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THE MULTISTEP AVALANCHE CHAMBER AS A DETECTOR IN RADIOCHROMATOGRAPHY IMAGING

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ABSTRACT

We have investigated the possibility of using the multistep avalanche chamber as in imaging detector in radiochromatography. High position resolution, better than 0.5 mm FWHM, combined with a high sensitivity give very satisfactory results. Measurements on two-dimensional distributions of β -emitting radionuclides such as ^3H , ^{14}C , and ^{35}S are presented and compared with results obtained from autoradiography.

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1. INTRODUCTION

Thin layer chromatography using β -ray emitting radionuclides as tracers is a powerful tool in biophysics, biochemistry, pharmacology, and clinical medicine, for the analytical separation of chemical compounds. Two-dimensional distributions of the compounds can be created by employing consecutively two different principles of chromatography or electrophoresis combined with chromatography on the same plate1). Commonly used tracers are 3H, 14C, 32P, 35S, and 131I. The different characteristics of the β -emission spectra of these isotopes may necessitate the use of different techniques in their detection. The classical method consists in the exposure of the chromatogram plate to an X-ray film (autoradiography). Compounds labelled with 32P or 131I often yield satisfactory pictures within a couple of hours, but for low intensity samples, especially those labelled with 3H, the exposure time may extend to several weeks. In the case of ³H the sensitivity of the method is often enhanced by adding a small amount of fluorescing component to the solvent used. Among other detection methods can be mentioned liquid scintillation counting and scanning of the chromatogram with one or several Geiger-Müller tubes in a row2). Both of these methods can be used in a quantitative determination of the amount of a certain compound on the chromatogram plate, but they are rather time consuming; a scan over a 20 × 20 cm² chromatogram takes typically 24 h. Spark chambers3, and hybrid spark chambers4, have been constructed as detectors for radiochromatography. As they use optical read-out with registration on film, the above-mentioned quantitative measurements are not easy to perform with these detectors. We have approached the problem in a different way by using the multistep avalanche mechanism in a gaseous detector, recently investigated in our group at CERN, as the basis for detection of the \beta-rays emitted from the surface of the chromatogram plate. We have used a purely electronical read-out of the detector with subsequent data handling done in a small computer.

2. THE MULTISTEP AVALANCHE CHAMBER

2.1 The photon mediated avalanche

It has been shown^{5,6)}, that with a proper choice of gas mixture A + B it is possible to develop an avalanche between parallel grids with the following characteristics: the lateral spread of the avalanche is such that a sizeable fraction of the resulting electron cloud can be extracted into a following transfer space. The transferred fraction is nearly independent of the position of the electron initiating the avalanche. This permits proportionality between the initial ionization and the final transferred charge and avoids modulation of the efficiency as a function of position.

It has been inferred that the emission and absorption of VUV photons play a role in the spread of the avalanche; however other processes, such as electron diffusion and the Penning effect, may also participate.

2.2 The localization effect

The charge multiplication that an electron in the preamplification stage gives rise to varies exponentially with its distance from the second preamplification grid. An electron cloud, obtained by preamplification and transfer of electrons in the ionization trail through the preamplification region, will therefore mainly localize the point where the ionizing particle crossed the first preamplification grid. The localization effect can easily be treated theoretically. Assuming infinitely long ionization tracks emitted uniformly in space from a point on the first preamplification grid one gets the following expression for the centre of gravity distribution of preamplified and transferred electron clouds

$$I(z) \simeq \frac{1}{1 + \left(\frac{\ln M}{\lambda} z\right)^2}$$

where ℓ is the distance between preamplification grids, M is the multiplication that an electron obtains over the distance ℓ , and z is the coordinate in a direction parallel to the grids. The formula is accurate within a few per cent for values of M > 50. In the case of no multiplication the expression is modified to

$$I(z) = \frac{1}{1 + \left(\frac{2}{\chi} z\right)^2} .$$

Figure 1 shows I(z) for $\ell=3$ mm and different values of M. In the case of M = 2000 we expect a position accuracy of about 1 mm FWHM.

In practice the ionization tracks that a β -emitting isotope gives rise to are neither straight lines nor infinitely long. A large amount of the tracks have a very limited range, less than a few hundred μm , in the detector gas thus enhancing the final position accuracy of the detector. This effect is specially pronounced in the case of 3H . For all the isotopes that we have used, a position resolution better than 0.5 mm FWHM was obtained.

3. EXPERIMENTAL SET-UP

The arrangement of wire planes in the detector is shown in Fig. 2. Two electrodes have been mounted on top of a standard multiwire proportional chamber (MWPC) to provide the preamplification and transfer regions. The upper preamplification electrode is a mesh of stainless steel wires, 30 μm in diameter and 400-600 μm apart. The lower electrode is a mesh of Ni wires, 10 µm in diameter and 100 µm apart. The distance between the two electrodes is 3 mm, and the distance between the lower mesh and the MWPC is 5 mm. The sample is mounted inside the detector, as close as possible to the upper preamplification electrode. The MWPC and associated electronics have been described elsewhere 7). Briefly, the anode plane is made of gold-plated tungsten wires, 10 μm in diameter and 1.27 mm apart. The two orthogonal cathode planes are made of Cu-Be wires, 100 µm in diameter and 1.27 mm apart. The anode-cathode distance is 5 mm, and the active area of the detector is $10 \times 10 \text{ cm}^2$. The bidimensional read-out of the MWPC is done with the centre-ofgravity method where, for each avalanche occurring on the anode, one records the positive induced charge distributions on the two cathode planes. The cathode wires are connected together 4 by 4 in strips, and each strip is connected to a charge preamplifier having a sensitivity of about 250 mV/pC, a rise-time of 10 ns, and a decay time of 50 ns*; the amplified signals are then transmitted via coaxial cables to a CAMAC-based 10-bit current integrating ADC**).

^{*)} Developed at CERN by J.C. Santiard and produced with a hybrid thick film technique by CIT-Alcatel, France.

^{**)} Lecroy, 12-channel ADC type 2249 A.

All anode wires are connected together; the signal from this plane is recorded in an ADC channel and provides also the gate for the ADC modules after a suitable discrimination and shaping. The gate width is 100 ns. A small on-line computer is used to organize the data acquisition and transfer to magnetic tape.

As a gas filling we have used a mixture of argon and acetone in the volume concentrations 98-2.

4. CALIBRATION AND DATA HANDLING

A detailed description of the procedure used to calibrate the chains of the preamplifier, coaxial cable, and ADC channel, is given in Ref. 7. The calibration allows us in software to correct for non-linearities in the chains as well as for dispersions between them.

Let us denote by \mathbf{q}_i the effective charge signal on strip \mathbf{S}_i , as obtained from the raw data using the above-mentioned calibration procedure. We define the centre of gravity of a cluster of adjacent induced charges as the quantity:

$$\bar{S} = \sum_{i} (q_i - b) S_i / \sum_{i} (q_i - b)$$
,

where b is a bias level, and the sum extends only to positive values of \mathbf{q}_i - b. A proper choice of bias level allows reduction of the influence of pick-up and electronics noise in the centre-of-gravity determination. In our case a value of b = 2% of $\Sigma \mathbf{q}_i$ gave satisfying results.

5. EXPERIMENTAL RESULTS

5.1 Introduction

We have made measurements on different samples marked with ³H, ¹⁴C, and ³⁵S. In the case of ³H and ¹⁴C, silica gels on glass plates were used as chromatogram plates; for ³⁵S a polyacryl-amide gel was used. It is important that the chromatogram plates be flat, so as to keep the distance to the upper preamplification electrode as small as possible over the whole surface of the sample. Silica gels on glass plates seem to be perfectly suited for our detector in that respect.

5.2 14C results

The 14C has an emission spectrum extending to about 150 keV, implying that most of the β particles have a range of many millimetres in the detector gas. The half-life of the isotope is 5600 y. The total activity of our sample was about 90,000 disintegrations per minute (DPM). We have measured the counting rate in the detector for different values of anode voltage and preamplification voltage. The result is shown in Fig. 3. It is seen that for high values of the preamplification voltage, we obtain the beginning of a plateau. A full plateau cannot be reached in our case, because the preamplification mechanism is not efficient enough to bring the lowest part of the β-emission spectrum over the electronic threshold. The distribution of regions with high activity of the sample is shown in Fig. 4. The upper part of the figure is the autoradiogram that was obtained after 48 h of exposure time; the lower part shows the response of our detector after about 20 min of data taking. The scales are 25 mm in both directions. The lower left corners of the pictures cannot be compared directly, because the most intense part of the sample was removed before placing it in the detector. Figure 5 is a comparison between a photometer scan of the autoradiogram (top) and the lower part of Fig. 4 projected on to the vertical axis. The resolutions of the two methods are comparable.

5.3 ³H results

The emission spectrum of 3H extends to about 18 keV, implying ranges of a few hundred μm for a considerable part of the β -rays. The half-life of the isotope is 12.3 y. The sample activity was about 600,000 DPM. After an exposure time of 48 h, the autoradiogram shows only the most intense line, see Fig. 6. The response of our detector after 5 s and 10 min of data taking, respectively, is shown in Fig. 7. The scales are 70 mm in both directions. In this case our method is indeed several orders of magnitude more sensitive than the autoradiography.

5.4 35 results

The emission spectrum of this isotope is similar to that of ¹⁴C, but the half-life is much shorter, i.e. 87 d. The upper part of Fig. 8 is the autoradiogram after 4 d of exposure time; the lower part is the response of our detector

after a few minutes data taking. The scales are 70 mm in both directions. In this case we do not resolve very well the detailed structure seen in the autoradiogram; after a few minutes more data taking a large part of the picture will be overexposed. We do not consider this as a drawback of the detector itself. The problem of creating good pictures can easily be solved using a more refined software, and grey-tone or colour display screen.

6. CONCLUSIONS

The multistep avalanche chamber seems to be a very useful tool in radiochromatography imaging. Compared with the traditional method of autoradiography it is extremely sensitive, especially in the case of ³H measurements. In a fast way it provides information about the two-dimensional distributions as well as intensities of the radionuclides. The method requires completely flat samples, but this is not considered as a serious problem; it is probably possible in many cases to adhere gels to support plates so as to obtain the desired geometrical tolerances.

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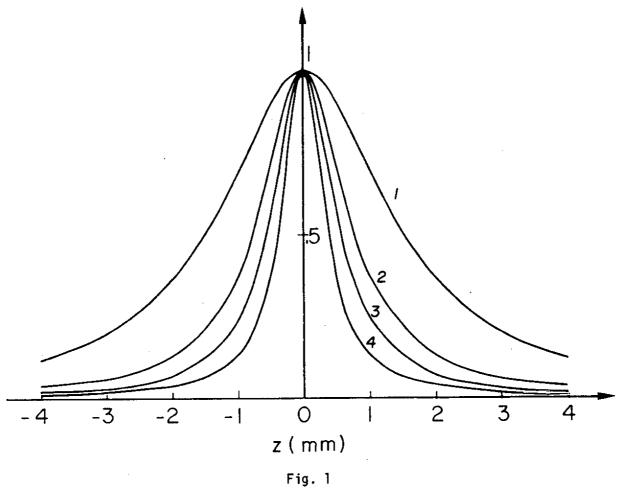
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Figure captions

- Fig. 1: Theoretical distributions of the centre of gravity of preamplified and transferred electron clouds for different values of the amplification factor M. The curves 1 to 4 correspond to the M values 1, 50, 200, and 2000.
- Fig. 2: Schematical view of the detector. F and G are mylar windows; A and B are the preamplification electrodes; C, D, and E constitute a standard MWPC. H is the sample, mounted inside the detector on top of the upper preamplification electrode.
- Fig. 3: Counting rates obtained with the ¹⁴C sample for different voltages on the chamber. The curves I, II, and III correspond to the anode voltages of 2.25 kV, 2.35 kV, and 2.45 kV, respectively.
- Fig. 4: Comparison between an autoradiogram of the ¹⁴C sample and the response of our detector. The scales are 25 mm.
- Fig. 5: For ¹⁴C: comparison between a photometer scan of the autoradiogram and the response of our detector. The resolutions are comparable, better than 0.5 mm FWHM. The most intense line of the sample was removed before mounting it in the detector. This cuts the high peak as shown on the top curve.
- Fig. 6: The autoradiogram of the ³H sample after 48 h of exposure time. Only the most intense line is visible.
- Fig. 7: The response of our detector for the ³H sample for 5 s and 10 min of data taking. The scales are 70 mm.
- Fig. 8: Distribution of ³⁵S labelled compounds. The upper picture shows the autoradiogram, the lower one shows the response of our detector. The scales are 70 mm.



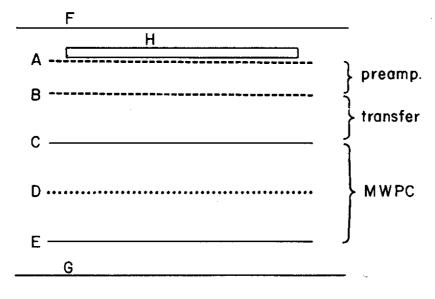


Fig. 2

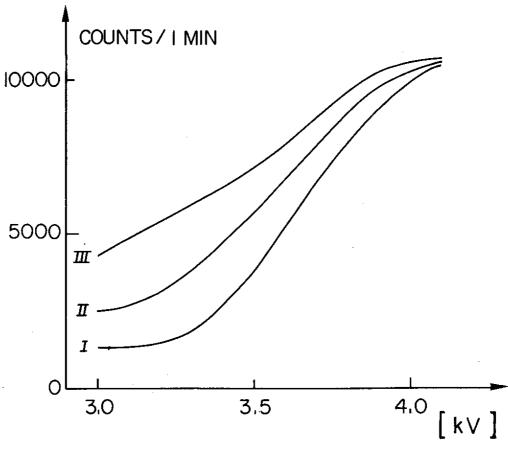
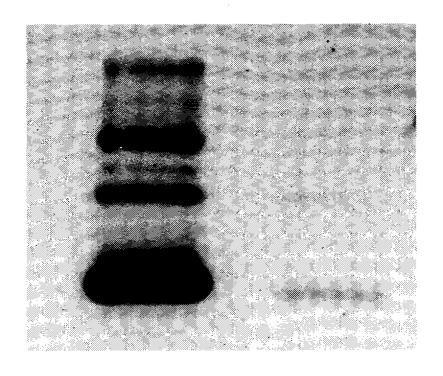


Fig. 3



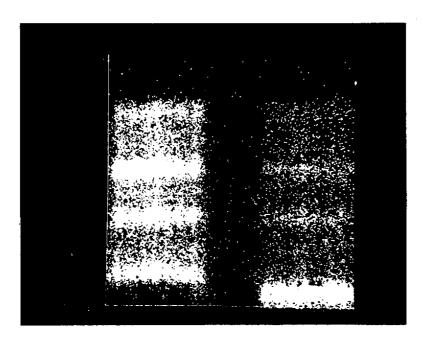
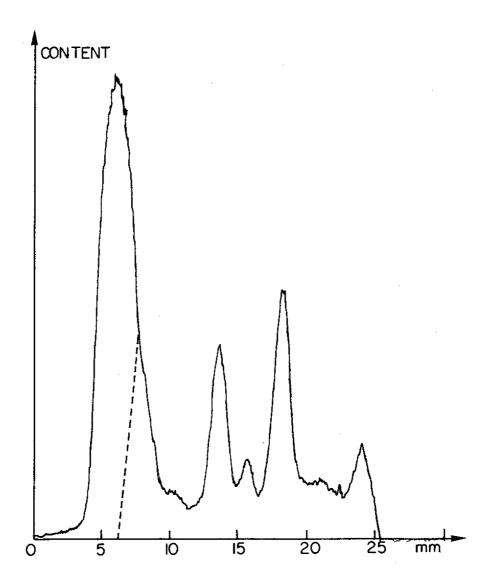
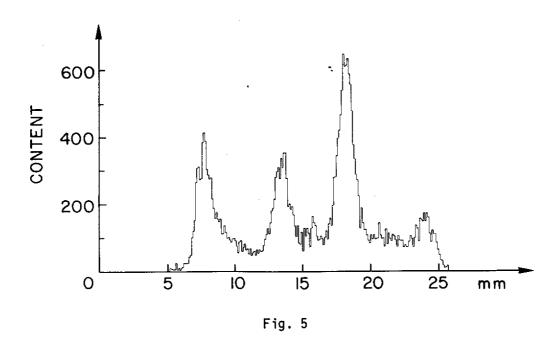


Fig. 4





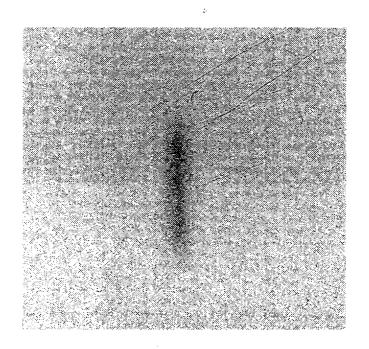
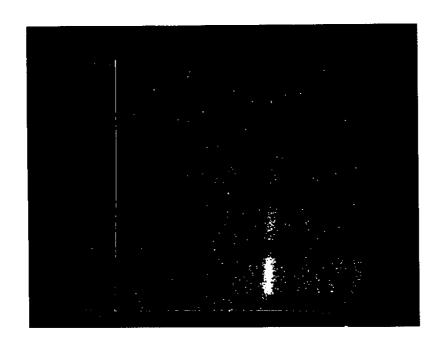


Fig. 6



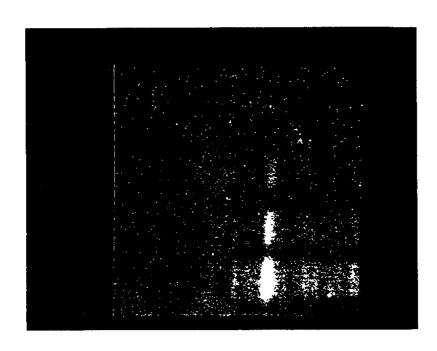
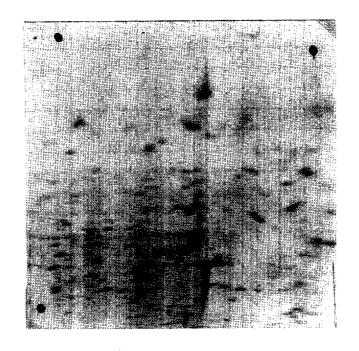


Fig. 7



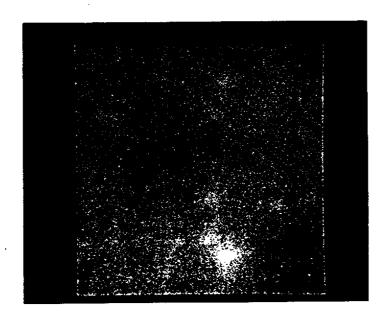


Fig. 8

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