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W. G. VERLY, S. BRICTEUX-GRÉGOIRE, G. KOCH AND E. DEMEY

Laboratoire des Isotopes des Services de Pathologie Générale et de Biochimie, Service de Chimie Médicale et Clinique Médicale A, Université de Liége

Reprinted from

UNITED NATIONS
PEACEFUL USES OF ATOMIC ENERGY

PROCEEDINGS OF THE SECOND INTERNATIONAL CONFERENCE, GENEVA, SEPTEMBER 1958

PERGAMON PRESS
LONDON . NEW YORK . PARIS
LOS ANGELES
1959

Reprint from 2nd UN Geneva Conference.
Printed by Pergamon Press, London.

Windowless Flow Geiger Counters and Measurement of the Soft Beta Radioactivity of Thick Solid Samples

By W. G. Verly, S. Bricteux-Grégoire, G. Koch and E. Demey*

The experiments described in this paper were performed with a Tracerlab SC-16 windowless flow counter operating in the Geiger region with a gas composed of 99% helium and 1% isobutane. Since 1952 we have observed, when measuring the C^{14} in a deposit of barium carbonate with this apparatus, that the variability of the results exceeded the statistical error to be expected in nuclear disintegration. Tracerlab, Inc. of Boston, USA, consulted in 1954, informed us that, in their opinion, the difficulty was due to static electricity which formed on the sample; to eliminate this electricity, they advised placing the barium carbonate directly on a conducting support and covering it with a fine conducting film. The first remedy proving insufficient and the second introducing an additional source of error, we preferred to keep the deposits on filter paper and perform the measurements with a Geiger tube having a thin mica window.

The problem of the gas-flow counter arose again when we wished to measure the radioactivity of solid samples tagged with tritium. The weakness of the radiation of this isotope ($E_{max} = 18$ kev) made it impossible to use a tube with a mica window. It was necessary, as Eidinoff and Knoll[1] and Banks *et al.*[2] found, to use a windowless counter. The method, however, did not seem usable for nonconductors. Banks *et al.*[2] seem to have solved the difficulty by mixing the samples with graphite, thus making them conducting.

The anomalies observed in the measurement of C^{14} with a windowless counter are greatly exaggerated for tritium, since the radiation of the latter radioisotope is much weaker. Tritium showed us that when a thick, nonconducting soft-beta-ray emitter was placed in our gas-flow counter, the main aberration was a progressive fall in the apparent radioactivity until equilibrium was established.

EXPERIMENTS AND DISCUSSION

The Tracerlab SC-16 counter is provided with a platform rotating about a vertical axis and containing three small recesses; one communicates with the exterior, the second completes the cathode of the detector tube and the third occupies an intermediate position. The sample, on an aluminum support, is put into the access hole; by a 120 deg turn the sample is turned to the intermediate or "preflushing" position where air is replaced by the counting gas; a second 120 deg turn places it under the anode of the counter tube which is at a potential of 1300 volts.

Experiment 1. A deposit of barium carbonate–C^{14}, 22.6 mg/cm² thick, was laid down on SS 589/3 filter paper. It was then placed on the aluminum support and, after the air had been replaced by the Geiger gas, was put under the anode; counting was begun immediately. Figure 1 shows the apparent variation of the activity of the deposit as a function of time. The ordinates are expressed in both counts per minute and percent (the mean activity measured during the first time interval is arbitrarily taken as 100%). The standard error ($N^{\frac{1}{2}}$) for each measurement is shown on the graph.

Experiment 2. By centrifuging, a layer of barium carbonate–C^{14} (22.3 mg/cm²) was deposited on an aluminum support. Figure 2 shows the apparent variation of the activity as a function of time.

Experiment 3. N-α-naphthylacetamide tagged with tritium was deposited by filtration on a filter paper (SS 589/3); the deposit was 3.26 mg/cm² thick. The apparent variation of activity as a function of time is given in Fig. 3.

Experiment 4. Figure 4 gives the results of measurements made on an 18.3-mg/cm²-thick deposit of N-α-naphthylacetamide tagged with tritium placed directly on a metallic disk. In all cases, a diminution of apparent activity with time was observed, which finally leveled off after 30–60 min. This was true for both C^{14} and tritium, whether the deposits were made on a metallic support or on filter paper.

It seemed that the diminution of apparent activity was due to a potential barrier which grew for a certain time before attaining an equilibrium value (V_{max}). The origin of this static electricity could be attributed to the nuclear disintegrations (electrons leaving the deposit, causing it to acquire a positive charge) and to the positive ions deposited on the sample with each discharge of the Geiger tube.

If the deposit was in the atmosphere, its surface

Original language: French.

* Laboratoire des Isotopes des Services de Pathologie Générale et de Biochimie, Service de Chimie Médicale et Clinique Médicale A, Université de Liège.

would be discharged by the ions produced by the beta radiation. But since it is in the counter tube, the anode attracts the negative charges, which are then no longer capable of neutralizing the positive charge on the surface of the deposit. This positive charge ought to discharge the cathode, which is grounded, but it is prevented from doing so by the high resistance of the sample or the filter paper. It is the transition from electrical equilibrium in air to electrical equilibrium in the counter tube which is observed during the first 30–60 min of counting.

The situation may be described as follows: Let I = the number of charges which appear on (or in) the deposit per unit time (expressed in amperes); I is proportional to the number of electrons emitted per unit time and hence is a measure (uncorrected for self-absorption) of the radioactivity of the sample; let R = the resistance to the flow of the charges to the cathode. When the deposit is made on filter paper, the resistance has two components; one due to the filter and the other to the sample itself. Equilibrium is reached when the current passing through R is equal to I. Hence the equilibrium potential $V_{max} = IR$.

For a given radioisotope the percentage of electrons counted, and hence the apparent activity, should decrease as the potential barrier increases. On the other hand, a given potential barrier will have a much greater effect if the energy of the impinging electrons is smaller. Comparison of the results of experiments 1–4 shows that the percentage fall in the apparent activity is much greater for tritium ($E_{max} = 18$ kev) than for C^{14} ($E_{max} = 155$ kev).

Since, according to our hypothesis, the percentage fall of the apparent radioactivity between the moment that the sample is introduced into the counter tube and the moment that the new equilibrium is reached is a function of V_{max}, it should also depend on I and R. We have been able to establish this for both C^{14} and tritium.

Experiment 5. A sample of barium carbonate mounted directly on aluminum foil gave 23,000 counts/min when equilibrium was reached. When a filter paper (SS 589/3) was slid under the aluminum foil, the apparent activity dropped to 18,400 counts/min. This agrees with our hypothesis because, R being increased, the potential barrier is higher when the filter paper is interposed.

Experiment 6. A sample of tritiated naphthylacetamide was placed directly on a metallic support and counted, and then placed on a filter paper and counted again. The apparent activity at equilibrium, which had been 6900 counts/min in the first case, fell to 3400 counts/min when the sample was placed on the filter paper.

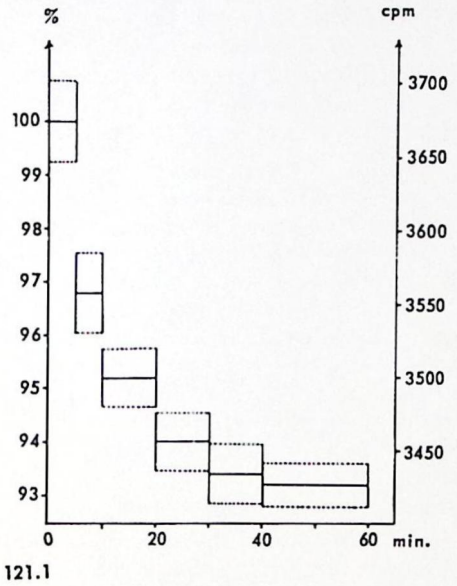

Figure 1

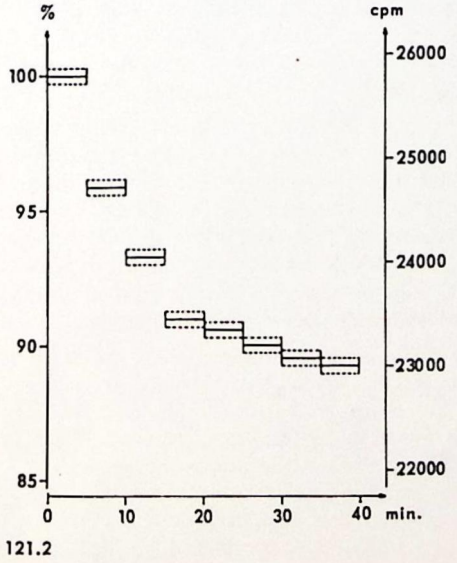

Figure 2

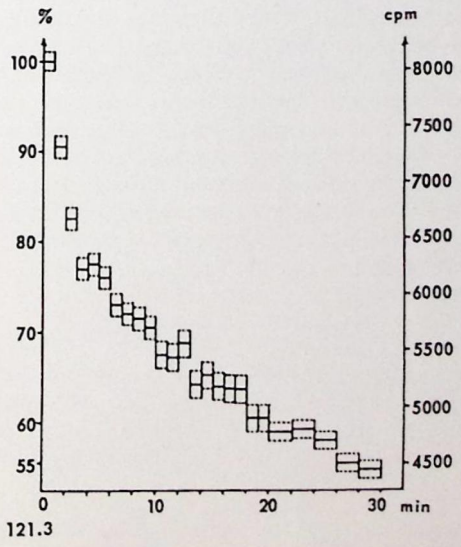

Figure 3

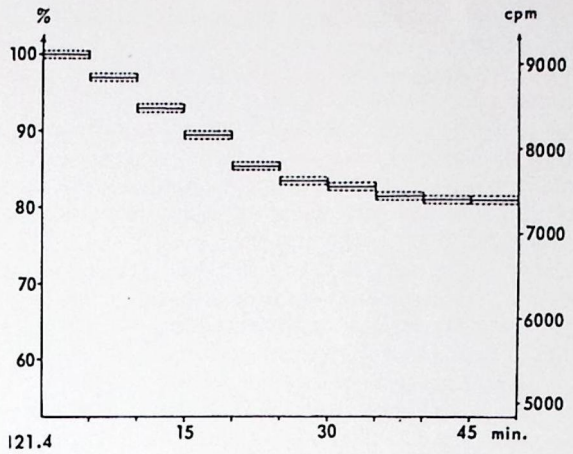

Figure 4

Experiment 7. Three samples of tritiated naphthylacetamide deposited on filter paper were used for the experiment. After 30 min of counting, the first sample showed a drop of 44% in apparent activity. The second sample, which had been prepared from the same tritiated naphthylacetamide diluted about 10-fold with untagged naphthylacetamide, showed a decrease of 35% in apparent activity after 30 min of counting. With the third sample, prepared from tritiated naphthylacetamide diluted 100-fold, the decrease in the same time was only 28% (Table 1). The effect of R on relative measurements of radioactivity can be eliminated by comparing only the equilibrium activities of deposits of the same substance having the same thickness and placed on identical supports. However, it is the effect of I on V_{max} and hence on the percentage of electrons that escape counting which is the principal objection to the gas-flow counter for precise measurements of the radioactivity of thick nonconducting samples emitting soft beta rays. In Table 1 the ratio of the activities measured during the first and the thirtieth minute for tritiated naphthylacetamide samples is compared with the theoretical values known from the dilution; it will be seen that the experimental ratios are wrong from the very first minute, that they vary with time and that they are lower than the true values. But it is ratios of this sort which form the relative measurements for which the counter is used.

If our hypothesis is correct, there should be no decrease in the apparent radioactivity with time after the sample is introduced in the counter tube if $R=0$. Experimentally, no significant variation in time of the apparent radioactivity is observed for deposits of

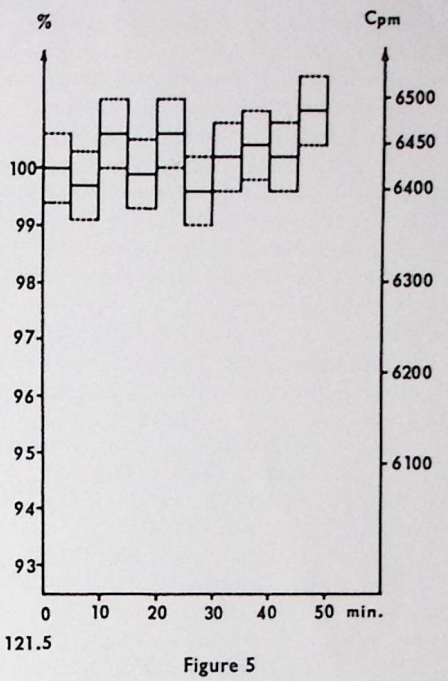

Figure 5

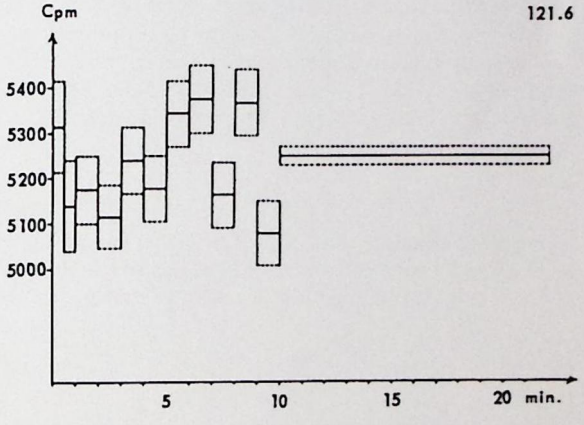

Figure 6

Fe^{59} on copper, for very thin deposits of tritiated naphthylacetamide on a metallic support or for a pellet of barium carbonate–C^{14} covered with a thin conducting coating.

Experiment 8. One part of the beta radiation emitted by Fe^{59} is due to relatively low energy electrons ($E_{max}=271$ kev). Figure 5 shows that the apparent activity of a sample of Fe^{59} deposited electrolytically on a copper disk does not change significantly during

Table 1

	Counts/min			*I/II*	*I/III*
	I	*II*	*III*		
First min.	8056 ± 90	943 ± 31	173 ± 13	8.5 ± 0.3	46.6 ± 4.0
30th min	4508 ± 67	610 ± 25	124 ± 8	7.4 ± 0.4	36.4 ± 2.8
Ratio by dilution				10.68	99.88

the 50 min after the introduction of the sample into the counter.

Experiment 9. Ten micrograms of tritiated naphthylacetamide was spread over an aluminum support with a surface area of 4.54 cm^2. The results of the radioactivity measurements made on this preparation are shown in Fig. 6. It will be seen that, contrary to what occurred in experiments 3 and 4, the apparent radioactivity does not vary during the first few minutes after the introduction of the tagged product into the counter tube.

Experiment 10. A pellet of barium carbonate–C^{14} placed on a metallic support was covered with a thin sheet of aluminum in peripheral contact with the support. The radioactivity of this sample, measured in our gas-flow counter, did not vary significantly during the first few minutes after its introduction into the counter tube. This experiment shows that when the pellet of barium carbonate is enveloped by a conducting substance, the equilibrium of the electric charge flux is not affected by the introduction of the sample into the windowless counter tube.

It was said in the introduction that the counting results for the same sample of barium carbonate were very irregular when the gas-flow counter was used. The experiments described above show that the counting rate is affected by the time elapsing between the moment when the sample is placed under the anode, which is at a potential of 1300 volts, and the moment at which the counting begins, as well as by the duration of the counting.

COUNTING

A gas-flow counter of the Tracerlab SC-16 type gives erroneous measurements of the relative activities of thick nonconducting samples which emit soft beta rays. The error is greater if the deposit is placed on a nonconducting support, which increases the resistance to the discharge of the static electricity that appears in the sample while it is in the counter tube. The error may be as much as 5–10% for C^{14} counted in the form of barium carbonate. Hence the method may still be used with this isotope if the measurement does not need to be very accurate. The error becomes intolerable in the case of tritium.

It is not certain that covering the nonconducting deposit with a conducting film is a completely adequate remedy, even though it stabilizes the counting rate. Moreover, it introduces an additional error in the estimation of the fraction of the radiation retained by this screen, and it cannot be used for tritium unless the film is extremely thin (several micrograms per square centimeter).

Only a conducting deposit gives completely accurate results. Banks *et al.*[2] mix their tritiated samples with graphite. For C^{14}, elementary carbon could be used.

It seems also that very thin deposits, with negligible resistance to the discharge of the electricity, could be used. But this method is not very sensitive when the specific activity of the products is small. With tritium the results contain a large error. Since the energy of the beta radiation of tritium is very weak, and since the deposit is not infinitely thin and has irregularities, part of the disintegration electrons, varying from deposit to deposit even where the weight of the latter is constant, are absorbed by the product itself.

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