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THE RADIOMETRIC ASSAY OF URANIUM AND THORIUM ORES

AN A. E. R. E. REPORT

by

E. FRANKLIN
R.K. BARNES

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E. Franklin and R. K. Barnes

ABSTRACT

Work on various methods of assay of uranium and thorium in ores by purely radiometric methods are described. Some of the methods are applicable only to ores containing either uranium or thorium separately, and some can be used when a mixture of the two is present in one ore. Methods are discussed also of dealing with ores in which the radioactive constituents are not present in their equilibrium proportions.

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CONTENTS

Page

CONTENTS (Cont'd.)

APPENDICES

ILLUSTRATIONS

ILLUSTRATIONS (Cont'd.)

1. INTRODUCTION

The work of the geophysics team at the Atomic Energy Research Establishment (A.E.R.E.), Harwell, is to develop equipment for prospecting for radioactive ores, geological mapping, mine control, and the radiometric assay of samples. A current descriptive list of equipment available as a result of this work is available on request to A.E.R.E.

The development of assay equipment includes the development of methods of assay which are applicable to the various types of ore and conditions of working, and the purpose of this report is to describe some of these methods. Users of the assay equipments should be able to plan the methods of working most suitable to their applications with the help of this report, and prospective users should find it useful, in conjunction with equipment lists, in deciding what type of apparatus to obtain. Some of the techniques and equipment described may be of interest to those faced with radioactivity assay problems concerning materials other than ores or minerals.

The aim has been to provide purely radiometric methods of assaying ore samples in the form of dry, untreated, powders. Such methods avoid the work involved in dissolving the samples and carrying out lengthy chemical analyses, hence if the number of samples is very large the saving is considerable. In some cases, with heterogeneous low grade samples, the consistency of the results is superior to that given by chemical analysis. This is probably due partly to the difficulties in obtaining strictly representative samples of the smaller size usually used for chemical analysis.

It is proposed to describe first of all the methods arrived at for assaying ores of which the radioactive content is made up either entirely of uranium and its decay products or entirely of thorium and its decay products, the decay products being present in equilibrium proportions in both cases. Methods appropriate to mixed uranium and thorium ores will then be discussed, and, finally, proposals made for dealing with ores in which the radioactive materials are not in secular equilibrium. A method of assaying uranium ores having the radium content in disequilibrium, but in which thorium is known to be absent, is described in A.E.R.E. Memo No. EL/M.76. The present report, particularly its appendices, contains material concerned with the statistics of nuclear counting, on which subject a written lecture is also available (A.E.R.E. Lecture No. EL/L7).

2. METHODS FOR UNMIXED EQUILIBRIUM ORES

2.1 Choice of the Type of Radiation to be Measured

In general, when choosing the type of radiation to measure, we have two conflicting requirements to weigh against each other. We want the radiation detector to have a high efficiency, so that the time required to make the measurements is as small as possible, and we want the radiation to be sufficiently penetrating to emerge from a sample of moderate size without appreciable absorption by the sample material itself. Absorption in the sample decreases the amount of radiation available for the measurement and therefore cuts down the sensitivity of the measurement, and it also introduces errors of measurement which have to be allowed for in some way.

Considering the three types of natural radioactivity in turn:-

2.1.1 Gamma-Ray Measurements

For a given sample size and activity, the self-absorption of the sample is much less for gamma-rays than for alpha- and beta-rays. Unfortunately the efficiencies of all existing types of gamma-ray counter are extremely low, being only about 1/3% for the normal Geiger Müller counter, over the range of gamma-ray energies of interest in ore assay work. This makes it necessary to use a much larger sample than would otherwise be required, which is often inconvenient, and may sometimes be impossible to produce. The larger sample also has greater self-absorption, and, since gamma-ray absorption effects are more complicated and more difficult to estimate or allow for empirically than beta-ray absorption effects, gamma-ray methods generally tend to compare unfavourably with beta-ray methods where low grade ores are concerned. For richer ores containing more than say 0.2% of uranium or 0.5% of thorium, gamma-ray measurements are probably preferable, provided that a sample of the order of 50 c.c. is available.

2.1.2 Beta-Ray Measurements

In general, beta-ray counters have efficiencies very close to 100%, which represents a very big advantage over gamma-ray counters. This advantage is only partly outbalanced by the greater self-absorption which occurs with beta radiation, and far greater sensitivity is attainable by beta counting than is possible with gamma counters of the G.M. tube type.

The greater self-absorption provides a greater error which has to be corrected for, but, due to the more simple nature of beta-ray absorption effects, the error is more accurately calculable.

2.1.3 Alpha-Ray Measurements

Here again we have counting efficiencies in the region of 100%, but the self-absorption which occurs in the sample is extreme. The ranges of the alpha particles in a sample of the atomic number and density of aluminium (the sample grain will generally be denser than this) vary from about 0.0015 to 0.0048 cms. Grinding the sample to a finer mesh than 200 would be laborious and inconvenient for routine work, and even this gives grains of about 0.006 cms in dia.

Thus, even if the sample were only one grain thick, it would in general act as a very thick source for alpha particles, and would present an extremely difficult problem in self-absorption correction, quite apart from the problem of producing a sample which was representative of the whole mass of ore. If all the grains of such a sample were exactly similar chemically and physically, an approximate correction could be made for self-absorption on the basis of the mean density and estimated mean atomic number of the whole sample. Generally, however, the sample is a collection of dissimilar grains and, in order to make a satisfactory correction, a considerable amount would have to be known about the distribution of the active content between grains and their relative numbers and densities. If a thin sample were prepared by evaporation of the dissolved ore, the errors due to dissimilar grains could be eliminated, but solution of an unknown ore is often a tedious and time consuming process, and the small amount of material in the sample would make counting times excessively long for any but the richest samples.

A further objection to alpha-ray counting for uranium arises from the relatively rapid rate of diffusion of radon from the surface layers of the sample into the atmosphere and the consequent loss of the alpha-ray activity of radon and its disintegration products. The effect is of the order of 10%, but is very variable with the type of material and the temperature at which the sample has been kept (see appendix I). The effect is not so serious with thorium, since the rapid rate of decay of thoron (half-life 54.5 secs, as compared with 3.83 days for radon) does not give time for any serious loss by diffusion.

2.1.4 Conclusions

For fairly high grade ores, i.e., those containing 0.2% of uranium or 0.5% of thorium or more, the low self-absorption with gamma-rays gives a convenient and accurate measurement provided that a sample of about 50 c.c. is available. For ores of lower grade and in cases where only a small amount of material is available, beta-ray counting gives better results.

Alpha-ray measurements on powdered samples are in general unreliable, and thin samples prepared by evaporation of a solution of the ore gives very low sensitivity. There are special cases, however, where alpha-ray counting is valuable and one or two of these will be mentioned.

2.2 Gamma-Ray Method

2.2.1 General Precautions Necessary in Gamma and Beta-Ray Assay Work

2.2.1.1 Disturbance of Secular Equilibrium by Grinding

Fine grinding of the ore leads to a loss of radon, thoron, and actinon gases to an extent which is determined by the fineness of grinding and the amount by which the grinding raises the temperature of the ore. The rate of diffusion of the gases out of the material increases rapidly with increase of temperature and it is difficult to assess the amount of disturbance likely to arise for a practical grinder. It can easily cause a change of count rate of several per cent and, in the case of radon, recovery of equilibrium requires a period of several times the four day half-life of radon.

Disturbance of the powdered ore subsequent to grinding for the purpose of making samples does not cause such large effects, but it is desirable to seal the sample holder against the loss of radon, particularly when counting beta-particles.

The effects of loss of thoron or actinon in this way are unlikely to be found troublesome owing to their very short decay periods (54.5 secs and 3.9 secs half-life, respectively.

2.2.1.2 Grain Distribution

The distribution of different types of grain affects the measured count rate, particularly when counting beta-particles. The sample should therefore be thoroughly mixed and pressed into the sample holder rather than shaken down by tapping or vibration. The larger and the less dense grains tend to rise with shaking.

When beta or gamma-ray samples are required in a fairly permanent form, it is often convenient to mould the powder into a solid block with some form of plastic, such as distrene. The grain distribution is then fixed, and the sample also is sealed against the escape of radon. The powder is mixed with a solution of the plastic in a solvent such as acetone, the whole dried, reground coarsely, mixed and moulded into the required shape at about 240°C.

The suitability of any procedure of mixing can be checked by re-mixing and re-counting a sample a few times, to see if any statistically significant variation occurs in the count rate observed.

2.2.1.3 Reliability of Geiger Müller Tubes

Measurements made with Geiger Müller counting tubes are always liable to errors resulting from spurious counts produced by the tubes themselves. It is found that one of the chief causes of plateau slope in Geiger Müller tubes is the variation in the rate of spurious count production with supply voltage, and that variations of apparent sensitivity with time and temperature also are largely spurious count effects. A large proportion of these spurious counts arise from conditions left in the tubes by previous counts, and it is therefore found that they tend to occur within a few hundred microseconds after a genuine count(1). It is to be expected therefore that they can be largely eliminated by rendering the tube inoperative during such periods, and it is found that great improvement can be effected by reducing the applied voltage to a value lower than the threshold voltage for three or four hundred microseconds after each pulse. This is therefore now standard practice, the facility being provided by a standard "quench" probe unit (Probe Unit Type 1014) designed to operate from the power pack of the scaling unit used for counting.

Of the various types of tubes used, the end window beta counters have been found to exhibit the greatest temperature effects. This is to be expected, of course, in view of the much greater end effects in this kind of tube. These have temperature coefficients which may be as high as 1% per °C., or more, when the full supply voltage is applied continually, but a reduction to about 0.2% per °C. results when the quench probe unit is used. The temperature coefficients of the cylindrical beta- and gamma-ray counters vary from type to type but generally speaking are about 0.06 - 0.1% per °C., when the quench probe is used.

Apart from variations with temperature and applied voltage, two other types of sensitivity change occur. As the tube ages, a gradual reduction of sensitivity begins to appear, slowly at first but rapidly towards the end of the tube's life. Superimposed on this we have a random variation of a few per cent, with no apparent cause. The latter type of variation is almost entirely eliminated by use of the quench probe, or its onset considerably delayed, but the former type is not affected. However, variations of this kind are slow, and their effect on assay results can be effectively eliminated by a suitable sequence of counting. A complete series of counts is made on the unknown sample, the standard sample and background, if this is not negligible, the time for each count being some short period, of the order of ten minutes. The whole sequence of three measurements is then repeated over and over again until the total count for each is at least that required to give the required statistical accuracy (standard deviation = $\sqrt{\text{number of counts}}$), the division of time between the three being chosen for maximum overall accuracy (see appendix VII). Any sensitivity changes which occur tend to affect all readings of each set to much the same extent, as the time per set is short, and the process of averaging to obtain the mean rates automatically allows for drifts.

2.2.2 The Apparatus (Assay Unit Type 1045A)

The powdered ore sample to be assayed is poured into a standard sample bottle to a standard level, being pressed down gently with a flat ended instrument sufficiently to prevent any appreciable change of level during subsequent normal handling. The results given apply to bottles of 2.3 cms diameter, with 0.7 mm. thick glass walls, filled to a height of 8 cms. A screwed cap provides sealing.

Four Cinema Television Type G.M.4B Geiger Müller tubes are used, these having cathodes of 2.2 cms dia. and length 11 cms. They are arranged at the corners of a square of side 6 cms and a stand is provided in the middle of the square for the sample bottle. An annular tray surrounds the sample bottle stand, and this is used to support annular radiation absorbers between sample and tubes, when rich samples are being assayed. The fixed shielding cylinder to which the clips supporting the G.M. tubes are attached ensures that no beta-particles from the sample are counted. The positioning of the sample bottle with respect to the G.M. tubes is not critical, since it is at the centre of an equidistant set of four and any slight change of position nearer to the tubes on one side is fairly accurately compensated for by the increase in distance from those on the other side.

The assay unit fits into a lead box (or castle, as it is generally called) and can be swung out on a pivot for changing samples, absorbers or G.M. tubes, or for making adjustments. The lead castle (Type 1065) is $5\frac{5}{8}$" x $5\frac{5}{8}$" x 9" high inside and has $1\frac{1}{2}$" thick walls which eliminate most of the gamma-ray background. The back of the castle has fittings to take the standard quench probe unit Type 1014 and this connects into standard counting equipment by a 6-way screened cable. The assay unit and lead castle are illustrated in Plate 5.

The standard counting equipment consists of the following units, which can be fitted conveniently onto a standard 3 ft. enclosed rack:-

(i) Scaling Unit Type 200 (or Type 1009)

(ii) Power Unit Type 1007 with Potentiometer Type 1007.
(or Power Unit Type 1082)

(iii) Timing Unit Type 1003.

The scaling unit consists of an electronic scale of a hundred, with neon indicators to indicate the count, followed by a mechanical counter which counts the hundreds on four digits. Six figure numbers can therefore be registered. The resolving time of the electronic circuits is 5 microseconds, but, when used with G.M. tubes, it can be increased artificially to a time which is variable between 300 microseconds and 1000 microseconds. It is normally set to 300 microseconds, while the length of the quenching pulse from the Probe Unit Type 1014 is set to the maximum of 500 microseconds. The paralysis time of the equipment is therefore defined solely by the quench probe unit and, when this has been determined accurately, paralysis time corrections to count rates observed can be made on the basis of the curve shown in Fig. 9 (see also appendix II).

Power Unit Type 1007 provides a stabilised supply of high voltage for operating the G.M. tubes. The power unit gives a fixed potential of 2200 volts, and this is reduced to that required by the tubes by the Potentiometer Type 1007. Up to ten potentiometer units can be supplied from one power unit if several sets of equipments are to be operated at once. Power Unit Type 1082 can be used alternatively to supply an individual counting set.

Timing Unit Type 1003 can be set to do one of two things. It can switch on the scaling unit for a preset time and record the number of operations of the mechanical counter in the scaling unit, or it can measure the time required for a preset number of counts to be registered by the scaling unit. The preset times are variable from 0.5 secs to 100 mins, and the preset counts from 10 to 10,000. If required, of course, counts can be timed by means of a stop watch.

A general view of a three channel counting system is shown in Plate 1.

2.2.3 Experimental Procedure

Before commencing assay measurements the stability of the equipment is first checked by taking a series of about ten one-minute counts with a rich sample. If the fluctuations are entirely statistical and not contributed to by defects in the equipment, then the probability that the sum of the squares of the differences of the readings from their average shall differ from the sum of the readings themselves by a ratio of more than 4 to 1 either way is only 4.6%. If the ratio is greater than this, or if the background reading with the four tubes inside the lead castle is greater than 200 per minute, some fault is suspected in the G.M. tubes or in the counting equipment. Any variation of sensitivity of the tubes or intermittent missing of the counting equipment would tend to make the sum of the squares of deviations from the mean too great, while any regularising effect, such as multiple pulsing of the G.M. tubes or multiple triggering of the counting equipment, would tend to make it too low.

If the equipment is found to be operating satisfactorily, measurements are commenced on the unknown sample, a suitable standard sample (prepared in the same way as the unknown), and background, if this is not negligible. The series of readings is repeated until the sum of counts in each one is sufficiently high to give the required statistical accuracy, and the mean rates are then calculated. It is then necessary to apply density corrections to the readings, as described in the next section, if the best available accuracy is required.

2.2.4 Density Corrections

Self-absorption of the gamma-rays in the sample has the effect of reducing the count rate by a variable amount which may be up to 15% and it is therefore necessary to make a correction for this, if the weights of the unknown sample and of the standard sample are different. This can be done by means of a correction curve such as that shown in Fig. 10, which gives the counting rate per per-cent uranium or thorium as the case may be, plotted as a function of the weight of sample filling the bottles. The curve was obtained by use of a series of samples of various densities which were produced by diluting a rich sample of pitchblende, in the case of uranium, and of monazite sand, in the case of thorium, with materials of various densities. The diluants used were the oxides of lead, iron, copper, silicon and magnesium.

The use of this method eliminates the effect of differences in closeness of packing, provided that the bottles are always filled to the same height. The bottles used for samples, both unknown and standard, must be similar as regards cross-section and wall thickness and must be filled to a standard height, otherwise corrections for radiation geometry must be made.

The density correction is not a perfect one, since self-absorption is a function not only of density and packing, but also of the atomic numbers of the materials present. This is due to the superposition upon the Compton recoil absorption, of photo-electric absorption and pair production, which take place to a disproportionate extent in elements of high atomic number. This modifies the self-absorption from that to be expected by consideration of density alone and results in an ultimate indeterminacy which may be as high as ± 3% of the count rate for the sample and G.M. tube disposition used. The effect was checked by combining two diluants in each sample to give a series of samples of constant richness and constant density, but with varying atomic number. The error can be reduced to a minimum by using a standard made up of an ore of the same general type as the unknown.

Having determined the mean count rates for the unknown sample, the standard sample and the background, and read off the density correction ordinates appropriate to the weights of the two samples, the uranium or thorium content of the unknown sample is given as:-

$$\text{Percentage in standard sample} \times \frac{\text{count rate with unknown} - \text{background count rate}}{\text{count rate with standard} - \text{background count rate}}$$

$$\times \frac{\text{density correction for the standard}}{\text{density correction for the unknown}}$$

2.2.5 Accuracy and Sensitivity Attainable

The normal background counting rate is about 100 per minute. This is equal to the increase of count rate obtained with a sample containing 0.1% of uranium or 0.25% of thorium.

If we allow an hour for completing all counting, including unknown sample, standard and background, an ore containing x% of uranium can be assayed as containing [x ± (0.03 x + 0.02)]%, e.g. (10 ± 0.3)%, (1 ± 0.05)% or (0.1 ± 0.02)%. The coefficient of x in the error represents the 3% error in density correction and the 0.02% constant error is the statistical error. The latter error could be reduced by increasing the counting time, but the time required increases rapidly when the error gets down to a small fraction of background. For thorium, an x% ore can be assayed as [x ± (0.03 x + 0.05)]% in one hour.

These figures apply to the particular geometry described and would vary considerably with tube and sample shape.

2.2.6 Possibilities of Improvement

Since the mean efficiency of G.M. tubes for detecting gamma-rays of the uranium and thorium series is only about 1/3%, there are very considerable possibilities for improving the sensitivity of gamma-ray assay methods by improvement in detection efficiency.

Gamma-ray scintillation counters, using naphthalene, anthracene, or some other type of scintillating medium, give efficiencies of the order of 10-20%. This would represent an improvement of 30-60 times in sensitivity, or, alternatively, would give an improvement by this ratio in the fixed component of the errors given for an ore of given richness within the same time limit.

2.3 The Beta-Ray Method, using End Window G.M. Tubes

2.3.1 Apparatus

Views of the lead castle (Type 1100) used for this work appear in Plates 1 and 2. The lead box is divided into horizontal sections for ease of carrying, each section weighing about 20 lbs. Each section is recessed into the one below to prevent any relative sliding during normal use.

The G.M. tube used is the standard G.E.C. type G.M.2 with an argon and alcohol filling. The circular end window of slightly under one inch diameter is of 0.001" copper foil which will pass beta-particles of energies in excess of 200 KeV.

The sample holder screws into a slide which can be slid into one of the pairs of slots in the compartment under the G.M. tube, and is arranged to come to rest immediately below the G.M. tube window.

A quench probe unit is attached to the back of the lead castle, as before, and connection made to the G.M. tube by means of a flexible polythene insulated wire passing through a hole in the lead wall. The remainder of the counting equipment used, i.e., scaling unit, power unit and timing unit, are exactly the same as those described for gamma-ray counting.

2.3.2 Choice of Form of Sample

In the arguments which follow, a sample whose depth is greater than the maximum range of the highest energy beta-particles emitted is referred to as a deep sample. Such a sample is equivalent to one of infinite depth, as far as the beta-particles are concerned. A sample whose depth is so small that the loss of beta-particles by internal absorption is negligible is called a thin sample. We are free to choose the depth of our sample at either of these extremes, or at any intermediate value, in relation to its effect on the accuracy and sensitivity attainable.

The accuracy attainable depends upon the extent to which the beta count rates per per-cent uranium or thorium are liable to be varied by factors which cannot be easily measured or taken into account. As mentioned when describing the gamma-ray method, one of the most important of these is the fact that the absorption properties of the sample do not depend simply on density, but also on atomic number distribution.

It can be shown that, if the mass absorption coefficient of the sample material is constant, the count rate per per-cent of active material is independent of density, if a deep sample is used (see appendix IV). However, the mass absorption coefficient does vary slightly with atomic number and this results in a counting error. The error could be largely eliminated by taking measurements on a range of samples of different thicknesses, but the length of time to obtain a result would then be very considerably increased.

It is found that for a thick sample, or for a sample of an intermediate depth sufficient to give a reasonable sensitivity, the error caused by variation of mass absorption coefficient is about ± 3%. This is the same in magnitude as that found in the gamma-ray assay work, but it is made up in a different way. The fractional change of mass absorption coefficient is far less in the beta-ray case but it occurs as an error on a much larger self-absorption correction than occurs in the gamma-ray case.

A further factor which is affected by sample depth is the modification of count rate resulting from non-uniformity of mixing of either the constituents of an artificially produced standard sample or of the constituent parts of an ore sample. This effect was measured by repeated counts and remixing of a few typical samples. It was found that the mixing was unsatisfactory for thin samples, giving a standard deviation of about ± 10% in count rate, but quite satisfactory in deep and intermediate samples.

In view of this, and also of the very long counting periods required with thin samples, the choice is narrowed down to one between thick and intermediate ones. There is little to choose between these two as far as straight beta assay is concerned, but the deep sample has been used as being more suitable for measurement of uranium/thorium ratio by the beta/gamma ratio method described in section 3.2. The sample depth is 2.54 cms and the holder volume is 13 c.c. The holders are made in the form of light anodised aluminium cups to facilitate accurate weighing of the sample and are sealed by a 0.001" aluminium foil disc laid on top of the powder and held in place by a screwed bezel ring. The presence of this aluminium foil between the sample and the G.M. tube reduces the count rate only very slightly.

2.3.3 Density Correction

Density effects were measured here in the same way as for gamma-ray assay and curves of mean density effect plotted against sample weight were obtained. As already mentioned, deviations of up to ± 3% from these curves occur due to variations of mass absorption coefficient. Curves a and b of Fig. 6 show the variation of count rate per per-cent uranium and per per-cent thorium plotted as a function of density. The curve marked "Monazite" gives similar figures for monazite sand containing 9% of thorium and 0.36% uranium. Curve b for thorium was derived from this by subtracting the appropriate count rate for the uranium content by the aid of curve a.

2.3.4 Experimental Procedure

A standard sample having an active content fairly near to that of the unknown sample is selected and the weights of standard and unknown filling the holders are measured. A sequence of counts, unknown, standard, and background, is made, taking about 5-10 minutes for each count and the sequence is repeated until the total counts for each number adds up to the value appropriate to the statistical accuracy required. The average count rates for the unknown and standard are then corrected for background and probe unit paralysis time (Fig. 9) and the apparent uranium or thorium contents of the two samples determined from the calibration curves (Fig. 6). The value of the active content of the unknown is then adjusted by the ratio by which the apparent content determined for the standard differs from the known content. This last step allows for the change of sensitivity since calibration.

2.3.5 Sensitivity and Accuracy

The background counting rate of the G.M. tube with no sample present is about 8 or 9 per minute and this is equivalent to the increase in count rate produced by an ore sample containing 0.01% of uranium or 0.05% of thorium.

If we allow up to about 3 hours to complete all counting, including unknown sample, standard sample and background, we can assay an ore containing x% of uranium to an accuracy of ± (0.03 x + 0.005)%. A thorium ore assayed in the same time would be subject to an error of ± (0.03 x + 0.025)%.

2.4 The Beta-Ray Method Using Cylindrical Thin Walled G.M. Tubes

2.4.1 Apparatus (Assay Unit Type 1044A)

As can be seen from section 2.3.5, the sensitivity of the end window beta-ray method is very little greater than that of the gamma-ray method, owing to the very poor geometry of the sample and G.M. tube arrangement used.

Very great improvement in sensitivity and reduction of counting times can be obtained by the use of a cylindrical thin walled G.M. tube surrounded by an annular holder containing the sample (see Fig. 5). The G.M. tube used is the B6E tube made by Messrs. 20th Century Electronics. It has a cathode of 1.6 cms dia. with a sensitive length of 6 cms. The glass wall has a thickness of 24 mgms/sq. cm., and the cathode is formed by an aquadag coating on the inside surface.

The sample holder and tube assembly are designed to fit into a standard lead castle (Type 1065) and can be swung out on a pivot for sample changing, etc. (see Plate 4). Two sizes of annular sample holder are used, one having a volume of 90 c.c., and the other a volume of 13 c.c. (same volume as the end window deep sample holder) (see appendix III f r notes on choice of shape of sample and G.M. tube). The small holders are generally used and the large ones reserved for the weakest samples. The small holders are placed on a carrier which rests inside one of the large holders retained for the purpose. A cylinder of 0.001" thick copper foil soldered down the seam, forms the window for beta-particles on the inside of the annulus. Diagonally slotted phosphor-bronze split rings seal the copper foil against the ends of the holders. The joints are water-tight without sealing and can be taped with p.v.c. tape to give a gas-tight seal. The holders have been designed for easy cleaning, but different ones are generally used for samples of very different orders of richness. A specially shaped funnel is used for filling, the centre being filled in so as to guide the powdered ore into the annular opening of the holder. An open mesh guard round the tube cathode protects the thin wall when withdrawing from the assembly and acts as a guide for re-insertion.

2.4.2 Experimental Procedure

Shaking or tapping down of the powdered sample has been found permissible in the case of the annular sample. This is undoubtedly due to the fact that the window is vertical, and therefore the tube views a vertical cross-section of all layers made dissimilar by gravitational separation.

The experimental procedure, density corrections, etc., are exactly similar to those for the end window equipment.

2.4.3 Sensitivity and Accuracy

The normal background count rate inside the lead castle with no active sample present is about 12 per minute. This is equal to the increase of count rate produced by an ore containing 0.0005%

of uranium or 0.002% of thorium when the 90 c.c. sample holder is used, and 0.001% of uranium or 0.005% of thorium when the 13 c.c. sample is used. The effect of tube and sample shape on sensitivity is discussed in appendix III.

If we again allow up to 3 hours for completing all measurements, we can assay:-

$$\left.\begin{array}{l}\text{an } x\% \text{ uranium ore as } x\% \pm (0.03x + 0.00025)\% \\ \text{or " } x\% \text{ thorium " " } x\% \pm (0.03x + 0.001)\%\end{array}\right\} \text{using the 90 c.c. holder.}$$

$$\left.\begin{array}{l}\text{and an } x\% \text{ uranium sample as } x\% \pm (0.03x + 0.0005)\% \\ \text{or " } x\% \text{ thorium " " } x\% \pm (0.03x + 0.0025)\%\end{array}\right\} \text{using the 13 c.c. holder.}$$

2.5 Alpha-Ray Counting Methods

2.5.1 Introduction

No methods of assay depending on straight alpha-ray counting have been developed, owing to the difficulties set out in section 2.1.3. However, a number of possible special applications of alpha-ray counting suggest themselves, and a certain amount of work has been carried out on them.

All alpha-ray counting is at present done with the scintillation counter. This consists essentially of a transparent screen, coated on one side with a phosphor, such as silver activated zinc sulphide, with a photo-electric cell and electron multiplier mounted on the other side. The alpha-particles strike the coated side of the screen and cause scintillations in the phosphor, and these are detected and amplified by the photo-multiplier to a pulse of a suitable size to feed into the standard counting equipment.

2.5.2 Examination of Thin Slices of Rocks

By cutting thin sections from a rock of such a thickness as to be transparent, the general structure of the rock can be examined. It would often be useful to measure the distribution of uranium or thorium in a relation to the visible features of the section and this may be done by means of a scintillation counter with a very small phosphor screen. The technique of autoradiography(5), whereby alpha particle sensitive emulsions are exposed against a polished rock surface and an enlargement of the result compared with a microphotograph, gives far greater definition, however.

2.5.3 Measurement of Locking Factor

In the process of mineral dressing a property of the ground ore which it would often be useful to know is the so-called "locking factor" of the particles of the active mineral concerned, i.e., the extent to which the particles of the uranium or thorium bearing constituents of the ore are enclosed by inactive material. This can sometimes be determined by microscopic examination of a number of grains, but the method would be tedious, as a routine practice.

The ratio of beta-ray counts to alpha-ray counts would give an indication of the active area of the average grain, as a function of the volume or total area, since the beta-particles would not be appreciably stopped by the enclosing inactive material but the alpha-count would come mainly from those parts of the grains which are active on the surface.

3. METHODS FOR MIXED URANIUM AND THORIUM ORES

3.1 Introduction

In order to determine the amounts of both the uranium and thorium in a mixed ore, at least two measurements must be made and the conditions must be such that the two readings are contributed to in different proportions by the uranium and thorium content.

In such a case, the accuracy is ultimately limited by the extent to which the ratio of readings is varied by the ratio of uranium to thorium, in relation to the extent by which it is modified by factors which cannot easily be eliminated or allowed for. Among these may be mentioned inaccuracies in density corrections, statistical fluctuations, etc.

If the response of the measuring equipment varies linearly with radiation intensity and the geometry of the system is standardised, the two readings can be expressed in the form:-

$$N = aU + bT + n \text{ and } F = cU + dT + f$$

where N and F are the two count rates observed, U and T are the percentages of uranium and thorium present, a, b, c, and d are functions of geometry, sample density, etc., and n and f are background count rates.

The constant a, b, c and d can be plotted as functions of density by measurements on a number of known samples and the simultaneous equations can then be solved for U and T.

A number of possible radiometric methods providing pairs of readings, N and F, offer themselves for consideration and some of these are described below, in the order in which they have been investigated.

The calculation of errors arising from statistical errors in N and F and from unpredictable errors in a, b, c and d are dealt with in appendix V.

3.2 Beta/Gamma Ratio Method

3.2.1 Introduction

The method consists essentially of measuring the amounts of beta and gamma radiation from the sample under standard conditions. The beta radiation is measured by the normal process described in sections 2.3 or 2.4 and then a measure of the gamma-ray emission is obtained by inserting an absorber between the sample and the counter to eliminate the beta-rays.

The apparatus used is the same as that used for the straight beta-ray assay methods of sections 2.3 and 2.4, and the same precautions are needed to avoid errors. The method was developed using the end window counting equipment and most of the figures given refer to this, but the use of the cylindrical thin walled tube and annular sample gives much improved sensitivity, or alternatively, much improved accuracy in a given time.

When considering this method, constants a and b (see above) represent beta plus gamma counts per second per per-cent of uranium or thorium, respectively, and c and d represent gamma counts alone per per-cent of uranium or thorium. Each of these constants is subject to variations due to self-absorption, and those are corrected for by taking the values of the constants appropriate to the sample density, i.e., weight of material filling the sample holder (see Figs. 6 and 7).

3.2.2 Density Correction

The effect of density on the constants a, b, c and d was determined by making measurements on a series of synthetic samples of different densities and constant richness. The active component of the samples was pitchblende in the case of uranium and monazite sand in the case of thorium, and the diluants used were the oxides of lead, iron, copper, silicon and magnesium. These gave a wide range of distribution on the atomic number scale and were therefore useful in indicating the error to be expected due to variation of mass absorption coefficient, i.e., errors not correctable on the basis of sample density. Such errors were investigated by combining two diluants in each sample to give a range of samples with constant richness and constant density, the densities chosen being 0.6, 1.5, 2.4 and 3.3. (Density here means weight of sample packed into the holder divided by its volume. The microscopic density of the powder would be greater.)

The results of these tests showed variations of a, b, c and d due to changes of mass absorption coefficient at constant density of ± 3%, ± 3%, ± 5% and ± 5%, respectively, errors of this order being observed over the whole range of densities. Mean curves of a, b, c and d plotted against density are given in Figs 6 and 7, for the end window counter, the values of b and d being corrected for the known uranium content of the monazite sand, on the basis of the curves of a and c obtained with the pitchblende samples.

It was found that by use of a source of only one tenth of the thickness of the deep holder normally used with the end window counter, the variation of c and d (the gamma ray constants) due to variation in mass absorption coefficient was reduced to ± 1%. However, the gamma-ray count rate was then reduced to an impracticably low level, and therefore the deep sample was retained.

The coefficients of correlation between the variations in the values of a, b, c and d (due to mass absorption coefficient variations) were measured. Variations of a and b showed 80% correlation and those of c and d about 50%. Virtually no correlation was displayed between any other pairs of the four variables.

In order to reduce the amount of calculation during an assay, the coefficients A, B, C and D in equations

$$U = AN - BF$$
$$\text{and} \quad T = -CN + DF$$

are calculated from a, b, c and d according to the following relations:-

$$A = \frac{d}{ad-bc}, \; B = \frac{b}{ad-bc}, \; C = \frac{c}{ad-bc} \; \text{and} \; D = \frac{a}{ad-bc}$$

These coefficients are plotted against sample weight in Fig. 8.

3.2.3 Experimental Procedure

A standard sample is chosen to represent the unknown ore as closely as possible, and the weights of the standard and unknown filling the holders are measured. Each is counted for an interval of not more than 10 minutes, with and without a filter, and the sequence of four measurements is continued until the total for each member is sufficiently high for the required statistical accuracy (probably about 5000). Average count rates are then determined and the appropriate corrections applied for paralysis time losses and background, and the apparent uranium and thorium contents determined by the aid of the calibration curves (A, B, C and D). The values for the unknown are then adjusted by the ratio by which the apparent contents of the standard differ from the known contents. This last stop allows for change of sensitivity since calibration.

3.2.4 Accuracy and Sensitivity

3.2.4.1 Using the End-Window Counting Tube

Using this equipment, with samples of density 2.4, the values of a, b, c and d at the time of calibration were 807, 158, 73 and 34 nett counts/min. per per-cent, respectively. If we insert these values into equation 10, appendix V, together with those of the errors, $\frac{a-a'}{a}$, etc., taking into account the mean effect of different possible sign combinations, where indicated by the lack of correlation, we have:-

$$\frac{U-U'}{U} \times 100\% = \sqrt{\left[\tfrac{1}{2}\left(7-2.7\,\frac{T}{U}\right)^2 + \tfrac{1}{2}\left(3.4+0.85\,\frac{T}{U}\right)^2\right]}\,\%, \qquad \ldots\ldots(11)$$

U and T being the uranium and thorium contents per cent and U' the apparent uranium content.

This equation gives the accuracy to which the uranium content can be determined for any given value of uranium/thorium ratio, provided that the uranium content is not less than 0.1%, and that sufficient counting time is available for the effect of statistical fluctuations to be overshadowed. Substituting T/U = 25, the accuracy is given as 50%, indicating that the uranium is just detectable when twenty-five times as much thorium is present. In a similar way it can be shown that thorium is just detectable when U/T = 10.

For practical purposes, the sensitivity limit for uranium in the presence of thorium can be taken as 0.1%, when the gamma-ray count rate due to uranium is equal to background count rate.

If thorium is known to be absent, or present in negligible proportions, no gamma-ray count is required and the sensitivity is then that obtainable with straight beta-ray counting, i.e., 0.01%. It may be observed in this connection that, if T/U is put equal to zero in equation (11), the accuracy for the uranium determination is given as 6%, whereas the accuracy in the case where thorium is known to be absent is 3% (see Section 2.3). This difference is due to the fact that, in one case, thorium is known to be absent, while, in the other case, it has to be established by experiment that it is present only to an extent not greater than a certain proportion. Its possible presence to a smaller extent adds to the ultimate uncertainty in the estimation of the uranium content.

If the counting time is limited, as it must be for practical purposes, we must add statistical errors given by equation (11). Assuming that the total counting time is limited to five hours (including counts on standards and background), an ore containing U% of uranium can be assayed as being:-

U% ± (0.06U + 0.015T + 0.05)%,

while an ore containing T% of thorium can be assayed as containing:-

T% ± (0.1U + 0.06T + 0.1)%

3.2.4.2 Using the Thin-Walled Cylindrical G.M. Tube

The ultimate accuracy attainable with this equipment is the same as that attainable with the end-window equipment, but the accuracy and sensitivity attainable in a limited time is far superior. This can be judged by consideration of the richness of ore required to give an increase in beta or gamma count rate equal to the background rate:-

		Richness for increase equal to background	
		Beta counting	Gamma counting
90 c.c. annular sample holder	(Uranium	0.0005%	0.005%
	(Thorium	0.002%	0.01%
13 c.c. annular sample holder	(Uranium	0.001%	0.02%
	(Thorium	0.005%	0.05%
13 c.c. end-window holder	(Uranium	0.01%	0.1%
	(Thorium	0.05%	0.2%

For practical purposes, the sensitivity limit is set by the richness of ore required to give an increase of gamma count equal to background, and this is twenty times better for the 90 c.c. annular sample holder than for the end-window one.

Thus for reasonable counting times (up to about 5 hours, including unknown, standard and background counts), the accuracies obtainable with the various forms of counting equipment can be summarised as follows:-

End-window tube with 13 c.c. samples holder:-

With thorium known absent, uranium can be assayed as
U% ± (0.03U + 0.0005)%.

With uranium known absent, thorium can be assayed as
T% ± (0.03T + 0.025)%.

In mixed ores uranium can be assayed as
U% ± (0.06U + 0.015T + 0.05)%.

In mixed ores thorium can be assayed as
T% ± (0.1U + 0.06T + 0.1)%.

Thin Walled Cylindrical Tube with 13 c.c. Annular Sample Holder:-

With thorium known absent, uranium can be assayed as
U% ± (0.03U + 0.0005)%.

With uranium known absent, thorium can be assayed as
T% ± (0.03T + 0.0025)%.

In mixed ores, uranium can be assayed as
U% ± (0.06U + 0.015T + 0.01)%.

In mixed ores, thorium can be assayed as

$$T\% \pm (0.1U + 0.06T + 0.025)\%.$$

Thin Walled Cylindrical Tube with 90 c.c. Annular Sample Holder:-

With thorium known absent, uranium can be assayed as

$$U\% \pm (0.03U + 0.00025)\%.$$

With uranium known absent, thorium can be assayed as

$$T\% \pm (0.03T + 0.001)\%.$$

In mixed ores, uranium can be assayed as

$$U\% \pm (0.06U + 0.015T + 0.0025)\%.$$

In mixed ores, thorium can be assayed as

$$T\% \pm (0.1U + 0.06T + 0.005)\%.$$

3.3 Absorption Methods

3.3.1 The Gamma-Ray Absorption Method

The method depends on the difference in mean hardness of the gamma-rays of the uranium series and the thorium series, as exhibited by the ratio of two gamma-ray counts, one without an absorber and one with a brass absorber of about 6.5 gms per sq. cm. placed between sample and G.M. tubes. The measurements are made with the apparatus described under Section 2.2.

The filter is made of brass in preference to lead, since experimental results show that it gives a greater change of absorption for a given change of mean energy. This is due to the swamping effect of the photo-electric absorption of the lower energy components of the radiation which occurs in lead. The thickness of the filter is such as to give a 50% reduction of count rate, approximately. A greater thickness would have given a greater discrimination between uranium and thorium radiations, but would have reduced the count rate obtained with weak samples to an impracticably low level.

The constants a, b, c and d (see Section 3.1) appropriate to this method represent the count rates per per-cent of uranium or thorium, with and without a filter (a and b with, c and d without).

The variation of coefficients a, b, c and d due to mass absorption coefficient were found to be about ± 3%, the total magnitude of self-absorption being 8%. The coefficient of correlation between the variations of a and b was 50% and similarly for c and d, the variations being almost invariably of the same sign. Less correlation was found between a and c and between b and d, the coefficient being only 25%. The values of the constants for a typical sample were:-

$$a = 1000,\ b = 367,\ c = 451 \text{ and } d = 196,$$

and therefore by applying equation (10) of Appendix V, we obtain:-

$$\frac{U-U'}{U} \times 100\% = 6\% + \frac{T}{U} \times 8\%.$$

Thus, if thorium is absent (but not known to be absent) the accuracy of uranium determination is 6%, and the maximum value of the thorium to uranium ratio for which uranium can just be established as being present is about 5 to 1.

If we take an ore containing 1% of uranium and 5% thorium, we obtain a value for U-U' of 0.5%, i.e. the ultimate indeterminacy is 0.5%. If we allow the statistical error to be about one third of this, a counting time of 90 minutes is given by equation (8) of Appendix V, and, allowing for counts on a standard sample and background, a total counting time of about 4 hours is required.

The sensitivity limit is set by the proportion of active material required to give an increase of count rate equal to the background rate of 100 per minute. For uranium, this is 0.1% without the absorber and 0.2% with it.

From these figures, it is clear that the gamma-ray absorption method is inferior to the beta-gamma ratio method.

3.3.2 The Beta-Ray Absorption Method

Since, in general, beta-ray measurements provide much higher count rates in relation to background and amount of sample than do gamma-ray measurements, it would appear advantageous to use a method of uranium/thorium determination in which the two measurements involve beta counting. A method involving the measurement of beta-ray absorption was therefore investigated, using the end-window type of G.M. tube. For reasons already discussed, the sample depth was again chosen to be effectively infinite, as far as beta particles were concerned.

The contribution of gamma-rays, even from the lower layers of the sample, is not greatly affected by self-absorption, and we can therefore not neglect their effect on the total count rate. The gamma-ray contribution must therefore be deducted from the beta-ray contribution or it must be regarded as an intrinsic part of the total effect.

Figure 11 shows the logarithm of the count rate plotted against filter thickness, the uranium and thorium curves being made to coincide at zero filter thickness by choosing samples of different richness. The samples were made up for the most part with the same diluant and equal weights used to fill the containers. By extrapolating the gamma-ray section of each curve and deducting from the combined beta and gamma section, the graph of Fig. 12 is obtained. This shows beta or gamma counts only, whichever preponderates for the particular filter thickness concerned. It will be seen that the slopes of the beta sections of the curves differ by only about 2%. The slopes of the gamma-ray portions also appear nearly equal when drawn on the same scale, but their difference is a much larger percentage. The difference between the beta/gamma ratios is reflected in the difference between the filter thicknesses for which the gamma-rays preponderate.

The difference between the slopes of the combined beta and gamma sections of the curves of Fig. 12 is seen to be due mainly to the difference in proportion of gamma rays, and the discrimination between thorium and uranium provided by beta-ray absorption alone is inferior to that obtainable by measurement of beta-gamma ratio.

3.4 Delayed Coincidence Method

3.4.1 Basis of Method

If an isotope A degenerates by emission of some form of radiation into an isotope B, which is itself radioactive and of half-life t, then the probability of a radiated particle from A being followed by a particle from B within a time of the order of t, is very high. This provides a possible method of recognising the radiation from isotope A, and, therefore, of estimating the amount of it which is present in a sample.

Obviously, for the method to be practicable, the time t must be short, i.e., a small fraction of a second, otherwise the chance of a particle of the type emitted by B arising from some other source than the recently degenerated atom of A within the same period becomes high, unless the total counting rate is extremely low. Such low count rates would make the time of measurement excessively long.

3.4.2 Possible Applications

There are four examples of this phenomena in the natural radioactive series, in which the half-life of the isotope B is sufficiently short to make the method applicable (see Figs 1, 2 and 3).

(a) In the actinium series, actinon decays by alpha-ray emission (7.3 MeV.) to actinium A, which is also an alpha-ray emitter (6.8 MeV.) of half-life 1.83 milliseconds.

(b) In the thorium series, thoron decays by alpha-ray emission (6.8 MeV.) to thorium A, which is an alpha-ray emitter (6.3 MeV.) of half-life 158 milliseconds.

(c) In the uranium 238 series, radium C decays by beta-ray emission (3.1 MeV.) to radium C', which is an alpha-ray emitter (7.7 MeV.) of half-life 145 microseconds.

(d) In the thorium series, 66.3% of the thorium C decays by beta-ray emission (2.2 MeV.) to thorium C' which is an alpha-ray emitter (8.7 MeV.) of half-life 0.3 microseconds.

No other correlated pairs of particles are connected by sufficiently short lives to produce any significant interference with observed delayed coincidences from these four.

Since alpha-ray measurements are involved, it is undesirable to make a single measurement of one type of delayed coincidence in order to measure merely uranium or thorium concentration in the ore. It is much better to make simultaneous measurements involving one type of coincidence from the thorium series and another from the uranium series, and compare their ratios, with a view to determining the ratio of thorium to uranium in the ore. Some other measurement, such as a total beta-ray count, could then be used to determine the actual amounts of each. This procedure also eliminates the unknown registration factors of the counting equipment for alpha and beta radiations, except in as far as they are different for the different energy particles emitted by uranium and thorium.

Suitable pairs of delayed coincidences would be (a) combined with (b) or (c) combined with (d), and the choice between the two pairs depends mainly on a consideration of the lengths of the half-lives concerned.

3.4.3 Description of Equipment

The equipment which is being used to investigate this method is shown schematically in Fig. 13. The sample is deposited in powder form with the aid of an adhesive, on the outer surface of a cylinder of copper foil. This is slid over a thin walled cylindrical G.M. tube and is surrounded by an outer transparent cylinder having the fluorescent screen for alpha-particle detection deposited on the inside surface. The beta particles are detected by the G.M. tube and the scintillations produced in the screen by the alpha-particles are detected by a ring of six photo-multiplier tubes.

The apparatus was later rearranged so that the powdered ore can be carried in a flat tray, so as to simplify sample preparation. The screen and photo-multipliers will be mounted above the tray, and a row of G.M. tubes below.

The pulses from all photo-multipliers are fed into a single amplifier, and then into a discriminator, which is so adjusted as to eliminate most of the pulses produced by noise in the photo-multiplier, but to pass most of those produced by alpha-particle scintillations.

The pulses from the G.M. tube pass via a quench probe unit into three channels:-

(i) They pass into a scaling unit, and are counted.

(ii) They pass into a coincidence circuit, which is so arranged that any pulses coming from the alpha-ray discriminator between the arrival of the G.M. tube pulse and a predeterminable time later are allowed to pass to the mechanical counter. This time is variable up to 5 microsecs for delayed coincidences of type (d) and is normally set to about 3 microsecs.

(iii) They pass through a delay circuit, which is so arranged that it triggers the square wave generator 5 microsecs after the arrival of the pulse. The triggered square wave generator opens the gate circuit for a period, which is variable up to 300 microsecs (for delayed coincidences of the type (c)), so that any pulses from the alpha-ray discriminator occurring during this period are passed onto a second mechanical counter.

Besides being fed to the coincidence circuit and the delayed coincidence circuit, the alpha-ray pulses are fed to a scaling unit for counting.

We thus get simultaneous counts of total alpha-rays, total beta-rays, alpha-rays occurring within 3 microsecs after a beta particle and alpha-rays occurring between 5 microsecs and 300 microsecs after a beta particle.

3.4.4 Accuracy and Sensitivity

Since, in both pairs of delayed coincidences (a and b, or c and d), the two half-lives concerned are very different, the two measurements for uranium and thorium can be looked upon as nearly independent. The accuracy of determination of the ratio of uranium to thorium is therefore not very dependent upon the ratio itself, as it is with the differential methods discussed in sections 3.2 and 3.3. The method is therefore likely to give greatly improved results when the ratio of the two active contents is high.

This is the real advantage of the method over those already discussed. The accuracy is not likely to be greater in cases where the ratio of the two active contents is such that their activities are of the same order, owing to errors arising from dissimilarity of self-absorption, counting efficiency, etc., for the different energy particles from uranium and thorium. Errors due to self-absorption of alpha-particles would be particularly high in cases where the uranium and thorium contents of the ore occurred in grains of different densities.

A further error which would arise in the case of the pair (c) and (d) is the effect of diffusion of radon from the surface of the sample. This error would be of the order of 10%. It could be calibrated out, to a certain extent, but a fair fraction of it would remain, due to the variation of the diffusion coefficient of radon with sample material and temperature.

A little consideration will show that, for a given geometrical arrangement and given counting efficiencies, there exists an optimum value for the weight of active material in the sample for which the counting time required for a given statistical accuracy is minimum. If a very small amount of active material is present, the count rate is very low, and therefore the required time is long. If a very large amount is present, the coincidence rates (short and long) are high, but so also is the probability of the random occurrence of extraneous pulses from other atoms within the open time of the short and long delayed coincidence circuits after a given beta particle. This means that a very large number of counts have to be obtained, in order to correct the observed coincidence rate accurately for these extraneous occurrences. Between these extreme amounts, a quantity of material can be chosen to give a shorter counting time.

The method of calculation of this optimum mass of material and the minimum time required for a given accuracy is shown in Appendix VI, and the following are a few examples of the accuracies obtainable under various conditions:-

For the purposes of these estimations, the registration coefficients for both beta- and alpha-particles is taken as 30%.
Alpha-alpha Delayed Coincidences (a) and (b):-

Uranium Content Expressed as a % of Total Radioactive Content	Mean Error Allowed (% of Total Radioactive Content)	Optimum Mass of Radioactive Content	Counting Time Required
30%	10%	0.07 mgm.	13 hours
10%	3%	0.17 mgm.	28 hours
5%	1.5%	0.07 mgm.	69 hours

Beta-alpha Delayed Coincidences (c) and (d):-

Uranium as % of Radioactive Content	Mean Error as % of Total Radioactive Content	Mass of Radioactive Content	Counting Time Required
5%	0.5%	4.5 mgm (Optimum)	20 mins
5%	0.5%	1.1 mgm (not ")	1 hour
1%	0.1%	4 mgm (Optimum)	3 hours
1%	0.2%	4 mgm (")	45 mins

The second example for beta-alpha delayed coincidences is given so as to indicate the order of increase of counting time required if the mass of radioactive material differs from the optimum mass.

It will be seen from these results that the second method involving beta-alpha coincidences is superior to the first, except for the errors introduced by radon diffusion from the sample. These were not included in the calculations.

The measurement of gate width and coincidence discrimination is not critical. Tolerances of ± 6 microsecs on the 300 microsecs gate and ± 1 microsec. on the 3 microsecs coincidence period do not affect the accuracy of the results to any detectable extent.

3.5 Other Time Spectrum Methods

A number of other methods depending on the time distribution of particle and photon emissions are possible.

The detection of high speed electrons produced by internal conversion of gamma-rays following a beta-particle emission is one possibility. The literature on the subject is very incomplete, but uranium X_1 is generally quoted as exhibiting a high conversion efficiency, and, if this is so, the measurement of uranium in ores which are not in secular equilibrium becomes fairly simple. Since uranium X_1 is the direct disintegration product of uranium 238 and is itself short-lived, such a measurement would be independent of the equilibrium unbalance produced by normal geological effects, and even in cases of very recent leaching, a measurement taken after a delay of a few weeks would be accurate.

Even if the UX_1 conversion electrons do not predominate, such measurements may give a useful indication of equilibrium unbalance, which could be used to correct other assay results.

Another method which suggests itself for measuring the thorium content is the measurement of gamma-gamma coincidences which occur in the thorium series. This would require a gamma-ray detector of high efficiency, and again the gamma-ray scintillation counter is a possibility.

3.6 Methods Involving Separation Processes

Methods involving chemical separation have not been investigated by the A.E.R.E. geophysics team, but brief mention is made of them here for completeness.

One of the most sensitive methods for determining uranium and thorium is by measurement of the gases radon, thoron and actinon. These gases can be driven from the sample by grinding and heating and measured by the aid of an ionisation chamber and electroscope. Measurement of thoron and actinon by this method is only very approximate, due to their very short half-lives (54 secs. and 3.9 secs. respectively), but the measurement of radon is much more satisfactory. The radon measurement is a very sensitive one, having been used to estimate the uranium content of ordinary rocks and minerals, not normally considered radioactive, but is not very accurate. Errors arise also from any lack of equilibrium occurring in the sample.

In connection with the measurement of extremely weak samples it should be mentioned that the presence of potassium could provide an additional source of interference, if a purely radiometric method of sufficient sensitivity were used. The naturally radioactive isotope, K^{40}, is present to an extent of 0.012% in natural potassium. It has a half life of 10^8 - 10^9 years and emits energetic beta and gamma radiations. Thus 0.001% of uranium could be equalled by about 5% of potassium in the mineral.

A method has been suggested(6) involving chemical precipitation, followed by a radiometric measurement, which will give a measure of the uranium content of an ore, independently of its thorium content or the presence of any lack of secular equilibrium.

A quantity of the powdered mineral expected to contain about one milligram of uranium is put into solution, and the uranium X_1 is then precipitated by the iodate process or the hypophosphate process. The beta activity of the uranium X_2 which builds up to the equilibrium proportion in a few minutes (see Fig. 1) is then counted. The thorium isotopes of uranium X_1, i.e., thorium and radio-thorium (see Fig. 3), are precipitated with it and therefore additional beta activity builds up with approximately the 3.6 day half-life period of thorium X. A further count after a few days, therefore, gives an estimate of the thorium content of the ore also.

Activation analysis (using an atomic pile), and the use of a fluorimeter, are two other methods involving chemical separations.

4. METHODS FOR ORES NOT IN SECULAR EQUILIBRIUM

4.1 Methods of Reducing Errors due to Equilibrium Disturbance in Normal Radiometric Measurements

Lack of equilibrium in an unknown sample produces errors in the uranium and thorium estimations only in so far as the equilibrium unbalance differs from that in the standard sample. This arises from the fact that the equilibrium unbalance changes the mean energy of the radiations and also the activity per unit weight of uranium or thorium.

Thus, if we use a standard which is of similar geological origin to the unknown, the errors due to equilibrium unbalance will be to a large extent eliminated. Such a standard may be very different from those used for the original calibration of the counting equipment and a measure of re-calibration is required. The form of this re-calibration will be described for the beta/gamma ratio method of uranium/thorium ratio determination, and its application to other methods will then be obvious.

Since, in the thorium series, only thorium itself has a long half-life, the likelihood of an equilibrium unbalance of the thorium series in minerals is generally small. In general, only the uranium series will be affected, and this will have the effect of modifying the constants a and c. Let this modification be such that the constants become ah and ck. The procedure is then as follows.

The standard chosen for its similarity with the unknown will be called the comparison standard, and the standard similar to those used for calibration will be called the calibration standard.

The average count rates are calculated and the paralysis time losses and background allowed for. The count rates observed for the unknown sample and the comparison standard are then adjusted by the ratio by which the readings for the calibration standard differ from those at the time of calibration. This allows for any change of sensitivity since calibration. Four figures are now obtained, namely, N_1 and F_1 for the comparison standard without and with a filter, and N_2 and F_2 for the unknown. From the calibration curves (Figs 6 and 7), the values a_1, b_1, c_1, d_1 for the comparison standard and a_2, b_2, c_2 and d_2 for the unknown are read off. The factors h and k are then calculated from the equations:-

$$h = \frac{N_1 - b_1 T_1}{a_1 U_1}, \quad k = \frac{F_1 - d_1 T_1}{c_1 U_1}$$

which are derived from the equations:-

$$N = ahU + bT, \text{ and } F = ckU + dT.$$

U_1 and T_1 are the known percentage contents of the comparison standard.

Finally the contents of the unknown are calculated from the equations:-

$$U = \frac{d_2N_2 - b_2F_2}{a_2d_2h - b_2c_2k} \text{ and } T = \frac{a_2hF_2 - c_2kN_2}{a_2hd_2 - b_2c_2k}$$

4.2 Other Purely Radiometric Methods

A method, developed at A.E.R.E., using (a) chemically separated uranium oxide and (b) an ore sample known to be thorium free and to have its uranium decay products in equilibrium, as comparison standards is described elsewhere(3). This is limited to equilibrium determinations of thorium free samples.

Another method, using a beta/gamma scintillation counter has also been developed. This takes into account the thorium content but is limited to high grade ores.

A further possibility is provided by counting U235 fission particles emitted while the sample is exposed to a source of neutrons.

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5. Report by S. H. U. Bowie, Geological Survey & Museum, London, entitled "The Geological Applications of Alpha-Particle Emulsions".

6. Report by J. V. P. Long, Chemical Research Laboratory, Teddington, entitled "The Determination of Uranium by Radiometric Measurement of UX1".

APPENDIX I

ERRORS ARISING FROM THE DIFFUSION OF RADON FROM THE SURFACE OF A SAMPLE

Let R be the number of grams of radon formed per unit volume of ore per second, and let u be the radon concentration at a depth z after a steady state has been reached. Then the nett number of grams of radon gained per second due to radioactive processes alone, in an elementary volume $S\delta z$ at depth z

$$= (R - \lambda u)\ S\delta z,$$

where λ is the disintegration constant of radon.

This must be equal to the nett loss by diffusion.

The amount diffusing out of the elementary volume towards the surface

$$= SD\ \left(\frac{du}{dz}\right)_z$$

where D is the coefficient of diffusion, and the amount entering

$$= SD\ \left(\frac{du}{dz}\right)_{z+\delta z}$$

Therefore, the nett loss by diffusion

$$= -SD\ \frac{d^2u}{dz^2}\ .\ \delta z$$

Therefore $\quad -D\ \frac{d^2u}{dz^2} = R - \lambda u$

of which the solution is

$$u = \frac{R}{\lambda}\ (1 - \varepsilon^{\sqrt{\frac{\lambda}{D}}\ z})$$

Hence, the variation with depth in a grain of the powder is exponential, the variation constant being $\sqrt{\frac{D}{\lambda}}$.

For alpha-ray assay the distance $\sqrt{\frac{D}{\lambda}}$ must be small as compared with the alpha-ray range, otherwise a large proportion of the alpha-rays counted will come from depths at which the radon concentration is noticeably lower than normal, due to diffusion losses.

The coefficient of diffusion of radon through solids is of the order of 10^{-14} cms^2/sec., at room temperature, and the disintegration constant is 2.1×10^{-6} disintegrations per second. Therefore, by substitution, we have $\sqrt{D/\lambda} = 0.0001$ cm.

The range of the alpha particles in a typical mineral is of the order of 0.002 cms., and this has to be compared with the depth $\sqrt{D/\lambda} = 0.0001$ cm.

If we take into account the fact that part of the alpha track must be outside the grain if it is to be counted, and also that some loss occurs from the other sides of the grain, the error involved is of the order of 10%. This value is very variable with the type of material and its temperature.

When measuring beta- or gamma-rays, we are concerned with activity of the whole volume of all of the grains, or a large proportion of them, and we must therefore consider the total loss of radon from the whole grain. If we consider that all radon diffusing to the outside surface of any grain is lost, we can calculate the minimum grain size allowable for a given required accuracy.

The contribution of radon and the short life products following it might be estimated roughly as about one half of the total.

Therefore, if the volume effect is to be 1%, we have

$$\frac{1}{2} \cdot \frac{\frac{4}{3}\pi\left[r^3 - (r - \sqrt{\frac{D}{\lambda}})^3\right]}{\frac{4}{3}\pi r^3} = \frac{1}{100}$$

Hence, the mean grain radius r = about 0.015 cms.

In practice, unless the sample is very thin, we shall not lose all the radon diffusing to the outside of every grain, but if the grinding is very fine it would be advisable to use the sample in a sealed container. This is also advisable from the point of view of contamination of measuring apparatus.

APPENDIX II

MEASUREMENT OF CIRCUIT PARALYSIS TIME

In order to cut down the time required to make this measurement, high counting rates are used, and, therefore, since the mechanical counter which counts the hundreds in the scaling unit will not operate reliably more than twice per second, two scaling units are run in series. The mechanical counter in the second unit therefore operates once for every ten thousand counts fed into the first scaling unit.

Two samples are selected which will each give count rates of 25,000 to 50,000 per minute, when suitably positioned near the G.M. tube. The count rate is first measured with one sample in position, then with both, without disturbing the position of the first, and finally with the second one only. Small powerful sources should be used, and they must be so arranged that the radiation from one is not affected by scatter or absorption in the other.

If P, Q and R are the three readings obtained, in that order, then the effective paralysis time can be shown to be given by

$$t = \frac{1}{Q}\left[1 - \sqrt{1 - \frac{Q}{PR}(P + R - Q)}\right] \qquad \ldots\ldots(1)$$

APPENDIX III

CHOICE OF SHAPE OF SAMPLE AND G.M. TUBE

These comments apply to the cylindrical thin-walled tube and annular sample holder.

The choice of sample thickness depends on such considerations as self-absorption, grain size, etc. As discussed in section 2.3.2, the sample should be either of intermediate or effectively infinite thickness for beta-particles, whilst for gamma-ray measurements it should be as thin or shallow as possible without reduction of volume. It is most convenient to use the same holder for both measurements, if this can be done without serious disadvantage, and a choice of thickness between 0.3 and 1.0 cms. has been found to satisfy these requirements.

The beta-particle count rate is approximately proportional to the window area and to the solid angle subtended by the sensitive volume of the tube at a point on the window. The second factor is sensibly constant for tubes having a length equal to about three times their diameter, or more, assuming that no large gap is left between the window and the tube. Variation of the window area, which may be taken as the cathode area, if the whole length of the tube is surrounded by the holder, also has a directly proportional effect on the background count rate. We cannot, therefore, gain in sample count/background count by change of window area, but, if both are increased, the counting time required is correspondingly reduced.

Taking the sample volume fixed at some value V and the thickness at X, the tube diameter, d, and the length, l, are then related by the equation

$$V = \pi(d + X) X l$$

The window area is given by π d l, and, if we substitute the value of l given by the equation above, this becomes

$$\frac{V}{X} \times \frac{d}{d + X}$$

Hence, the window area increases rapidly as the tube diameter increases, until the diameter is about twice the sample thickness, and then continues to increase gradually (asymptotically). The required counting time is therefore reduced by choosing a G.M. tube with the largest diameter which gives the required sample depth and volume, without making the length less than about three times the diameter (this limitation to the maximum value of d/l is desirable also from the point of view of G.M. tube characteristics).

The gamma-ray count rate is approximately proportional to the sample volume and the cathode area, and inversely to the square of the mean distance between elementary sections of the sample and the cathode. Again, it may be shown that the best geometrical arrangement is similar to that for beta-particle counting, provided again that the same sample volume and depth are pre-selected on other grounds.

The larger of the two annular holder sizes gives the maximum sensitivity available with the B6E G.M. tube, whilst the 13 c.c. holder is more convenient for general use.

APPENDIX IV

DEPENDENCE OF SAMPLE SELF-ABSORPTION ON DENSITY AND MASS ABSORPTION COEFFICIENT

Figure 4 shows the variation of the logarithm of the count rate with thickness of absorber placed in front of a thin pitchblende sample. The part corresponding mainly to beta-ray absorption is approximately straight, which indicates that the absorption is exponential.

Considering an elementary layer at a depth x, its contribution to the count rate is given approximately by

$$n_x \, \delta x \;=\; n_0 \, \varepsilon^{-\mu x} \, \delta x$$

where μ is the absorption coefficient, and n_0 is the value which the count rate would have per unit layer without self-absorption.

n_0 is proportional to the activity per unit volume, i.e., to $U\rho$, where U is the percentage of uranium (or thorium) and ρ the density. If m is the mass absorption coefficient, μ/ρ, we therefore have

$$n_x \, \delta x \;=\; K \, U\rho\varepsilon^{-m\rho x} \, \delta x$$

Integrating for a thick sample, the total count rate is given by

$$n \;=\; K \, U \, \rho \int_0^{\infty} \varepsilon^{-m\rho x} \, dx$$

$$=\; \frac{kU}{m} \qquad \ldots\ldots(2)$$

Hence the approximate expression for the count rate shows it to be independent of density for a thick (infinite) sample. The expression is only approximate since the effect of the gamma rays present, the back scatter from successive layers and the change of solid angle of acceptance with depth of sample are all neglected. It shows, however, that changes of mass absorption coefficient have a direct effect on the count rate.

APPENDIX V

CALCULATION OF ERRORS ARISING IN MIXED ORE ASSAY

1. Statistical Errors

Let N and F be the two count rate readings obtained (see Section 3.1), then we have

$$N = aU + bT + n, \quad F = cU + dT + f \qquad \text{.....(3) and (4)}$$

where U and T are the percentages by weight of uranium and thorium, a, b, c and d are functions of sample density, geometry, etc. and n and f are background count rates.

Solving these equations simultaneously, we have

$$U = \frac{d(N - n) - b(F - f)}{ad - bc}, \quad T = \frac{a(F - f) - c(N - n)}{ad - bc} \text{.....(5) and (6)}$$

When statistical fluctuations are dominant, the errors in U and T are due to the mean errors in count rate

$$\sqrt{\frac{N}{t_1}}, \sqrt{\frac{F}{t_2}}, \sqrt{\frac{n}{t_3}}, \text{ and } \sqrt{\frac{f}{t_4}}$$

where t_1 and t_2 are the times for the two counts N and F, and t_3 and t_4 are those for n and f.

The mean error in the sum or difference of two uncorrelated quantities is equal to the square root of the sum of the squares of the two individual mean errors. Therefore, the mean error in U is given by

$$\Delta U = \sqrt{\left[d^2 \left(\frac{N}{t_1} + \frac{n}{t_3}\right) + b^2 \left(\frac{F}{t_2} + \frac{f}{t_4}\right)\right]} \Big/ (ad - bc) \qquad \text{.....(7)}$$

and a similar expression can be deduced for the error in T.

If the background is negligible, we can re-express this error in terms of the active contents as follows:-

$$\Delta U = \sqrt{\left[\frac{d^2}{t_1} (aU + bT) + \frac{b^2}{t_2} (cU + dT)\right]} \Big/ ad - bc \qquad \text{.....(8)}$$

$$\Delta T = \sqrt{\left[\frac{c^2}{t_1} (aU + bT) + \frac{a^2}{t_2} (cU + dT)\right]} \Big/ ad - bc \qquad \text{.....(9)}$$

2. Non-Statistical Errors

Let us suppose that the counting times allowed are such that statistical errors become negligible, but that unknown factors introduce unsuspected modifications to a, b, c and d, such that they take up new values a_1, b_1, c_1 and d_1.

We then have $N = a_1U + b_1T + n$ and $F = c_1U + d_1T + f$.

Hence, by substitution in equations (5) and (6), we have

$$U_1 = \frac{d(a_1U + b_1T) - b(c_1U + d_1T)}{ad - bc}$$

$$T_1 = \frac{a(c_1U + d_1T) - c(a_1U + b_1T)}{ad - bc}$$

Taking the case of uranium and re-expressing in terms of relative percentage errors

$$\left(\frac{U-U_1}{U}\right) \times 100\% = \left\{ \left[\frac{1}{bc} \left(\frac{a-a_1}{a}\right) - \frac{1}{ad} \left(\frac{c-c_1}{c}\right) + \frac{1}{ac} \cdot \frac{T}{U} \left(\frac{b-b_1}{b} - \frac{d-d_1}{d}\right) \right] \Bigg/ \left(\frac{1}{bc} - \frac{1}{ad}\right) \right\} \times 100\% \qquad \text{.....(10)}$$

APPENDIX VI

CALCULATION OF OPTIMUM MASS OF RADIOACTIVE MATERIAL AND MINIMUM COUNTING TIME FOR THE DELAYED COINCIDENCE METHOD

As an example, let us take the case of the pair of alpha-alpha coincidences, (a) and (b), of Section 3.4.2; the extension to other cases will be obvious.

Let the total number of alpha-particles produced per gram of uranium per second be a and that produced per gram of thorium per second be b. Also, let the masses of uranium and thorium present in the sample be x and y, respectively.

Then the total number of alpha-rays counted will be $(axr_1 + byr_2)$, where r_1 and r_2 are the mean registration efficiencies of the counting system for the alpha-rays from uranium and thorium, respectively.

Suppose the delayed coincidences (a) are counted through a gate circuit which is opened at time t_1 and closed at time t_2 after the initiating alpha-ray, and the corresponding times for the delayed coincidences (b) are t_3 and t_4. If the total time during which the gate circuits are open is small as compared with the total closed time, the number of chance occurrences of random pulses during open periods during a counting time t will be

$$(axr_1 + byr_2)^2\ (t_2 - t_1)t \text{ and } (axr_1 + byr_2)^2\ (t_4 - t_3)t$$

Let λ_1 and λ_2 be the two decay constants appropriate to the coincidence delays in cases (a) and (b), and let the number of genuine delayed coincident pulses for the two cases per gram of uranium or thorium per second be c and d.

The number of delayed coincident pulses counted in the t_1 to t_2 gate in time t will be equal to

$cxtr_3r_4$ x probability of the second pulse lying between the gate limits.

$$= cxtr_3r_4 \int_{t_1}^{t_2} \lambda_1 \varepsilon^{-\lambda_1 t}\, dt$$

$$= cxtr_3r_4\ (\varepsilon^{-\lambda_1 t_1} - \varepsilon^{-\lambda_1 t_2})$$

where r_3 and r_4 are the registration factors for the first and second alpha-rays of the correlated pairs, respectively.

For case (b) the number counted

$$= dytr_5r_6\ (\varepsilon^{-\lambda_2 t_3} - \varepsilon^{-\lambda_2 t_4})$$

Since the alpha-ray energies concerned are very comparable we can probably put $r_3 = r_4 = r_5 = r_6$ without introducing any serious error.

If, therefore, we obtain three readings A, B and C for the total alpha-ray count, the total count in the $(t_1 - t_2)$ gate and the total count in the $(t_3 - t_4)$ gate, we will have

$$A = (axr_1 + byr_2)\ t \qquad \text{.....(12)}$$

$$B = (axr_1 + byr_2)^2 (t_2 - t_1) t + cxtr_3^2 (\varepsilon^{-\lambda_1 t_1} - \varepsilon^{-\lambda_1 t_2}) \quad .....(13)$$

$$C = (axr_1 + byr_2)^2 (t_4 - t_3) t + dytr_3^2 (\varepsilon^{-\lambda_2 t_3} - \varepsilon^{-\lambda_2 t_4}) \quad .....(14)$$

From which we obtain the uranium/thorium ratio

$$x/y = \frac{Bt - A^2 (t_2 - t_1)}{Ct - A^2 (t_4 - t_3)} \times \frac{d}{c} . \frac{(\varepsilon^{-\lambda_2 t_3} - \varepsilon^{-\lambda_2 t_4})}{(\varepsilon^{-\lambda_1 t_1} - \varepsilon^{-\lambda_1 t_2})} \quad .....(15)$$

The absence of r_1, r_2 and r_3 from this expression implies that, if all three readings are available, no knowledge of solid angle of acceptance or efficiency of the counter, or of the alpha-ray absorption is required. However, these factors affect the minimum time of experiment required, in a particular case.

It will be noted also, that if the background can be considered as random, and thus merely an addition to the total random count, it is also automatically taken into account as part of A and does not affect the result.

If we cannot assume $r_3 = r_4 = r_5 = r_6$, we merely have the result multiplied by a factor $\frac{r_5 r_6}{r_3 r_4}$. This is constant for a given geometry and for all ratios x/y, but is varied to some extent by change of atomic number distribution. The important thing is to eliminate those registration factors which appear as coefficients of x and y and this is seen to be possible when the total alpha count A is available. If we used the beta-alpha delayed coincidences (c) and (d) of Section 3.4.2, it would be necessary to take also a total beta count, in order to eliminate these factors.

In order to calculate the error introduced into the ratio x/y by statistical errors in A, B and C, the following statistical laws are made use of:-

(1) The standard deviation (i.e., the R.M.S. value of the deviation from the mean) occurring in the number of random occurrences in a given time is equal to the square root of the mean number occurring in the same time.

(2) The standard deviation in the sum or difference of two quantities is equal to the square root of the sum of the squares of the standard deviations in the individual quantities.

(3) The relative standard deviation (standard deviation divided by the quantity concerned) in the product or quotient of two quantities is equal to the square root of the sum of the squares of the relative standard deviations of the two quantities.

(4) The relative standard deviation error in the square of a quantity equal to twice the mean standard deviation in the quantity itself. The case of a square is not of course covered by the case of a product of two quantities carrying independent errors, since the errors in the two factors of a square vary together.

Let us assume that we wish to determine the ratio x/y to an accuracy p, i.e. to determine the ratio as being $(\frac{x}{y} \pm p)$ 100%.

Referring to equation (15), $\frac{x}{y}$ will have its required relative standard deviation $(\frac{py}{x})$, if

$$(\frac{py}{x})^2 = \frac{Bt^2 + 4\Lambda^3 (t_2-t_1)^2}{[Bt - \Lambda^2 (t_2-t_1)]^2} + \frac{Ct^2 + 4\Lambda^3 (t_4-t_3)^2}{[Ct - \Lambda^2 (t_4-t_3)]^2} \qquad \ldots\ldots(16)$$

Substituting for Λ, B and C from equations (12), (13) and (14), and writing ky for x, this may be rewritten in the form

$$t = \frac{jy^2 + \ell y + n}{y} \qquad \ldots\ldots(17)$$

where $$j = 4 \frac{f^3k^2}{p^2} \left[\frac{(t_2-t_1)^2}{g} + \frac{(t_4-t_3)^2}{h} \right]$$

$$\ell = \frac{f^2k^2}{p^2} (2\frac{t_2-t_1}{g^2} + \frac{t_4-t_3}{h^2})$$

$$n = \frac{k^2}{p^2} (\frac{1}{g} + \frac{1}{h})$$

in which $f = (akr_1 + br_2)$, $g = ckr_3^2 (\varepsilon^{-\lambda_1 t_1} - \varepsilon^{-\lambda_1 t_2})$,

$$h = dr_3^2 (\varepsilon^{-\lambda_2 t_3} - \varepsilon^{-\lambda_2 t_4})$$

Differentiating (17), the counting time is minimum when

$$y = \sqrt{\frac{n}{j}}$$

or when the total active content is

$$(x + y) = (1 + k) \sqrt{\frac{n}{j}}$$

Substituting $y = \sqrt{\frac{n}{j}}$ in (17), the minimum counting time is given by

$$t = \ell + 2 \sqrt{jn}$$

APPENDIX VII

DIVISION OF COUNTING TIME BETWEEN INDIVIDUAL COUNTS IN AN ASSAY

Case 1, When Only a Sample and Background are to be Counted

Let N be the total count obtained with sample and background in time t.

Let B be count obtained with background only in time t_b.

Let n be the count rate per second for sample and background = N/t.

Let b be the count rate per second for background only = B/t_b.

The radioactive content of the sample is then obtained in the form

$$(n - b)C$$

where C is a constant depending on apparatus, etc.

The standard deviation of the count N will be $\sqrt{N}$, or, expressed as a proportion of N, $\frac{1}{\sqrt{N}}$.

The standard deviation of count rate will therefore be

$$n \times \frac{1}{\sqrt{N}} = \sqrt{\frac{n}{t}}$$

Similarly, the standard deviation of b will be $\sqrt{\frac{b}{t_b}}$

$\therefore$ Standard deviation of (n - b)

$$= \sqrt{\frac{n}{t} + \frac{b}{t_b}}$$

Let T be the total time allowable for both counts, N and B, then

$$t + t_b = T$$

Therefore, if $t = kt_b$

$$t = Tk/(1+k) \quad \text{and} \quad t_b = T/(1+k)$$

The square of the standard deviation of (n - b) may therefore be re-expressed as

$$\frac{(1 + k)}{T} \left(\frac{n}{k} + b\right)$$

We now require to find the value of k for which this is minimum.

Differentiating the expression with respect to k and equating to zero for a minimum, we have

$$\frac{1}{T}\left(\frac{n}{k} + b\right) - \frac{n}{k^2} \cdot \frac{(1+k)}{T} = 0$$

$$\therefore \; k = \sqrt{\frac{n}{b}}$$

Or $$t/t_b = \sqrt{n/b}$$

Thus, the smaller the background count the smaller is the proportion of time for which it should be counted, in order to give maximum statistical accuracy.

Case 2, Sample and Standard to be Counted

Here, the answer appears in the form n_1C/n_2, where C again is a constant and n_1 and n_2 are the count rates obtained with the unknown sample and the standard sample, respectively.

If the times for the two counts are, respectively, t_1 and t_2, the standard deviation of n_1 is

$$\sqrt{\frac{n_1}{t_1}}$$

or, the relative standard deviation is

$$\sqrt{\frac{n_1}{t_1}} \bigg/ n_1 = \sqrt{\frac{1}{n_1 t_1}}$$

Similarly, the relative standard deviation of n_2 is

$$\sqrt{\frac{1}{n_2 t_2}}$$

If $t_1 + t_2 = T$, and $t_1 = kt_2$, then we have

$$t_1 = Tk/(1+k) \text{ and } t_2 = T/(1+k)$$

Therefore

$$(\text{Total Relative Standard Deviation})^2 = \frac{1}{n_1 t_1} + \frac{1}{n_2 t_2}$$

$$= \frac{(1+k)}{T}\left(\frac{1}{n_1 k} + \frac{1}{n_2}\right)$$

Differentiating this expression with respect to k and equation to zero, we have

$$\frac{1}{T}\left(\frac{1}{n_1 k} + \frac{1}{n_2}\right) - \frac{(1+k)}{T}\left(\frac{1}{n_1 k^2}\right) = 0$$

Therefore $$k = \sqrt{\frac{n_2}{n_1}}$$

or $$\frac{t_1}{t_2} = \sqrt{\frac{n_2}{n_1}}$$

In this case, therefore, we give a longer time to the slower count rate.

If N_1 and N_2 are the total counts obtained in times t_1 and t_2

$$\frac{N_1}{N_2} = \frac{n_1}{n_2} \times \frac{t_1}{t_2} = \frac{n_1}{n_2} \times \sqrt{\frac{n_2}{n_1}}$$

$$= \sqrt{\frac{n_1}{n_2}}$$

Case 3, Sample, Standard and Background

Here the result for the sample strength relative to the standard is obtained in the form

$$\left(\frac{n_1-b}{n_2-b}\right) C$$

and we require to find the values of t_1, t_2, and t_b to give the minimum relative standard deviation (R.S.D.) of this expression, if allowed a total count time T.

The standard deviation (S.D.) of (n_1-b) is again

$$\sqrt{\left(\frac{n_1}{t_1} + \frac{b}{t_b}\right)}$$

and its R.S.D. is therefore

$$\sqrt{\left(\frac{n_1}{t_1} + \frac{b}{t_b}\right)} \Big| (n_1-b)$$

Similarly, the S.D. of (n_2-b) is

$$\sqrt{\left(\frac{n_2}{t_2} + \frac{b}{t_b}\right)}$$

and its R.S.D. is

$$\sqrt{\left(\frac{n_2}{t_2} + \frac{b}{t_b}\right)} \Big| (n_2-b)$$

The S.D. of the background count rate alone is $\sqrt{(b/t_b)}$. In the present case account must be taken of the fact that the numerator and denominator of the quotient whose R.S.D. is required, namely $(n_1-b)/(n_2-b)$, are correlated with one another by virtue of the fact that a common background reading has been deducted from both.

If the coefficient of rectilinear correlation between any two quantities is r and their respective R.S.Ds. are R_1 and R_2 then the R.S.D. of the quotient is $\sqrt{(R_1^2 + R_2^2 + 2r\, R_1\, R_2)}$. The coefficient, r, expresses the proportion of variation in the two quantities which is common to both. In the case of two quantities which are correlated by virtue of the fact that they have had a common quantity deducted from them, and which were uncorrelated prior to such deduction, the applicable formula giving r is $r = S_3^2/S_1\,S_2$, where S_1 and S_2 are the S.Ds. of the two quantities concerned and S_3 is the S.D. of the common quantity deducted from both. Therefore the R.S.D. of the quotient becomes $\sqrt{(R_1^2 + R_2^2 + 2S_3^2\,R_1\,R_2/S_1\,S_2)}$.

In the case in point we have seen that

$$R_1 = \sqrt{\left(\frac{n_1}{t_1} + \frac{b}{t_b}\right)} \Bigg/ (n_1-b) \qquad R_2 = \sqrt{\left(\frac{n_2}{t_2} + \frac{b}{t_b}\right)} \Bigg/ (n_2-b)$$

$$S_1 = \sqrt{\left(\frac{n_1}{t_1} + \frac{b}{t_b}\right)} \qquad S_2 = \sqrt{\left(\frac{n_2}{t_2} + \frac{b}{t_b}\right)} \quad \text{and} \quad S_3 = \sqrt{\left(\frac{b}{t_b}\right)}$$

Substituting these values for R_1, R_2, S_1, S_2 and S_3 and putting $t_1 = k_1 t_b$, $t_2 = k_2 t_b$ and $t_b = T - (t_1 + t_2)$ the R.S.D. of the quotient $(n_1-b)/(n_2-b)$ becomes

$$\sqrt{\left[\frac{k_1 + k_2 + 1}{T}\left(\frac{n_1/k_1+b}{(n_1-b)^2} + \frac{n_2/k_2+b}{(n_2-b)^2} + \frac{2b}{(n_1-b)(n_2-b)}\right)\right]}$$

By partial differentiation with respect to k_1 and k_2 it is found that this is a minimum when

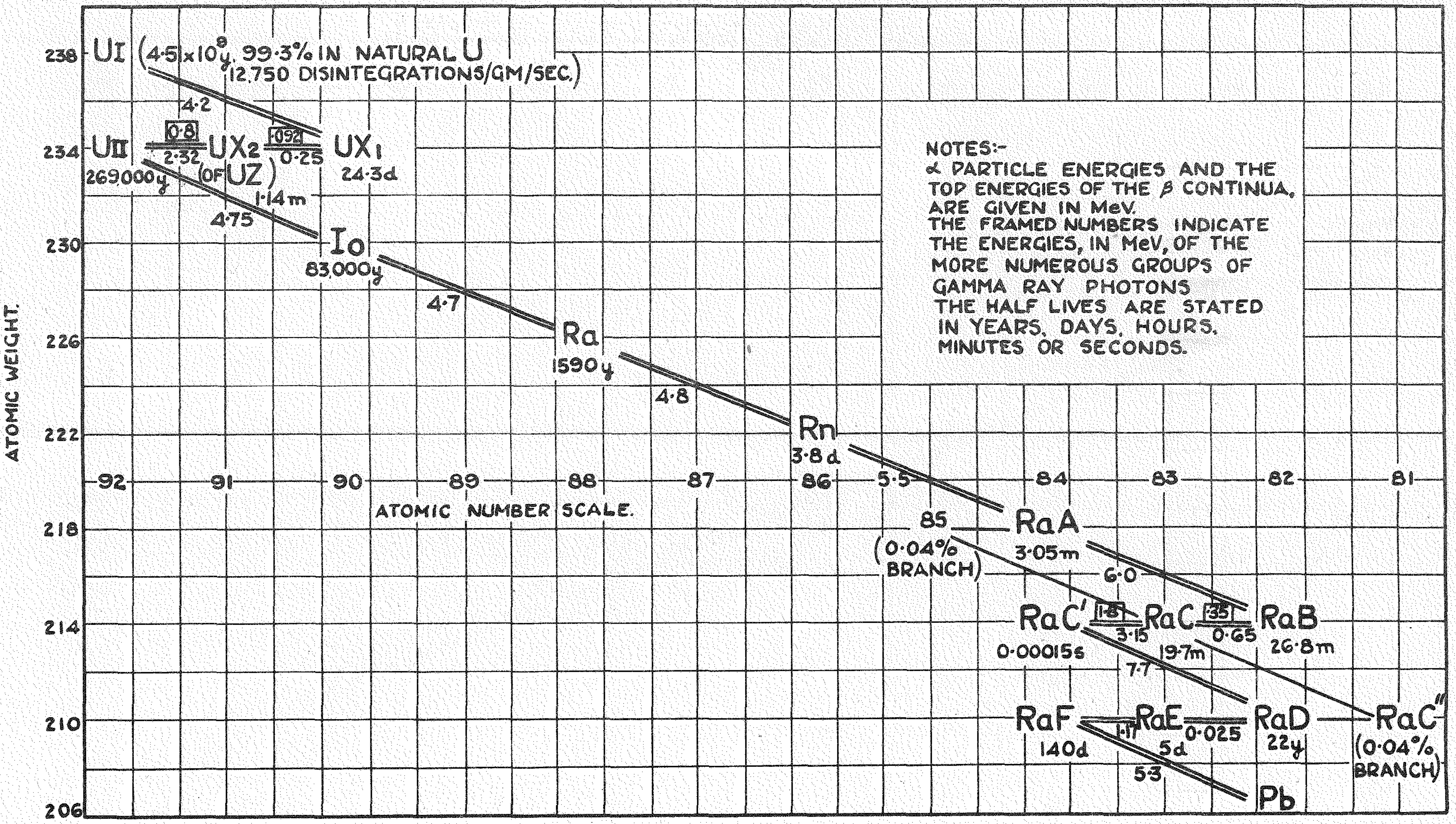
$$k_1 = \frac{n_2-b}{n_1 + n_2 - 2b}\sqrt{\left(\frac{n_1}{b}\right)} \quad \text{and} \quad k_2 = \frac{n_1-b}{n_1 + n_2 - 2b}\sqrt{\left(\frac{n_2}{b}\right)}$$

Hence the optimum subdivision of the total available counting time, T, between the unknown sample, the standard, and the background is given by

$$t_1 = \frac{\sqrt{n_1}\,(n_2-b)}{D}\,T, \quad t_2 = \frac{\sqrt{n_2}\,(n_1-b)}{D}\,T, \quad t_b = \frac{\sqrt{b}\,(n_1 + n_2 - 2b)}{D}\,T$$

where $D = \sqrt{n_1}\,(n_2-b) + \sqrt{n_2}\,(n_1-b) + \sqrt{b}\,(n_1 + n_2 - 2b)$

It is useful to note that if $n_1 = n_2$ then $\dfrac{t_1}{t_b} = \dfrac{t_2}{t_b} = \dfrac{\sqrt{n}}{2\sqrt{b}}$



NOTES:-
α PARTICLE ENERGIES AND THE TOP ENERGIES OF THE β CONTINUA, ARE GIVEN IN MeV.
THE FRAMED NUMBERS INDICATE THE ENERGIES, IN MeV, OF THE MORE NUMEROUS GROUPS OF GAMMA RAY PHOTONS
THE HALF LIVES ARE STATED IN YEARS, DAYS, HOURS, MINUTES OR SECONDS.

FIG. I. THE U238 NATURAL DECAY SERIES.

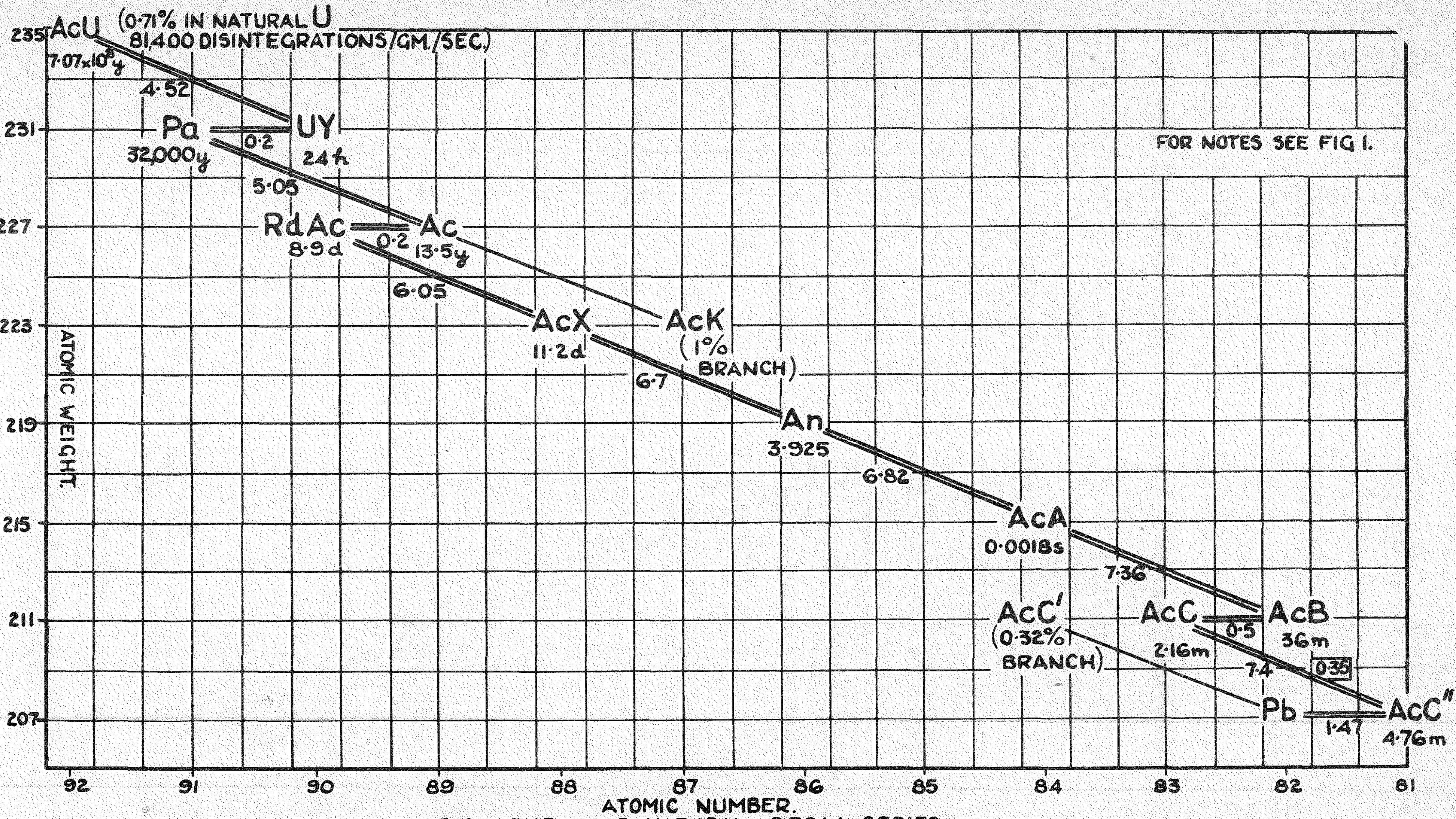


FIG. 2. THE U235 NATURAL DECAY SERIES.

FOR NOTES SEE FIG. 1.

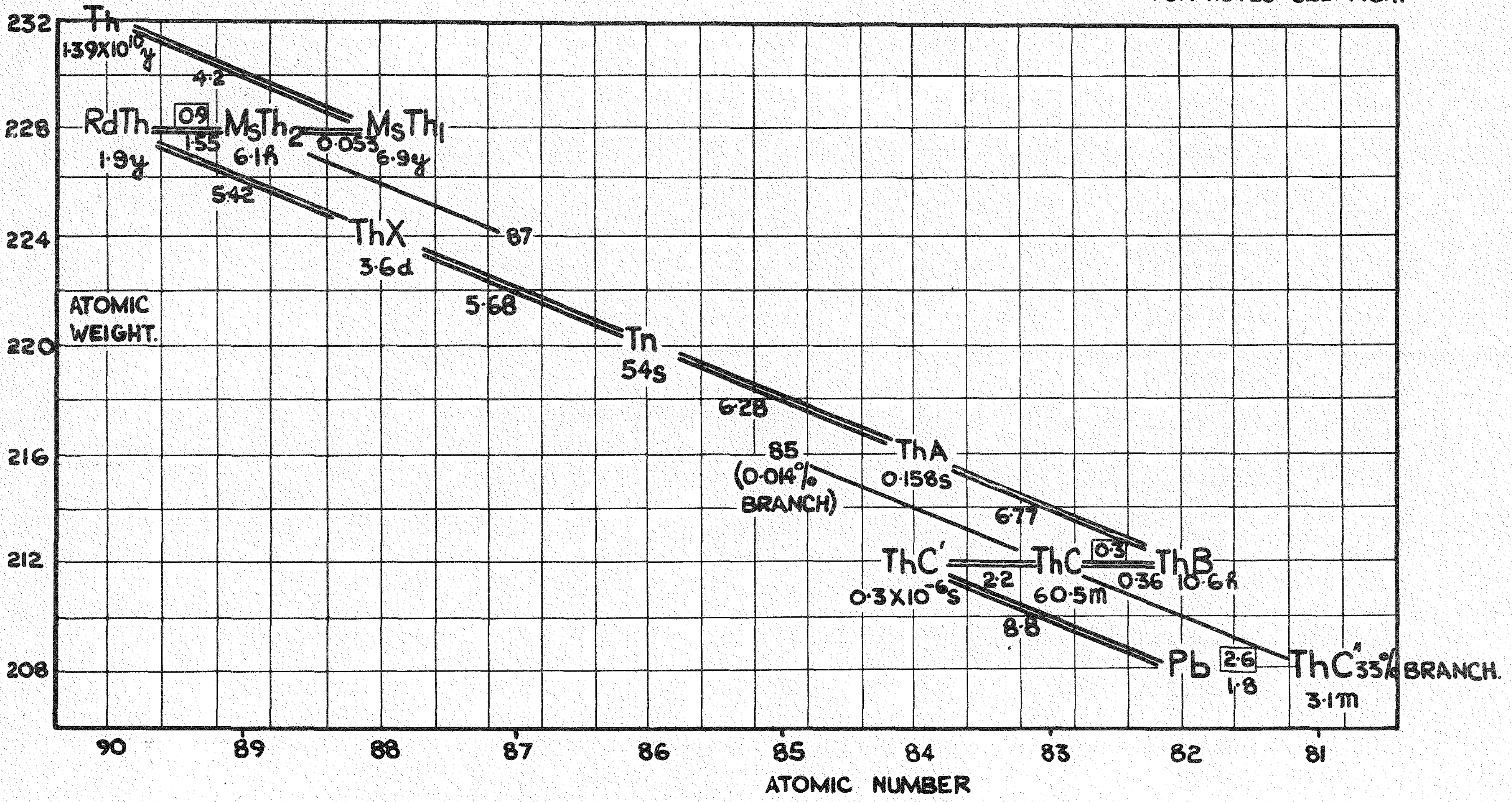


FIG. 3. THE THORIUM NATURAL DECAY SERIES.

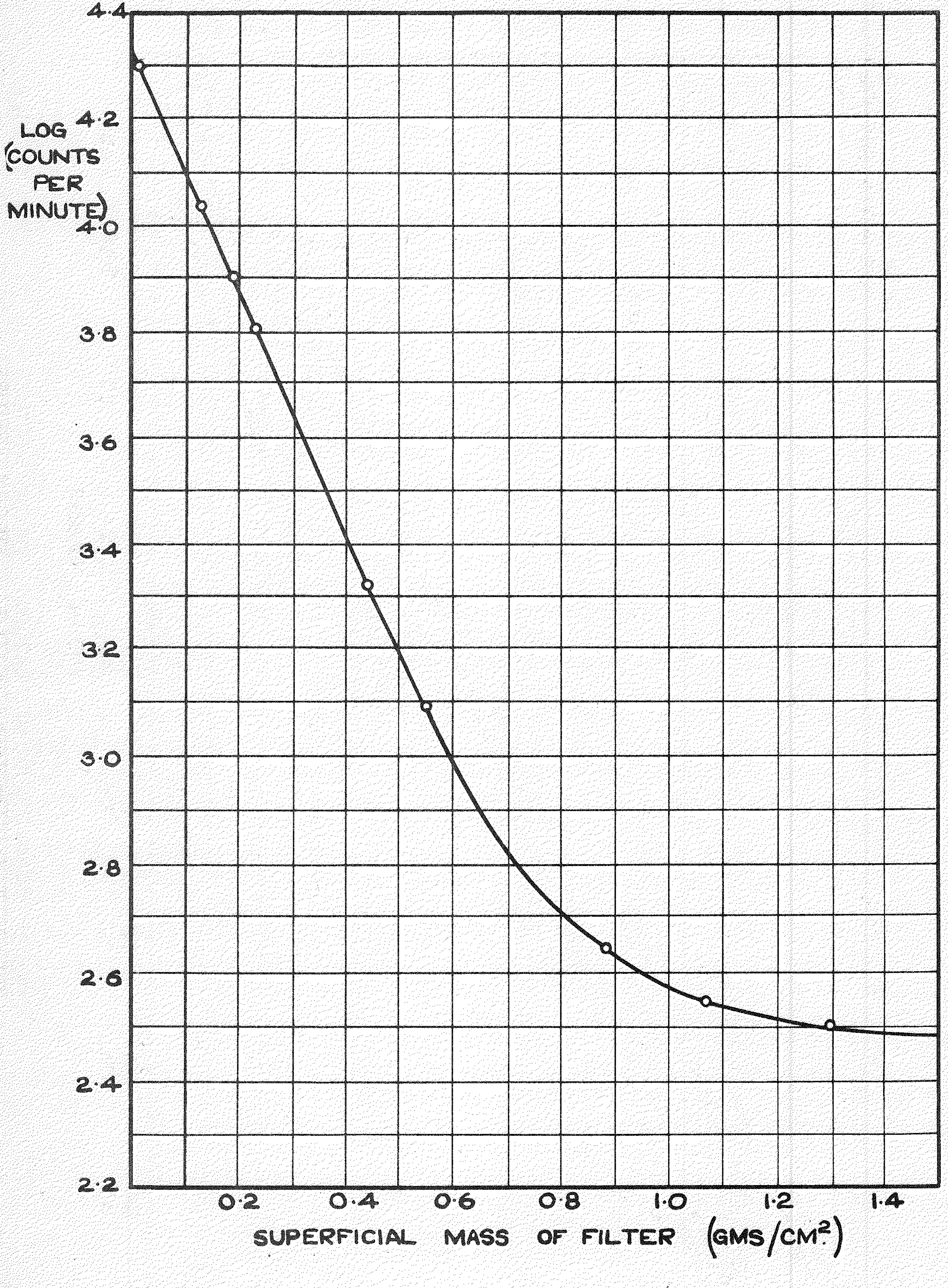


FIG. 4.

ABSORPTION CURVE FOR RADIATION FROM A THIN PITCHBLENDE SAMPLE.

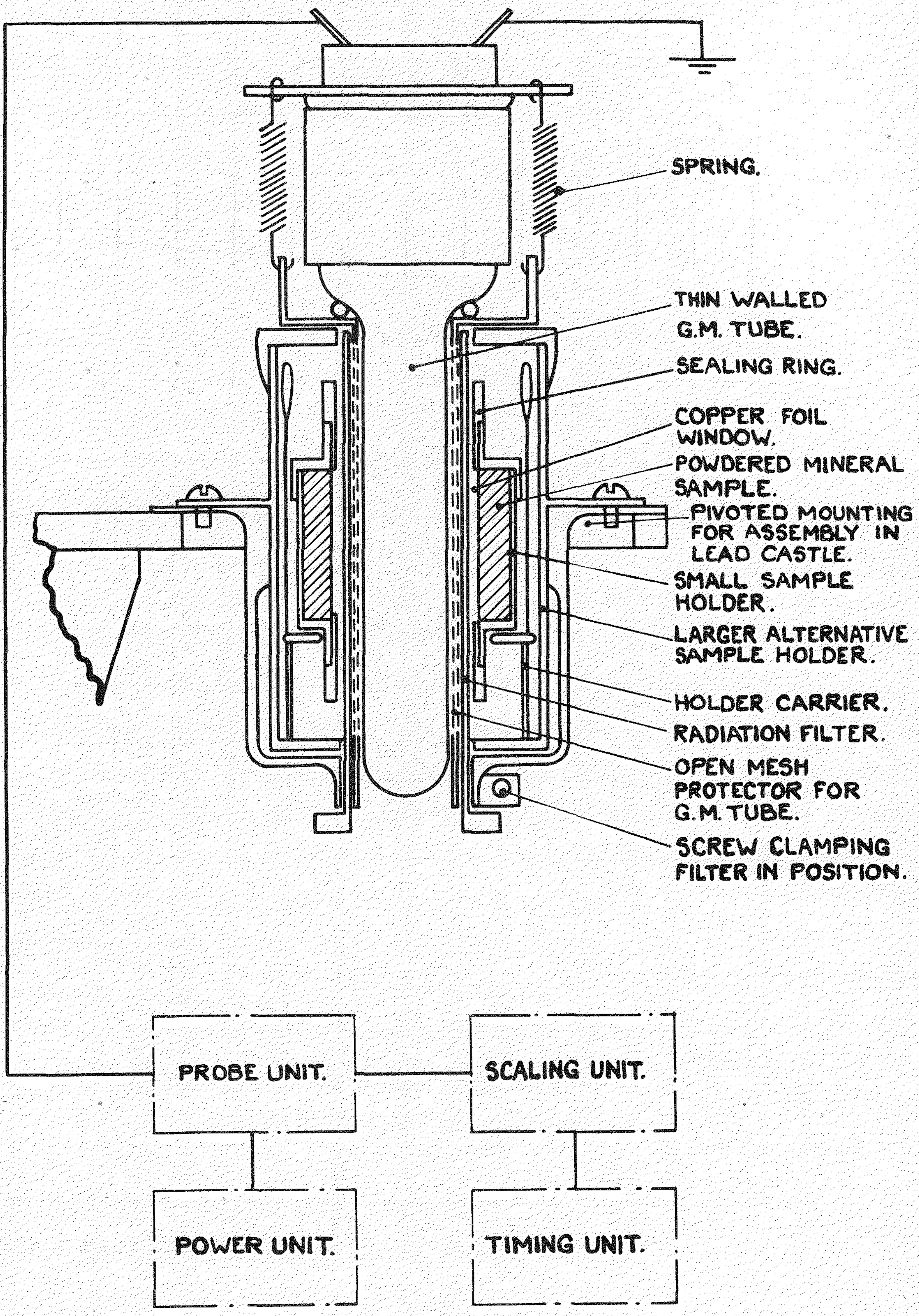


FIG. 5. ANNULAR SAMPLE HOLDER ASSEMBLY.

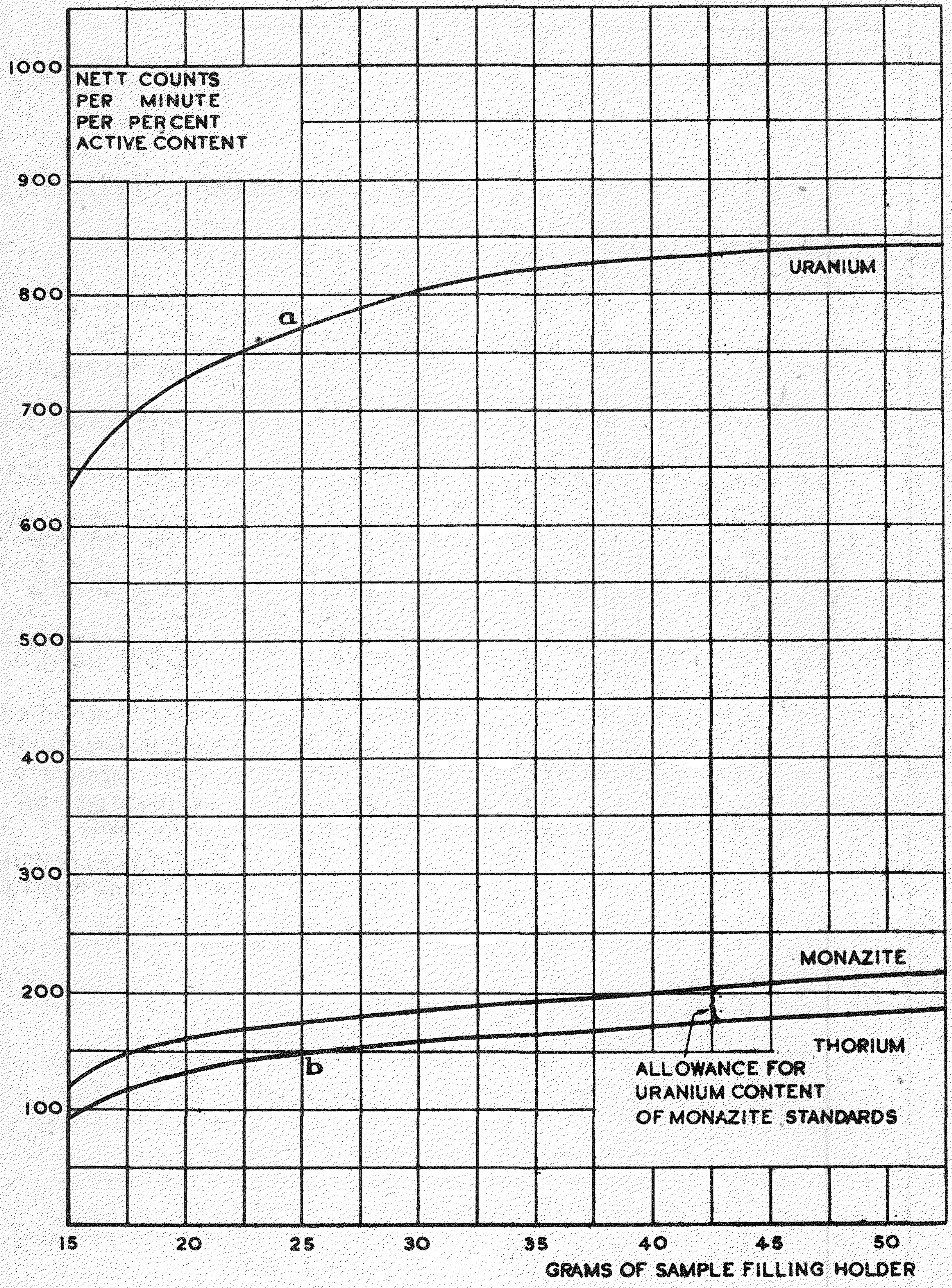
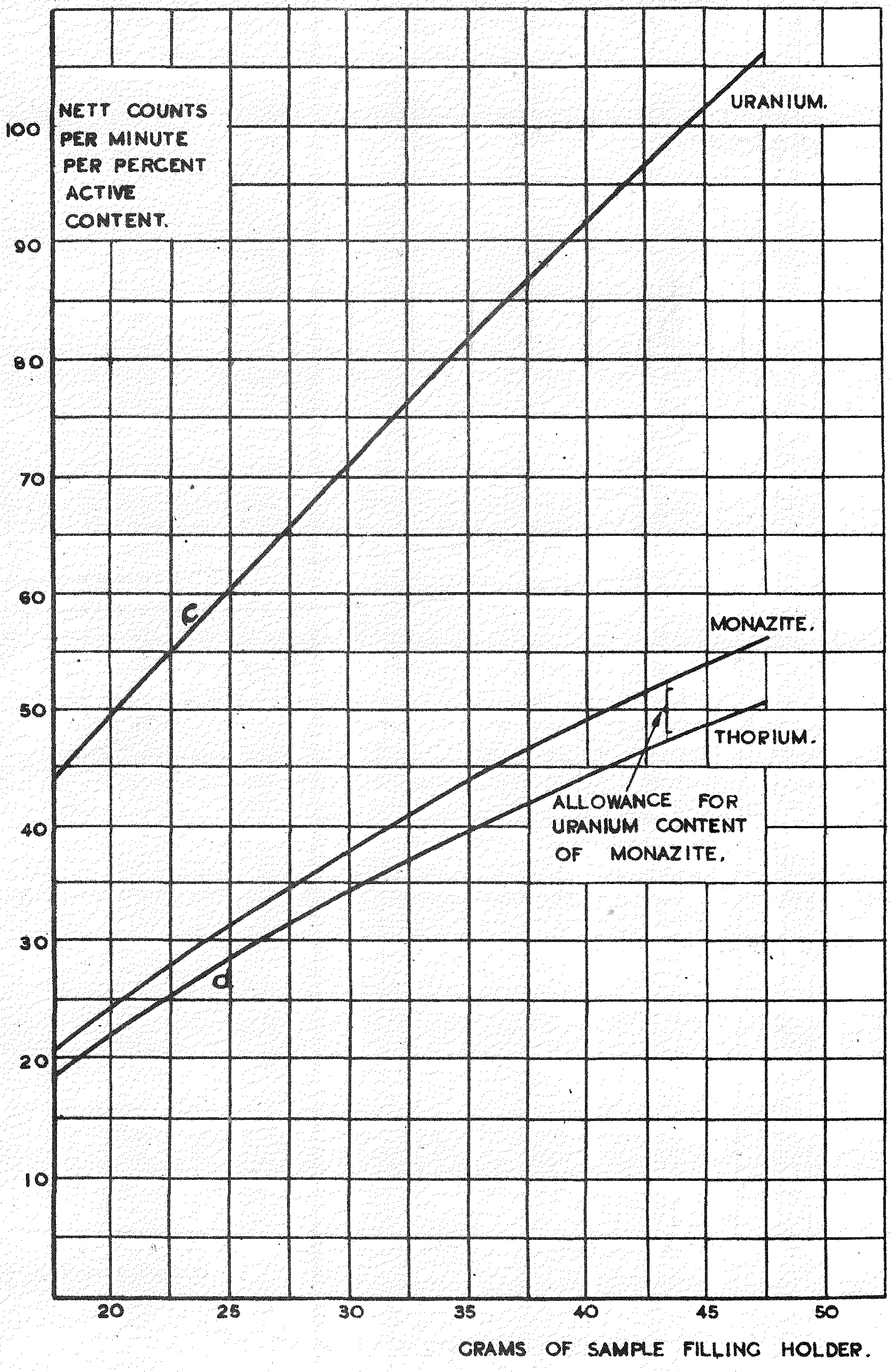


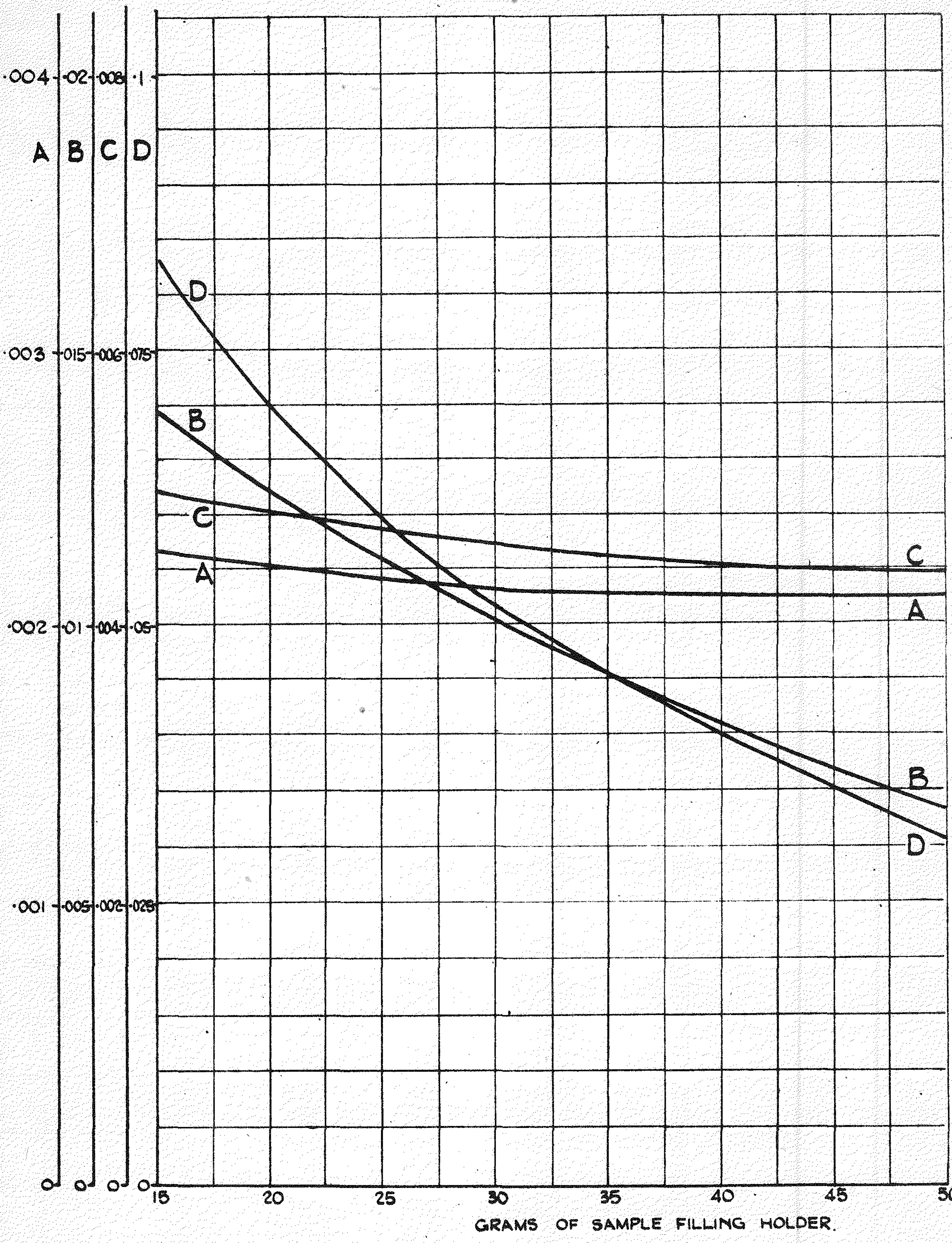
FIG. 6

COUNT RATE CURVES (END WINDOW HOLDER, NO FILTER)
— AVERAGE CURVES.



FILTER : Nº 10 GAUGE ALUMINIUM (890 MGMS / CM^2)

FIG. 7.
COUNT RATE CURVES (END WINDOW HOLDER WITH FILTER.) AVERAGE CURVES.



URANIUM CONTENT = (+AN − BF)%

THOIUM CONTENT = (−CN + DF)%

N = NETT COUNT RATE WITHOUT FILTER, F WITH No. 10 GAUGE ALUMINIUM FILTER.

FIG. 8.

CURVES FOR MIXED ASSAY. WITH END WINDOW COUNTER

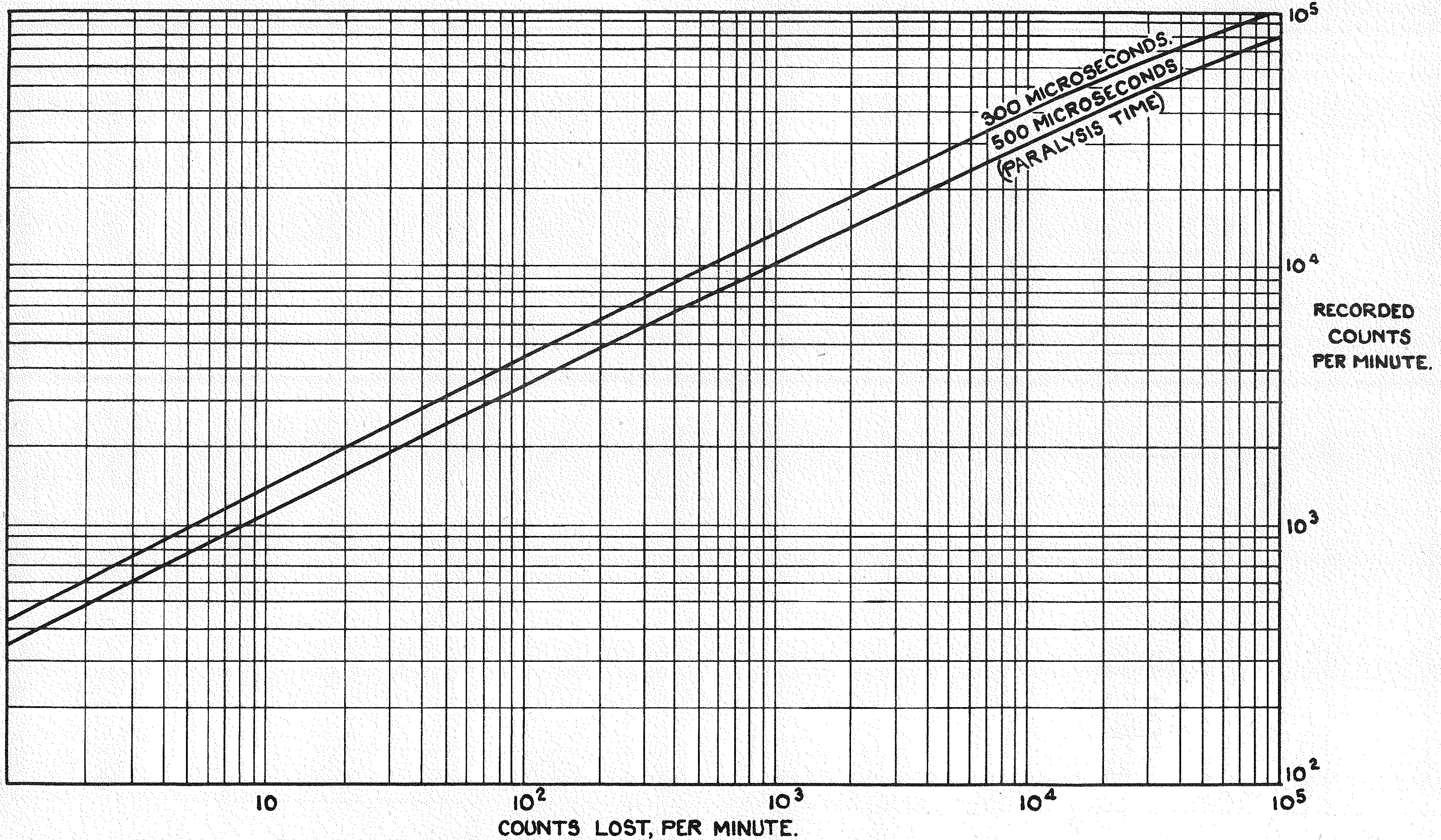


FIG. 9. CORRECTION CURVES, FOR PARALYSIS TIME LOSSES.

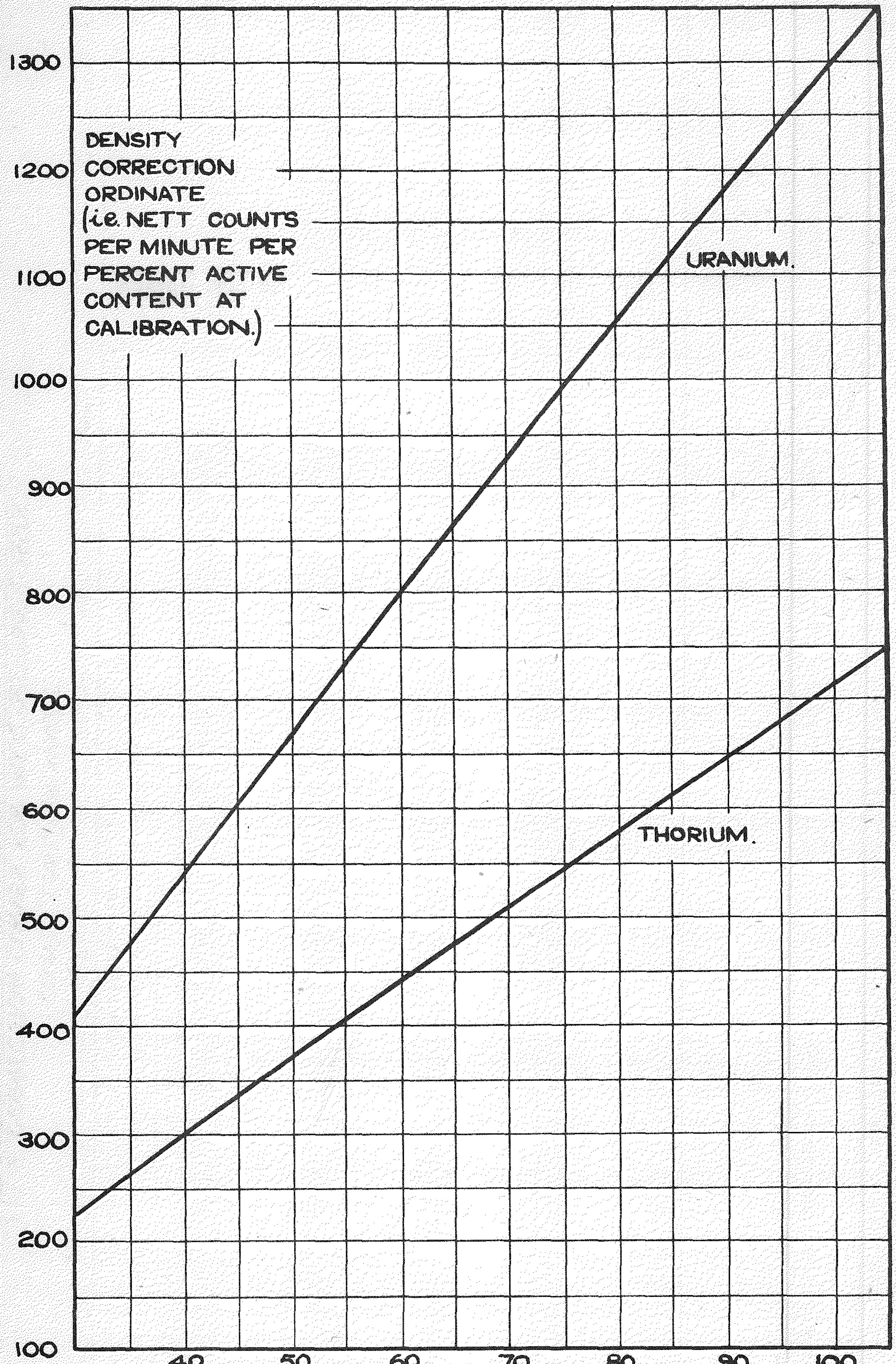


FIG. 10.

CALIBRATION OF GAMMA RAY ASSAY UNIT. (WITHOUT FILTER)

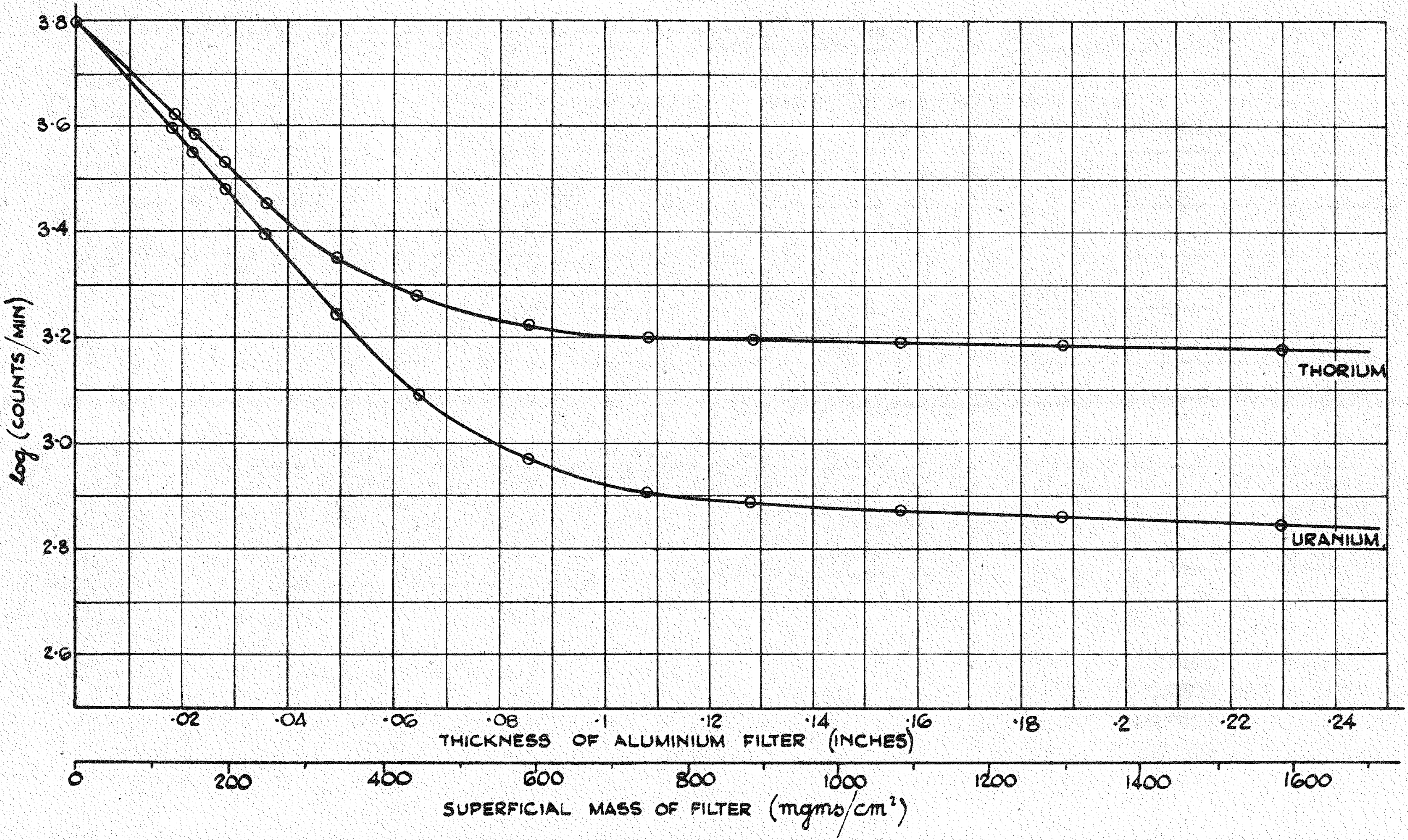
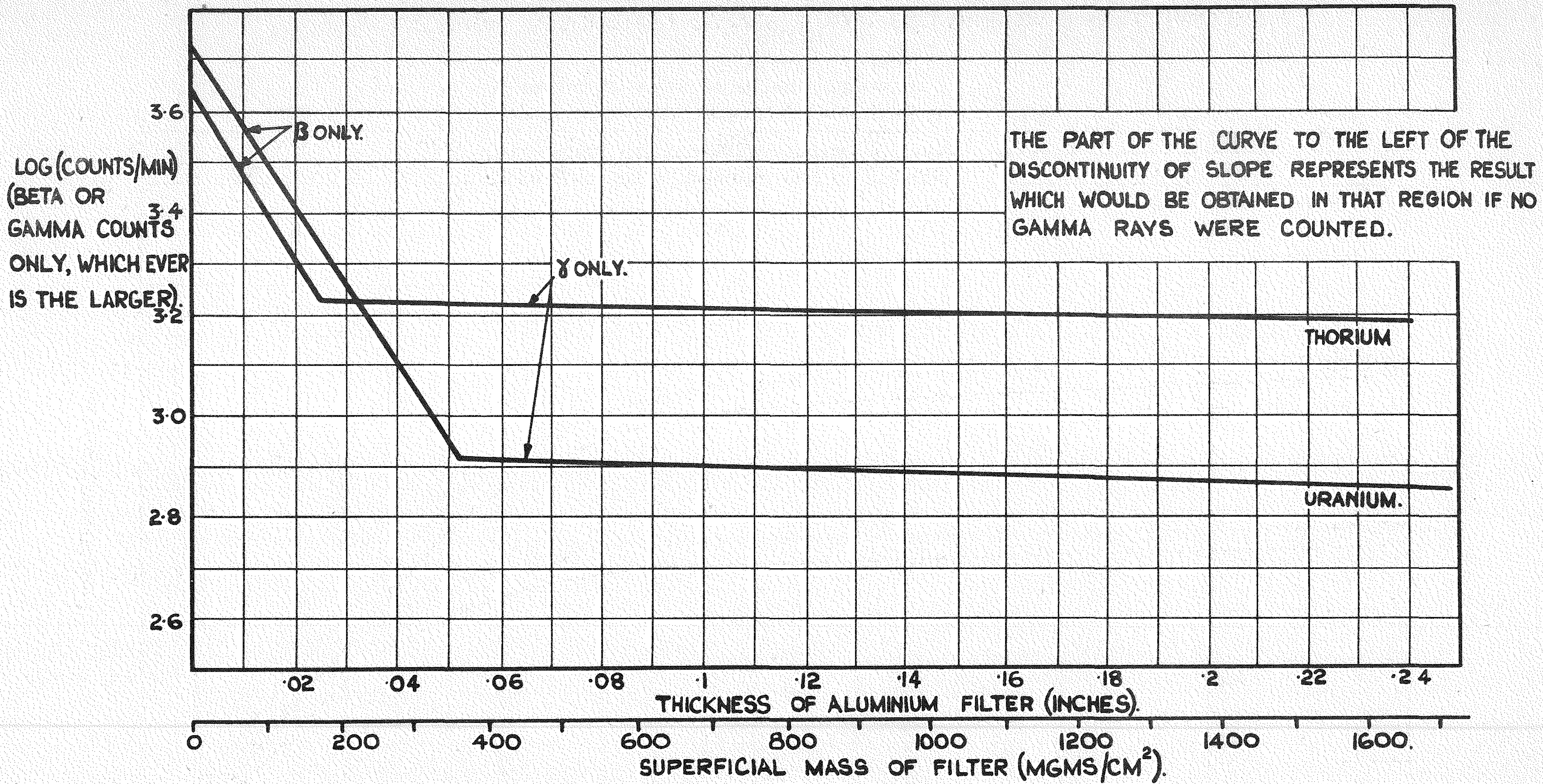


FIG. II.

EXTERNAL ABSORPTION CURVES (DEEP END WINDOW SAMPLE HOLDER)



THE PART OF THE CURVE TO THE LEFT OF THE DISCONTINUITY OF SLOPE REPRESENTS THE RESULT WHICH WOULD BE OBTAINED IN THAT REGION IF NO GAMMA RAYS WERE COUNTED.

FIG. 12. EXTERNAL ABSORPTION CURVES (COMPENSATED) (DEEP END WINDOW SAMPLE HOLDERS).

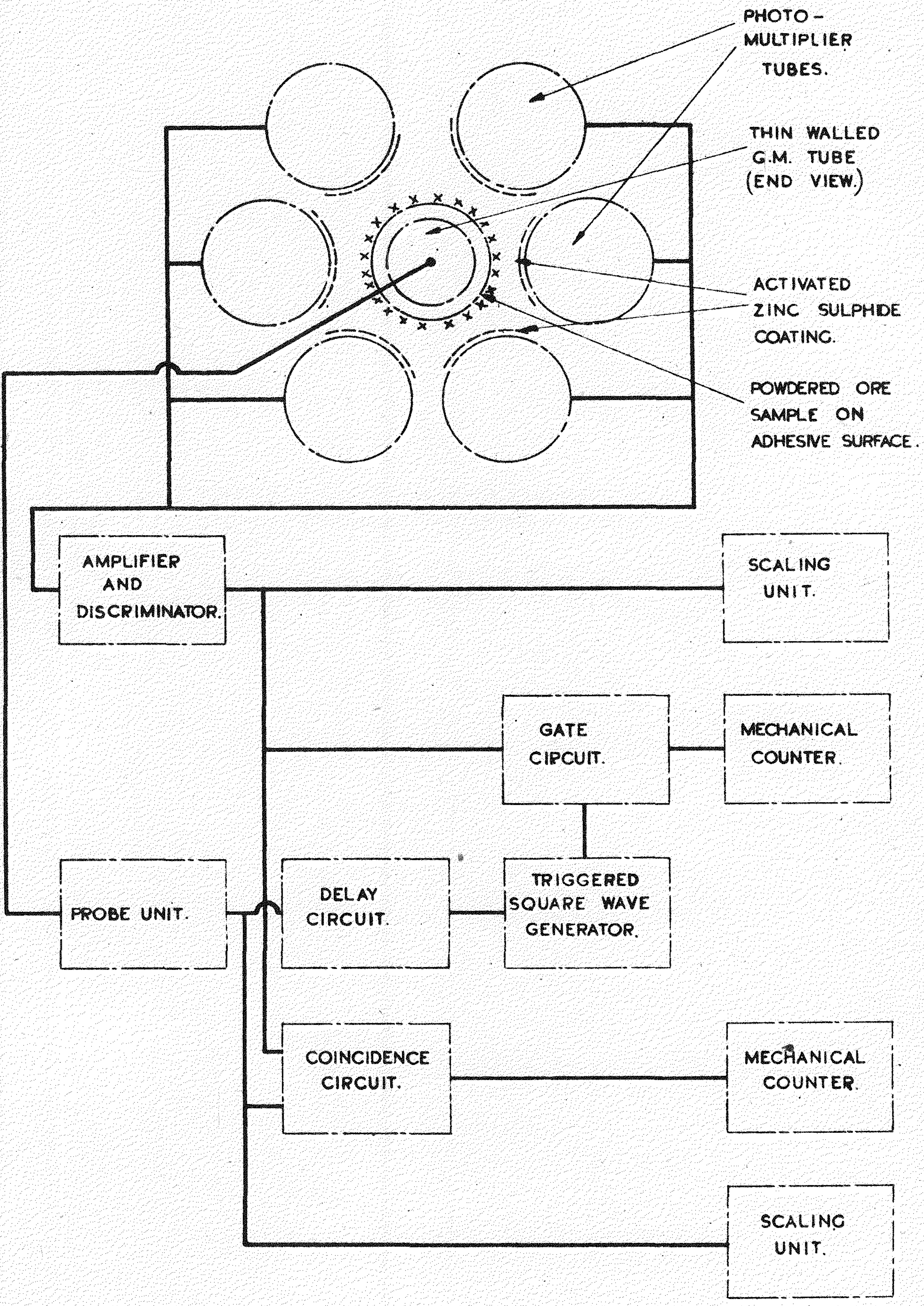


FIG. 13.
THE BETA-ALPHA TIME SPECTRUM METHOD.

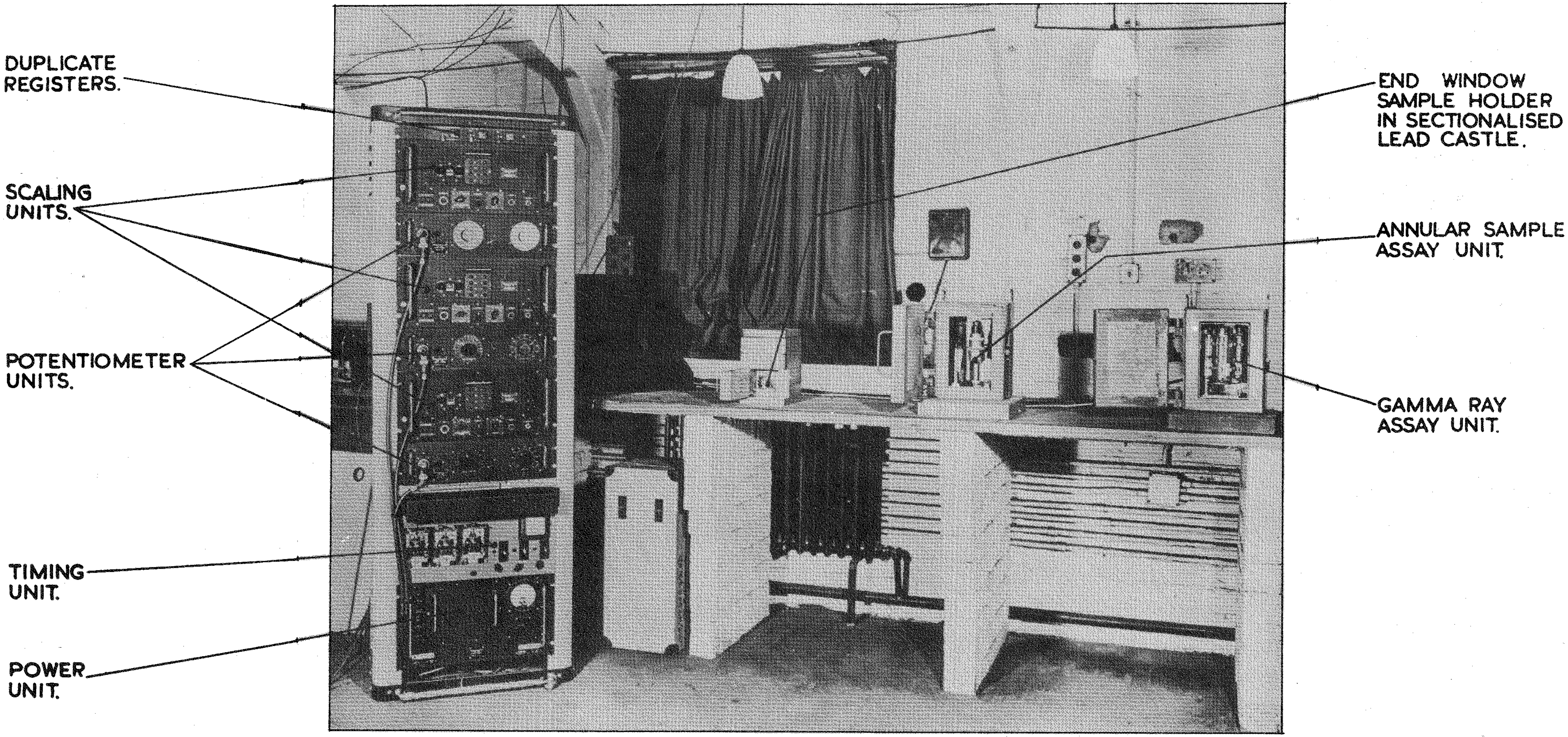


PLATE I. RADIOMETRIC ASSAY EQUIPMENT.

END WINDOW ASSAY UNITS

PLATES 2 & 3

PLATE 2

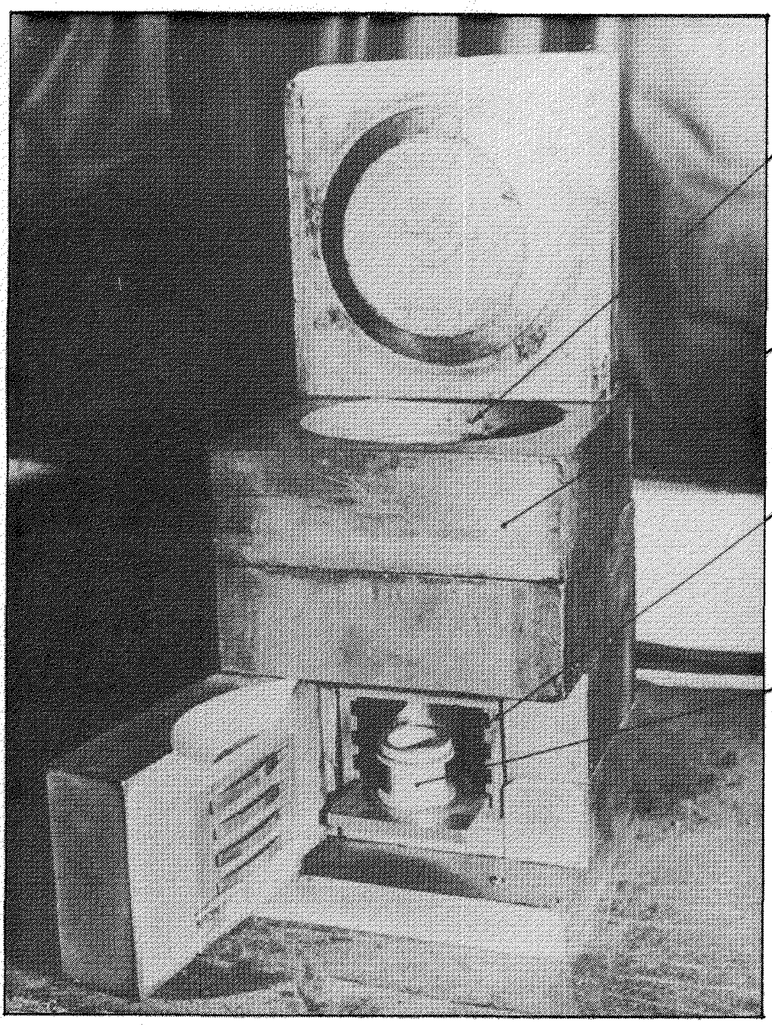
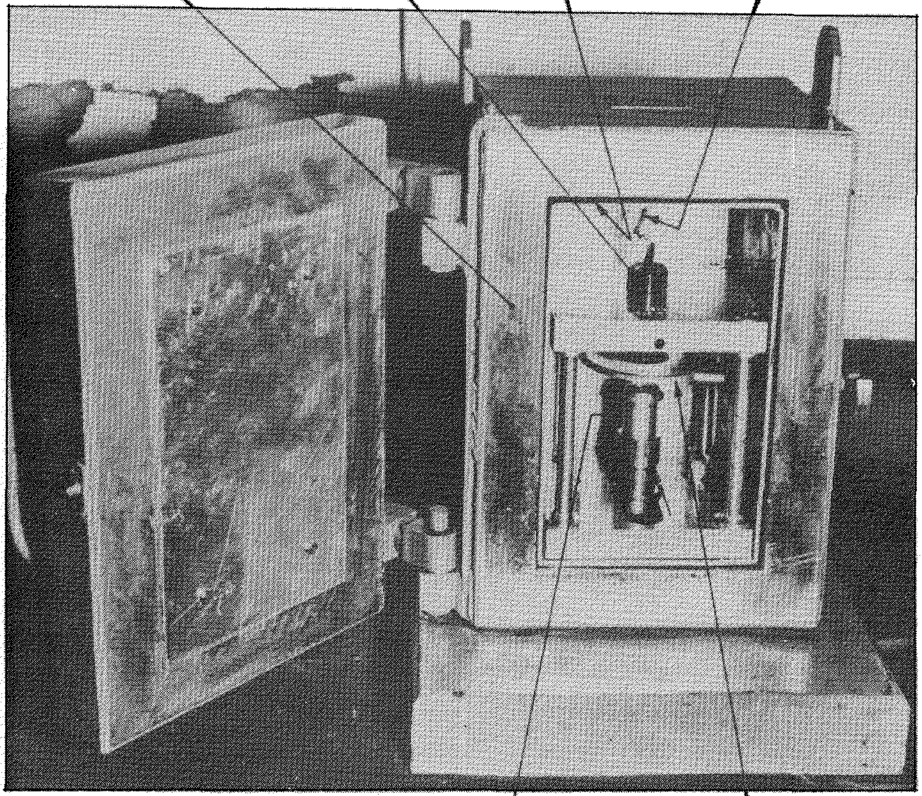


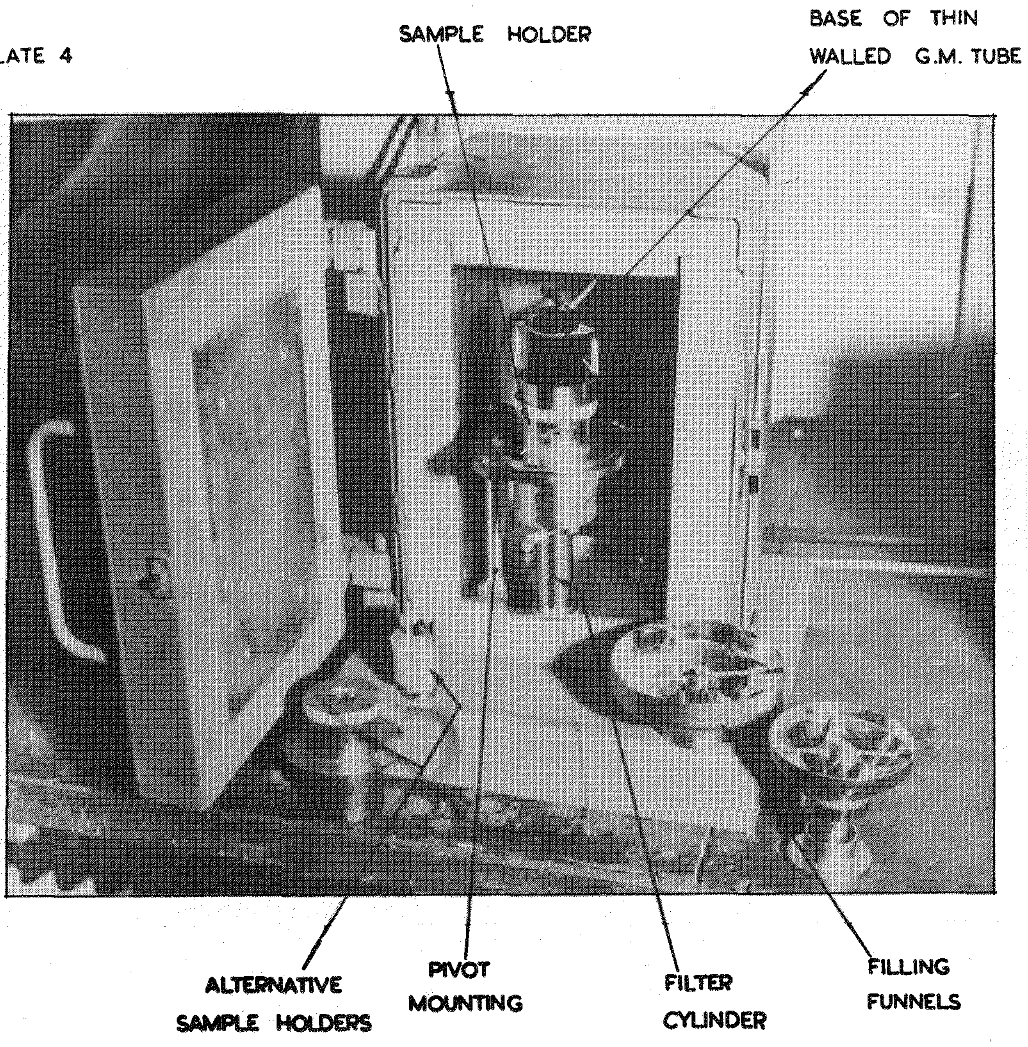
PLATE 3



PLATES 4 & 5

ANNULAR SAMPLE ASSAY UNIT

PLATE 4



GAMMA RAY ASSAY UNIT

PLATE 5