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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

AN IMPROVED METHOD OF QUANTITATIVE
AUTORADIOGRAPHY
FOR INDUSTRIAL PURPOSES

by

L. MATTEOLI and P. LOGI
(Istituto di Ricerche Breda)

1966



Report prepared by the
Istituto di Ricerche BREDA, Milan - Italy

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By comparison of the photographic densities of the standard and the sample autoradiographs, taking into account the correction factors, a correlation is calculated, at first between photographic density and radioactivity of the tracer, then between photographic density and the concentration of the tagged constituent in the sample.

Series of radioactivity standards have been prepared for ten beta-emitting radioisotopes; the relative photographic efficiency has been determined, so as radioactivity standards of radioisotopes differing from those under examination could be used.

Working conditions for reproducible exposures and developments have been determined; several photographic emulsions have been tested.

A method to make thin sections of metals and minerals specimens has been developed.

Quantitative autoradiography was then applied in five studies, that are reported as examples of possible applications. They refer to the examination of components liquation in alloys, to the tagging of a mineral, to the absorption on steel surface of a corrosion inhibitor.

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SUMMARY

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INTRODUCTION

Radioactive tracers are widely used in a great variety of scientific research processes.

Autoradiography is a method of detection used wherever it is desired to determine the distribution as well as the presence of the radioactive tracer (labelled compound or element).

In most cases this method is used only to reveal the qualitative distribution of the tracer; but the findings are, of course, more useful when the technique is refined so as to give quantitative results, thus showing not only the relative variation in tracer concentration from one point to another, but also the actual concentration value.

From a review of the literature¹⁾ on research into the application of the autoradiography method, or on cases where it was used as a research tool, it emerges that the restriction to purely qualitative results often prevented further progress in exploring the problems in hand. For this reason, various attempts were made to obtain fuller information from the autoradiograph, i.e. to convert the qualitative determinations. In most instances this operation to

¹⁾ See Appendix for bibliographical references on the subject
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obtain quantitative results from autoradiography was devised to meet a specific problem, i.e. to provide a means of determining a given tracer in a given matrix according to the particular requirements of the case in question.

The Radioisotopes Laboratory at the Breda Research Institute has in the past carried out a number of research projects in the field of metallurgy and metallography, using radioactive tracers and detecting them by autoradiography, and on more than one occasion it was apparent that it would be highly useful to obtain quantitative results from the autoradiographs. In such cases special techniques were adopted which from time to time yielded the desired results; that is to say, the techniques employed were designed and developed each time to meet the individual case.

Since the method of investigation by autoradiography is by now relatively well developed and frequently used in metallurgy and metallography - a field which specifically interests the Breda Research Institute - it was decided that there would be practical advantages in launching a project aimed at the development of a quantitative autoradiography method, to be relatively simple and as widely applicable as possible. This method, via the preparation of radioactivity standards and relative comparison data, would rule out the need for repetition of the initial preparatory operations required for each new autoradiographical application, a procedure so complicated and time-consuming that it often militates against the use of this sometimes invaluable research tool.

Investigation procedure

It will perhaps be useful to start with a summary of the programme of work which was effected. The research was aimed at developing a technique for the quantitative determination of radio-tracers by means of measurements performed on their autoradiographs.

For this purpose a relative measuring method was adopted. The method applied calls for simultaneous autoradiographs of standard sources and of the tracer-containing test specimen; the concentration and distribution of the tracer is derived from a comparison of the photographic densities of the autoradiographs obtained. To render

the method suitable for more general application, a relative photographic efficiency factor must be determined between one radioisotope and another, so that use can be made of standard sources of radioisotopes other than that contained in the test specimen.

This factor is particularly useful in cases where it is intended to apply the quantitative determination method to short-lived tracers and long-term standards cannot be prepared.

The exposures are calculated to produce a visible blackening of the autoradiographs, and determinations are effected by comparison of the optical densities, measured with a microdensitometer.

To produce a quantitative process, the following factors were taken into account :

- Standard sources

The standard sources were all produced by the same method; they consist of thin films of photographic gelatine containing a soluble salt of the radioactive isotope. Standards of varying activities were prepared for each radioisotope. Their characteristics are uniform, and all standards can be used in the same exposure conditions.

- Specimens

The specimens labelled with radioactive tracer were prepared for thin-section examination. In thin section the specimen presents the best geometrical conditions for higher resolution of the autoradiographic image and also for calculation of the corrections required for self-absorption.

- Carriers

The carriers employed were all of the same material, selected, of course, from materials composed of elements of low atomic weight in order to reduce back-scattering effects.

- Exposure and development conditions

The geometrical exposure conditions are particularly important. Care was taken to have thin specimens and standard sources, to reduce to a minimum the distance between source and photographic emulsion, and to use emulsions with a thin sensitive layer. These

conditions were made reproducible.

To avoid fading, exposures were effected at low temperature and in an inert atmosphere; whenever two autoradiographs had to be compared, they were obtained by simultaneous exposure with emulsions taken from a single plate or film, and developed simultaneously.

Development was carried out in strictly controlled and reproducible conditions.

- Photographic material

Various samples of photographic material were tried, the two finally selected being Kodak V 1056 plates and Kodak Microtex film for radiography.

When all the above preparations had been carried out, autoradiographs of the standards were prepared in substantial quantities, on the principle that a large number of measurements would dispose of the occasional variations still present. The photographic efficiencies of the radiations from the various radioisotopes tested were then compared and the efficiency ratios were determined. This made it possible, when determining tracer distribution in a specimen by quantitative autoradiography, to use a standard of some other more convenient radioisotope.

Lastly, five examples of applications of quantitative autoradiography were studied; three were applications to metallurgical problems, one to an investigation of corrosion inhibitors, and one to sand labelling.

Preparation of the standard sources

A technique was devised, by which sources of any radioisotope could be prepared according to a satisfactorily reproducible set procedure. Of the various methods tried, it proved most convenient to use a photographic gelatine containing the tracer in a soluble salt form. Gelatine was found to be an exceptionally effective medium for dispersing the radioactive tracer, since it makes it possible to control the tracer concentration in the sources produced, the distribution uniformity and the source thickness, while it can also be used with any tracer available in soluble salt form.

The radioactive sources had to be in thin film; they were prepared by a conventional technique, used in photographic sensitometry to obtain gelatine films of increasing thickness (1) and also in the preparation of radioactive gelatine wedges employed in quantitative autoradiography in the field of biology (2).

For each radioactive tracer studied, 6-8 sources of different activities were prepared. A source-activity variation scale had to be selected; it could have been scaled according to a law of linear variation, with the factor $K = 1, 2, 3, 4$, etc. The specific activity/photographic density graphs would then have been plotted from a set of radioactive exposure values distributed linearly on the abscissae. There was an obvious drawback, however, in that only a rather narrow interval of possible tracer concentrations would be covered. For this reason it was thought preferable to prepare the sources with activities scaled according to a factor $K = 1, 2, 4, 8, 16, 32$. In that way, with a limited number of standards, a broad interval of concentrations could be covered in accordance with requirements.

Under the programme six standard radioactive sources were to be prepared; in the course of work, however, owing to the requirements of the scheduled specimen applications (see below) ten standards in all were prepared, namely, H3, C14, S35, Ca45, Pm147, Cl36, Y91, Sb124, P32, Sr-Y90. These are beta-emitting radioisotopes, ranging in β -energy from tritium (H3) up to Sb124.

The maximum energy in MeV and the half-life $T_{1/2}$ for each of the above-named isotopes are as follows :

| Isotope | max. beta E | MeV | $T_{1/2}$ |
|---------|-----------------------------------|-----|-----------------------|
| H3 | 0.018 | | 12.26 years |
| C14 | 0.155 | | 5760 " |
| S35 | 0.167 | | 87.2 days |
| Pm147 | 0.22 | | 2.6 years |
| Ca45 | 0.25 | | 165 days |
| Cl36 | 0.714 | | 3×10^5 years |
| Y91 | 1.53 | | 58 days |
| P32 | 1.71 | | 14 " |
| Sr-Y90 | 0.54-2.25 | | 28 years |
| Sb124 | 0.22-0.61-0.95- 1.59-1.66-2.31 | | 60 days |

In line with the specific activities available for the chosen tracers, the attempt was made to prepare the standards of the various elements in such a way that they would all be comprised within a single specific activity interval. It was found, however, that there are limits to the preparation of standards with high specific surface activity for those tracers which are only available with low specific activities, owing to a ceiling for saline concentration in the photographic gelatine, above which gelatinization deteriorates or fails completely.

The isotopes of lower specific activity employed include Ca^{45} and Cl^{36} , used in the form of $\text{Ca}(\text{NO}_3)_2$ and NaCl respectively.

The greatest concentrations of these salts, compatible with proper jelling of the 12% gelatine solution used in preparing the standards, were 0.15 - 0.20 mol/litre.

Method of operation to prepare the radioactive films

The method chosen for preparing the radioactive films was as follows.

A mother solution of the radioactive tracer is prepared, with a concentration of approximately 4 microcuries/cc (or higher for short-lived tracers, for which it is expedient to prolong the useful life of the standard sources).

This solution is neutralized before addition to the gelatine and, where under neutral conditions it tends to yield a precipitate, a gelatine concentration of 0.1 - 0.2% is added before neutralizing.

Next, six small crucibles are prepared, containing an equal weight of dry gelatine; by micropipette a portion of radioactive solution is added in proportions of 1,2,4,8,16 and 32, and then water to bring the six solutions up to an equal weight and the gelatine to 12%. The crucibles are sealed and placed in a thermostatic incubator at 85°C for the time required for complete homogenization; normally 2 hours will be long enough for this.

Meanwhile frames must be prepared, in which to form the films. Each frame requires two 50 x 70 mm slides of optical glass selected for flatness.

The upper slide is washed with a chromic mixture, then mounted on a rotating plate, covered with a colloidal solution of albumin and centrifuged at about 400 rpm. After drying, the glass will be covered with a uniform thin film of albumin (about 0.5 micron) completely insoluble in water. The colloidal solution of albumin is prepared with the white of one egg, diluted in 6 parts of distilled water to which 0.5% phenol has been added; the whole is boiled in a water-bath.

The lower slide is treated with a solution of silicone oil or grease so as to cover it with a fairly thin film.

The object of these coatings is to make it easier to detach the upper slide from the gelatine film when it is still wet after jelling, and the dry film from the lower slide. A frame is formed by mounting these two glass slides one above the other, with the coated sides facing one another and separated by spacers placed along three sides, so as to leave the fourth side open with a useful central area of about 50 x 50 mm. The three spacer inserts are 0.2 mm thick.

When prepared, the frames and a set of Pasteur pipettes are kept in a thermostatic incubator at 65°C until required.

Each of the radioactive gelatine solutions already prepared is transferred by pipette into a frame, until the space between the two slides is full. The frame is rapidly cooled to 4-5°C to hasten the setting of the gelatine, which occurs in 5-15 minutes. The upper slide is then detached and the film is left to dry for 12 hours at 5°C. The drying process must not be too rapid.

When the film is dried out, it is cut along the edges and lifted off the second slide. The finished films have a thickness of 20 microns, and if the frames and the radioactive gelatine solutions have been prepared correctly, the maximum thickness variation will be 1-2 microns over an area of 50 x 50 mm. Films prepared by this method should preferably be stored in a cool place, with humidity controlled at 40-60%.

Uniformity of tracer distribution in the films, as checked by autoradiographs of the prepared standards, proves very satisfactory;

the relative deviations from the pre-established activity ratios are 15% at the most. It should be noted in this connection that a divergence from the pre-established concentration scale is of no practical importance, since all the standards are subsequently classified according to their absolute surface activity.

Absolute measurement of activity of standard sources

The six radioactive films, of scalar activity for a given radioisotope, prepared by the foregoing method, are trimmed down to one disc each of 5 mm diameter. These six discs are used to prepare a set of standard sources of that radioisotope.

The first step is to measure the absolute radioactivity emitted from one face of the disc.

This is performed with a Geiger counter rigged for 4π geometry measurements on a source located inside the counter. The counter, constructed in the Laboratory, is a revamped version of a familiar model.(3)(4). Fig. 1 shows an operating diagram of the counter, which when in use is filled with a mixture of argon and 10% ethyl alcohol, at a pressure of 100 mmHg. For measurements on standard sources, the radioactivity emitted in a 2π semispace is determined. The measurement error is estimated to be 3%.

Table 1 gives the activities measured for the different standard source sets produced. Table 2 shows a comparison between 2 sets of standard sources of a single isotope.

Table 1 - Radioactivity of the sets of standard sources

| Radioactivity in B/mm ² | | | | | | | | | | | |
|---|---|----------------|-----------------|-----------------|------------------|-------------------|------------------|-----------------|-------------------|-----------------|--------------------|
| Element | | H ³ | C ¹⁴ | S ³⁵ | Ca ⁴⁵ | Pm ¹⁴⁷ | Cl ³⁶ | Y ⁹¹ | Sb ¹²⁴ | P ³² | Sr-Y ⁹⁰ |
| Sources | 1 | 355 | 425 | 355 | 132 | 520 | 1060 | 810 | 72 | 980 | 1750 |
| | 2 | 110 | 280 | 196 | 86 | 285 | 480 | 370 | 29 | 470 | 815 |
| | 3 | 66 | 126 | 100 | 38.5 | 145 | 265 | 172 | 14.7 | 275 | 395 |
| | 4 | 19.5 | 53 | | 17.5 | 66 | 120 | 80 | | 114 | 220 |
| | 5 | | 22 | | 10 | 36 | 58 | 37.5 | | 50 | 100 |
| | 6 | | 14.5 | | 4.7 | 16 | 36.5 | 14 | | 32 | 53 |
| Date on which measurement was performed | | 3.3.65 | 11.9.64 | 13.4.65 | 21.5.64 | 29.12.64 | 15.5.64 | 24.8.64 | 8.7.65 | 9.7.64 | 27.11.64 |

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Table 2 - Comparison between pairs of sets of standard sources

| Radioactivity in B/mm ² | | | | | | | |
|---|---|-----------------|---------|------------------|----------|-----------------|----------|
| Element | | C ¹⁴ | | Sr ⁹⁰ | | Y ⁹¹ | |
| Set | | 1 | 2 | 1 | 2 | 1 | 2 |
| | 1 | 425 | 423 | | 1750 | 176 | 186 |
| | 2 | 280 | 270 | 830 | 815 | 80 | 84 |
| | 3 | 126 | 126 | 405 | 395 | 37.5 | 30 |
| | 4 | 53 | 53 | 207 | 220 | 17.4 | 17.2 |
| | 5 | 22 | 19.3 | 102 | 100 | 8.2 | 8.4 |
| | 6 | 14.6 | 14.1 | 53 | 53 | 3.1 | |
| Date on which measurement was performed | | 11.9.64 | 21.5.64 | 15.6.64 | 27.11.64 | 30.12.64 | 30.12.64 |

Mounting the sets of standard sources

In order to have readily usable sets of standard sources it is necessary to provide for suitable mounting of the discs obtained from the films of radioactive gelatine.

This is done by sticking the radioactive film discs on to a cellulose-acetate backing 0.2 mm thick by means of "Scotch" tape which is adhesive on both sides.

The discs are arranged in a row and are so spaced that, even with sources of the highest radiation energy, there can be no interferences between sources in the formation of the autoradiographic image.

When the discs have been attached to their backings the latter are given a coating of Saran in order to provide mechanical protection of the source and to exclude moisture; this also serves to cover up the still exposed portions of the adhesive tape.

The Saran coating is obtained either by centrifuging a 2% solution of Saran in methyl ethyl ketone on glass at 800 rpm, or by causing a single drop of 7% Saran solution in cyclohexanone to spread over a surface of distilled water and collecting the film that forms. This protective film has a thickness of about 0.3 micron.

A protective coating cannot be used in the case of low-energy radiation sources (e.g. tritium); in such cases talcum powder is applied to the exposed portions of the adhesive tape.

Photographic material employed

Various types of photographic emulsion were tested and their possibilities for use in autoradiography examined.

The following types were investigated:

- Kodak emulsions

1. Fine Grain Autoradiographic Stripping Plates AR 10
2. Fine Grain Autoradiographic Stripping Plates AR 50
3. Experimental Scientific Plates V 1056
4. Microtex X-ray Film
5. Cristallex X-ray Film
6. Industrex X-ray Film
7. Kodirex X-ray Film

- Ilford emulsions:

8. Nuclear Research Plates K 5
9. Nuclear Research Plates G 5

Types 1, 2, 3, 8 and 9 are specially designed for laboratory use and the detection of nuclear radiations.

Types 4, 5, 6 and 7 are emulsions for industrial use and X-ray detection.

Characteristics of the photographic material

Types 1 and 2 are sensitive emulsions in a gelatine medium, applied to a glass plate from which they must be removed before use.

To prepare them for use, the sensitive film is stripped off the plate and placed on a water surface, on which it swells and spreads out, it is then transferred to the surface of the sample to be studied or to another backing, to which, when dry, it adheres without shrinking.

Emulsions of this type are convenient for biological experiments in which it may be desirable to have the autoradiograph mounted directly on the sample in order to compare the optical and autoradiographic images; in metallographic studies, however, this kind of mounting gives rise to difficulties on account of the reactivity of the metal surfaces with respect to the wet emulsions which are deposited on them.

Furthermore, for quantitative applications, when it is desired to determine the density of the autoradiographic image, the emulsion must have a transparent backing. In such cases a glass plate is used.

Type 3 consists of the same emulsion as the AR 10, but the emulsion is permanently attached to the plate. Since this type is ready for immediate use, whereas the AR 10 has to be prepared, the former was preferred for the purpose of the present investigation. The data obtained on the V 1056 (type 3) are, of course, also applicable to the AR 10.

Tests were then carried out with the four types of X-ray film. Fig. 2 shows the autoradiographs of three Ca^{45} sources exposed on Microtex, Cristallex and Industrex films respectively; the sensitivity increases approximately as 1 : 4 : 8, but the Cristallex and Industrex

emulsions are subject to more fogging than Microtex and have a coarser grain, which reduces the resolving power. Cristallex film is about 16 times as sensitive as Microtex but has such a coarse grain that it is unsuitable for autoradiographic work, even when only a moderate resolving power is required.

Emulsions 8 and 9 were compared, and Fig. 3 shows the autoradiographs obtained for a simultaneous exposure with Ca^{45} standards. The sensitivity of emulsions 8 and 9 is higher than that of type 1 but their resolving power is lower.

On the strength of a comparison of all the characteristics of the various emulsions tried, it was considered advisable to confine the investigation to two types, viz:

1. Kodak Experimental Scientific Plates V 1056
(similar to AR 10, but with fixed backing)

For the fineness of grain and the thickness of emulsion this type of plate gives the highest resolution. Its disadvantages are a low sensitivity and a pronounced tendency towards spotting due to the formation of images corresponding to blemishes or points where the emulsion has come into contact with other materials.

Consequently, these plates are eminently suitable for applications in which good resolution is required, there is sufficient tracer activity in the sample under study and it is possible to employ an accurate technique.

2. Kodak Microtex X-ray film

The sensitivity of this emulsion is greater than that of the above-mentioned plates and similar to that of the Kodak AR 50; it has, however, a finer grain than the latter and is rather easier to handle. Since this is really an industrial film the sensitive layer is protected and therefore largely unaffected by pressure, impacts or other spurious causes of blackening. The plastic backing makes it easier to use than V 1056 and in view of its sensitivity it is preferable to the latter whenever a particularly high resolving power is not essential. Faster emulsions will be resorted to only in cases where, owing to the specific activity available, it is not possible to obtain reasonably long exposures with Microtex, and where a measurable amount of blackening occurs.

It should be noted that on X-ray films the emulsion is applied to both sides of the plastic backing, which is 17 mm thick. Consequently, if the radiations are of sufficiently high energy, the use of these films as such results in blackening of both sensitive coatings. In such cases the image formed on the emulsion not in contact with the sample is hazy, of extremely low resolution and difficult to interpret for quantitative purposes. In our investigation, therefore, whenever we used a double emulsion we took care, after developing the autoradiograph, to obliterate the image formed on the emulsion not in contact with the sample. For this purpose we used Farmer's reducer, which was applied to the relevant surface until the image had entirely disappeared.

Exposure and development conditions - reproducibility

It is well known that there are numerous factors which can cause variations in the optical density resulting from an autoradiographic exposure. We shall therefore consider in detail all the conditions that can give rise to undesirable and uncalculable variations in the photographic efficiency of two standard sources to be compared.

Variations in the thickness of the sources

As described in a previous section, the thickness variation in the sources was within $\pm 5\%$ over a surface of $2,500 \text{ mm}^2$ and is therefore a great deal less over the area of a standard source of 19.5 mm^2 . In order to ascertain the effect of thickness variation in the sources, at the start of the preparation procedure a study was made of the difference in photographic effect for sources of increasing thickness and a constant vol/vol concentration of radioactive tracer (wedges); the tracer used was Ca^{45} . In Fig. 4 are reproduced the autoradiographs and the corresponding density records for two such Ca^{45} wedges; the thicknesses vary from 5 to 50 microns. Fig. 5 shows the graph for photographic density versus source thickness: it can be seen that, in the case of a Ca^{45} source, a thickness variation of about 5% gives rise to a density variation of 3%.

Exposure conditions - fading

In the interests of good resolution it is necessary to ensure good contact between the emulsion and the standard source or the sample to be autoradiographed. Use was therefore made of clamps whereby the film or plate is kept in contact with the source by means of a rubber pad which presses on it.

Another factor of particular importance is the study of fading, or obliteration of the latent image. It emerges from the relevant literature (5, 6, 7) that the causes of fading are to be sought primarily in the atmosphere with which the emulsion has been in contact, and in particular that fading is due to the presence of oxygen and moisture during exposure. Consequently, the exposures were carried out in a dry inert atmosphere and - since the phenomenon is also affected by the temperature - at a low temperature.

The exposure procedure to be adopted is therefore as follows: the clamps containing the sources and the exposed films are placed in opaque black-walled drier, which is then evacuated and filled with dry nitrogen, in the presence of a drying agent. Throughout the exposure the drier is kept in a refrigerator at a temperature of 5-7°C. When it is time to develop the films, the drier is taken out of the refrigerator and left to stand at ambient temperature before the clamps are removed.

The fading in these conditions was determined by two types of test. In the first test three Sr⁹⁰ standard sources having activities scaled in the proportion 1 : 1/16 : 1/32 were exposed to an AR 10 emulsion which had been transferred to a rigid backing; the exposure times were of decreasing length, with the longest exposure for the weakest source, and were so calculated as to enable equal blackening effects to be obtained under the action of the developer. Developing was done simultaneously at the end of the three exposures. Fig. 6 shows the fading percentage as a function of the time elapsed between the start of a given exposure and the development of the emulsion. It can be seen that the tread is very nearly linear and that the fading values are fairly high.

With the aim of ascertaining the importance of the phenomenon independently of the compensatory effect of simultaneous exposure of the emulsion to radiation, a second series of tests was performed in which Microtex films were exposed to the same Sr⁹⁰ source for one hour. After exposure the films were kept in dry nitrogen at 5-7°C until they were developed. An analysis was made of six autoradiographs, corresponding to six different time intervals between the end of exposure and development.

Figs. 7 and 8 respectively show the photographic density variation and the fading percentage as functions of the time interval between the end of exposure and development.

The standard error due to the measurements proved to be rather high and is shown in the above-mentioned diagrams. The fading phenomenon can be assumed to increase linearly up to intervals of 29 days between

exposure and development. This fading of the latent image during the exposure period will obviously have to be taken into account in quantitative autoradiographic calculations whenever the sample under study and the reference standard cannot be exposed simultaneously and for the same length of time.

Still with the object of providing the optimum reproducibility conditions, each series of exposures with samples or with standards for intercomparison was performed with emulsions obtained from the same plate or film.

Development conditions

The development procedure employed was as follows:

- (a) Conditioning of the emulsions at development temperature;
- (b) Development for 5 minutes in Kodak D 19 b developer;
- (c) Arresting for 5 seconds in 5% acetic acid solution;
- (d) Fixing for 5 minutes in Kodak F5 fixer;
- (e) Prolonged washing in water, followed by washing in distilled water.

The developing and fixing solutions were kept in an inert atmosphere; the temperature of the baths was ultrathermostatically controlled to within $\pm 0.2^{\circ}\text{C}$. The baths were renewed for each development operation.

A study was then made of the optimum conditions as regards the developing-bath temperature. For this purpose, autoradiographs of a single Sr^{90} source were made with Microtex film, each one being developed at a different temperature.

Table 3 gives the characteristics obtained by developing at 18, 20 and 22°C ; in all three cases the degree of fogging is very slight indeed and cannot be recorded as a measure of the density; in order to get an estimate we counted the number of grains developed per unit of surface, this measurement being proportional to the photographic density. The value: number of grains/surface of $2,900 \text{ micron}^2$ represents the average of 20 counts.

Table 3. Influence of the developing-bath temperature on autoradiographs of an Sr^{90} standard source taken on Microtex film

| Developing-bath temperature | Photographic density | Fogging: No. of grains/ $2,900 \mu^2$ |
|-------------------------------|----------------------|--|
| $18 \pm 0.02^{\circ}\text{C}$ | 0.205 | 18 |
| $20 \pm 0.02^{\circ}\text{C}$ | 0.27 | 25 |
| $22 \pm 0.1^{\circ}\text{C}$ | 0.325 | 26.8 |

Method of preparing thin-section samples

In order to obtain quantitative data from the autoradiograph it is usually necessary for the sample to be in the form of a thin section. It must in fact be possible to assume that the tracer is evenly distributed throughout the thickness of the section, and it is necessary to calculate all the corrections for self-absorption of the radiation. Moreover, in the interests of good autoradiographic resolution it is essential that the labelled sample should have a thin section.

For the preparation of a thin-section sample the following points must be taken into account:

- it must be possible to predetermine and measure the thickness of the sample;
- the thickness must be uniform over a relatively large area (of the order of 1 cm² in many investigations);
- the section must have good planarity in order that it may be correctly exposed;
- the material of which the sample is composed must not be impaired.

The methods of preparing thin sections vary according to the material to be studied. For the applications described below, a method suitable for metallurgical and mineralogical specimens was developed. Other techniques are described in the references quoted (8, 9).

The following description relates to the method tried; this was applied to copper samples, but it is also suitable for other metallic substances provided that they are neither too hard nor too soft.

By means of a rotary saw a section 0.5 - 1 cm² in area and 0.2 - 0.4 mm thick is cut from the metal sample. Thinner sections than this are difficult to cut. Two steel cylinders 20 mm high and 30 mm in diameter are prepared and their ends ground flat and parallel. One of these cylinders is provided at one end with a seat 0.4 mm deep and 20 mm in diameter.

The section taken from the sample is soldered into the seat of the cylinder.

The specimen section thus mounted on the cylinder is surface-finished with a lapping machine equipped with a rotating plate to which can be fixed paper discs of varying abrasiveness; a micrometer comparator is used to check that the surface is perfectly flat and parallel with the other end of the cylinder.

The face which carries the specimen section is then cemented to one end of the second cylinder with epoxy resin (Araldit I), care being

taken to ensure that the resin forms a thin, even layer with no voids. The coplanarity of the whole assembly is checked with the aid of the comparator and the thickness of resin added is thus determined.

When the resin has polymerized, the first cylinder is detached by mechanical means, after which the rotating-disc lapping machine is used to reduce the thickness of the section which is still attached to the second cylinder.

By using the comparator to check the thicknesses removed and the planarity, it is possible to obtain sections of 1 cm² the thickness of which varies by less than 2 microns over the entire surface. For sections of copper specimens the minimum thickness obtained without undue difficulty was 10 microns. For the most part we used sections with a thickness of between 15 and 25 microns. The sections can be used for exposure as such mounted on the cylinder, or can be detached from the supporting cylinder by heating.

In order to measure the thickness of the section at predetermined points during the thickening operation, apart from measuring with the comparator the total thickness made up of cylinder plus resin plus sample, an ingenious trick was resorted to: this consisted in making Vickers hardness indentations of adequate depth in the first lapped face of the sample, measuring them before mounting the sample on the second cylinder and then checking on the second face first the appearance of the indentations and then their size, until this no longer corresponded to the desired thickness.

This method can always be employed for mineral samples by cutting a section approximately 1 mm thick with a diamond tool and using the epoxy resin to stick the section both to the first and to the second cylinder. Thickness reduction is effected with abrasive powder of varying granulometry, on flat glass and with the addition of water.

The method is not suitable for very soft metals such as lead; in this case thickness reduction by means of abrasive paper impairs the structure of the metal. Since among the examples of applications described elsewhere in this report an examination of a lead sample was also envisaged, a technique was devised for the preparation of thin sections from that metal.

A cylinder of the lead in question is lapped by removing various thicknesses with a microtome; the lapped face is then glued with epoxy resin to a steel cube the faces of which have been ground flat and parallel. Next, the sample is replaced on the microtome and successive layers are removed until only a thin section of the desired thickness remains attached to the sample-holder.

Correlation of radioactive exposure and photographic effect for the various radioisotopes

Radioactive exposure versus photographic density curves were plotted for the autoradiographs obtained with the various radioisotopes under study, using Kodak V 1056 and Microtex emulsions; the photographic efficiencies of the radiations emitted by the various isotopes were compared so as to establish comparative values between the various standards.

All the comparisons were between autoradiographs of standards and specimens exposed and developed simultaneously, since despite all precautions taken to ensure reproducibility of the photographic process the blackening results obtained in a number of successive exposures are not always the same, even when the exposure conditions are identical; this is due to variables which cannot be eliminated, e.g. films taken from different batches, renewal of the developing baths, non-controllable variations in the ambient conditions.

By carrying out the comparisons between autoradiographs made simultaneously it was possible to eliminate most of the variations that occur between autoradiographs taken at different times.

The comparison of the photographic effects of two different radioisotopes was done by averaging the results of numerous autoradiographic exposures so as to reduce any casual errors due to a single erroneous result.

In this way we made about 70 series of exposures, thus obtaining a like number of autoradiograph groups with which to carry out comparisons between two or more sets of standards.

Photographic density measurements

For measuring the photographic density use was made of a Zeiss microdensitometer on which a few modifications had been carried out in order to adapt it to the specific requirements of the investigation. In particular, fixed-aperture windows were constructed which corresponded to the following surfaces of analysis on the autoradiograph:

| | |
|----------|-------------|
| Window 1 | 100 μ^2 |
| Window 2 | 400 " |
| Window 3 | 900 " |

The type of window was chosen from case to case and in accordance with the characteristics of the autoradiographs. In fact autoradiographs taken with relatively coarse-grained emulsions need windows of larger aperture; moreover, the standards which show uniform blackening over a length of 5 mm can be examined with windows of relatively large aperture.

For autoradiographs of samples which, in addition to a fine-grained image, require a high resolution owing to sharp variation of the tracer concentration in the sample - and hence also of the blackening - type 1 windows were used.

The instrument measures the intensity of the light transmitted through the autoradiograph and from this it derives the photographic density in accordance with the formula

$$D = \log \frac{I_0}{I}$$

It is obvious that in the case of high photographic densities corresponding to a low value of I (i.e. the light intensity transmitted), the lack of precision in the determination of I gives rise to a large measuring error. It can be assumed that the standard error is 4% on measurements of $D = 1$ and 10% on measurements of $D = 2$. It will consequently be advantageous to perform the microdensitometry determinations in conditions such that the blackening of the autoradiographs is about $D = 0.5$.

Fig. 9 shows an autoradiograph of a standard source and the corresponding density record.

Density curves

By way of example Figs. 10a and 10b show the characteristic curves for three radioisotopes, as obtained with Microtex and V 1056 emulsion respectively.

It can be seen that:

- (a) the relation between optical density and incident radiation is a linear one, at least for density values not exceeding 1 in the case of Microtex and 0.5 in that of V 1056, i.e. within the range of normal operations;
- (b) the V 1056 emulsion (also the corresponding AR 10) is more sensitive to radiations of lower energy, as is confirmed by the literature;

- (c) the Microtex emulsion is more sensitive when the maximum radiation energy increases from 0.15 to 0.25 to 0.7 MeV, and thus from C¹⁴ to Ca⁴⁵ to Cl³⁶. An attempt will be made to explain this apparently anomalous behaviour.
- (d) The Microtex emulsion is approximately ten times as sensitive as the V 1056.

In view of the linearity of the characteristic curves in the sector of practical interest, it was deemed expedient not only to consider the curves themselves but also to regard as representative, in the case of a given radioisotope and a given photographic emulsion, that radiation exposure value required in order to obtain a constant photographic density value in the autoradiograph.

Table 4 gives the radiation exposure values, as derived from the density curves, which are necessary in order to get a density of 0.5 on autoradiographs made with Microtex emulsion. Table 5 gives the corresponding values for a density of 0.25 and V 1056 emulsion.

Table 4 - Summary of the various exposures made with Microtex emulsion and standard sources of the various radioisotopes

| Exposure No. | Standard sources | | | | | | | |
|--|---------------------|------------------|-------------------|---------------------|---------------------|---------------------|---------------------|-------------------|
| | C ¹⁴ | Ca ⁴⁵ | Pm ¹⁴⁷ | Sr ⁹⁰ | Cl ³⁶ | Y ⁹¹ | P ³² | Sb ¹²⁴ |
| Radiation exposure (in R/mm ² x 10 ³) necessary to give D = 0.5 | | | | | | | | |
| 2 | 770 [±] 20 | | | | 665 | | | |
| 4 | 740 [±] 10 | 700 | | | 665 [±] 30 | | | |
| 5 | 730 [±] 20 | | | 640 | 650 [±] 20 | | | |
| 9 | 760 | 705 | | | 670 | | | |
| 12 | 735 [±] 15 | 700 | | | 630 [±] 20 | | | |
| 14 | 780 | 740 | | 650 | 700 [±] 20 | | 650 | |
| 15 | | | | | | | 685 | |
| 24 | 900 | | | 740 [±] 20 | 710 | | 810 | |
| 26 | 850 | | | | 740 [±] 10 | 720 [±] 20 | 770 [±] 10 | |
| 29 | 895 | | | 780 [±] 15 | | 770 | 740 | |
| 31 | 750 [±] 50 | | | | 680 | | | |
| 32 | 880 [±] 20 | | | 760 [±] 30 | 785 | 730 [±] 20 | 800 | |
| 33 | 860 [±] 10 | | | 750 | 760 [±] 30 | 750 | | |
| 35 | 860 [±] 10 | | | 750 | 745 | | | |
| 37 | 840 [±] 40 | | | 750 | 760 | | | |
| 38 | 860 [±] 30 | | | 750 | 760 | | | |
| 38 | 870 [±] 20 | | | 740 | 750 [±] 5 | | | |
| 42 | 830 [±] 10 | | | 700 | 730 [±] 20 | | | |
| 55 | 880 [±] 10 | | | 760 [±] 5 | 780 [±] 20 | | | |
| 61 | 850 | | 830 | 750 [±] 70 | 780 [±] 20 | | | |
| 78 | 890 [±] 10 | | | | | | | 730 |

Table 5 - Summary of the various exposures made with V 1056 emulsion and standard sources of the various radioisotopes

| Exposure No. | Standard sources | | | | | | | | | |
|--------------|---|-----------------|-----------------|------------------|-------------------|--------------------|------------------|-----------------|-----------------|-------------------|
| | H ³ | C ¹⁴ | S ³⁵ | Ca ⁴⁵ | Pm ¹⁴⁷ | Sr-Y ⁹⁰ | Cl ³⁶ | Y ⁹¹ | P ³² | Sb ¹²⁴ |
| | Radiation exposure (in $\beta/\text{mm}^2 \times 10^4$) necessary to give D = 0.25 | | | | | | | | | |
| 1 | | 310 | | 335 | | | 495 | | | |
| 3 | | 325 | | 395 | | | 520 | | | |
| 16 | | 245 | | | | 375 | 390 | | 430 | |
| 21 | | 250 | | | | 380 | 390 | | 360 | |
| 22 | | 245 | | | | 425 | 350 | | | |
| 40 | | 713 | | | | | | 1280 | | |
| 43 | | 495 | | | | | 720 | | | |
| 51 | | 410 | | | | 625 | 680 \pm 10 | | | |
| 52 | | 340 \pm 10 | | | | 515 \pm 10 | 550 | | | |
| 53 | | 290 | | | | 480 | | 510 \pm 5 | | |
| 54 | | 300 | | | | 410 \pm 5 | 410 \pm 10 | 440 | | |
| 59 | | 335 \pm 10 | | | 300 | 460 | 440 | 510 \pm 10 | | |
| 62 | 575 | 265 | | | | | | | | |
| 65 | 545 | 260 | | | 305 | | 410 \pm 10 | | | |
| 66 | 650 \pm 25 | 345 | | | 345 | | 520 | | | |
| 68 | | 285 | 280 | | | | | | | |
| 74 | | 295 | 295 | | | | | | | |
| 76 | | 240 | 295 | | | | | | | 355 |

As far as the Microtex emulsion is concerned it was found that its radiation-sensitivity at first increases with the increase in the maximum radiation energy β and then drops again.

To explain this behaviour, which was noticed in all the exposures made, it must be assumed that this type of emulsion is not sensitive to radiations of very low energy - possibly on account of the absorption due to a protective layer of inert gelatine placed on top of the sensitive gelatine - and that it acts as an absorber of these radiations. It was found in fact that tritium standards fail to produce any image at all on this type of emulsion.

From the radiatopm exposure values needed to give photographic densities of 0.5 and 0.25 respectively on Microtex and V 1056 emulsion with the various radioisotopes, can be derived the photographic efficiencies of these radioisotopes by taking one of them as a basis for comparison. Table 6 lists the relative photographic efficiency with respect to C^{14} of each radioisotope studied and for the two emulsions tested. The error affecting these values is estimated at about 10%.

Thus a convenient standard source such as the C^{14} source can be used, with sufficient accuracy, as a reference for comparing the quantitative autoradiographs of specimens labelled with one of the other radioisotopes.

It is, of course, possible to take as the reference source one of the other long-lived radioisotopes employed (Cl^{36} , $Sr-Y^{90}$).

These relative efficiency values were used in the five experimental studies described below as examples of applications.

Table 6: Relative photographic efficiencies of the various radioisotope sources with respect to the C¹⁴ standard source

| Emulsion | Ratio $\frac{B/mm^2 \text{ of radioisotope } Z}{B/mm^2 \text{ of } C^{14}}$ which gives a constant photographic density | | | | | | | | |
|----------|---|-----------------|------------------|-------------------|--------------------|------------------|-----------------|-----------------|-------------------|
| | R a d i o i s o t o p e s | | | | | | | | |
| | H ³ | S ³⁵ | Ca ⁴⁵ | Pm ¹⁴⁷ | Sr-Y ⁹⁰ | Cl ³⁶ | Y ⁹¹ | P ³² | Sb ¹²⁴ |
| V 1056 | 2.05 | 1.05 | 1.15 | 1.10 | 1.5 | 1.55 | 1.65 | 1.40 | 1.50 |
| Microtex | | | 0.95 | 0.95 | 0.85 | 0.90 | 0.85 | 0.90 | 0.80 |

Examples of the application of quantitative autoradiography

Distribution of the phosphorus in a copper/1% phosphorus alloy

Phosphorus in the form of copper phosphide is commonly used in the deoxidation of molten copper whenever it is necessary to eliminate the brittleness caused by cuprous oxide and when no importance attaches to the reduction in the conductivity of the copper which results from this treatment. The phosphorus content in the copper is usually kept within 0.05%.

In some practical cases it has been found that the ingots made from copper which has been deoxidized with phosphorus contain macro- and micro-segregations which can cause the phosphorus content to vary considerably from one zone of the ingot to another.

It was therefore considered worthwhile to determine the various phosphorus distributions that occur under different conditions of solidification, annealing and plastic working of the copper.

A copper/14% radioactive phosphorus mother alloy was prepared by reacting a mixture of electrolytic copper filings and red phosphorus at 400°C under vacuum in a quartz phial.

The phosphorus was activated by in-pile irradiation and had an activity of 2 millicuries/gram at the time the experiment began.

A melt was then made of about 4 kg of electrolytic copper; the molten copper was deoxidized by adding 28.5 g of the mother alloy so that the resulting phosphorus content was approximately 0.1%.

When mixing was complete, one half of the copper was poured into a cast-iron mould; cooling was rapid and solidification took place in 15-20 seconds. The remainder of the melt was allowed to solidify slowly in the furnace. A thermal analysis showed that solidification had taken 20 minutes.

From a first series of autoradiographs of sections of two ingots it emerged that in the case of the rapidly solidified melt a fairly uniform phosphorus distribution is obtained, whereas the slowly solidified melt showed appreciable interdendritic segregation of the phosphorus.

From the slowly solidified ingot were taken three samples which were treated as follows:

- No. 1 sample was subjected to four cold-working treatments, each of which gave a 30% reduction in cross-section, with intermediate annealing at 750°C for 1 hour;

- No. 2 sample was annealed at 800°C for 24 hours;
- No. 3 sample was annealed at 950°C for 8 hours.

From each copper sample, both in the as-melted and in the annealed or cold-worked state, a section was taken which was reduced to the required thinness by the method described on Page 17.

The five thin sections prepared were as follows:

1. a sample taken from the rapidly solidified ingot (15 sec);
2. a sample taken from the slowly solidified ingot (20 min);
3. as sample 2, subjected to 4 successive cold-working operations with intermediate annealing at 750°C for 1 hour;
4. as sample 2, annealed at 800°C for 24 hours;
5. as sample 2, annealed at 950°C for 8 hours.

The thin sections from the 5 copper samples were mounted on plastic holders by means of the technique employed for the mounting of the standards, and were exposed together with sets of standards, using Microtex emulsions. The simultaneously exposed standards were those of C¹⁴ and P³². Microdensitometry determinations were then carried out on the autoradiographs obtained, and in the case of the autoradiograph of the samples the maximum blackening variation curves were traced.

Examination of the autoradiograph of the P³² standard gave the density curve shown in Fig. 11. From this is derived a value of 760×10^3 B/mm² for D = 0.5, which value will serve as a basis for the determination of the phosphorus concentration in the samples.

If, owing to its short half-life, the phosphorus standard had not been available, the simultaneously exposed carbon-14 standard would have enabled the above-mentioned value to be calculated. In fact, the simultaneously exposed autoradiograph of the C¹⁴ standard indicates that for this radioisotope and this exposure there must be 870 B/mm² in order to obtain a photographic density of 0.5; table 6 shows that the photographic efficiency ratio for P³² and C¹⁴ is 0.9; consequently, it can be calculated that for the phosphorus a radiation exposure of 780×10^3 B/mm² is necessary in order to give D = 0.5.

This is in good agreement with the actual value determined experimentally by densitometry of the phosphorus standard.

The above-quoted values must then be corrected for the self-absorption and self-scattering factor of the P³² radiations in the thin section, this factor being derived from the formula

$$f_s = \frac{\mu t}{1 - e^{-\mu t}}$$

in which

μ = absorption coefficient of copper for the P^{32} radiations, expressed in cm^2/mg ,

t = thickness of the sample, expressed in mg/cm^2 .

The value of μ , as found by experimental measurements, is $0.009 cm^2/mg$. Fig. 12 shows the relationship f_s /thickness of the thin section.

A correction factor $f_b = 0.92$ was then introduced for the back-scattering of the β -radiations by the plastic source-holder, as well as a factor $\times 2$ to take account of the 2π geometry used for the exposure, and the number of β per mg of sample was calculated on the basis of the thickness of each sample and the densitometry data corrected as indicated above.

An absolute count carried out on the radioactive phosphorus used in the experiment showed, when extrapolated to the moment the experiment started, that the activity was 21.25×10^5 dpm/mg. For the 975 minutes' duration of the exposures the emission is thus found to be 20.39×10^8 B/mg.

Table 7 gives an example of the values obtained for samples with a thickness of $20 \mu a$.

Table 7

| Autoradiograph | | Sample | | |
|----------------------|---|---|------------------------------|-------|
| Photographic density | Incident β/mm^2 ($\times 10^3$) | β/mm^2 corrected for f_s , f_b and geometry ($\times 10^3$) | β/mg ($\times 10^3$) | P (%) |
| 0.05 | 76 | 152 | 854 | 0.042 |
| 0.1 | 152 | 305 | 1714 | 0.084 |
| 0.2 | 304 | 610 | 3427 | 0.168 |
| 0.3 | 456 | 914 | 5135 | 0.252 |

From the densitometry data were calculated the concentrations and the concentration variations for the five samples studied.

Figs. 13-17 show, for the five samples in question, the micrographs, the corresponding thick- and thin-section autoradiographs and the respective microdensities expressed in % phosphorus concentration.

Sample 1 is found to be the most homogeneous; its average phosphorus concentration is calculated at 0.085%. Chemical analysis of a sample taken from the same ingot shows an average phosphorus content of 0.076%; there is thus a difference of about 10% between the values as determined by autoradiography and by chemical analysis. In view of the number of variables involved in the autoradiographic determination (the thickness of the thin sections, for example, can vary by 10%), it is considered that the results are in satisfactory agreement.

If we examine the differences between the maximum and minimum phosphorus concentrations in the various samples, we find them to be as follows:

| Sample No. | Difference between maximum and minimum concentration, expressed in % P |
|------------|--|
| 1 | 0.025 |
| 2 | 0.20 |
| 3 | 0.10 |
| 4 | 0.07 |
| 5 | 0.04 |

It is obvious from the above figures that slow solidification (Sample 2) produces an unevenness of phosphorus distribution which is greater by one order of magnitude than that resulting from rapid solidification. The segregation of the phosphorus present in Sample 2 is only partly attenuated by plastic working (Sample 3), and even with prolonged annealing at high temperature (Samples 4 and 5) it is not possible to achieve complete homogenization of the phosphorus in the sample.

The obvious inference from the foregoing is that for practical purposes rapid solidification of the ingot is more effective in producing a uniform phosphorus distribution than are the treatments to which the ingot may be subjected after slow solidification.

Distribution of Ca^{45} on the surface of quartz granules labelled with that tracer

In connection with a research project on foundry sands and their contribution to exogenous inclusions in steel, the said project forming part of an investigation to be carried out with the aid of radioactive tracers, it was desirable to look into the possibility of labelling sand in a stable manner with Ca^{45} .

The method tried consisted in wetting the sand with a soluble salt of the tracer ($\text{Ca}(\text{NO}_3)_2$), drying it and then heating it to a high temperature (1000°C) so that the nitrate decomposes and the CaO formed is stably fixed on the silica.

A quantitative autoradiographic examination was performed in order to determine the concentration and distribution of the tracer in the individual grains of the siliceous sand.

Quartz granules which had been labelled in the above manner were reduced to thin sections by the technique described on page 18. The thickness of the section obtained was 20 ± 1 microns. The next step was to expose, together with C^{14} standards, the preparation containing the thin sections of the granules. The emulsion used was Kodak V 1056, since the highest possible resolution was required in this case. For the exposure, the grains of sand were left mounted on the steel cylinder that had served for the preparation of their thin section.

Microdensitometry of the C^{14} standard source gave $410 \pm 5 \times 10^4$ B/mm^2 for $D = 0.25$.

In order to perform the quantitative calculations from the microdensitometry determinations on the autoradiograph, it is necessary to ascertain the correction factors to be introduced, that is to say the difference in photographic efficiency between the standard source employed and the radioisotope in question, the corrections for the exposure geometry, for back-scattering and for self-absorption.

Given 1.15 as the ratio between the relative photographic efficiencies of C^{14} and Ca^{45} with V 1056 emulsions, it follows that for the Ca^{45} $470 \pm 5 \times 10^4$ B/mm^2 are necessary in order to get a photographic density D of 0.25. The geometric correction factor is 2, since the exposure is done with 2π geometry.

The back-scattering factor, as measured in an internal-source counter with a Ca^{45} source on an iron carrier, was found to be $fb = 0.87$.

The self-absorption can be calculated from the formula

$$dm = \frac{m_0}{t} e^{-ux} dx$$

in which

m and m_0 are the counts with and without absorption,

t is the thickness of the section in mg/cm^2 ,

μ is the absorption coefficient in cm^2/mg .

The coefficient μ can be calculated with the aid of absorption measurements in an internal-source counter, using the formula

$$\mu = \frac{1}{x} \lg \frac{m_0}{m}$$

in which x is the absorber thickness in mg/cm^2 .

The absorber element used for the determination of μ was aluminium.

From the measurements performed it emerged that μ varies with the thickness of the absorber. Fig. 18 shows the graph for absorption coefficient versus absorber thickness. From this graph is derived the relation

$$\mu = 0.427 x^{-0.408}$$

By substituting this value of μ in the self-absorption formula and integrating graphically, we obtain the correction factor for self-absorption $f_s = \frac{m_0}{m}$. In Fig. 19 this factor is plotted against the thickness of the m source.

The absolute radioactivity measurement of the $\text{Ca}(\text{NO}_3)_2$ used for labelling the sand gives an activity, referred to the moment the exposure started, of 5.86×10^5 dpm/mg Ca.

To a photographic density of 0.25 on the autoradiograph there corresponds a value of 470×10^4 incident β/mm^2 ; taking account of the previously calculated factors in respect of geometry, back-scattering and self-absorption, and taking the duration of the exposure as 10,320 minutes, we obtain a value of $1560 \beta/\text{mm}^2/\text{min}$ emitted into 4π by the tracer contained in the entire thickness of the thin section in order to give a photographic density $D = 0.25$.

Having regard to the 20μ thickness of the section of the quartz granules, their density and the absolute activity of the Ca^{45} employed, we find that the Ca concentration corresponding to a density of $D = 0.25$ is 4.8%.

Fig. 20 shows the autoradiograph of two quartz granules, enlarged 30 times, and also the microdensitometry scaled in photographic density and in calcium concentration.

Penetration of the tracer into the granules is confined to a very thin layer, owing to the relatively short time in which the tracer reacts with and diffuses in the matrix; as regards the analysis of the concentration curves obtained from autoradiographs in the case of sharp variations in the concentration of a tracer in a sample, the reader is referred to a previous study carried out at this Laboratory (10).

Investigations on corrosion inhibitors: distribution of S³⁵-labelled thiourea on the surface of steel samples

Within the framework of the investigations on the corrosion inhibitors studied by the Electrochemistry and Corrosion Laboratory of the Breda Research Institute, a preliminary experiment was conducted with the object of determining the absorption mechanism, on the surface of a steel sample, of thiourea used as corrosion inhibitor, in a solution acidified with H₂SO₄.

The conditions of attack by the active solution were as follows : solution containing 5% of H₂SO₄; thiourea inhibitor at 0.1% concentration; temperature 65°C; steel specimens which had been surface-roughened with an abrasive of grain-size 600.

A determination of the corrosion-inhibiting capacity of thiourea under these conditions gave the following values (obtained from an average of six samples) for specimens of mild steel exposed for two hours to attack by inhibitor-containing active solutions :

| | Loss of weight in mg/mm ² |
|---|--------------------------------------|
| - samples in a solution with inhibitor | 0.54 |
| - samples in a solution without inhibitor | 2.4 |

As a specific case of the application of radioactive tracers, a study was made of thiourea absorption and distribution on the surface of an AISI 347 steel sample, over a section which included a weld.

in which

m and m_0 are the counts with and without absorption,

t is the thickness of the section in mg/cm^2 ,

μ is the absorption coefficient in cm^2/mg .

The coefficient μ can be calculated with the aid of absorption measurements in an internal-source counter, using the formula

$$\mu = \frac{1}{x} \lg \frac{m_0}{m}$$

in which x is the absorber thickness in mg/cm^2 .

The absorber element used for the determination of μ was aluminium.

From the measurements performed it emerged that μ varies with the thickness of the absorber. Fig. 18 shows the graph for absorption coefficient versus absorber thickness. From this graph is derived the relation

$$\mu = 0.427 x^{-0.408}$$

By substituting this value of μ in the self-absorption formula and integrating graphically, we obtain the correction factor for self-absorption $f_s = \frac{m_0}{m}$. In Fig. 19 this factor is plotted against the thickness of the m source.

The absolute radioactivity measurement of the $\text{Ca}(\text{NO}_3)_2$ used for labelling the sand gives an activity, referred to the moment the exposure started, of 5.86×10^5 dpm/mg Ca.

To a photographic density of 0.25 on the autoradiograph there corresponds a value of 470×10^4 incident β/mm^2 ; taking account of the previously calculated factors in respect of geometry, back-scattering and self-absorption, and taking the duration of the exposure as 10,320 minutes, we obtain a value of $1560 \beta/\text{mm}^2/\text{min}$ emitted into 4π by the tracer contained in the entire thickness of the thin section in order to give a photographic density $D = 0.25$.

Having regard to the 20μ thickness of the section of the quartz granules, their density and the absolute activity of the Ca^{45} employed, we find that the Ca concentration corresponding to a density of $D = 0.25$ is 4.8%.

Fig. 20 shows the autoradiograph of two quartz granules, enlarged 30 times, and also the microdensitometry scaled in photographic density and in calcium concentration.

Penetration of the tracer into the granules is confined to a very thin layer, owing to the relatively short time in which the tracer reacts with and diffuses in the matrix; as regards the analysis of the concentration curves obtained from autoradiographs in the case of sharp variations in the concentration of a tracer in a sample, the reader is referred to a previous study carried out at this Laboratory (10).

Investigations on corrosion inhibitors: distribution of S³⁵-labelled thiourea on the surface of steel samples

Within the framework of the investigations on the corrosion inhibitors studied by the Electrochemistry and Corrosion Laboratory of the Breda Research Institute, a preliminary experiment was conducted with the object of determining the absorption mechanism, on the surface of a steel sample, of thiourea used as corrosion inhibitor, in a solution acidified with H₂SO₄.

The conditions of attack by the active solution were as follows : solution containing 5% of H₂SO₄; thiourea inhibitor at 0.1% concentration; temperature 65°C; steel specimens which had been surface-roughened with an abrasive of grain-size 600.

A determination of the corrosion-inhibiting capacity of thiourea under these conditions gave the following values (obtained from an average of six samples) for specimens of mild steel exposed for two hours to attack by inhibitor-containing active solutions :

| | Loss of weight in mg/mm ² |
|---|--------------------------------------|
| - samples in a solution with inhibitor | 0.54 |
| - samples in a solution without inhibitor | 2.4 |

As a specific case of the application of radioactive tracers, a study was made of thiourea absorption and distribution on the surface of an AISI 347 steel sample, over a section which included a weld.

The experiments involving a radioactive tracer were carried out with the aid of thiourea- S^{35} the activity of which, as determined by measurement in a 4π counter with internal source, was found to be 0.196 mc/mg on the date of exposure of the autoradiograph.

From the various experiments performed it emerges that the thiourea is rapidly absorbed on the surface of the sample from the very first moment of its immersion in the solution containing the inhibitor. By increasing the residence time in the active solution it is possible to detect a penetration in depth by the tracer, especially in the weld zone, over a depth of about ten microns.

In an initial experiment the steel sample was immersed in the active solution for about 2 minutes.

Absorption of the tracer is thus confined entirely to the surface. The sample is then carefully washed in water and alcohol and given a protective coating of Saran, which is prepared by the method described on page 11. The next step is to make the exposure on Kodak V 1056 emulsion, together with C^{14} standards.

Fig. 21a shows the autoradiograph of the sample and Figs. 21 b and 21 c the microdensitometry records along the lines A-A and B-B indicated in Fig. 21a.

In order to calibrate the microdensitometry records in S^{35} concentrations, and hence in thiourea concentrations, consideration must be given to the correction factors to be applied.

If we assume that the distribution of the labelled inhibitor is confined entirely to the surface, there is no need to take account of any self-absorption factor.

On the other hand there is an appreciable radiation back-scattering effect which is due to the sample itself.

Measurements of the back-scattering due to S^{35} radiations, performed by means of an internal-source counter, showed that the radiation emitted from the surface outwards has increased by 22-22.5% as a result of back-scattering. Consequently, it is necessary to introduce a correction factor $f_b = 0.82$ for the calculation of the radiation effectively emitted into the external half-space.

The ratio between the photographic efficiency of the C^{14} used as reference standard and that of the S^{35} is practically equal to 1; in fact the various values obtained in the comparative exposures of the standards gave results which do not deviate from 1 for a value exceeding the estimated error. Hence the radiation necessary to produce a photographic density of 0.25 can be taken as being 285×10^4 B/mm², the value obtained for the C^{14} standard. Since the samples were exposed for a shorter time than was the standard, there is a correction factor of $ff = 0.96$ for fading.

Given this value, the previously mentioned correction factors, an exposure time of 360 minutes and the specific activity of the thiourea used, it follows that the surface concentration of S^{35} -labelled thiourea which corresponds to a photographic density of 0.25 is 3.8×10^{-7} mM per mm² of sample surface. On the basis of this value the thiourea surface concentration value shown in Figs. 21b and 21c was introduced.

As stated above, longer exposure of the sample to attack by the solution results in diffusion of the tracer inside the sample.

The same sample as in the preceding autoradiograph was reground and again placed in contact with the active solution in the presence of the labelled inhibitor, this time for 60 minutes.

Fig. 22a shows the autoradiograph of this sample and Fig. 22b the microdensitometry record, likewise scaled in thiourea surface concentrations.

In this case, in order to ascertain the correction factor it is necessary to introduce not only the back-scattering factor but also the factor for self-absorption due to the S^{35} . The tracer concentration is not constant throughout the sample, but decreases with increasing depth. The self-absorption coefficient was calculated on the assumption that the tracer distribution decreases towards the interior in accordance with an exponential law.

This distribution was calculated from the densitometry records of the autoradiographs of the sample as such and after removal of a surface layer 5 microns thick; if we now take into account a coefficient of 0.195 mg/cm^2 for the absorption of the S^{35} radiations in iron - this value having been determined experimentally for an absorber thickness of 4.5 mg/cm^2 - we can approximately calculate a self-absorption factor $f_s = 1.54$.

By introducing this value, the value of the radiation exposure necessary to give a density of 0.25 (equal to $260 \times 10^4 \text{ B/mm}^2$), the above-mentioned back-scattering factor, the exposure time of 300 minutes and the tracer activity of 0.3 millicuries per mg of thiourea at the moment of exposure, we calculate that in order to have $D = 0.25$ the thiourea surface concentration must be $4.3 \times 10^{-7} \text{ mM per mm}^2$ of surface. On the basis of this value the surface concentration of inhibitor can be plotted on the ordinates of the densitometry diagrams in Figs. 22b, c, d.

The results of this preliminary investigation show that the method can be applied to the study of the surface and in-depth absorption kinetics of thiourea in steel samples; in particular, it would appear of interest to ascertain whether it is a case of physical absorption of thiourea by the surface, or whether it is not rather a reaction accompanied by the formation of a different product; the literature shows that it is worth while to pursue this investigation further by comparing the results obtained with the use of thiourea labelled with S^{35} and with C^{14} (11,12).

Antimony distribution in a copper/antimony alloy

In the three applicational examples reported above a study was made of the distribution of an element or a substance labelled with a purely beta-emitting radioisotope which emits a single beta radiation.

In the two examples that follow we shall study the distribution of an element, namely antimony, whose radioisotope Sb^{124} emits beta- and gamma-radiations of different energies. In view of the low photographic efficiency of gamma- as compared with beta radiations, there are no particular difficulties as regards either the resolution of the autoradiographs or the determination of the photographic efficiency of the antimony sources which will be considered as beta-emitters for the purpose of producing autoradiographic images.

The first of these experiments related to the determination of the antimony distribution in a copper/0.5% antimony alloy in the as-cast state.

The radioactive antimony was prepared from a hydrochloric solution of SbCl_3 from which metallic antimony was precipitated by reduction with tin. The concentration and the quantity of the solution were so calculated as to give 0.75g of antimony; 0.40 millicurie of Sb^{124} was added to the solution.

The metallic antimony in powder form was collected and melted under vacuum in a sealed phial; the pellet of antimony thus obtained was added to a melt of approximately 150 g of copper. When homogenization was complete the copper ingot was poured and cooled.

A test-piece taken from the centre of the ingot was reduced to a thin section by the customary method described on page 17; the thickness of the section obtained was $20 \pm 1 \mu$.

The autoradiographic exposures of the thin section were made with Microtex emulsion, together with C^{14} standards. Fig. 23 a shows the autoradiograph.

Qualitative evaluation reveals that there is considerable segregation of the antimony, which accumulates predominantly in the primary interdendritic zones.

Fig. 23 b shows the densitometry of the autoradiograph along the line indicated in Fig. 23 a. On the ordinate is plotted the antimony concentration as calculated according to a procedure similar to that used in the preceding examples. The following paragraphs describe in detail the operations to be carried out for the quantitative calculation of the antimony concentration from the autoradiograph.

The simultaneously exposed C^{14} standard required an exposure of 760×10^3 B/mm² in order to get a density of 0.5; the relative photographic efficiency factor between Sb^{124} and C^{14} is 0.8. The correction factor for back-scattering, as determined experimentally under the exposure conditions with the source mounted on a steel cylinder, was found to be $f_b = 0.86$.

The self-absorption factor, calculated on the basis of a number of measurements performed on thin sections of the Cu/Sb¹²⁴ alloy of varying thickness, was $f_s = 1.2$.

The exposure time of the thin-section sample was 1450 minutes. On the basis of the data and correction factors stated above we find that in order to obtain a photographic density of 0.5 it is necessary for the tracer present in the thin-section sample to emit 4850 B/min/mg of alloy.

This value is derived from:

$$- \text{activity of tracer in the sample which gives } D = 0.5 = \frac{\text{Exp}C^{14} \cdot \text{Eff } Sb^{124}/C^{14} \cdot f_b \cdot f_s \cdot 2}{T \cdot d \cdot x}$$

$\text{Exp}C^{14}$ = exposure of the C^{14} standard which gives $D = 0.5$;

$\text{Eff } Sb^{124}/C^{14}$ = relative photographic efficiency factor between the radioisotope contained in the sample and that of the standard;

f_b = correction factor for back-scattering by the carrier;

f_s = correction factor for self-absorption;

T = exposure time;

d = density of sample;

x = thickness of sample

Since the antimony used in making the alloy had an activity of 1.17×10^6 dpm/mg, it follows that a photographic density of 0.5 on the autoradiograph in Fig. 23 a represents an antimony concentration of 0.41% in the alloy. As the concentration of labelled antimony in the alloy, the resulting activity and the degree of blackening produced on the autoradiograph are directly proportional, it is possible to calculate the density corresponding to each antimony concentration and to plot it on the ordinate in the microdensity diagram.

Antimony distribution in a lead/antimony alloy

An experiment was devised for determining the antimony distribution in a lead/2% antimony alloy. This experiment was conducted in a similar manner to that described in the preceding case.

4g of metallic antimony labelled with 0.2 millicurie of Sb^{124} was prepared and added to a 200 g melt of lead; when mixing was complete, the melt was slowly cooled to solidification.

A thin section of the lead sample was prepared by removing with a microtome successive layers from the sample mounted on a steel support, in accordance with the technique described on page 18. The resulting section had a thickness of $22 \pm 2 \mu$.

The autoradiograph of this section is shown in Fig. 24a and the microdensitometry in Fig. 24 b.

The antimony concentrations in the alloy are plotted on the ordinate of the densitometry diagram. The calculations for deriving the concentration of labelled element from the photographic density are performed in the same way as in the preceding case, by introducing for the various factors the appropriate values obtained for this application.

The relevant data were as follows:

The C^{14} standard exposed simultaneously with the sample under study required 1050×10^3 B/mm² to give a density of 0.5 on Microtex emulsion.

The photographic efficiency ratio between Sb^{124} and C^{14} is 0.8.

The back-scattering factor as ascertained by experimental determinations is $f_b = 0.86$.

The self-absorption factor in the thin section for the Sb^{124} radiations was calculated, on the basis of measurements carried out on thin sections of varying thickness, at $f_s = 1.4$.

The exposure time of the autoradiograph in Fig. 25 was 2465 minutes.

On the basis of the above data it is found that in order to get a photographic density of 0.5 on the autoradiograph, the tracer present in the sample must have an activity of 3360 B/min/mg of alloy.

Since the specific activity of the labelled antimony used for the melt was 1.1×10^5 dpm/mg, it follows from a comparison with the above value that a photographic density of 0.5 represents a 3.5% concentration of labelled antimony in the alloy. The density values corresponding to the various antimony concentrations were calculated and plotted on the microdensitometry diagram in Fig. 24 b.

CONCLUSIONS

A quantitative autoradiography technique was developed which, though adapted in particular for metallurgical applications, can also be used without difficulty in other scientific and industrial research fields for investigations involving autoradiographic detection of radioactive tracers. The method provides for the use of standard radioactivity sources which are exposed simultaneously with thin sections of the sample containing the tracer to be examined, followed by microdensitometry determinations of the autoradiographs obtained. By taking account of the following factors: exposure geometry, self-absorption and back-scattering of the radiations from the sample, fading, photographic efficiency of the radioisotope employed as tracer as compared with that of the standard source, it is possible to derive the tracer concentration in the sample from the photographic density of the autoradiograph. The relative photographic efficiency factors of the following radioisotopes were calculated: H^3 , C^{14} , S^{35} , Pm^{147} , Cl^{36} , Y^{91} , P^{32} , $Sr-Y^{90}$, Sb^{124} , using Kodak V 1056 and Microtex emulsions; the values for other radioisotopes can subsequently be calculated in accordance with future research programmes.

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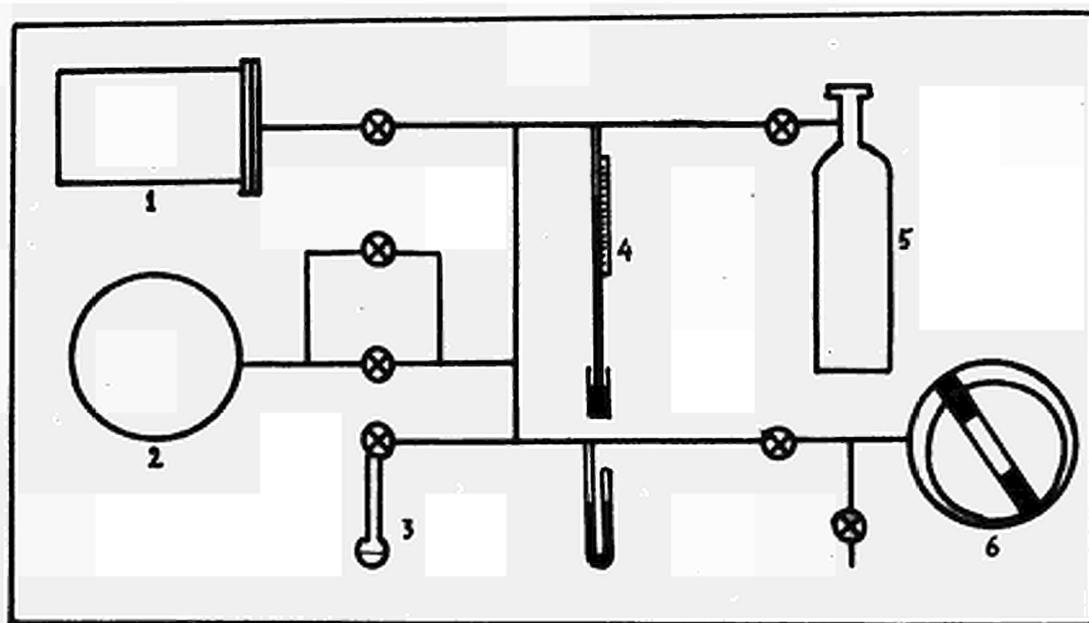


Fig. 1 - 4π Geiger counter with internal source

- 1 - Counter
- 2 - Reservoir containing a mixture of argon and 10% ethyl alcohol for filling the counter.
- 3 - Ethyl alcohol.
- 4 - Manometer.
- 5 - Argon
- 6 - Pump

Method: A mixture of argon and 10% ethyl alcohol at 300 mm Hg is prepared in the reservoir. The radioactivity source to be measured is placed in the counter. The counter is evacuated and the gaseous mixture is admitted at a pressure of 100 mmHg.

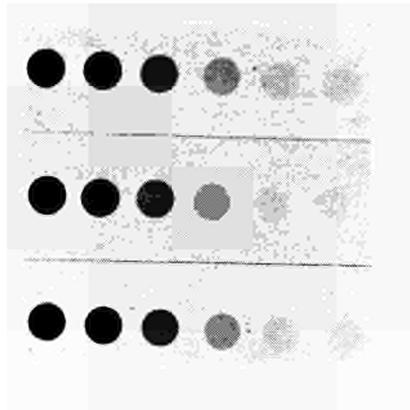


Fig. 2 — Comparison of Kodak X-ray emulsions :
Exposure with Ca²⁵ standard

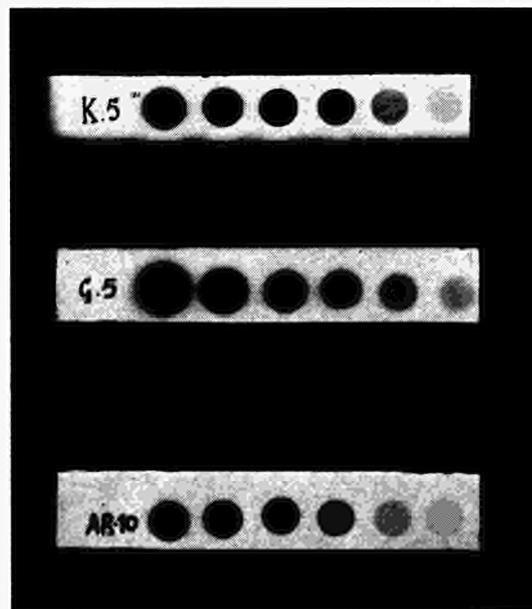
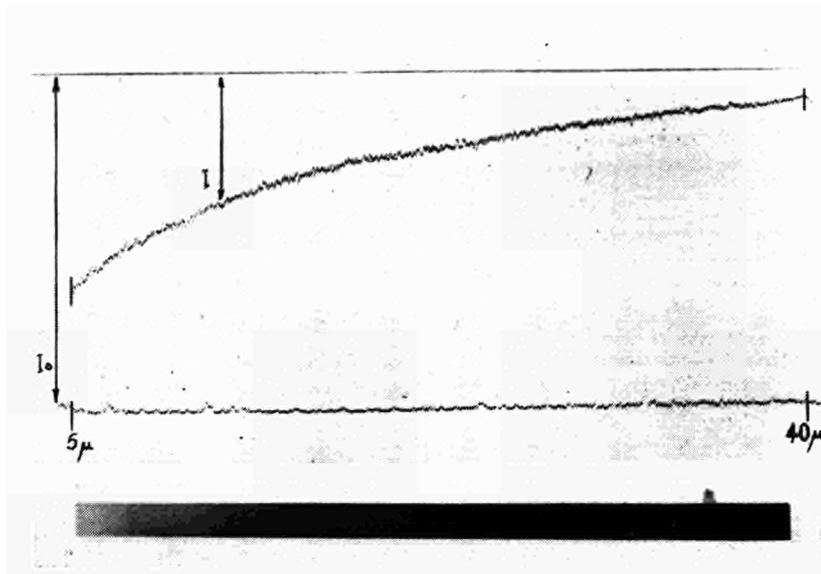
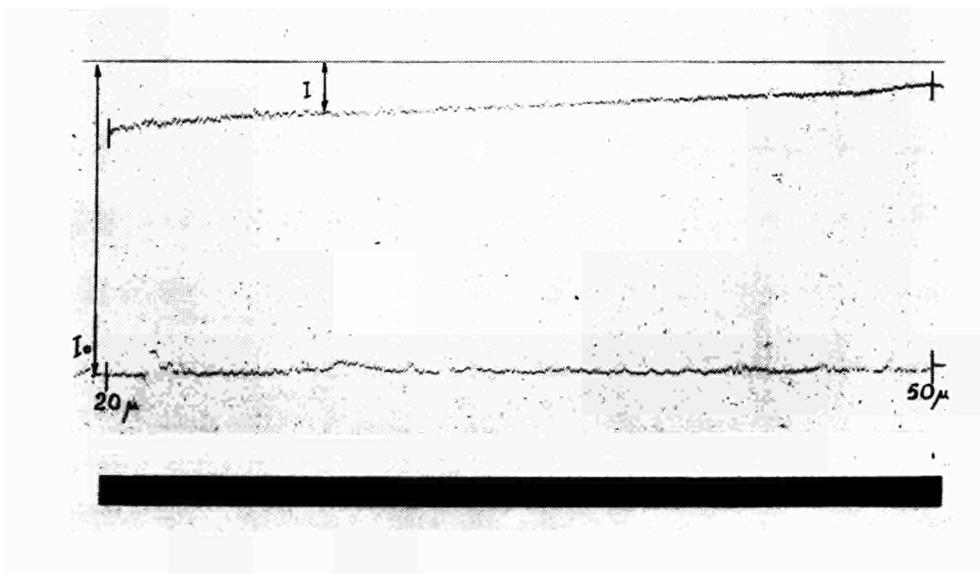


Fig. 3 — Comparison of Ilford Nuclear Research
Plates K5 and G5 and Kodak Fine Grain
Autoradiographic Plate AR10 -
Exposure with Ca²⁵ standard



a) Wedge No. 1



b) Wedge No. 2

Fig. 4 — Autoradiographs of two Ca^{45} sources of increasing thickness and the corresponding densitometry curves : a) thickness increasing from 5 to 40μ ; b) from 20 to 50μ

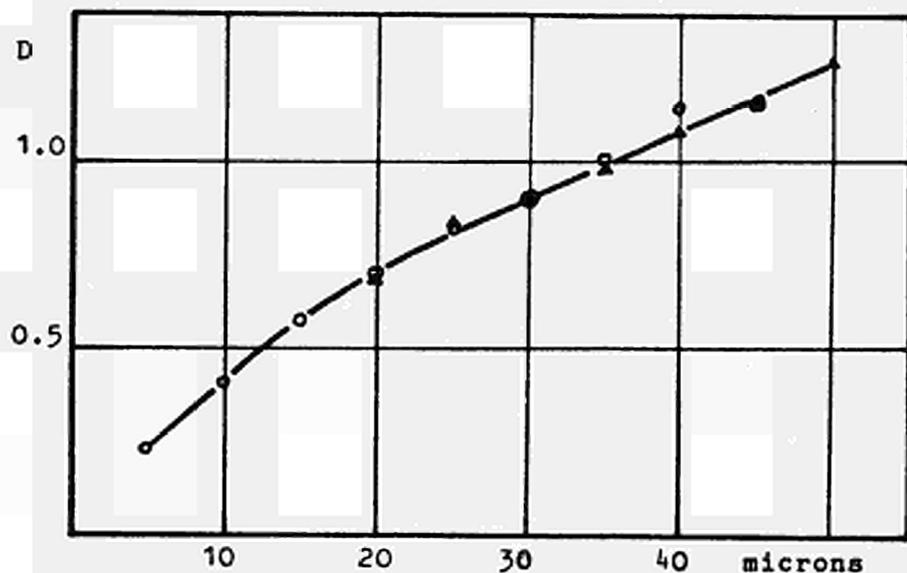


Fig. 5 - Diagram showing photographic density versus thickness of Ca^{45} standard source, derived from the densitometry curves in Fig. 4.

- wedge No. 1
- △ wedge No. 2

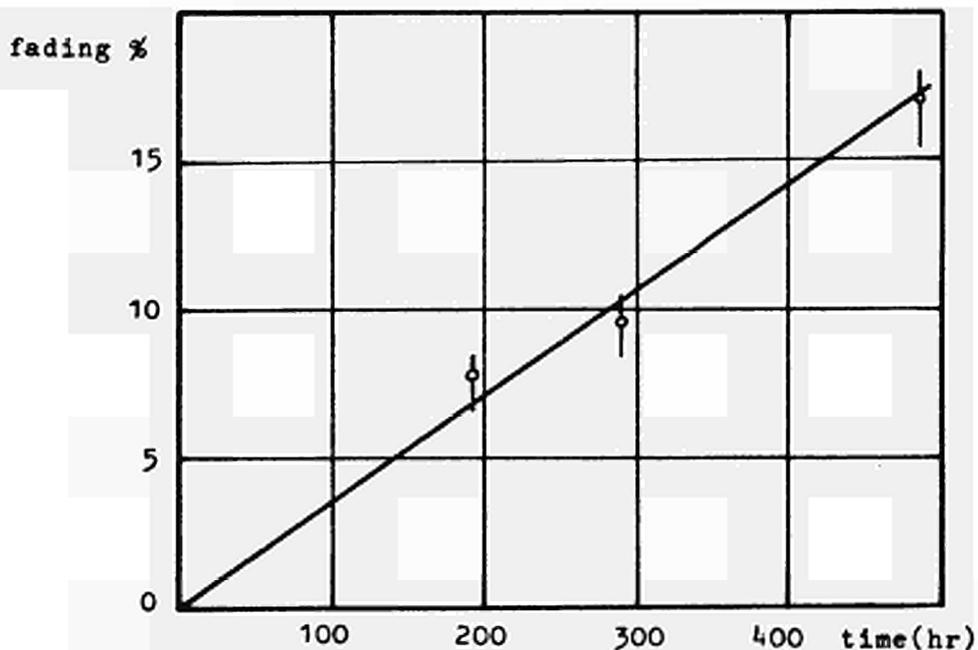


Fig. 6 - Percentage variation in the fading as a function of the exposure time

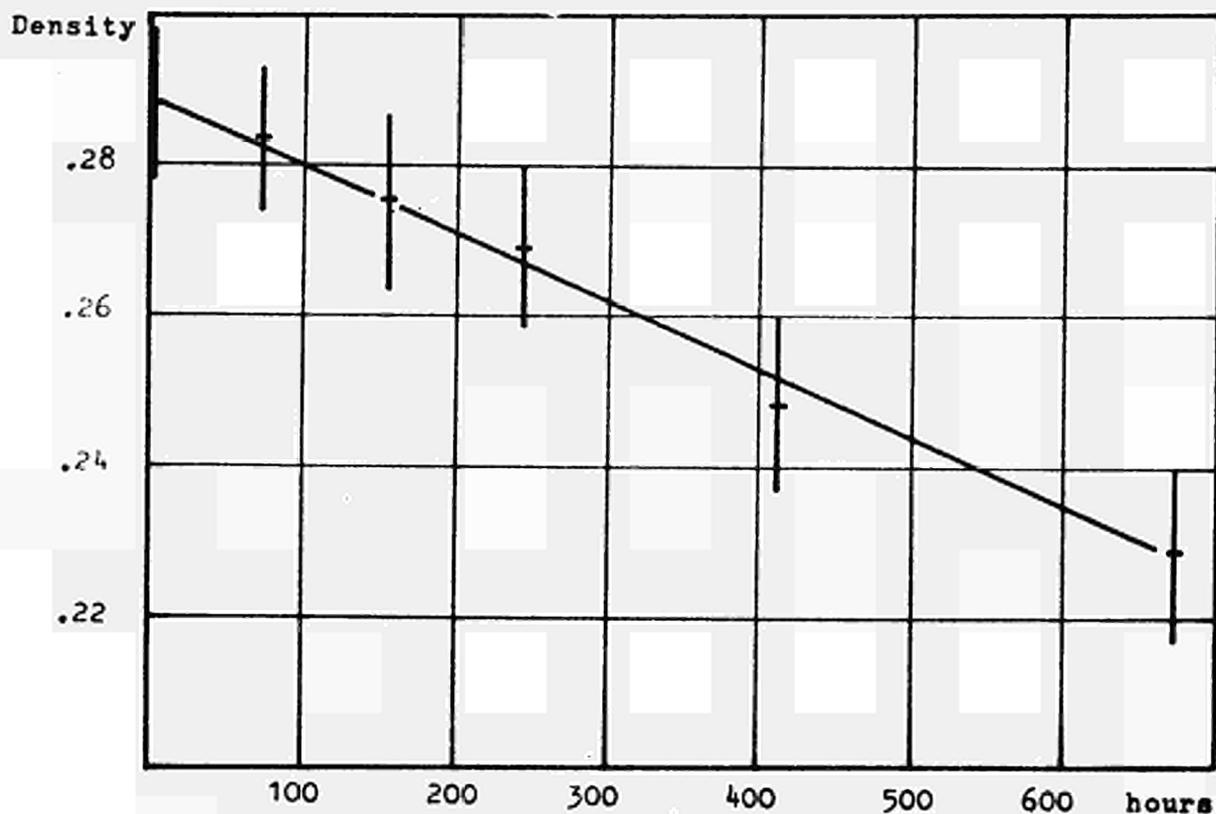


Fig. 7 - Variation of the blackening density as a function of the time elapsed between exposure and developing

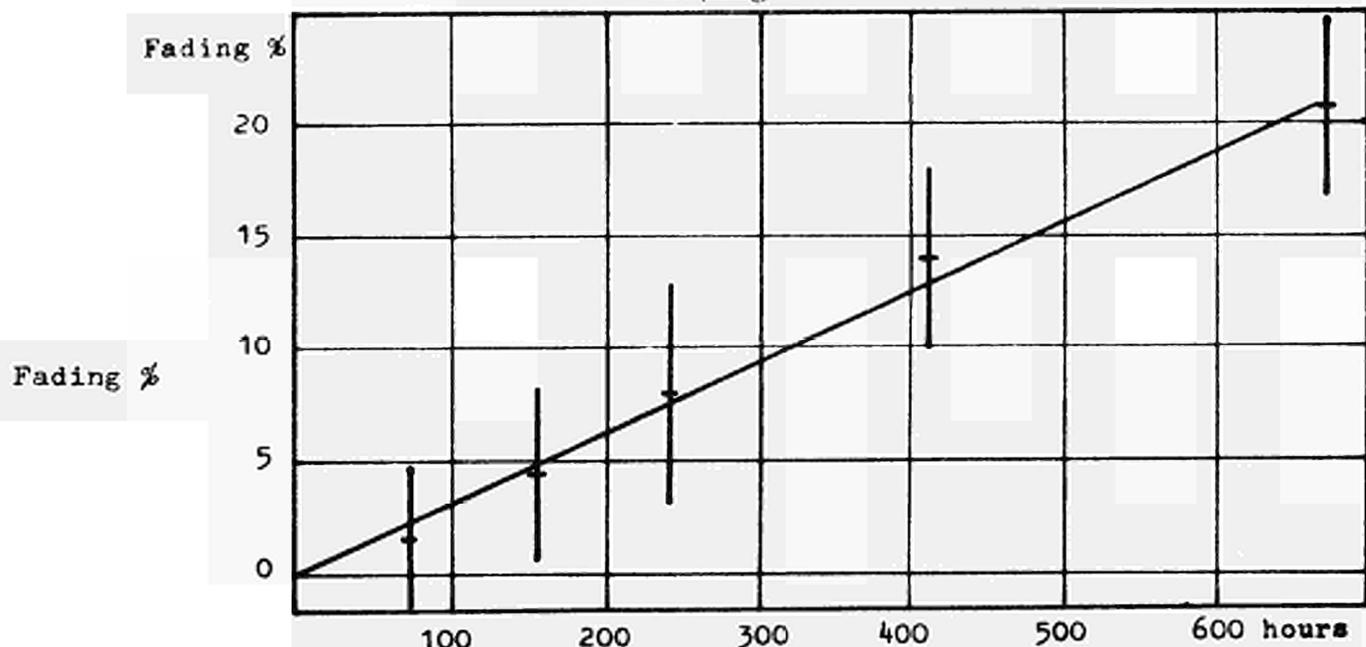


Fig. 8 - Percentage variation of fading as a function of the time elapsed between exposure and developing

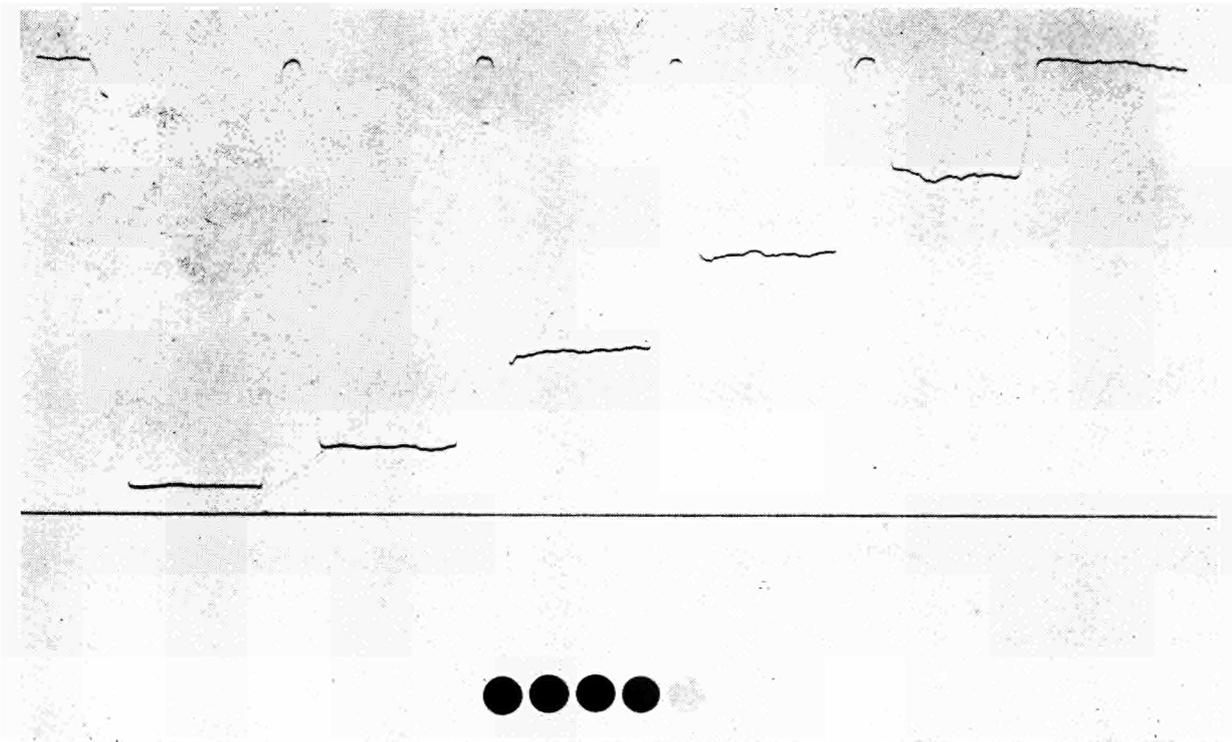


Fig. 9 — Autoradiograph of a Ca^{45} standard source, scaled in 5 gradations, on Kodak AR 50 emulsion exposed for 50 hours, and densitometry curve of the source itself. The transmitted intensities are shown on the ordinate.

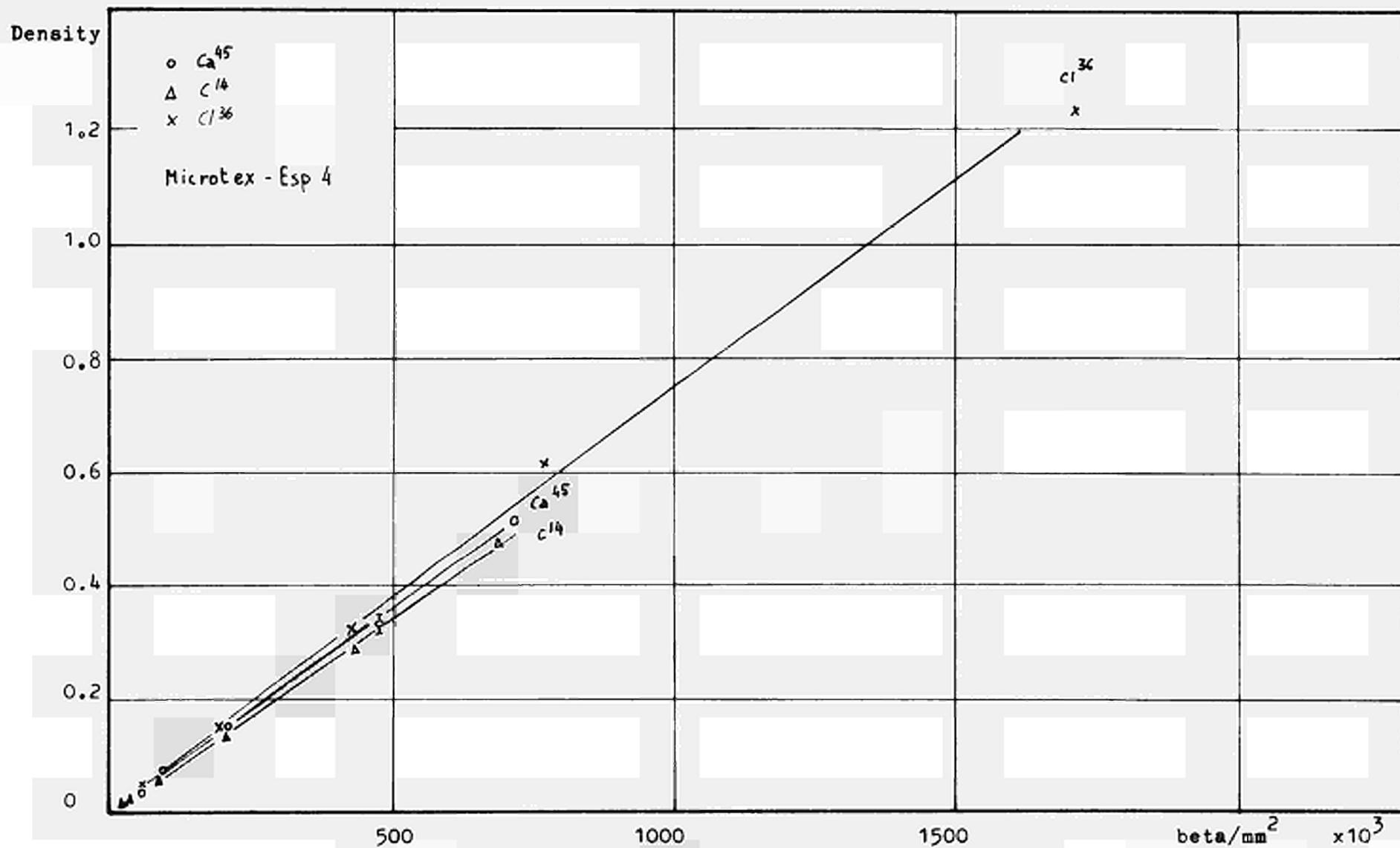


Fig. 10a — Photographic density versus specific surface activity curves obtained with sets of C^{14} , Ca^{45} and Cl^{36} standards, using single emulsion Microtex film ; exposure No. 4

Emulsion V 1056

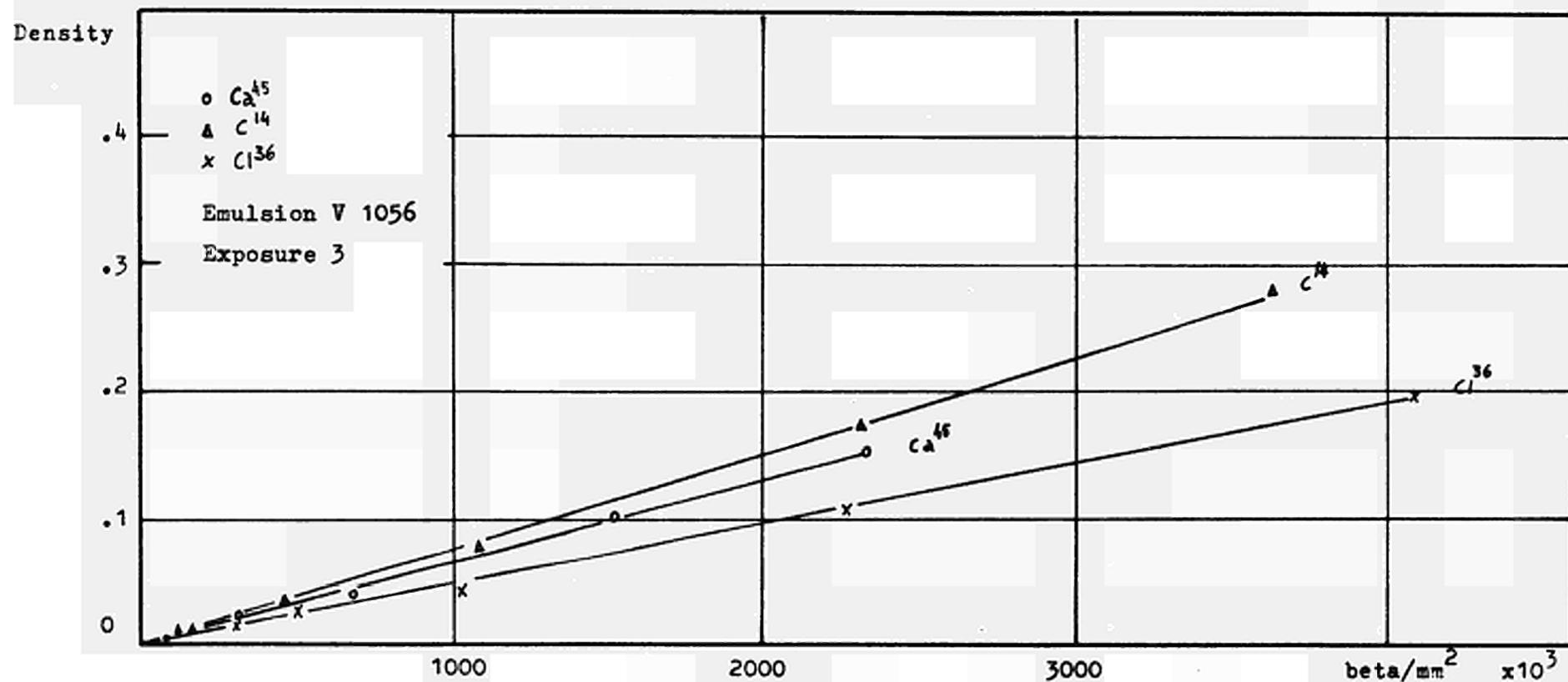


Fig. 10b - Photographic density versus specific surface activity curves obtained with sets of C^{14} , Ca^{45} and Cl^{36} standards, using V 1056 emulsion; exposure No. 3

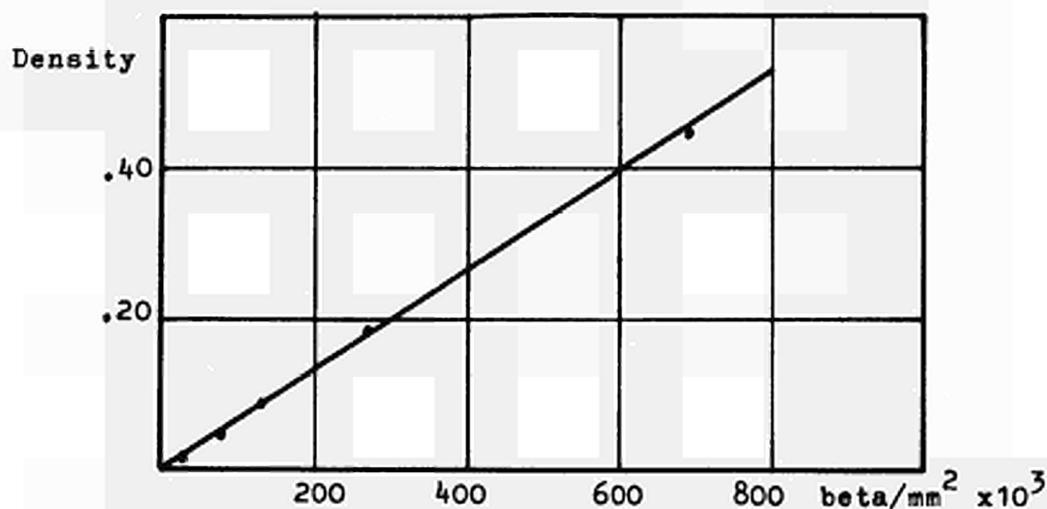


Fig. 11 - Graph showing photographic density versus number of β -particles/mm² for a set of P^{32} standards exposed simultaneously with the thin sections.

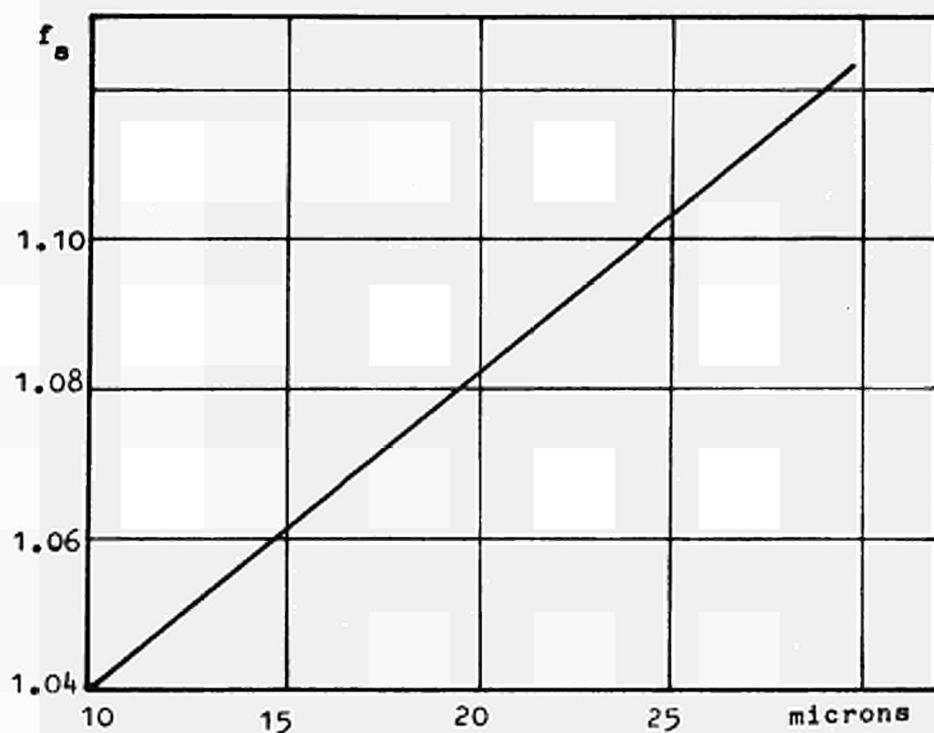
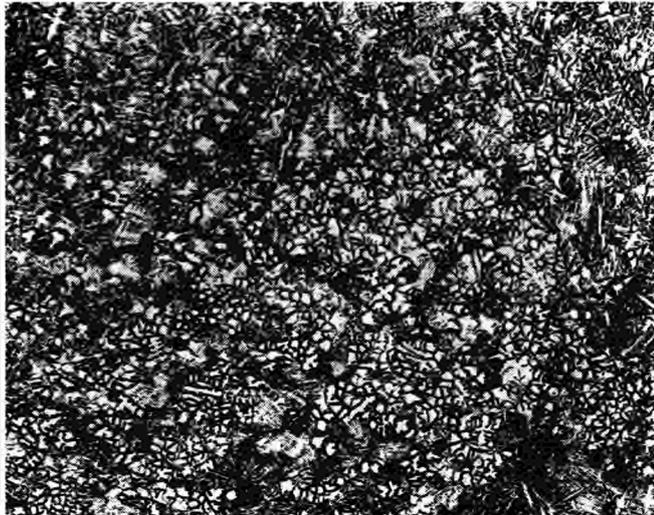


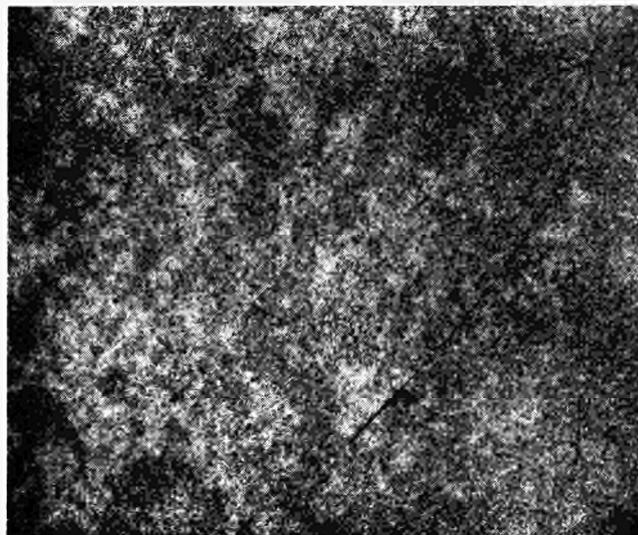
Fig. 12 - Graph showing the self-absorption factor (f_s) versus the thickness of the section for P^{32} radiations in copper



(809)

x 10

Fig. 13a — Micrograph of sample No. 1



(816)

x 10

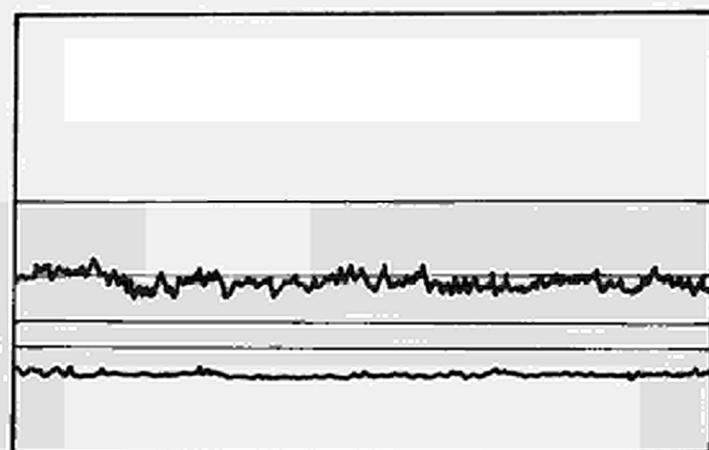
Fig. 13b — Autoradiograph of sample No. 1
(same position as Fig. 13a)

Phosphorus %

Density

0.20
0.10
0.05
0.025
0

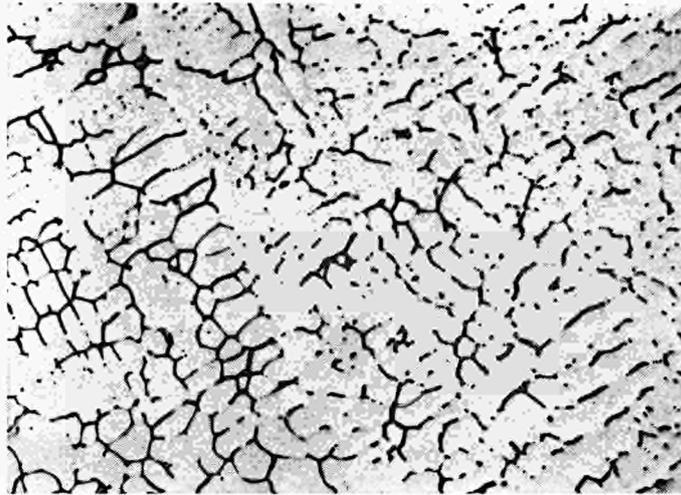
0.3
0.2
0.1



Distance x20

Fig. 13c — Densitometry of the autoradiograph of a thin section taken from sample No. 1

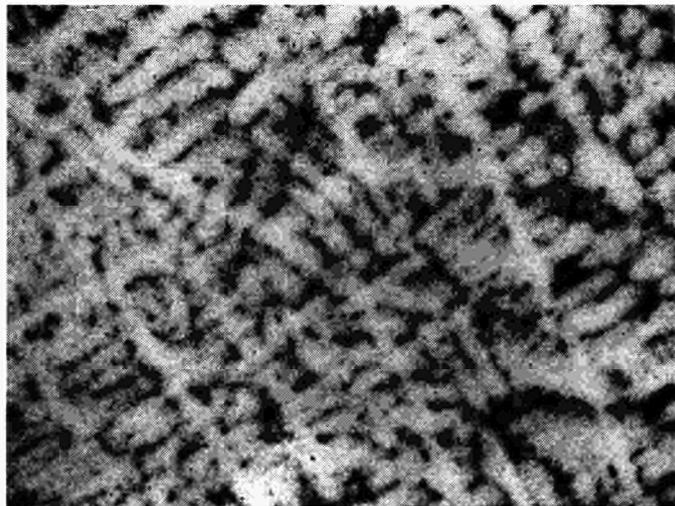
Note : The autoradiograph (not reproduced here) shows a uniform grey field.



(806)

x 10

Fig. 14a — Micrograph of sample No. 2



(814)

x 10

Fig. 14b — Autoradiograph of sample No. 2
(same position as Fig. 14a)

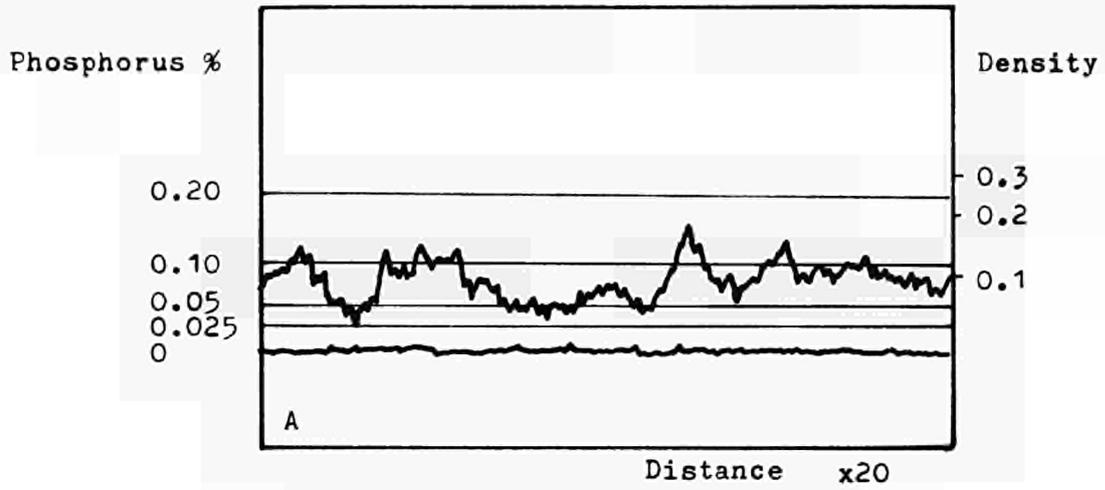


Fig. 14c — Densitometry of the autoradiograph shown in Fig. 14e, along the line A-A

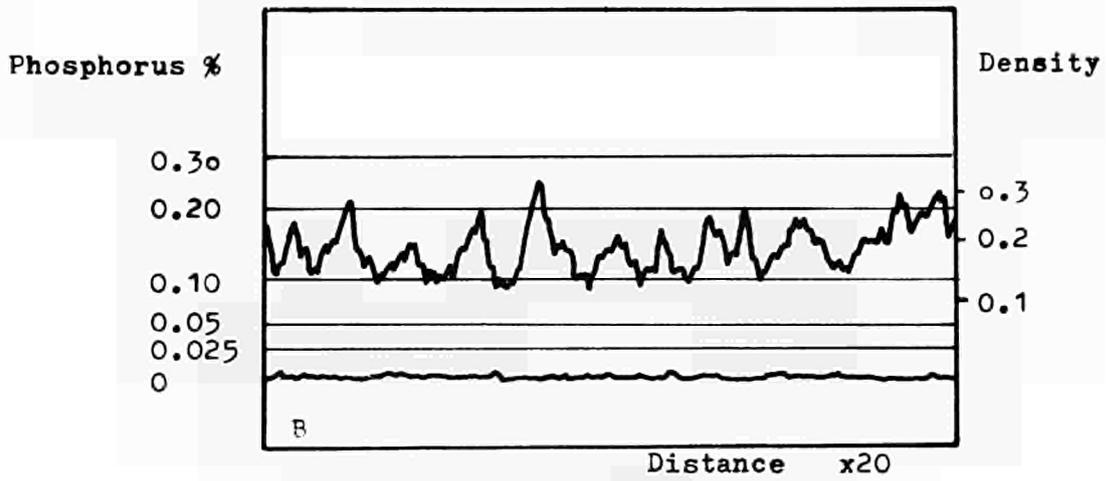


Fig. 14d — Densitometry of the autoradiograph shown in Fig. 14e, along the line B-B

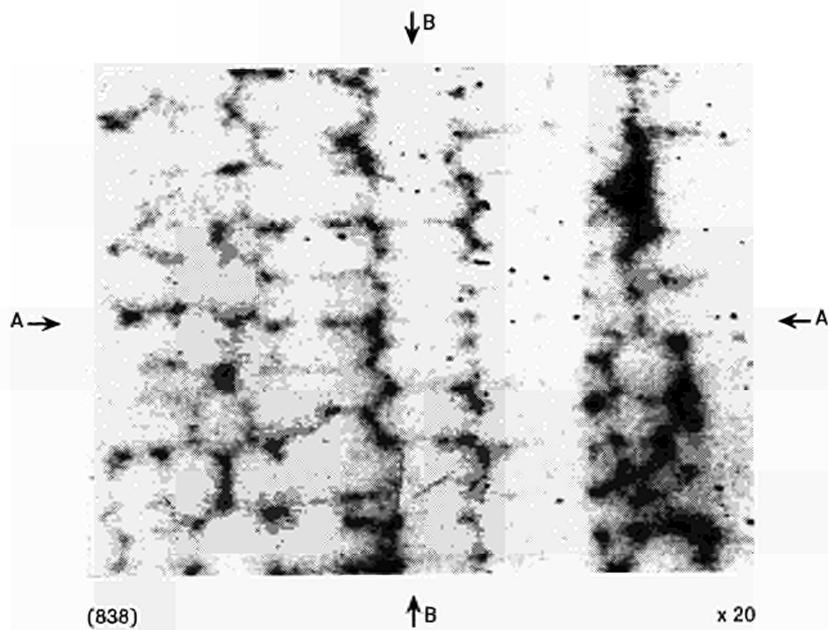
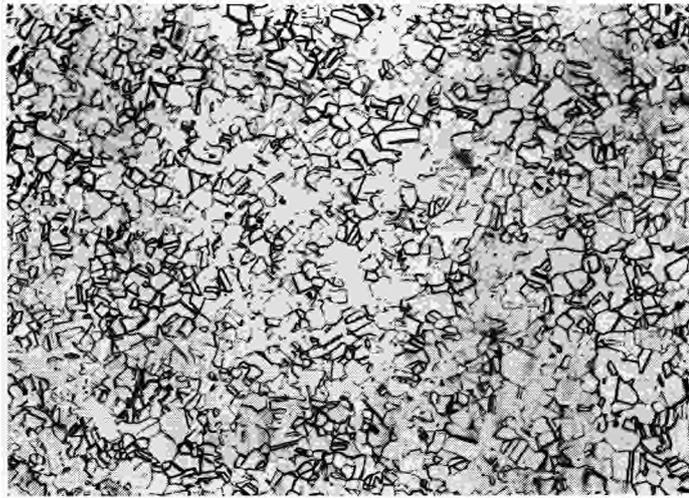


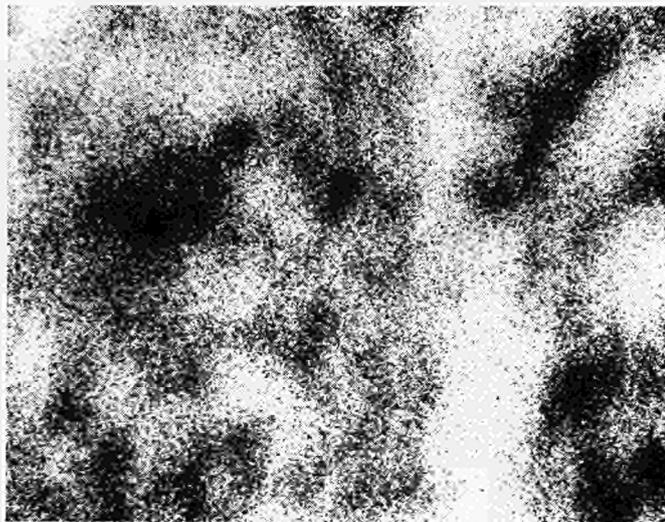
Fig. 14e — Autoradiograph of a thin section (22μ) taken from sample No. 2



(804)

x 30

Fig. 15a — Micrograph of sample No. 3



(805)

x 30

Fig. 15b — Autoradiograph of sample No. 3
(same position as Fig. 15a)

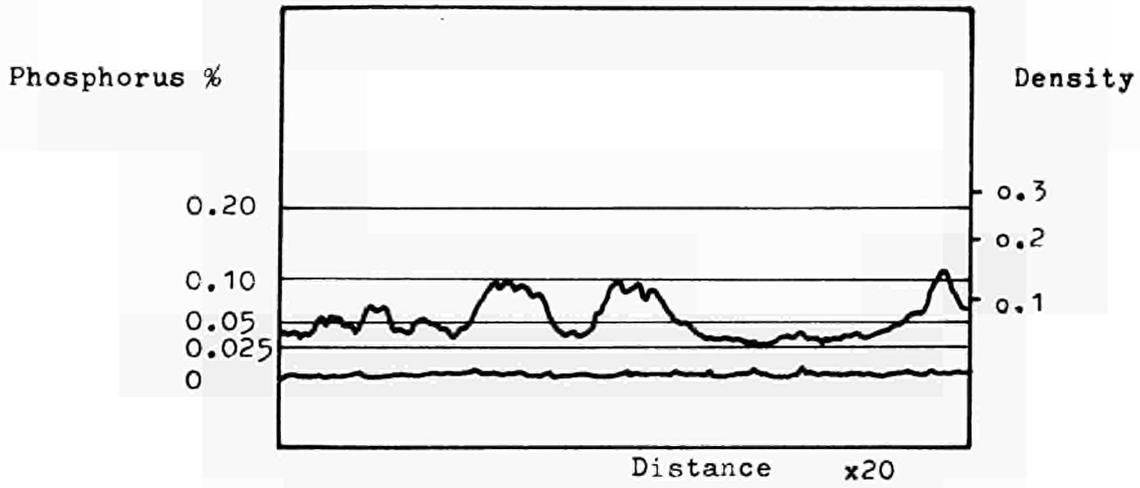
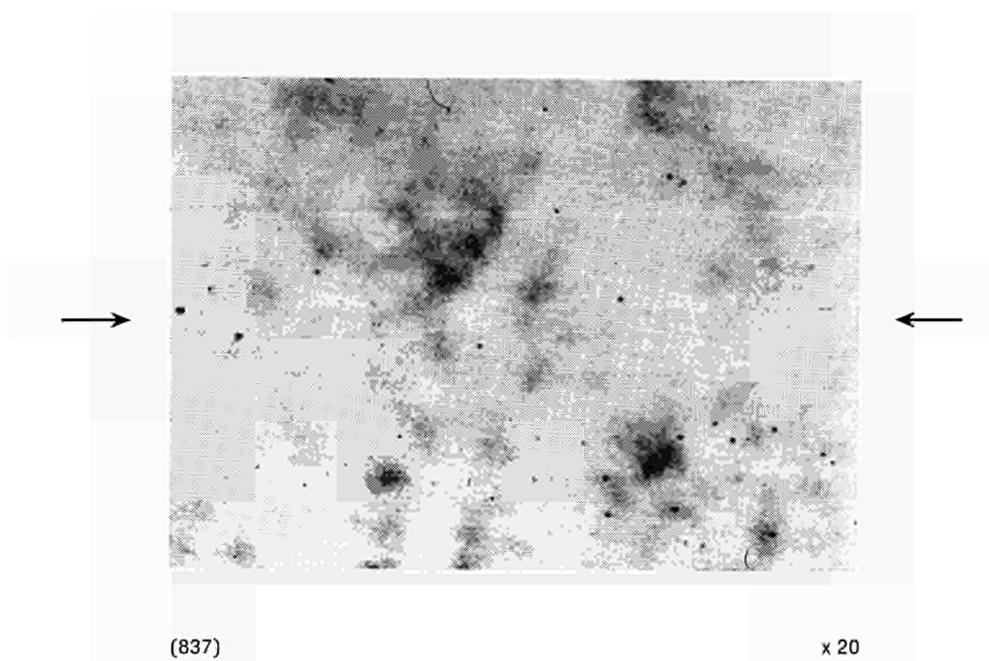
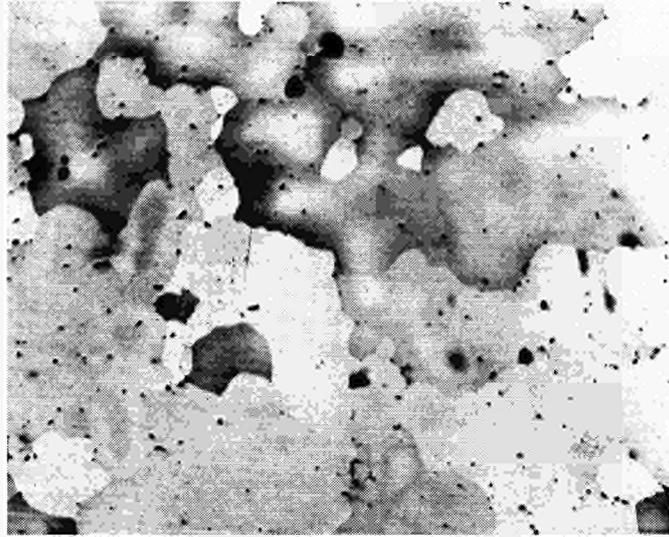


Fig. 15c — Densitometry of the autoradiograph shown in Fig. 15d



(837) x 20

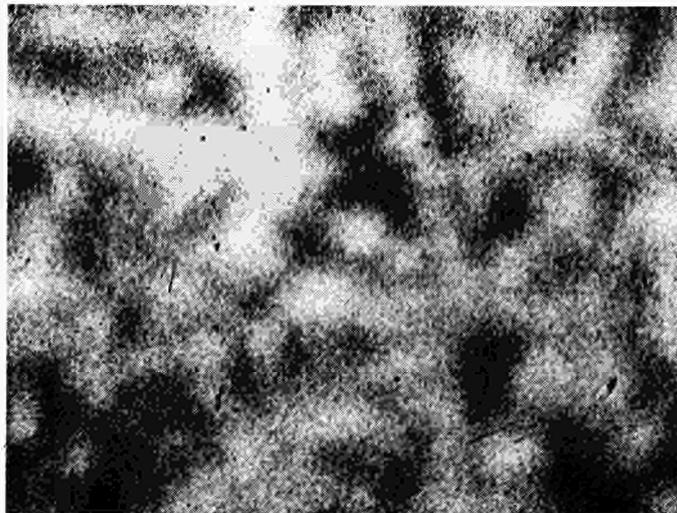
Fig. 15d — Autoradiograph of thin section (22 μ) taken from sample No. 3



(810)

x 10

Fig. 16a — Micrograph of sample No. 4



(815)

x 10

Fig. 16b — Autoradiograph of sample No. 4
(same position as Fig. 16a)

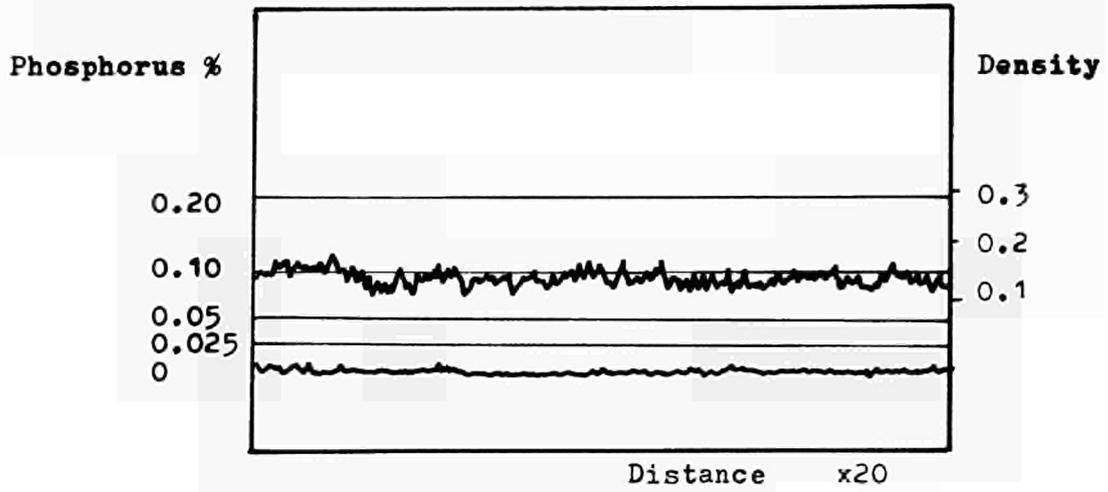


Fig. 16c — Densitometry of the autoradiograph shown in Fig. 16d

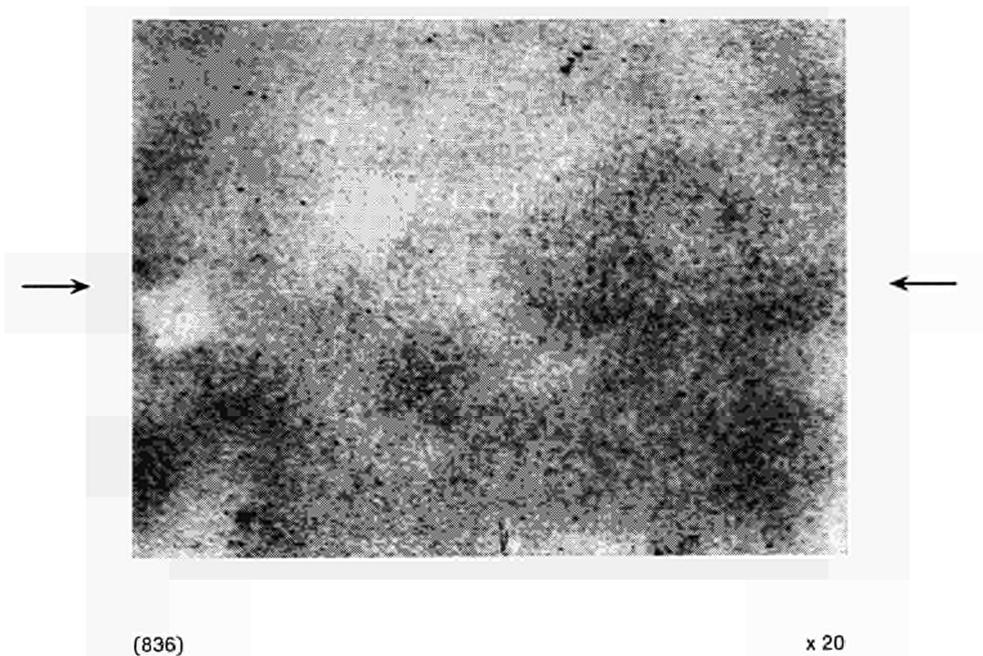
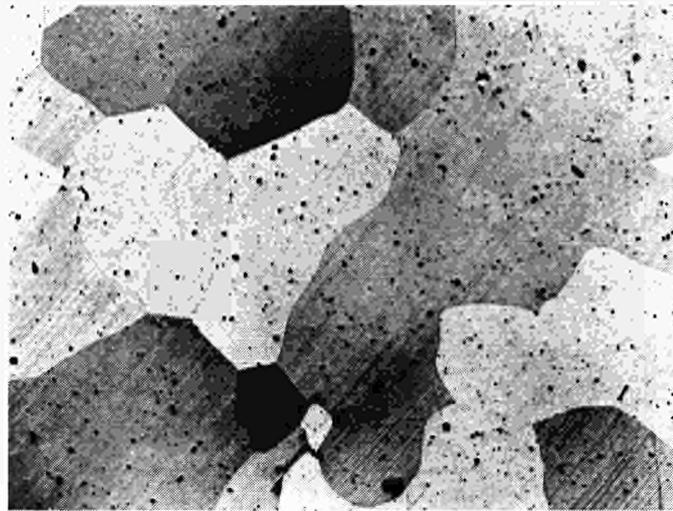


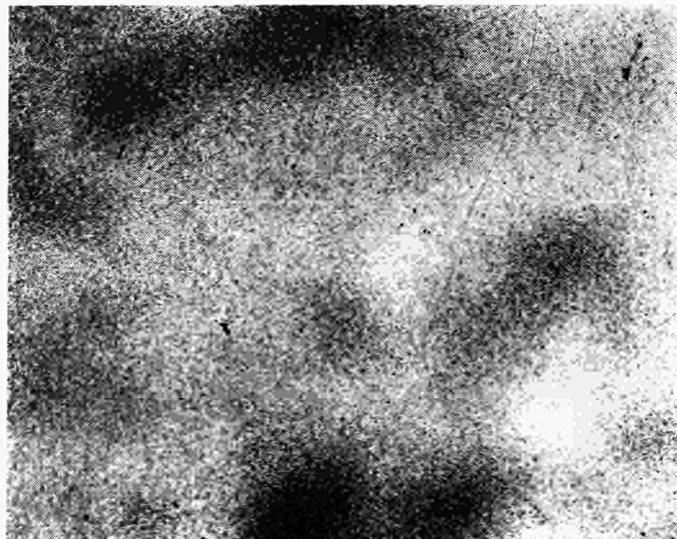
Fig. 16d — Autoradiograph of a thin section taken from sample No. 4 (22 μ)



(807)

x 10

Fig. 17a — Micrograph of sample No. 5



(808)

x 10

Fig. 17b — Autoradiograph of sample No. 5
(same position as Fig. 17a)

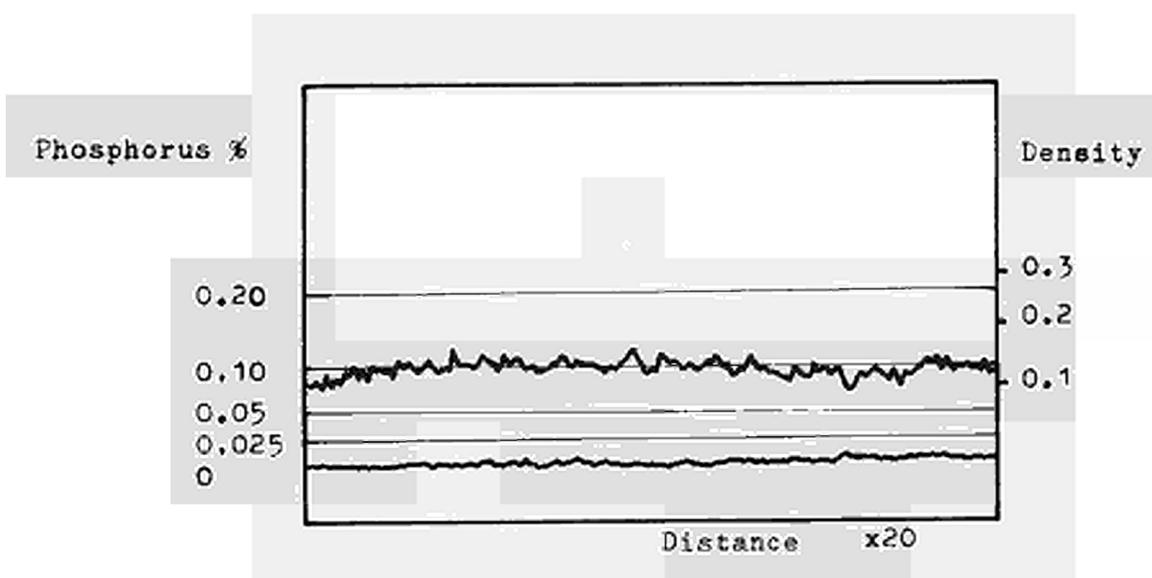


Fig. 17 c — Densitometry of the autoradiograph of a thin section taken from sample No. 5

Note : The autoradiograph (not reproduced here) shows a uniform grey field

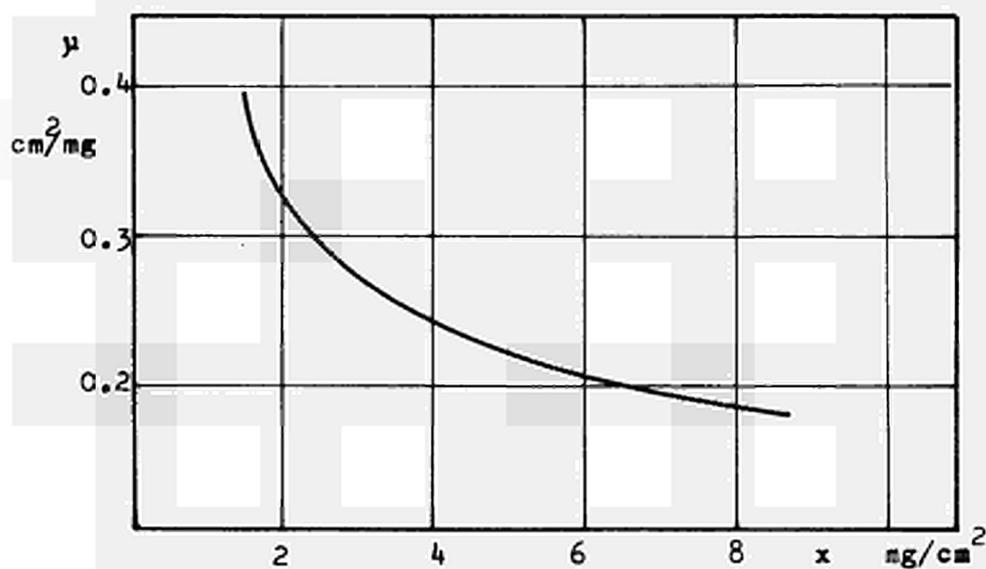


Fig. 18 — Ca^{2+} radiations : graph showing absorptiar coefficient versus thickness of absorber (Al)

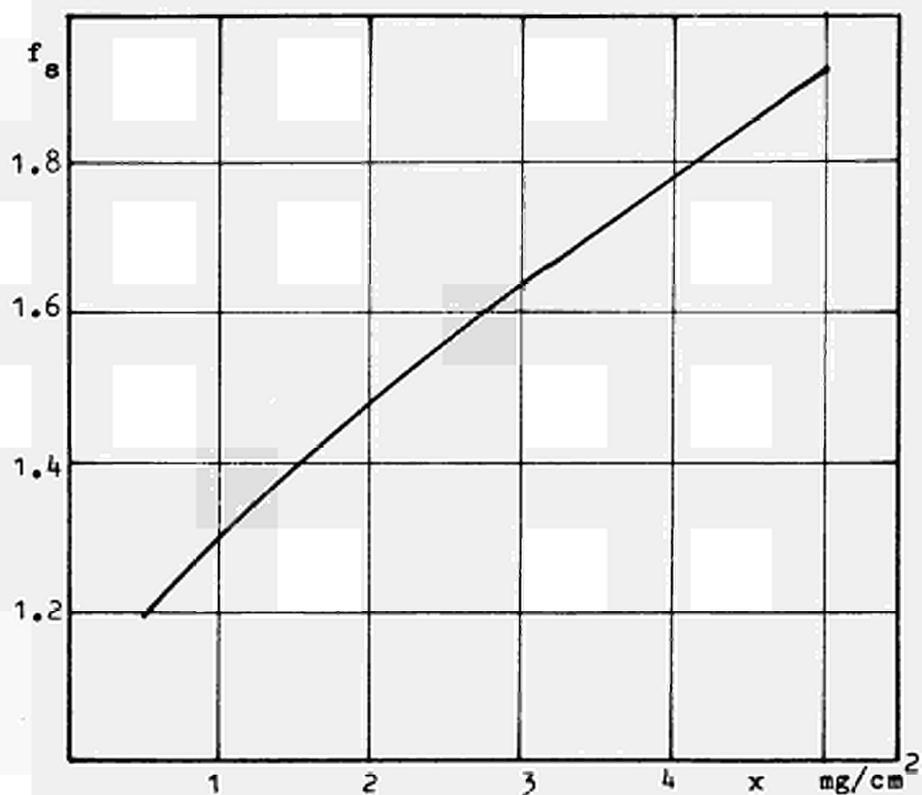


Fig. 19 — Ca^{2+} radiations : graph showing self-absorption coefficient versus thickness of absorber (Al)

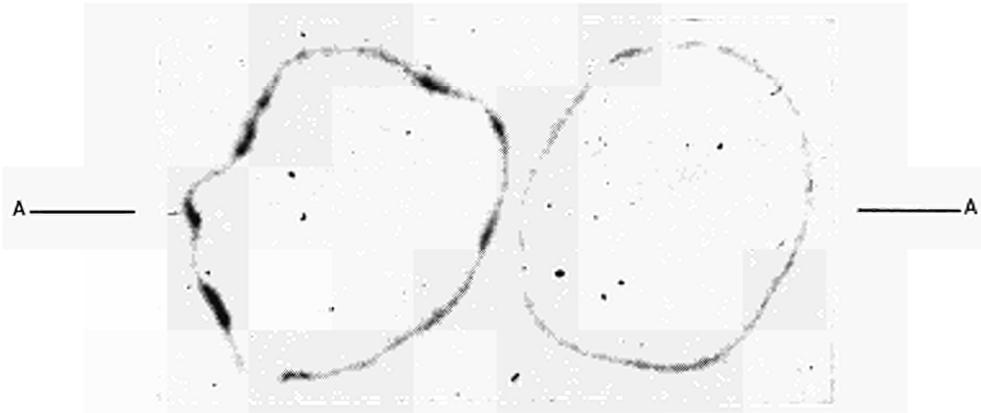


Fig. 20a — Autoradiograph of a thin section of quartz granules labelled on the surface with Ca^{45}

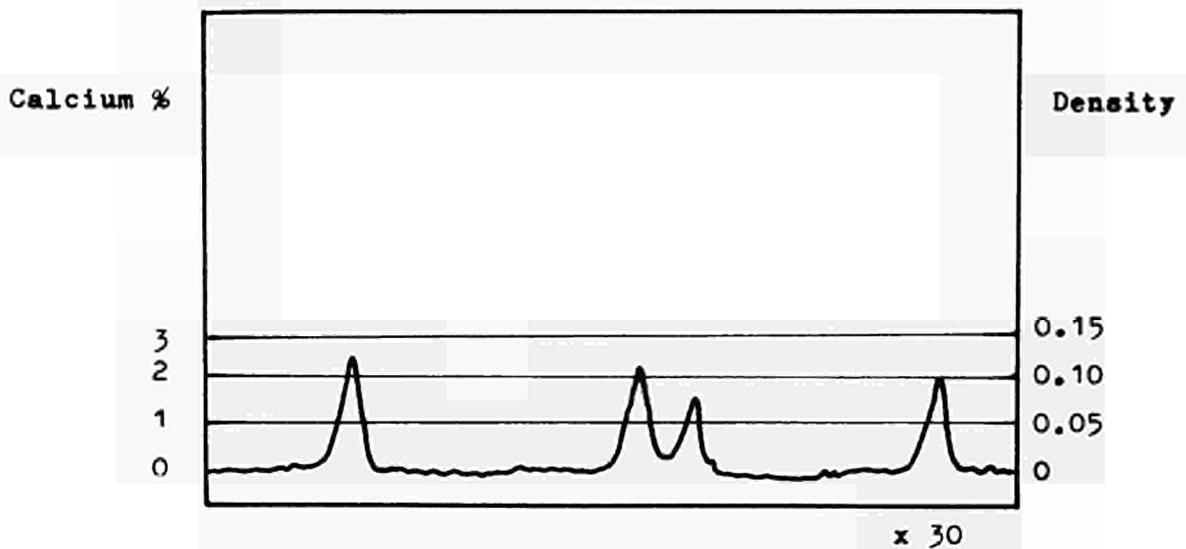


Fig. 20b — Densitometry of the autoradiograph shown in Fig. 20a (scaled in Ca %)

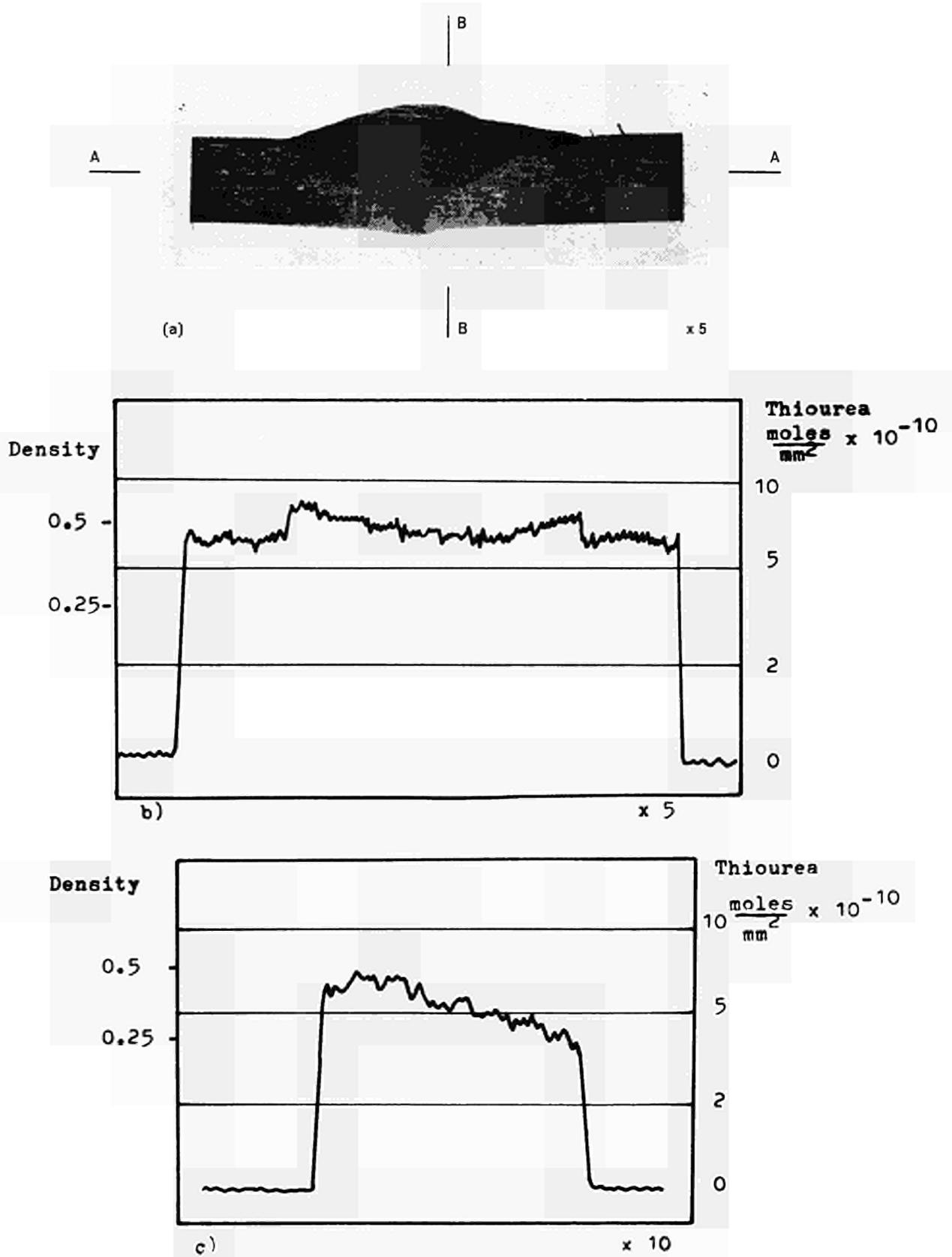


Fig. 21 — (a) Autoradiograph of the distribution of thiourea-S³⁵ on the surface of a steel sample immersed for 2 minutes in an inhibitor-containing active solution
(b) Microdensitometry along the line A-A
(c) Microdensitometry along the line B-B

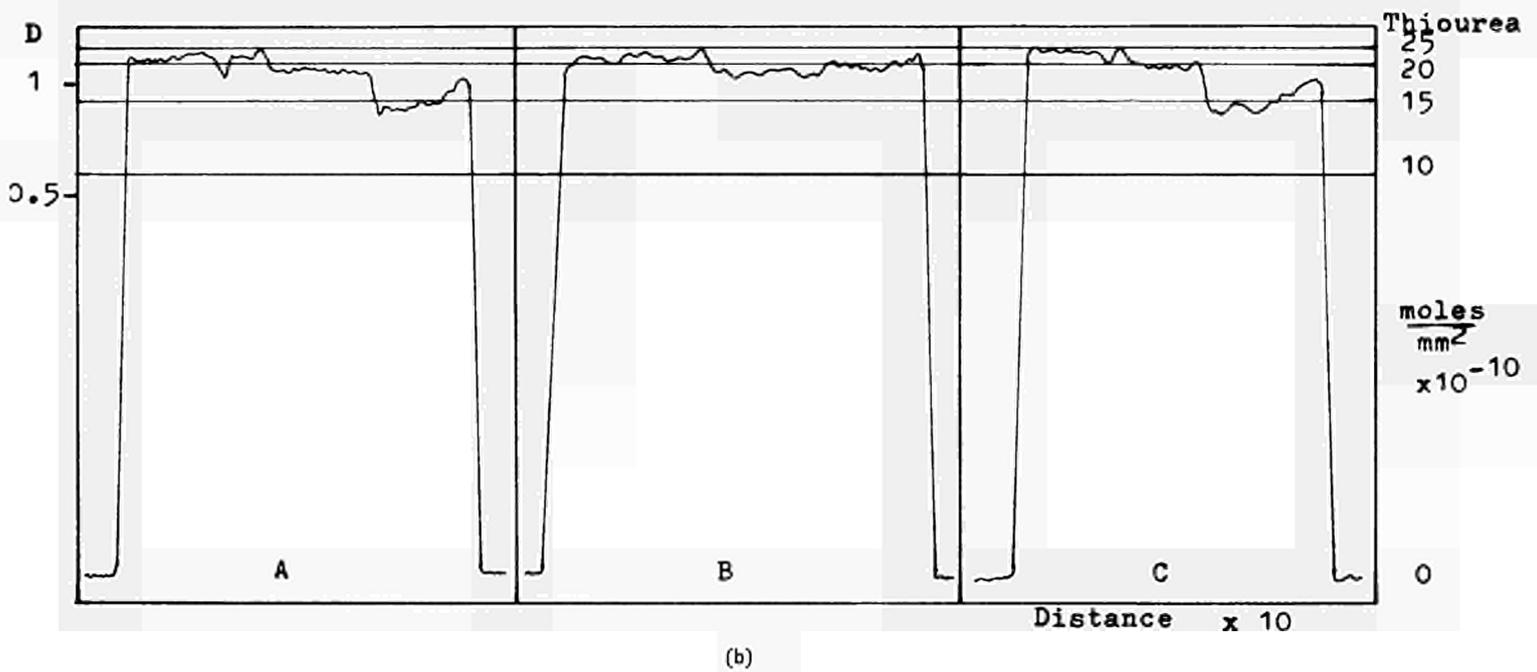
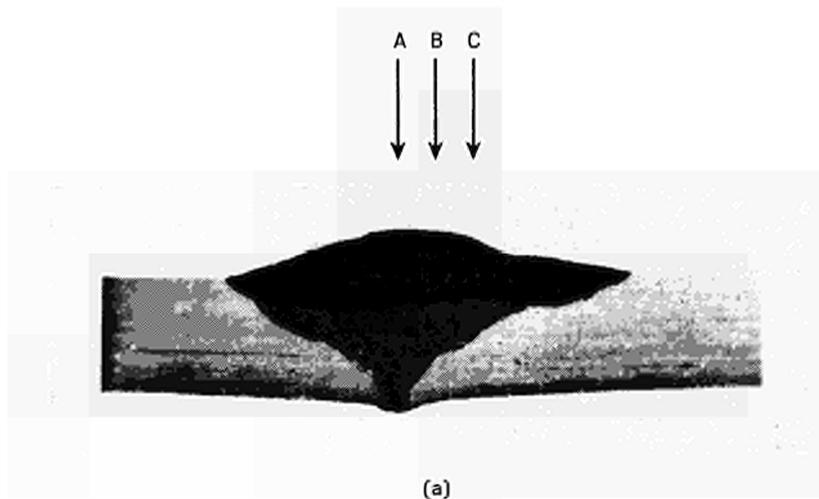


Fig. 22 — (a) Autoradiograph of the distribution of thiourea-S³⁵ on the surface of a steel specimen immersed for 60 minutes in an inhibitor-containing active solution

(b) Microdensitometry along the lines A, B and C.

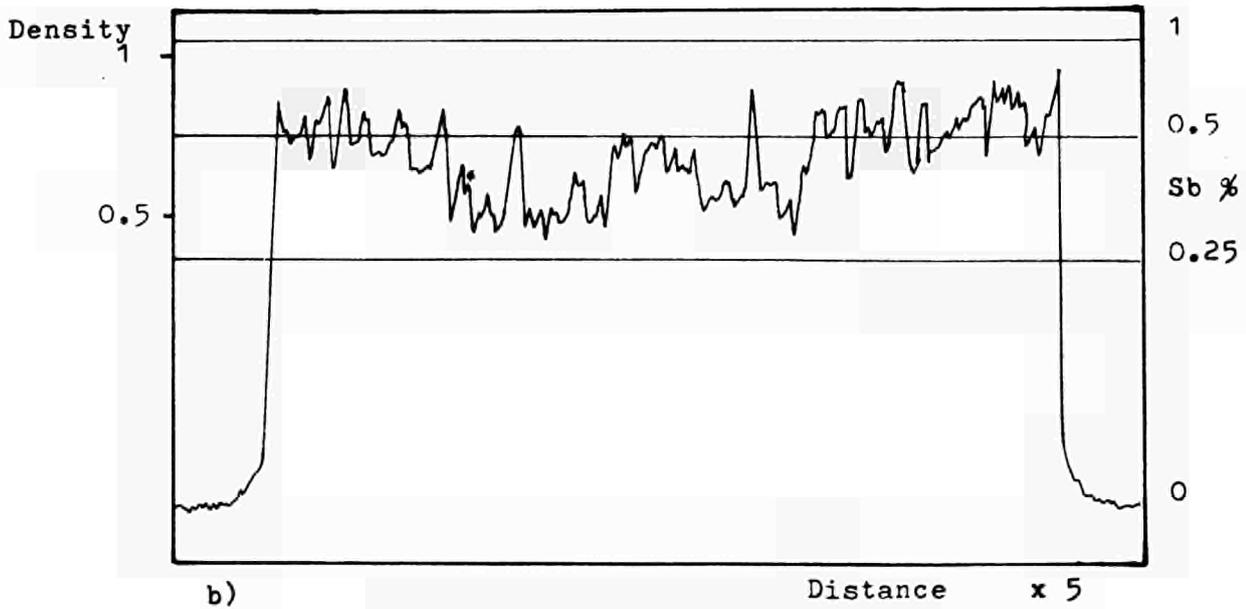
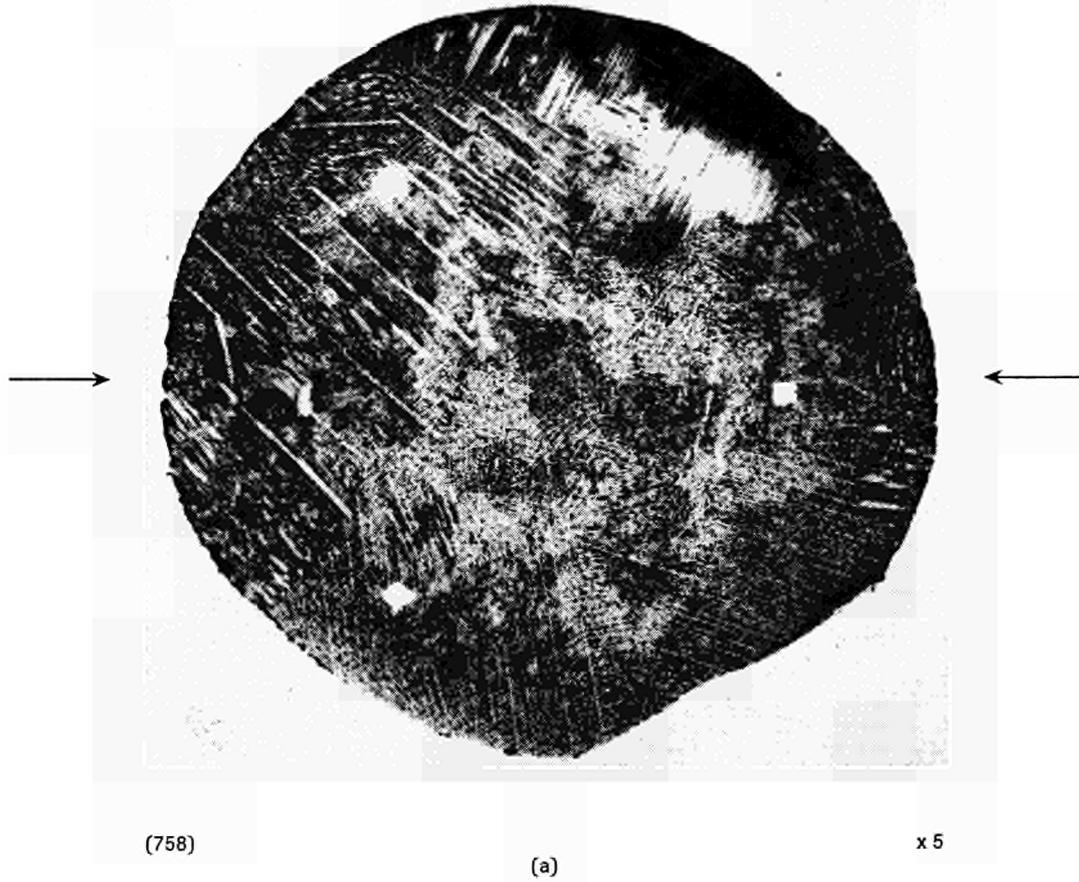


Fig. 23 — (a) Autoradiograph of a thin section (20μ) of a Cu-0.5 % Sb alloy labelled with Sb^{124}

(b) Microdensitometry along the line indicated ; the calculated antimony concentration is shown on the ordinate.

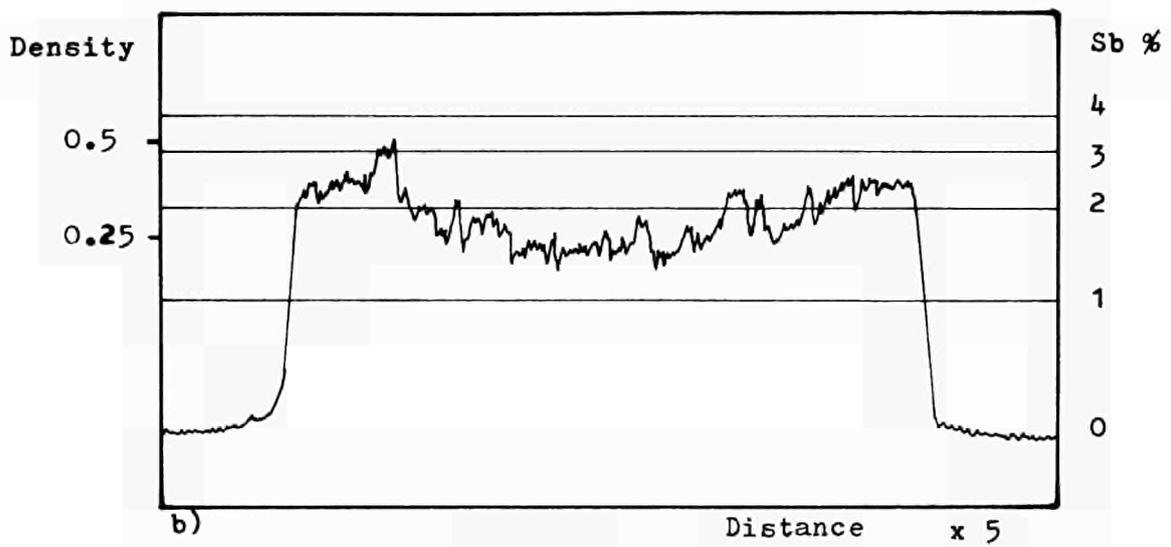
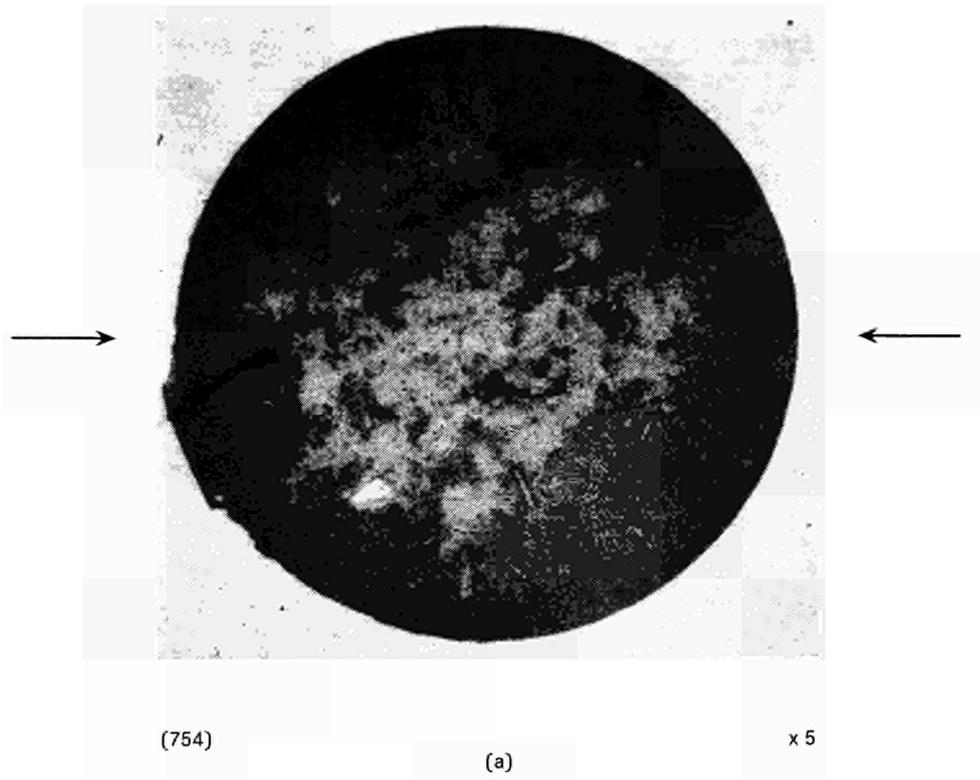


Fig. 24 — (a) Autoradiograph of a thin section (22μ) of a Pb-2% Sb alloy labelled with Sb^{124}

(b) Microdensitometry along the line indicated ; the calculated antimony concentration is shown on the ordinate.

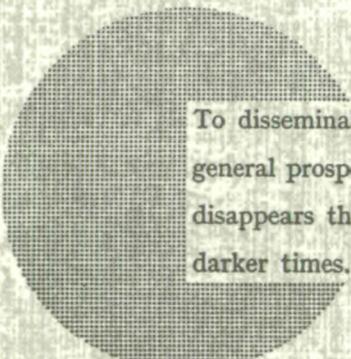
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Alfred Nobel

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