ABSOLUTE MEASUREMENT OF THE ACTIVITY OF RADIONUCLIDES

II — THE USE OF A 4π PROPORTIONAL COUNTER

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RESUMO

Neste trabalho os autores expõem as técnicas usadas no Laboratório de Medidas Absolutas dêste Instituto para a medida precisa da atividade absoluta de emissores alfa e beta, utilizando um detetor proporcional 4π.

Algumas das vantagens dêste método são que o fator de geometria é igual a 1 e as correções de absorção no suporte da fonte podem tornar-se bastante pequenas no caso de serem utilizados filmes plásticos muito finos.

São descritas também as técnicas de preparação de fontes e é apresentado o problema da auto-absorção.

Pode-se obter, se as condições forem favoráveis, precisão de cêrca de 1% na medida absoluta da atividade dos radioisótopos.

RÉSUMÉ

Dans ce travail on expose les techniques qui sont utilisées au Laboratoire de Mesures Absolues de cet Institut pour la mesure precise de l'activité absolue des emétteurs beta et alpha en utilisant un compteur proportional 4π.

L'un des avantages de cette méthode est que le facteur de géometrie est égal a 1 et les corrections d'absorption dans le support de la source peuvent devenir très petites si l'on utilise des films plastiques très minces.

On présente aussi les techniques de preparation de sources et le problème de l'auto-absorption.

Dans des conditions favorables on peut obtenir avec le compteur 4π des précisions d'environ de 1% dans les mesures absolues de l'activité des radioéléments.

ABSTRACT

This paper presents a discussion on the techniques used at the Absolute Measurements Laboratory of the "Instituto de Energia Atômica" for a precise evaluation of the activity of beta or alpha emitting isotopes by means of a 4π proportional counter.

One of the advantages of this method is that, since all particles emitted within a 4π solid angle can be detected, there is no correction for the geometry factor; at the same time, the use of very thin metallized source support films eliminates or makes very small the contribution due to errors arising from self--scattering and absorption through the supporting source material.

As a result, when due care is taken, the absolute activities can be measured with a precision of the order of 1%.

A discussion is also presented on the different techniques used in the preparation of sources for absolute measurement.

I - INTRODUCTION

The use of a 4π proportional counter is, practically the only method available for measuring the activity of a pure beta (or alpha) emitter with a precision of the order of 1%. For nuclides in which the particle emission is followed by a single gamma ray or by a gamma-ray cascade, the use of the coincidences between the particles and gamma rays affords a simple and almost straightforward method of evaluating the absolute activity, provided that the disintegration scheme of the nuclide is known.

Since the method of using a 4π proportional counter is devised to allow a precision as close to 100% as possible to be reached, its results will depend on the characteristics of the counter design, of the electronic circuitry, and of the source under measurement; it is also important to know the energy spectrum and the energy or the energies of the particles which are being counted.

Another advantage of the use of a 4π proportional counter is that since many correction factors can be eliminated, it is possible to make the contribution of the others sufficiently small to allow its influence to be practically negligible as a source of error in the absolute activity determination.

Another important characteristic of a 4π counter is its intrinsic high efficiency, due to the fact that no absorbers do exist between the source and the active area of the counter and its ability to count any particle, emitted under a 2π angle in each hemisphere, giving rise to more than a pair of primary ions inside the counter volume. Spurious counts, such as the multiple discharges which occur in a Geiger Müller counter are inexistent.

The advantages outlined above points out to another important application of such counters when associated with one or two scintillation counters, in the determination of the absolute counting rate of sources which emit either beta or alpha rays in cascade with gamma rays, since it is well known that the method of coincidences allows a higher degree of precision to be reached when one of the detectors has an intrinsic efficiency of about 100%.

4π counters can be used either as proportional counters or as Geiger Müller counters. In this paper we will be concerned essentially with the proportional counter, since it is known that it presents several advantages over the Geiger Müller counter. Amongst the disadvantages of the use of Geiger Müller counters, we should point out the long dead time and the difficulty in establishing the correction due to the formation of multiple discharges. Although the latter phenomenon can be evaluated by the slope of the plateau, and criteria are available for extrapolation of the measured counting rate to its "true" value, the instability of the phenomenon due to changes in the properties of the cathode surface and the presence of small impurities in the gas introduce sources of error which are difficult and tedious to be taken into account. One of the important advantages of the proportional counter is that whenever a pulse appears, there is no question that it was originated from a certain number of ion pairs liberated inside the counter by an ionization process. Besides, when a suitable gas filling is used, the proportional counter presents a very small dead time, and its time lags are also smaller than in a conventional Geiger Müller counter.

The main disadvantage of the use of proportional counters is the necessity of using a very stable linear amplifier; this is a problem, however, which presents no serious difficulties when such an apparatus is designed to present characteristics which make them almost independent of either the tubes or transistor parameters, by the use of negative feed-back and by a conservative design and choice of components.

The true counting rate of a source, as measured by a 4π counter, is given by

$$N = N_A + N_B - C - B_g$$

where N_A and N_B are the counting rates of counters A and B, C is the coincidence rate due to cosmic rays which have their path through both chambers A and B and to particle which give rise to a discharge in both counters and B_g is the background counting rate of both chambers or counters.

The factor most difficult to take into account is C. Although the number of coincidences due to cosmic rays traversing both counters can be readily measured, a large difficulty is presented when one wants to evaluate the contribution due to particles from the source which give rise to a discharge in both counters. Such discharges can be present whenever a particle, emitted in one hemisphere, is scattered back and can penetrate the other chamber either through the source film support or through the metal diaphragm used as the film holder. Another source of such coincidences is due to the real coincidence between a particle which traverses one counter and a secondary electron produced by a gamma ray in cascade with the disintegrating electrons.

Although scattered electrons from one chamber to the other could be absorbed by using a source supporting diaphragm sufficiently thick, its use would present the disadvantage of being an efficient converter to the gamma rays associated with the particle emission. It is a good rule, therefore, to build the proportional counters with a low atomic number material - such as aluminium - and use a light diaphragm on the same material. The contribution due to scattered particles from one chamber to the other can be evaluated by a comparison of the activities of a source, mounted on a thin film, and the decrease of the number of the observed coincidences when the diaphragm is changed into a infinitely thick one (in relation with the range of the most energetic particles) and the source is backed by an absorber of the same thickness as the diaphragm. Of course those measurements should be carried out by using a pure beta ray emitter.

The effect of coincidences due to electrom-gamma rays can be evaluated by means of a comparison between the observed counting rates due to a pure beta emitter and one disintegrating through a complex scheme. A knowledge of the sensitivity of the equipment is necessary, in order to make those correction independent of the shape of the spectrum and of the disintegration energy of the nuclide under consideration.

II - DESCRIPTION OF THE DETECTOR

The 4π proportional counter used at the Laboratory for Absolute Measurements of the IEA is a modification of the original model built by Tracerlab (fig. 1). The main modifications produced by us are in the anode structure and in the pre-amplifier built in a position adjacent to the counter chamber.

It consists essentially of two cylindrical aluminium chambers 7,5 cm in diameter and 9,3 cm high, shielded by thick lead absorbers. The anodes we are using are loops 0,076 cm in diameter made of stainless steel wire 2 mills in diameter. They are placed symmetrically in relation to the source so that the distance between the center of the loop and the source is 2,8 cm. A provision for piping the flowing gas which leaves the counter to the external atmosphere was also incorporated, in order to avoid undue contamination of the counting atmosphere which is air-conditioned.

Figure 2a is a block diagram of the counting system.

III - COUNTING CORRECTIONS

As it was pointed out before, many corrections which have to be taken into account with other methods of absolute measurement are either eliminated or their influence reduced when a proportional 4π counter system is used. In the following lines, the influence of the different sources of errors will be discussed.

a) Geometry.

The influence of the geometry factor can be neglected when the counter is properly designed. Any deviation from the theoretical 4π geometry can be due only to the loss of particles emitted tangentially to the plane of the source, or, to the inability of particles emitted almost tangentially to produce ion pair due to a short geometrical path between their emission point and the counter wall. The influence of the above causes of error can be minimized by using a very thin source mounting diaphragm and by using a source of a diameter small as compared with the counter diameter. On the other hand, the influence of the thickness of the ring used to support the source film, can be made sufficiently small by using film supporting frames of a fairly large diameter. To avoid the formation of charged points on the film which could give rise to spurious discharges, the practice of covering the films with a thin conducting (metallic) layer which is evaporated in vacuum. As it is well known, unless a conducting film is provided, the source mounting can become charged with positive charge which gives rise to a disturbance of the electric field in the neighbourhood of such points; the net result of such a phenomenon is the decrease of the ionization probability provoked by the decrease of the electron mobility and the consequently larger probability of recombination.

b) Absorption in the source mounting support.

By using plastic resins now available(1), it is possible to make films for supporting the sources which are only a few micrograms/cm^2 thick; for those mounting films and "invisible" sources (which can be prepared from carrier free isotopes), the corrections due to absorption are very small and can usually be neglected.

In case a higher degree of precision is wanted, the influence of this factor can be evaluated and its effect taken into account in the final activity calculation.

c) Self-absorption

Since the corrections for self-absorption in a thin source are usually smaller than 0,2%, the corrections on the counting rate can be assessed under some approximations.

If the mass of the source is known and its area is measured approximately (with a ruler, for instance), the mean source thickness can be deduced.

Let t be the half thickness for the beta particles in the source and a the mean source thickness, both measured in mg/cm^2; if I is the true activity, the measured or apparent intensity I_m is given by

$$I_m = \frac{I}{a} \int_0^a \left(\frac{1}{2}\right)^{\frac{a}{t_{1/2}}} d\,a$$

whence:

$$\frac{I_m}{I} = \frac{t_{1/2}}{a} \; \frac{1 - \left(\frac{1}{2}\right)^{\frac{a}{t_{1/2}}}}{\log_a 2} = \frac{1 - \left(\frac{1}{2}\right)^{\frac{a}{t_{1/2}}}}{0{,}693 \, \frac{a}{t_{1/2}}}$$

Now, if we assume that a << t, we can write

$$\frac{I_m}{I} \sim 1 - 0{,}346 \, \frac{a}{t_{1/2}}$$

IV - OPERATION CONDITIONS

In order that any particle emitted by the source be counted by the registering unit, it is necessary that:

1) any particle will give rise (in the sensitive volume of the proportional counter) to a number of ion pairs equal to or larger than x;

2) that the probability of such ion pairs giving rise to a pulse of sufficient amplitude to be registered is equal to 1.

In the case of a proportional counter, the minimum number of pairs of ions (produced by the incoming particle) required to produce a pulse dV depends both on the gas amplification and on the distributed capacity of the input circuit of the counter

$$dV = \frac{Q}{C} = \frac{n\,e\,A}{C}$$

where

Q .... is the total collected charge by the counter anode

C .... is the distributed capacity of the anode

n .... is the number of ion pairs produced by the primary particle

A .... is the gas amplification factor

For proportional counters carefully built, n can be as small as 1, a value between 1 and 10 being usual.

If one assumes as 35 eV the average energy required to form an ion pair, the upper limit of 10 ion pairs corresponds to an energy loss of about 350 eV - what is equivalent to say that under those experimental conditions no electrons of an energy smaller than 350 eV will be counted.

The apparent advantage of a Geiger Müller counter in being able to give rise to a pulse when only one pair of ions is formed is offset by the uncertainties in the correction of the effect of multiple discharges and by the circumstance that the number of rays which have an energy below this value (350 eV) is very small for the normal beta emitters and its number can be evaluated with a fairly good degree of accuracy.

If one examines the energy distribution spectrum of the several nuclides, one can realize that the fraction of particles which have an energy between 0 and 350 eV as compared with the number of particles of an energy above 350 eV can be neglected in most measurements, whenever the maximum energy is fairly large - as in the case of P^{32}, Au^{198}, Sr^{90} etc..

In the case where the disintegration energy is small one has, for instance:

Nuclide	E max.	% loss
S^{35}	0,17 MeV	0,4 %
C^{14}	0,15 MeV	0,6 %

The conclusion is that for the accurate evaluation of activities of beta emitting isotopes, the influence of this correction must be taken into account only when low energy isotopes are under measurements and only where a high degree of accuracy is wanted.

V - INFLUENCE OF THE GAS

There are several gases which can be used sucessfully for filling proportional counters.

The influence of the gas nature on the slope and length of the plateaux, on the rising time of the pulse and on the dead time of the discharge are subjects which have been carefully studied by several investigators.

As a rule, the general choice seems to favor the use of polyatomic gases, since it is observed that the gas amplification factor changes slowly with the potential difference across the counter; as a result, long and flat plateaux are obtained resulting in a high degree of stability and reprodutibility of the system; the main disadvantage of their use is the relatively high potential difference required (of the order of 4.10^3 Volts) which imposes a careful design of the anode lead in insulators and special connecting cables and connectors which are not of a standard use.

In our experiments several gases and gas mixtures were used or tried and our final choice resulted in a compromise between the ideal characteristics, the availability from local manufacturers and the simplicity of the gas purifying system - if any.

Very good results were obtained with a mixture of helium and isobutane (1,3%) - the so called Q-gas. Although its use provides long plateaux (of the order of 300 volts), with a slope not larger than 1% (fig. 3) and requires a comparatively low operating voltage, its main disadvantage is the long dead time (65 μs) the high cost and the availability (imported gas).

Very good results are being obtained through the use of pure propane (furnished by ULTRAGAS S.A.)(fig. 3-4). Traces of sulfur compounds and water vapour are eliminated by a simple system which consists essentially in the use of activated charcoal and a dehumidifier which consists of dried anionin resin. The gas, freed of its most unwanted impurities is fed to the counter through a silica gel tube provided with some glass wool whose purposes are of giving a visual indication of the efficiency of the dehumidifier system and of avoiding any dust particles from entering into the counter.

The obtained plateaux are usually about 800 volts long and flat; their length and slope are influenced by the presence of water vapour and impurities so that a reactivation of the dehumidifier and of the charcoal trap should be made every two months or so - depending on its size and the use of the equipment.

VI - ELECTRONIC SYSTEM

The continuous distribution - (this continuous distribution of pulses arises from the continuous distribution of energy of the emitted beta particles, from fluctuations of the number of primary ions, from the length of the trajectory of the particle inside the counter and from the effective value of the electrical field strength in the positions where such ions are formed) - of pulse amplitudes originated from a proportional counter imposes severe requirements in the design of the linear amplifiers and on the pre-amplifiers used, since it implies the existence of pulses (originated from particles which have produced a small number of primary ions) whose amplitude may be smaller than that due to the noise from the amplifying system. Since in absolute measurements every emitted particle must be counted, it is an important requirement to use a detecting system so designed that the overlapping of noise pulses with those due to particles should be as small as possible, in order that only pulses originated from ion pairs produced by real particles would be selected by a suitable pulse amplitude discriminator.

In order that such low amplitudes pulses originated in the counter can be counted, it is an important requirement to use a pre-amplifier designed in such a way that it should show a sensitivity as high as possible under a minimum noise amplitude (fig. 2b). The overall gain of the pre-amplifier plus the linear amplifier system should be as high as reasonably possible, in order that the proportional counter could be operated in a region of low gas amplification. These requirements impose very difficult problems in the design of the amplifier, if a constant gain is wanted for all the pulses originated in the counter (as would be the case, e.g., for beta spectra measurements).

When the greatest interest is in counting all the pulses of an amplitude equal to or larger than a pre-determined value (as set by the noise figure of the system), the amplifier design is less critical; since however pulses from a wide range of amplitudes do occur, the amplifier should be designed in such a way it should be not overloading and should be of a sufficiently high frequency response to avoid piling up of pulses and losses due to such effects and hysteresis effects of over or undershooting of the amplifier gain from preceding pulses.

In our 4π counting system, the pre-amplifiers which are being used are charge sensitive and have a very low noise; the low input capacity and low grid current noise are obtained by using an electrometer tube in the first stage of the pre-amplifier. The amplifier and scaler used are built by Nuclear Chicago Co. (Ultrascaler, Model 192-A) and was modified to increase its dynamic range by the use of solid state diodes introduced in feed-back loops$^{(2)}$. A rotary switch installed in the input circuit (which was modified) allows pulses from a single chamber or from both chamber to be counted.

VII - SOURCE PREPARATION

The preparation of active sources for absolute measurements of the activity is one of the less usual, the most difficult and challenging techniques; unless the utmost care is taken in the preparation of the supporting film, in the process of metallizing the film, in the source deposit on such film, and in the source mass measurement, large errors can occur which would make meaningless the use of special counting equipment for such purpose.

In the following lines, the main outlines of the techniques which have evolved in our laboratory during the last two years will be presented.

1. Preparation of the source supporting film

Although several plastic films were used in ourlaboratory, we will concern our attention only to the properties of VYNS, since the best results were obtained with its use.

Due to its properties, the use of VYNS films is an almost general practice in all tye laboratories engaged in this field of activity.

1.a) Properties of the VYNS films

Until a few years ago, one of the largest sources of error in absolute measurements with 4π counters was the impossibility of using supporting films sufficiently thin and physically resistant for the deposition of either alfa or low energy beta sources. Through the use of synthetic resin films, which are now used, the problems arising from the energy loss of the particles traversing such source supporting films can be made so small that its influence can be neglected in a large number of cases, since films with a thickness of the order of micrograms/cm^2 can be used.

The resin used in our laboratory (VYNS) is an acetate of polyvinylchloride, fabricated by the Bakelite Co., New York, USA.

Some of the most important properties of those films are the following:

a) Films of a few micrograms/cm^2 can be made easily and the energy loss of particles through them can be neglected in most cases.

b) The low atomic number (equivalent) makes the corrections due to the scattering of the particles negligible.

c) VYNS has a remarkable chemical resistance, being attached practically only by acetonic compounds and by ethers. As a result, such films resist the action of the chemical agents which quite often are used in the preparation of the source.

d) Thermal resistance - The sources deposited on VYNS films can be dried with an infra-red light and show no deterioration. No observable changes are noticed on the film structure and properties, even after it has undergone the influence of the high temperature of the tungsten filament during the metallization process.

e) Since it can be observed that the films are not affected by atmospheric influences, they can be prepared in batches and stored for use.

The remarkable resistance properties of those films to atmospheric agents makes them the ideal supporting material for reference sources, which will show no observable change in physical properties after long time of use.

1.b) The manufacture of source supporting films

VYNS is usually obtained from the manufacturer as a white powder which can be dissolved in cyclohexanone.

Since such a solution of VYNS is stable and lasts for several months, our procedure is to prepare a concentrated solution of one volume of powder dissolved in two of cyclohexanone. For the manufacture of films the concentrated solution is dissolved again in cyclohexanone, using three volumes of the solution and two volumes of cyclohexanone. Although the concentration of the final solution has some influence on the film thickness, it is observed that the final thickness depends on the spreading of the solution drop on the collecting surface which usually is water.

To prepare the film, it is usual to let from one to three drops of the solution fall near to one edge of a large plastic basin filled with distilled water: the drop solution spreads over a large area of the water surface. In order to obtain a better uniformity of the film thickness one can use a wooden stick, placed near to the basin edge. As the drops will fall between the basin edge and the wood stick (closer and along the wood stick), this is displaced over the water surface with a constant speed. The uniformity of the film deposit can be judged from its inability to show interference of the light by reflection.

The film thus formed can be transferred to the final frame simply by placing such frame (in our case an aluminium disc 0,1 mm thick, of 3 cm of external diameter and a central punched hole of 1,5 cm diameter) on the region of the spread film which is chosen; the excess of the film over the edges of the disc is cut simply by touching the edges with ones finger. Once this is done, the frame can be withdrawn from the water, without affecting the film, by tilting and drawing it out slowly.

It is necessary that the water surface and the air in the room should be free from dust, which might otherwise deposit on the one of the film surfaces.

1.c) Measurement of the film thickness

The measurement of the film thickness can be made either by gravimetric, by absorption or by optical methods.

The gravimetric method involves the use of microbalances and is currently used in several laboratories, although it presents some disadvantages when compared with other methods:

1. Weighing with microbalances is a lengthy and tedious procedure.

2. In order that the measurements be significant, a careful appraisal of the film distribution on the frame is required, since an excess or lack of film over the supporting metallic frame will give rise to errors; this requirement involves special care in the procedures used in picking up the film from the water surface.

3. In the case where the film is unhomogeneous, the calculated thickness corresponds to an average value which may be significantly different from the true thickness of the central part of film - which is obviously the only important region.

4. Since the films will be laid on a frame whose weight is previously measured, special care in handling the frame is required to avoid changing its mass by the presence of impurities which might arise in the handling process.

Although those disadvantages are notorious, the long term stability and the simplicity of the operations involved in checking the operation conditions of the microbalance are advantages which completely offset the incovenients pointed out above.

1.d) Procedures for metallizing the film

Although some authors believe that the presence of a small area of a non-conducting material inside a 4π counter does not disturb the electric field distribution, this is only true when such area is negligible as compared with the total cathode area of the counter. In the great majority of counters, whose dimensions are reasonable, the disturbance introduced by the presence of such materials on the behaviour of the counter can become very important and make the use of a conducting film imperative.

The usual procedure consists simply in covering the film with a thin layer of acquadag or of an evaporated or sputtered metal, in high vacuum.

In our laboratory the technique of vacuum evaporation of metals in used routinely and although there are several metals which can be used for such purpose, our experience is that the use of gold always gives the best results. Although metals such as aluminium, copper, silver etc. can be used, the chemical stability of the metal layer plays a very important role in the final properties of the film; this was observed to be the case when aluminium was used, since its oxidation leads to a change in the properties of the VYNS films which become wrinkled andbreak easily and expontaneously after a few days. The same behaviour should be expected with copper and a similar effect with silver, due to its ability to absorb traces of hydrogen sulfide usually present in air.

The apparatus used for the metallization of the films was built in the laboratory and is shown in figure 5.

The procedure used is start the evaporation only when the pressure in the system falls to about 10^{-5} mm Hg. It takes about 15 minutes to reach such a pressure after the oil diffusion pump is on; since our pump can start with a forepressure 10^{-2} mm Hg, the total time required to reach the appropriate vacuum pressure for metallizing takes only about 20 minutes.

The metal to be evaporated is wrapped around a tungsten filament and its temperature is raised until the evaporation starts.

Since the distribution of evaporated particles is practically isotropic, the films are disposed at equal distances from the filament, in what would be the equivalent of a spherical distribution.

This precaution is recommended in order that all the films should show an evaporated metal deposit of the same thickness.

The evaporation of the metal is done slowly, not only in order to allow a precise control of its thickness but also with the purpose of decreasing the amount of heat radiated from the filament in order to avoid damage of the films.

The thickness of the deposit is controlled by observing the colour of the deposit by reflected light on the aluminium supporting frame and it was found that a dark blue deposit is quite suitable.

The true thickness of the metallic deposit can be evaluated by weighing the films before and after the evaporation process. The precise knowledge of the metal deposited on the film is important in order that its absorption can be computed - and such an absorption can be significant when low energy beta ray sources are being counted.

Although sometimes both faces of the film should be metallized, it is known that when thin films are used, the same conducting effect can be achieved by depositing, on one side only, an amount of metal slightly larger than what would be required if both faces were metallized: it is found, for instance, that when gold is used, a layer of about 15 micrograms/cm^2 is sufficient for that purpose.

Since the length and the slope of the plateaux are seriously affected by the presence of non conducting areas inside the counter, its observation allow a good criterion for evaluating both the minimum required thickness and the uniformity of those deposits.

2. Source preparation

2.a) The preparation of a source suitable for absolute measurement with a 4π counter is one of the most delicate and critical operations involved in the determination of the specific activity. For such a measurement it is important to know precisely not only the true weight (or mass) of the isotope under measurement, but an experimental procedure must be followed to assure that no lumps or crystal are formed which would disturb the ideal uniform distribution. Once an examination of the deposit under a microscope will assure that the source material is evenly distributed, the precise knowledge of the area under which it is spread and its total weight will allow an accurate estimate of the self-absorption factor to be taken into account.

Since in the great majority of absolute activity determinations only carrier free isotopes are used, the sources are usually prepared by taking an aliquot from the isotope solution and by spreading it afterwards over the central region of the supporting film. It is usual to use just one or a few drops from the solution, to which a wetting agent, such as insulin or ludox is added, with the purpose of obtaining an uniform deposition.

It is found that unless a suitable wetting is added, during the evaporation process which the drop is submitted to (with the help of an infra red lamp, vacuum etc.), there is a large tendency to crystal formation around the edges of the initial drop, where most of the radioactive atoms will be concentrated.

Ludox is used instead of insulin, since this protein, as obtained commercially, usually has large amounts of phenolic compounds and glycerin which will leave an observable residue after evaporation. On the other hand, since ludox is a colloidal solution of silicon at 4%, no such effects are observable and very homogeneous deposits are obtained.

In our Laboratory insulin is being used 70% diluted.

2.b) Determination of the source mass

In order to determine the specific activity of a solution it is required that the mass of the deposited aliquot on the film be known accurately. This is often a difficult problem since it involves small weights which must be measured with the highest possible precision. Many difficulties arise due to problems of evaporation that occur during the experimental procedures of diluting, depositing and weighing the sample. Another factor which plays an important role is the adsorption of the radioactive atoms by the micropipet internal walls.

In our laboratory, use is made of an analytical micro-balance made by Sartorius Werke (Göttingen) (fig. 6) with a precision of 5 micrograms and a sensitivity of 1 microgram, with a maximum load of 20 grams.

As it is usual with such equipment, the balance is installed on a vibration free support in an air conditioned room which maintains both the temperature and the humidity constant. As it is known, an extreme cleanliness and periodic tests of its sensitivity are required. The model used is provided with an special device which brings the weighing plate support to the outside of the balance housing by means of a manipulator which opens momentarily a window whenever the plate reaches the housing front surface; this avoids undue contact with the external atmosphere during the operations of loading and unloading the balance, helping to maintain the constant temperature and humidity of its components.

2.c) Methods used to determine the source mass

The isotope whose activity is to be measured is usually provided as a solution in a sealed container and there are several methods which can be used for the processes of extracting, diluting, depositing and weighing a sample from such a solution. Micropipets, microburetes and picnometers may be used successfully.

The procedure used more often are:

1. Polyethylene picnometer.

The mass is determined by weighing previously the picnometer, then introducing the solution in it and weighing again, in order to determine the weight of the solution. After one or more drops are deposited on the film support, the picnometer is weighed again in order to determine the mass of aliquot. This method gives an accuracy of about 0,15%.

2. Determination of the weight of the film plus the solution deposited on it.

As in the preceding technique, the solution is diluted with a micropipet, previously treated with an inactive solution of the material which is to be deposited. In a second step, the micropipet is used to deposit the solution. By measuring the weight of the film and the solution drop at different time intervals, (in order to correct for the losses in mass due to the water evaporation), a plot of the weight values against time will provide the mass at zero time by extrapolation of the straight line thus obtained.

This method is more tiresome than the former and provides the same degree of accuracy (about 0,15%).

2.d) Correction for self-absorption

The crystals which are sometimes found on the source supporting films are due to chemical carriers or to other substances used in the preparation of the radioactive sources and which cannot be eliminated. The presence of such aggregates are important whenever a source of low energy beta rays is measured. It can be shown that for isotopes which emit beta rays of maximum energy of about 1 MeV or higher, the effect of absorption on such unwanted materials can be neglected. Their influence however, must be taken into account when the maximum energy is smaller than about 1 MeV, even when an uniform deposit is obtained through the use of a wetting agent as discussed in § 2.a.

The decrease in activity due to the presence of absorbing materials in a source of thickness d, is given by

$$a = a_o / \mu d \; (1 - \exp. - \mu d)$$

where μ is the mass absorption coefficient.

This formula is approximate, since it does not taken into account the crystalline structure of the sample and has a meaning only when d is the true thickness of the source, whereas the experimental value of d, as determined experimentally, is its mean value. Similar considerations hold for μ, since it is known to depend on the source thickness, the shape of the beta spectrum and the geometry of the counting system (due to the influence of the self-scattering of the beta rays in the source).

2.e) Methods of measurement of the self-absorption

1. Method of the successive dilutions

With sources of an increasing degree of dilution, a graph is plotted of the variation of the activity against the concentration and the graph is extrapolated for an infinite dilution (or zero concentration); such a plot show the effects of the self-absorption for various concentrations. One should remember however, that by increasing the dilution the size of the crystals is not diminished, the increase in their mean distance being the main factor responsible for the observed phenomenon.

2. Correction by the shape of the spectrum

From the knowledge of the source thickness, the cut--off energy can be calculated, providing an estimate of the fraction of the energy distribution spectrum which is not being counted. The measured results are then corrected accordingly.

3. Method of the added tracer

One way to correct for the self-absorption consists in incorporating to the source, during its preparation, a known amount of another isotope of short half life and of a similar energy spectrum. Since the number of particles emitted by such an isotope is previously known, a comparison between the actual and the expected counting rates gives the correction factor wanted.

The accuracy which can be obtained through the use of such method depends of course rather critically on the precision with which the added activity is known, since one can assume with a good approximation that the atoms of the isotope which one wants to measure and those from the added isotope will show the same degree of dispersion in the microcrystals which appear on the supporting film.

4. Correction by means of known experimental data

Since curves for self-absorption are already known for several isotopes, use can be made of such data once the mass of the source is known.

5. Correction of the self-absorption through the knowledge of the size of the crystals

The size of the crystals on the supporting film are measured by means of a microscope. An usual procedure for such measurements consists simply in determining the "radius" of the crystals.

Once those values are known, use can be made of the experimental data due to Meyer, in which curves are presented showing the variation of the product between the absorption coefficient and the mean crystal "radius", against the percentage of the particles which undergo self-absorption in such crystals.

The absorption coefficient can be obtained from the same curves as a function of the maximum energy of the beta rays emitted by the nuclide under study.

VIII - ROUTINE TESTS AND CHECKS

Since the absolute determination of activities requires measurements of a high degree of precision, there are several tests and checks which should be carried out periodically in order that the obtained results should be considered trustworthy. Some of the most proeminent amongst such procedures are the following:

1. Determination of the plateau from both counters with source under measurement. Determination of the symmetry of both counters (by rotating the plane of the source by 180º). Determination of the stability of the gas mixture and the counting system.
2. Determination of the pulse height distribution and losses due to pulses of a small amplitude.
3. Periodic measurements of the resolution time (with an oscilloscope and source, to determine the recovery time of the counters and by means of a double pulse generator to check the circuit losses).
4. Determination of the threshold energy for beta counting.
5. Statistical distribution of pulses.
6. Study of the counting system long term stability.
7. Measurement of the gas amplification factor.

Our procedure for such measurements (fig. 7-8) consists in calculating A (the gas amplification factor) by means of the formula

$$A = C \, dV / n \, e$$

where

C ... total distributed capacity between the charge collecting electrode and ground.

dV ... pulse height.

n ... number of primary ions produced by the particle.

e ... electron charge.

The number of primary ions can be computed readily whenever only particles of a well defined range are used, such as the alpha particles from a thin source of Po^{210}. In this case, the number of ion pairs is given by E/W, where E is the alpha particle energy and W the energy required to produce one ion pair in the gas.

The gases most commonly used in our laboratory are either propane or a mixture of helium and isobutane (Q-gas). For the measurement of the distributed capacity an L-C Tektronix capacity bridge was used.

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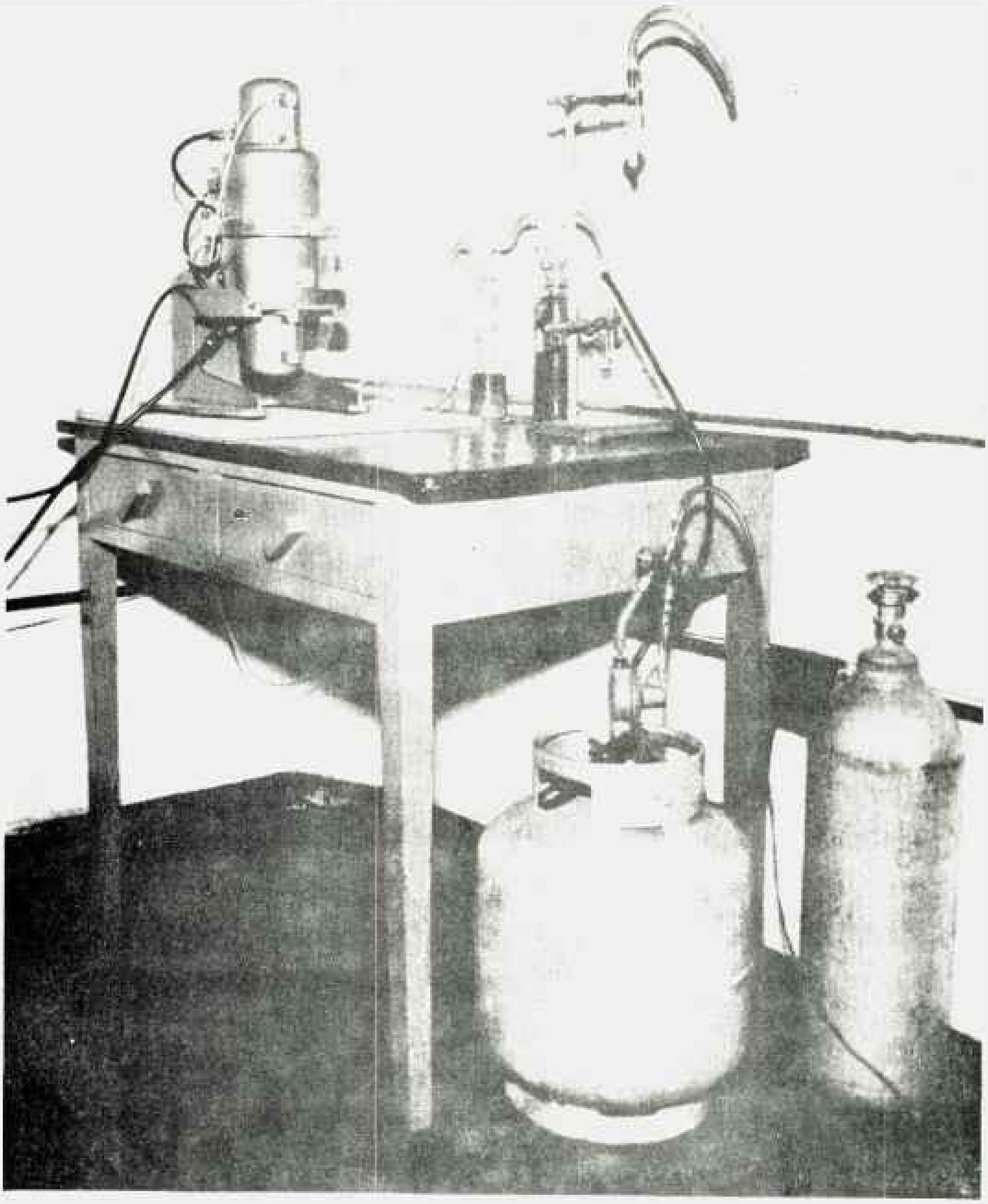

Fig. 1 a - 4π counter assembly

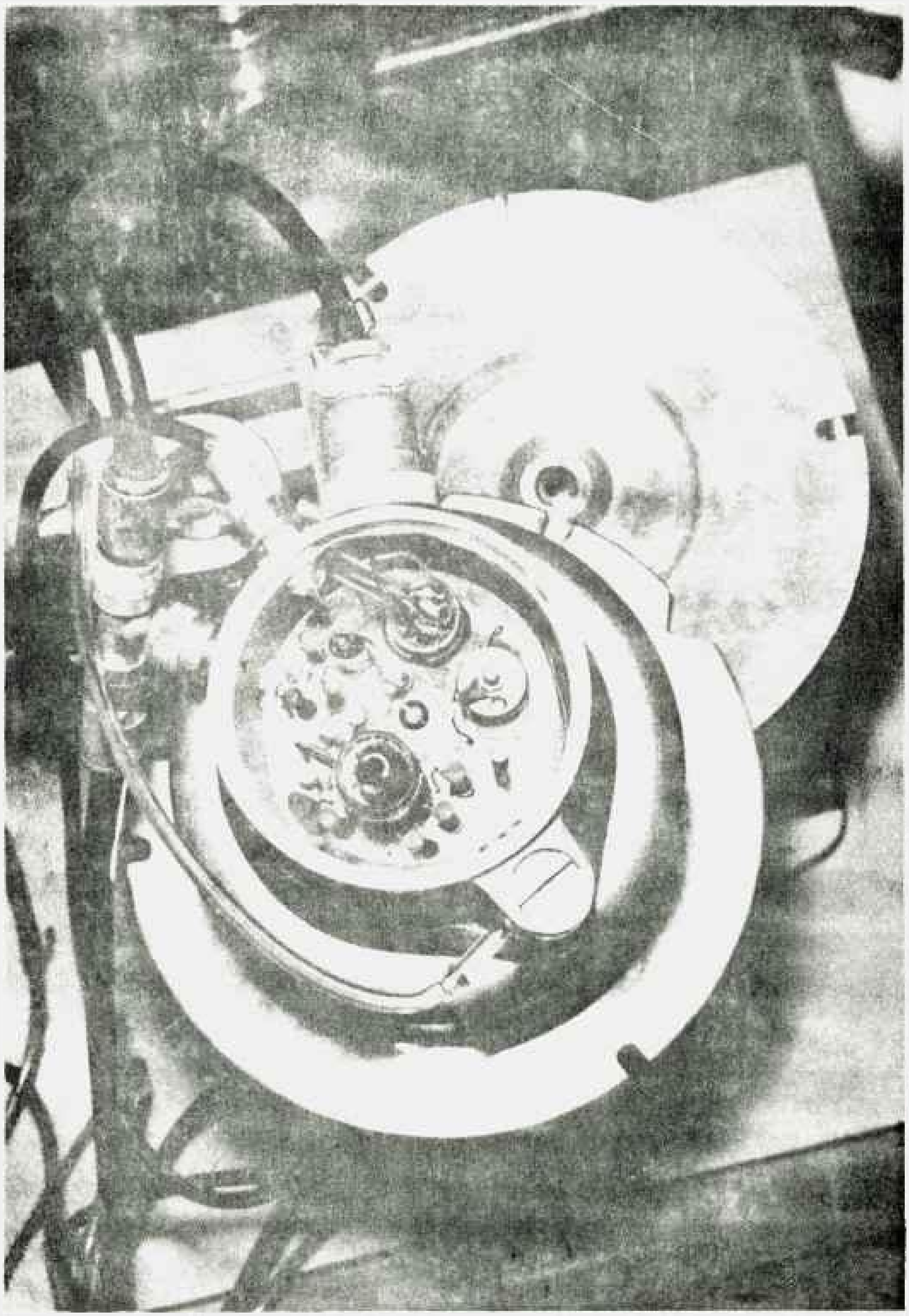

Fig. 1 b - Pre-amplifier and source support

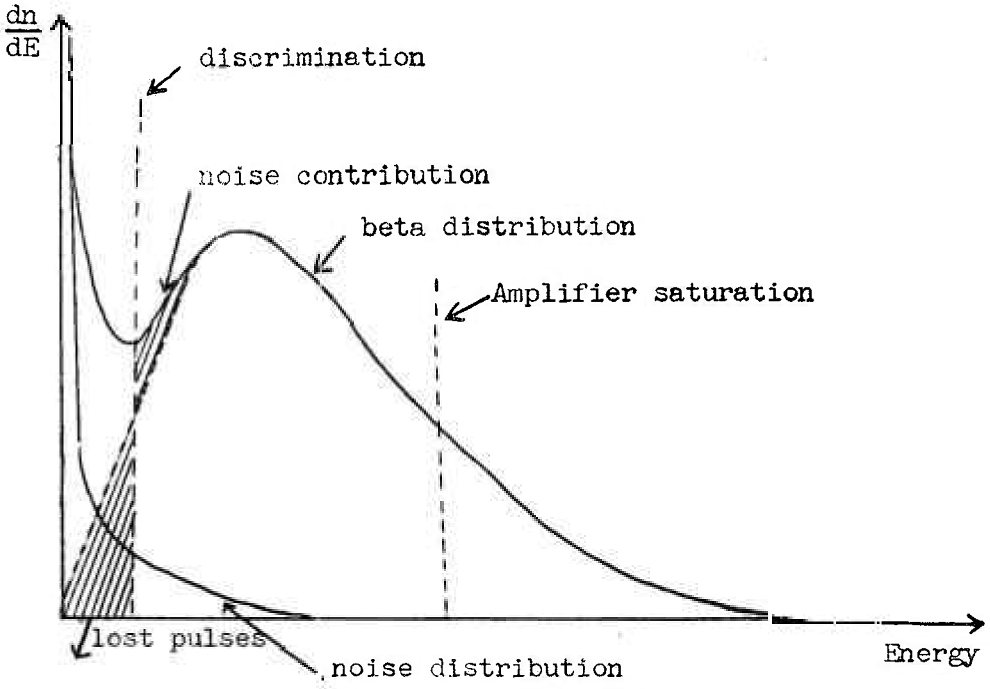

Fig. 2b - noise effect in the measurement of β -emitters

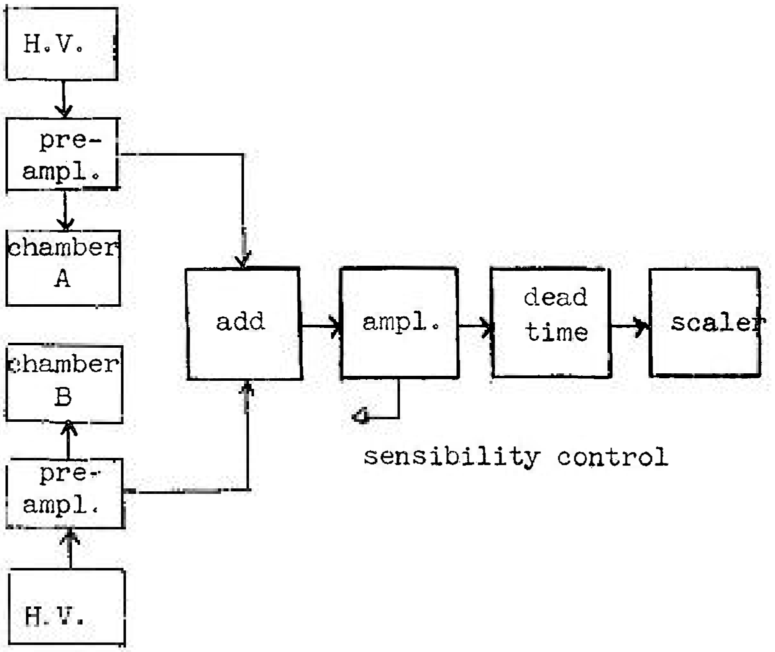

Fig. 2a - Block diagram of the 4π electronic system

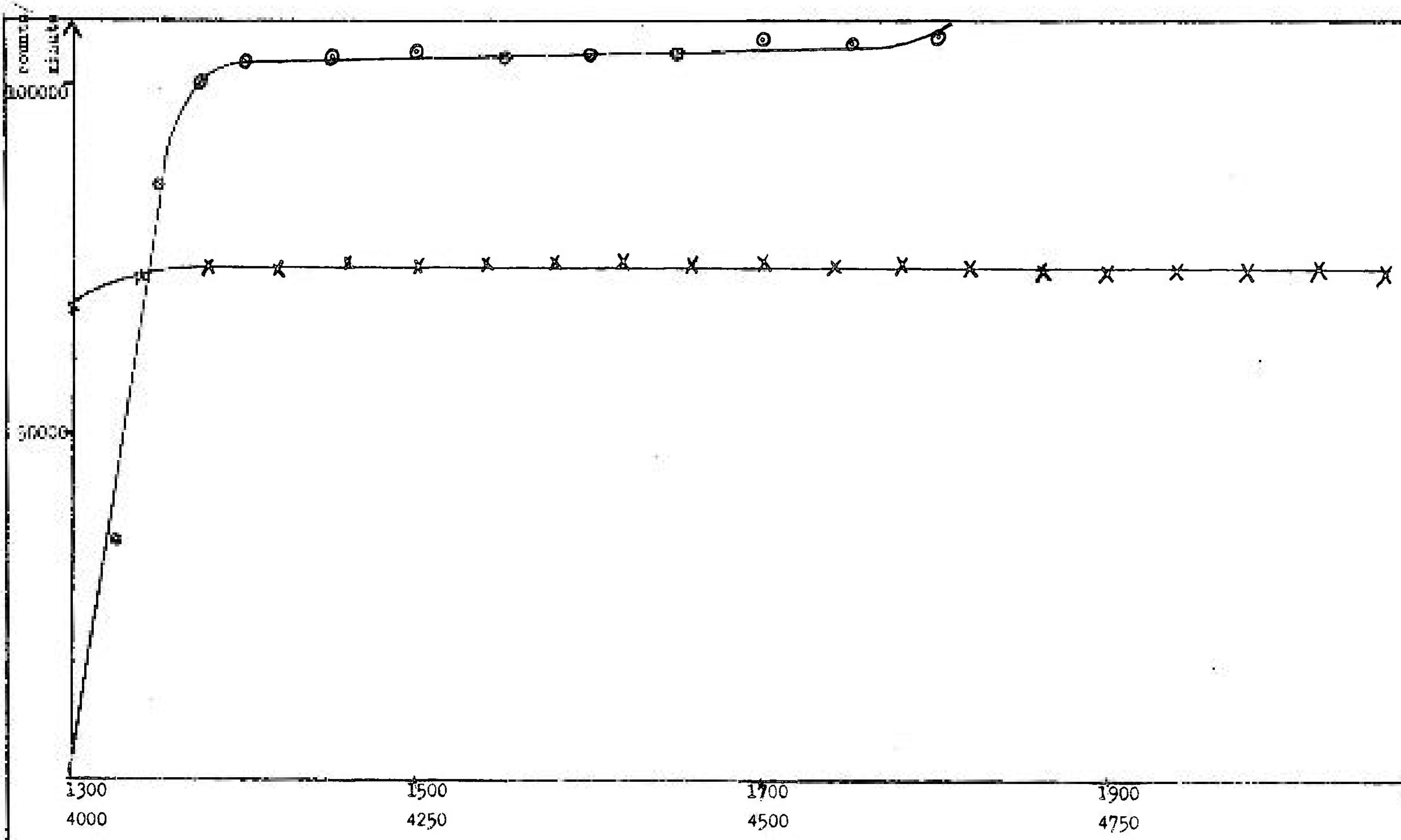

Fig. 3 - Plateaux of the 4π proportional

Source; Co^{60}

Gas: 1,3 % of butane in helium (x)- abcissa: lower scale

propane (⊙) - abcissa: upper scale

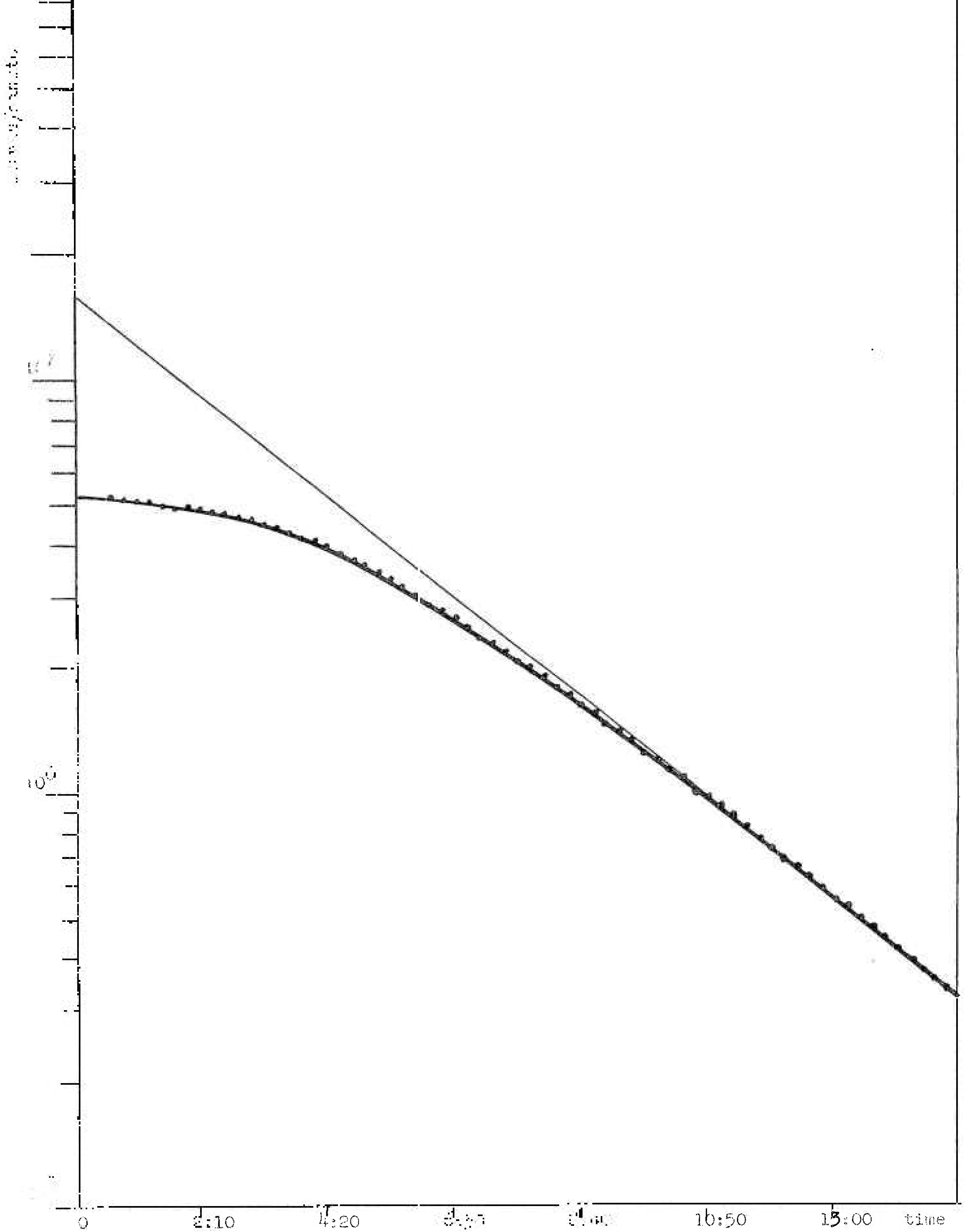

Fig. 4 - Measurement of the dead time
Method: decay of the Mn^{56} activity
Gas: propane

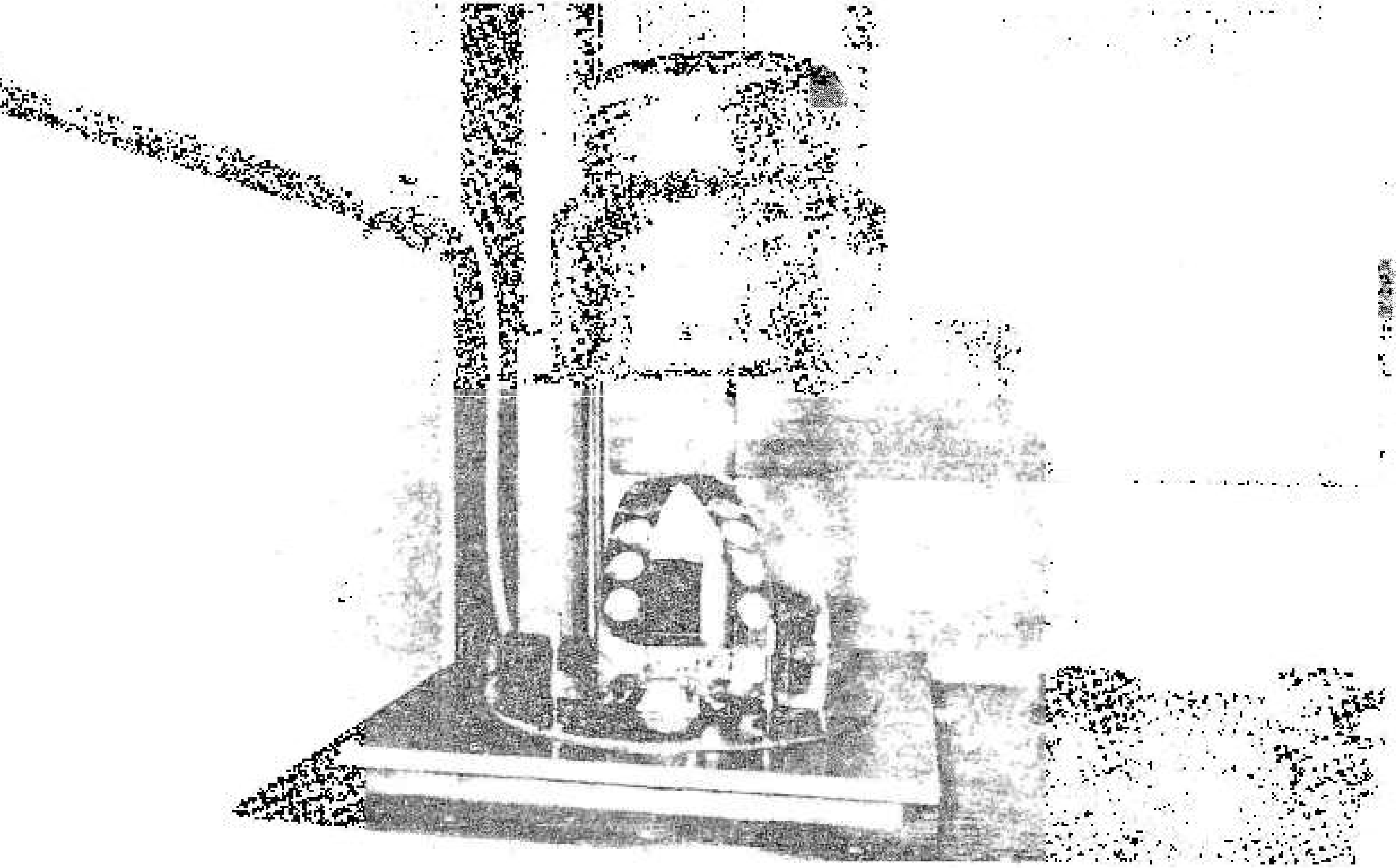

Fig. 5 a - Metallization Chamber

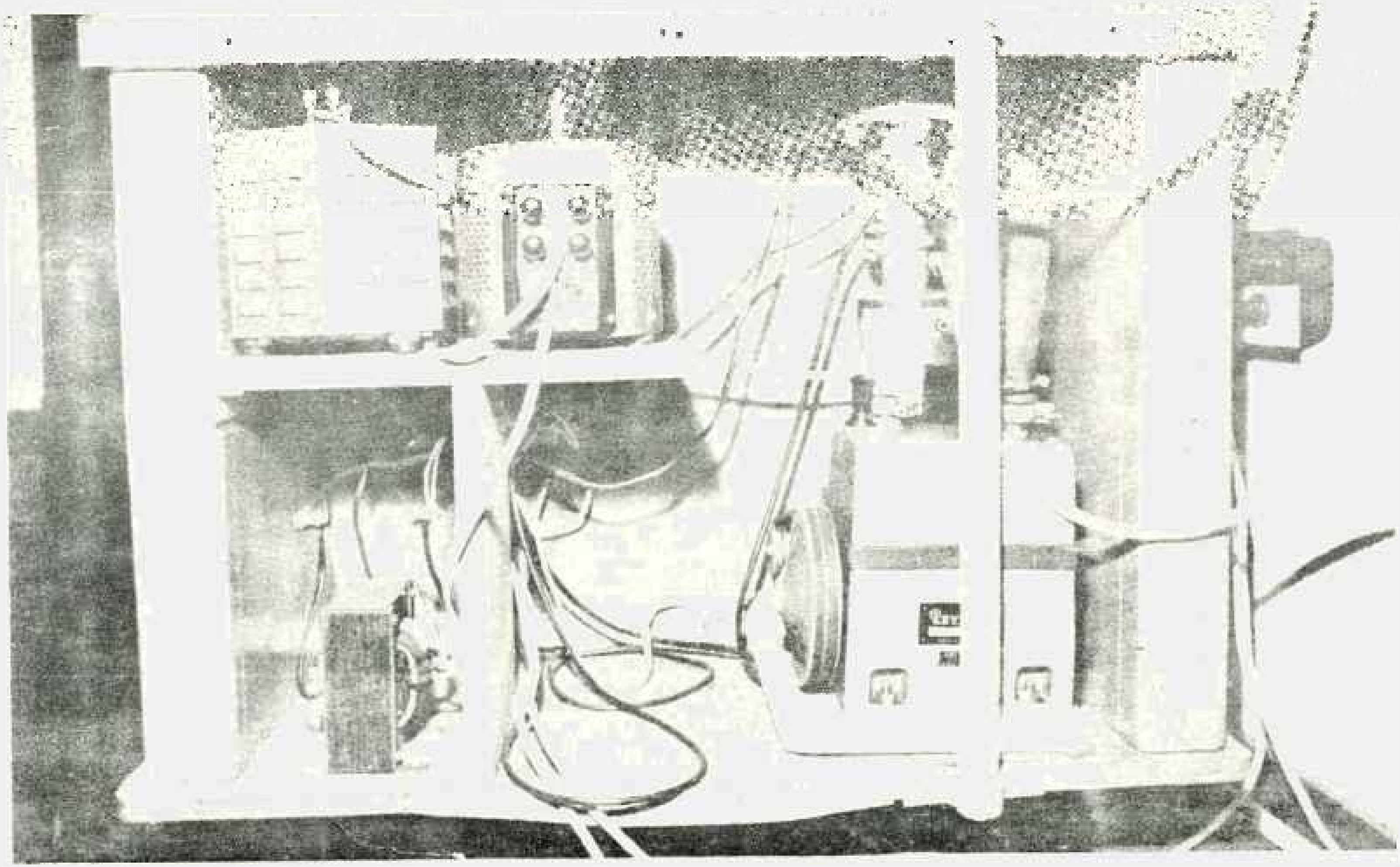

Fig. 5 b - System for metallization of the supporting films in high vacuum

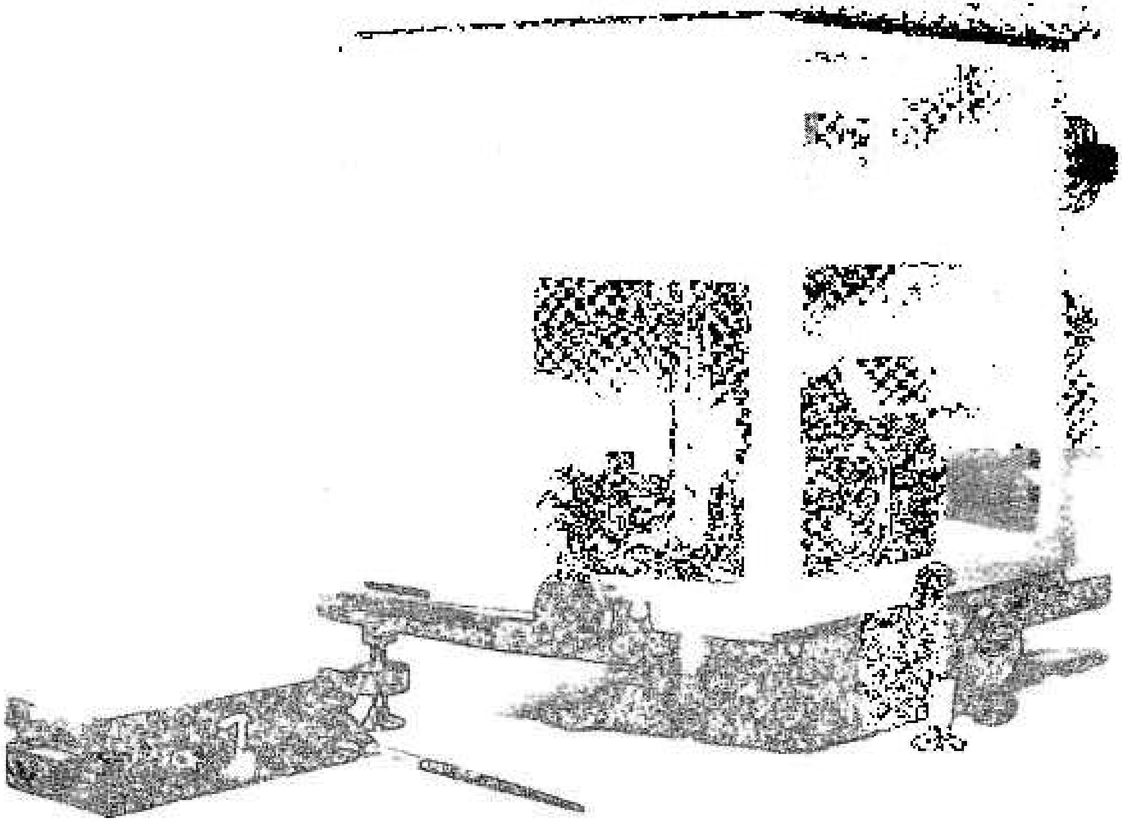

Fig. 6 - Sartorius Werke [illegible]

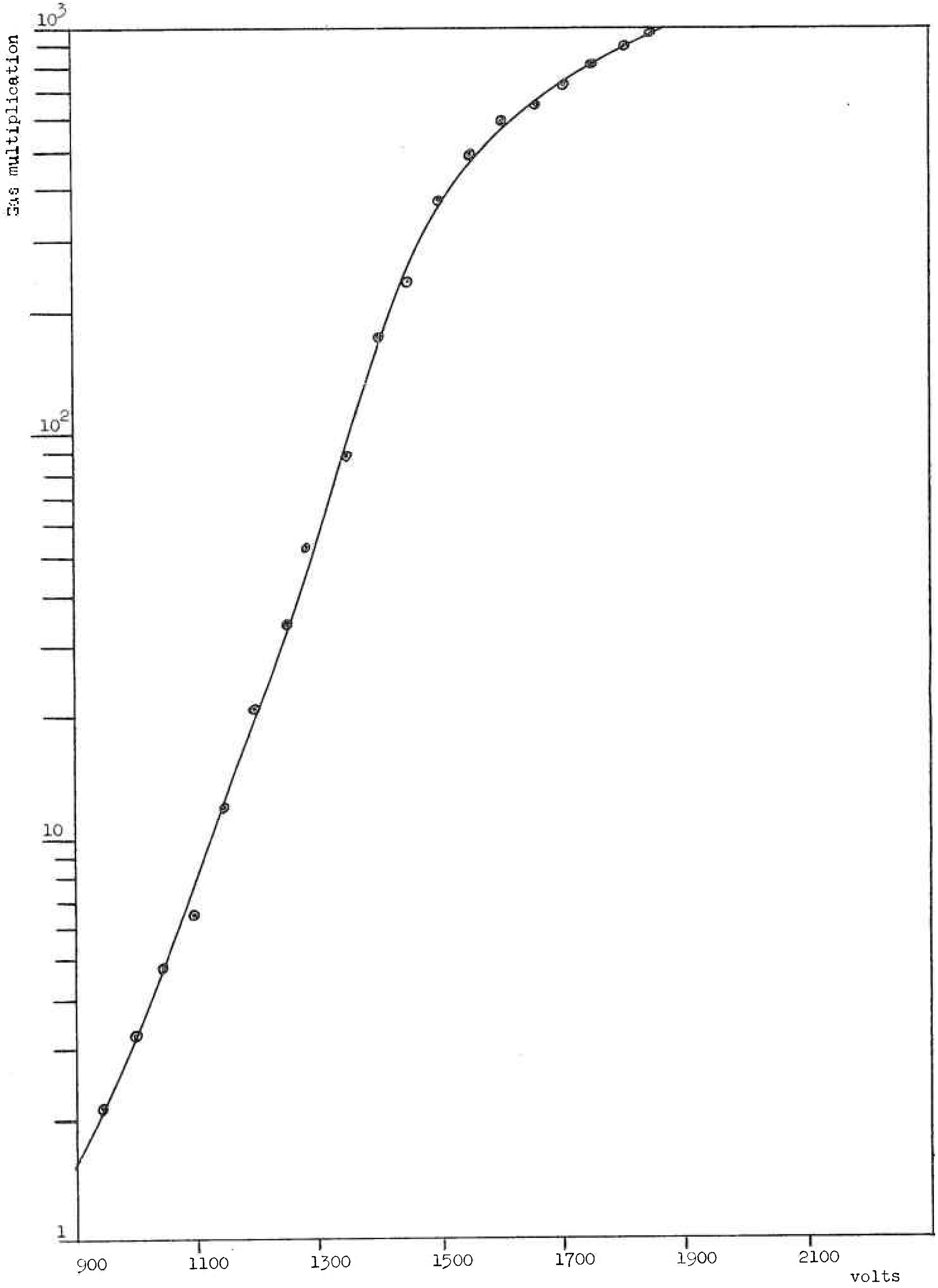

Fig. 7 - Curve of gaseous amplification

Gas: 1,3 % of butane in helium

Source: Po^{210}

Pressure: 1 atmosphere

Counter: 4π proportional

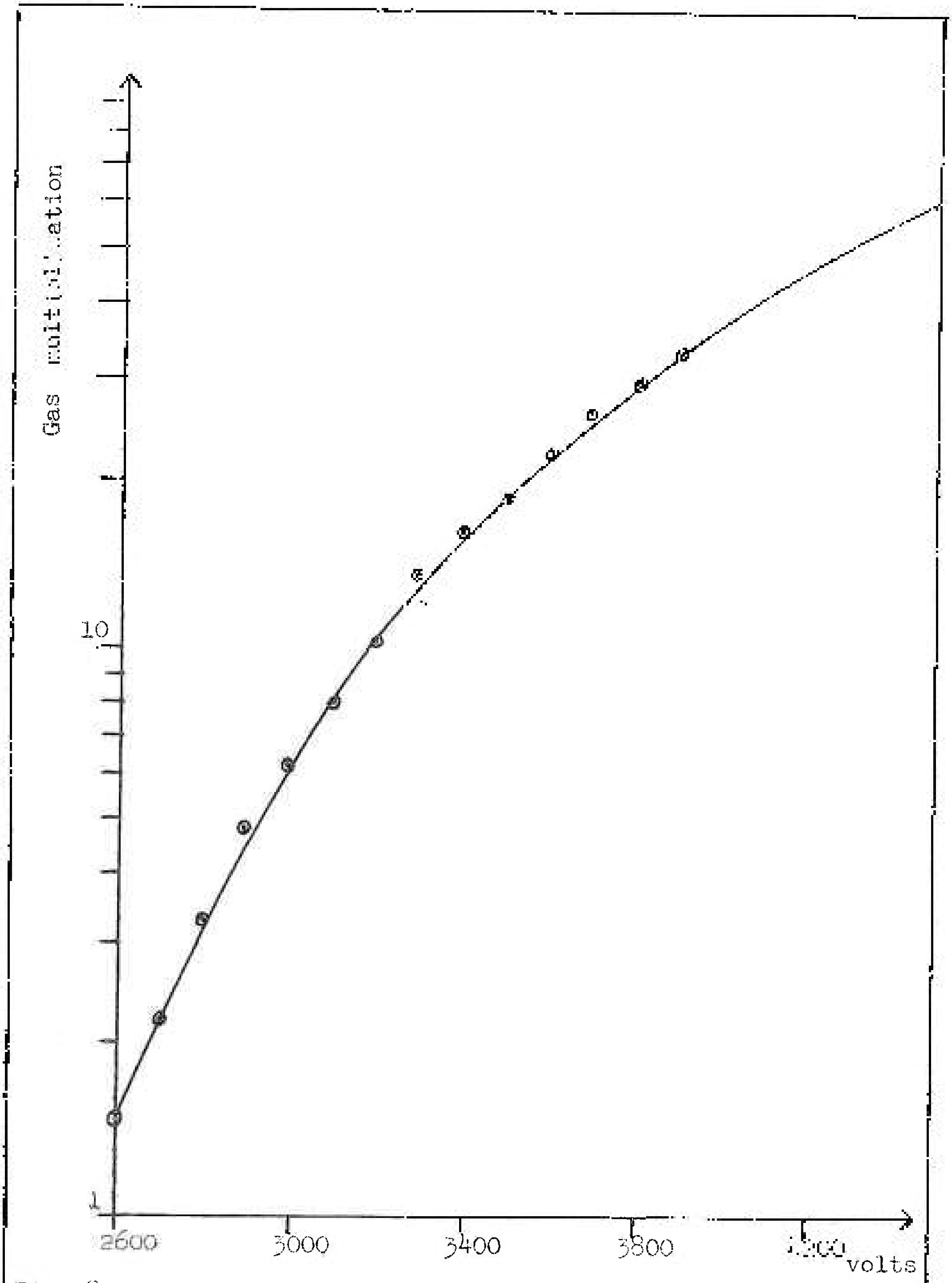

Fig. 8 - Curve of gaseous amplification

Gas: propane Pressure: 1 atmosphere

Source: Po^{210} Counter: 4π proportional