder beta radiation (Fig. 18).

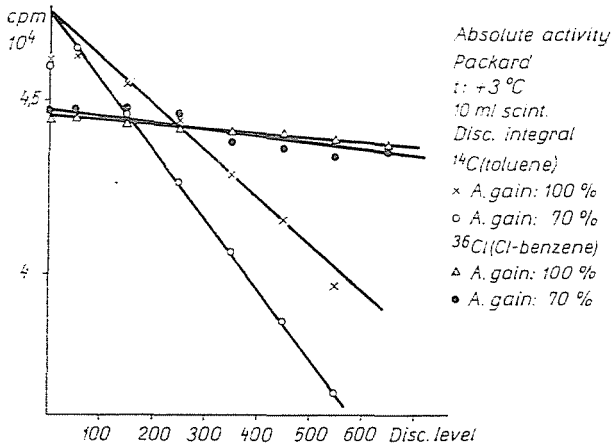


Fig. 18

In the case of Cl-36 the extrapolated values correspond within 0.8%, with the absolute activity, and even at relatively low (70%) amplifier gain, the deviation of the scaled data is within 1.4% as compared to the straight.

Using carbon-14 the extrapolated value is only 92,5% of the true activity.

Samples were scaled made with 2, 5, 10 ml scintillator in order to determine whether the extrapolated value is markedly influenced by the amount of the scintillator (Fig. 19). Owing to the impurities of the applied toluene the extrapolated values are between 76.5% and 75.3%, that is: within the measured range the amount of the scintillator does not influence the results, the maximum and minimum values are between the error limits.

When using 0.3% PPO in toluene as scintillator the result — 76.8% — is between the error limits similarly to the former ones (Fig. 20). On the figure the necessity of the linear extrapolation can be unambiguously seen, because of the spectrum curvature at low discriminator levels.

Though the absence of the POPOP did not modify the extrapolated value, the more rapidly declining curve indicates, that the pulse heights are lower i.e. the efficiency is smaller.

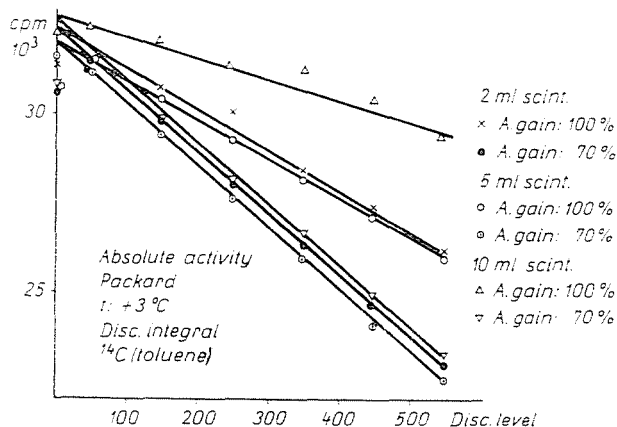


Fig. 19

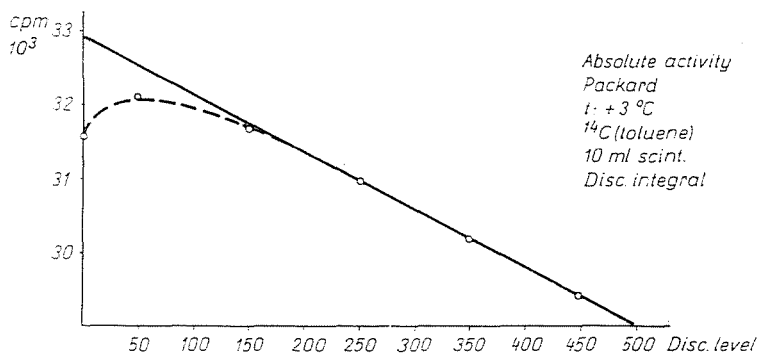


Fig. 20

According to these results we may say that at optimal working conditions (using chemicals of scintillator grade) in the case of carbon-14 the extrapolated value is about 92% of true activity, and within limits it is not influenced by the amplifier gain, by the amount of the scintillator and wavelength shifter. With the available scintillators the extrapolated values are between 75–77%, that is, we obtained $76\% \pm 1\%$ of the absolute activity.

Working with the Polish apparatus (10 ml Gamma scintillator and 0.1 ml active standard) using amplifier gains of $100\times$ and $500\times$ and various high voltage settings near the operating point the extrapolated values deviate around 73.2% (34900 cpm) within $\pm 1\%$ (Fig. 21).

Regarding that — operating with different instruments, with different scintillators and with different quenching — the extrapolated values are

between 72.5—77% with carbon-14, we found it possible in this way to determine the true activity, when we obtained $75\% \pm 2\%$ of the absolute activity, that is, the following correction must be applied:

$$A = \frac{A_{\text{ext}}}{0.75} \quad (10)$$

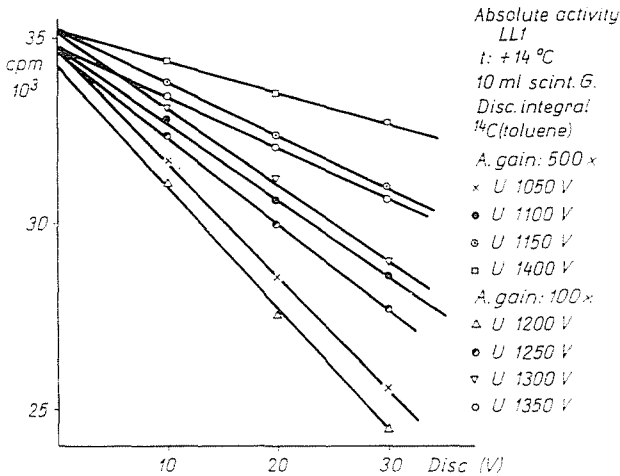


Fig. 21

5. Summary

In our investigations we determined first the high voltage, and temperature dependence of the photomultiplier.

We established that the so obtained curves can be divided into two linear sections, and the bend of the curve between them indicates the maximal high voltage to be used. The position of this knee is influenced by other counting parameters (amplifier gain, discriminator level) but the second section has always a linear shape, i.e. in our case the average current amplification factor is directly proportional with the high voltage, and the direction tangent of the straight line is an unambiguous function of the temperature. Thus the temperature dependence of the dark noise and the high voltage dependence of the average current multiplying factor, these two significant data of the photomultiplier can be measured without any special auxiliary instruments. The importance of this method is, that in the catalogues only approximate data can be found, the exactness of which are often not sufficient for using as selection criteria.

According to our scalings besides the Richardson—Dusham formula concerning the thermionic emission, a cold emission current should be taken into consideration too, to describe the temperature dependence of the dark noise. We reported its numerical value, but our data are not sufficient to set up an equation of universal validity, because of the small temperature and high voltage interval.

On the basis of our results related to the determination of the operating point, it can be fixed most rapidly by the simultaneous consideration of the signal/noise $= f(U)$ and $\eta = f(U)$ curves, and at the same time, these curves are giving overall and descriptive informations on the photomultiplier's operating area, which thus give a possibility to mark out "irregular" operating points for scales of special requirements.

According to the results obtained with the extrapolation method for absolute activity determination, at optimal counting parameters the extrapolated values are 92% of the real activity.

Using commercial pro anal. toluene the extrapolated values are $75\% \pm 2\%$ of the absolute activity with both the Packard, and the "Polish" apparatus, that is, the application of correction formula (10) is needed.

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Pro spectuses

- I. Z. W. E. "Eltra" Bydgoszcz scaler Typ. LL—1 Mod. B, time switch Typ. PS—1 Mod. B
- II. Scintillation counter Typ. SK—2 (The Research Institute of Nuclear Instruments Poland).
- III. Packard Tri-Carb Liquid Scintillation Spectrometers, Packard Instrument Company Inc Downers Grove, Illinois U.S.A.

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