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Report on task FIN A845 on the Finnish support programme to IAEA safeguards

R. Pöllänen, A. Kansanaho, H. Toivonen

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ABSTRACT

Autoradiography was used to study radioactive particles that may be released from the nuclear fuel cycle. Autoradiography suits for pre-screening of the samples. Radioactive particles can be located accurately and detached from the sample for subsequent analyses. A digital scanner and tailored software allow to estimate the activity of the particles by a factor of two to three. High-energy beta emitters as well as pure beta emitters can be identified. A particle with activity of 0.1 Bq can be detected in five days of exposure. More than 10^5 disintegrations are needed to detect a black spot on the autoradiography film. Total activity of beta active nuclides can be evaluated if the number of disintegrations is smaller than 10^6. The diameter of the black spot is then below 1 mm. The edge of the black spot receives a beta dose of approximately 10 mGy. Particular emphasis was placed on method development from the point of view of in-field applications.

CONTENTS

1 INTRODUCTION

Autoradiography is a simple method that can be used in searching radioactive particles from different surfaces. It was widely used in monitoring radioactive fallout after the nuclear weapon tests in the 1950's and 1960's (Mamuro et al., 1962; Sisefsky, 1961, e.g.). It was widely used after the Chernobyl accident, too (Vapirev et al., 1994; Kolb, 1986; Keck et al., 1987; Baltensperger et al., 1987; Toivonen et al., 1987; Mattsson et al., 1986, Devell et al., 1986). Autoradiography was used in Finland for searching the signatures of radioactive particles after the incident at Sosnovyy Bor nuclear power plant in Russia (Toivonen et al., 1992).

In autoradiography radioactivity is revealed by photographic emulsions. An individual radioactive particle is identified on the film as a circular black spot considerably larger than the particle itself (Fig. 1). The spot size is a function of the number of disintegrations during the film exposure. This relationship can be used as a measure of the activity of the particle. Microscopic radioactive particles can be localized and detached from the sample for subsequent analyses.

Beta radiation originating from radioactive particles is the main cause of the blackening of the film (Fig. 1). X-rays and gamma rays may have some influence on the blackening, especially outside the maximum range of the beta particles, but usually their contribution is negligible. Alpha particles may cause heavy blackening of the film if there is no absorbing material between the active material and the film. However, in practice the beta active nuclides mask the effects of alpha particles.

An elementary particle travelling through the film releases some of its energy in collisions with the emulsion crystals which darken in the chemical development process. If the amount of darkened crystals is large enough a black spot appears on the film. Radioactive materials may be also evenly distributed in the sample. Then a blackened area, rather than a circular spot, is detected on the film.

Autoradiography can be used in environmental monitoring of airborne radioactive particles (or deposited particles) provided that novel equipment and methods are used. The procedures of film exposure, film processing and film analysis should be automated for simple, flexible and fast treatment of the samples.

Autoradiography is suitable for pre-screening of the samples. The procedure could be as follows:

(1) Swipe samples or air filters are put in a lightproof cassette for a certain period of time. This may be performed in a hotel room at night or in other places where dark room conditions are available.
(2) If necessary, the cassettes as such can be sent directly to a laboratory for further processing and analysis. However, the film processing can be performed in the field provided that dark room conditions are available.
(3) If blackened areas or black spots are detected on the film the samples should be sent to laboratory for detailed analyses.

The relatively slow procedure of sample preparation and measurement is a disadvantage of the autoradiography method. Thus, it is often used only to support other methods. Lack of information about its usefulness in particle activity measurements and, perhaps, its reputation as an old-fashioned method are other 'disadvantages'. An advantage is that the particle nature of possible radioactive releases from a nuclear facility can be easily detected. Impactors can ce used for this purpose, too, but they do not identify single particles. In addition, autoradiography is a simple and cheap method.

In this study the method of autoradiography is developed for field applications. An other objective is to assess the activity of the monitored particles. The instruments and the methods are developed and standardized in such a way that thy can be used either in laboratory conditions or in the field.

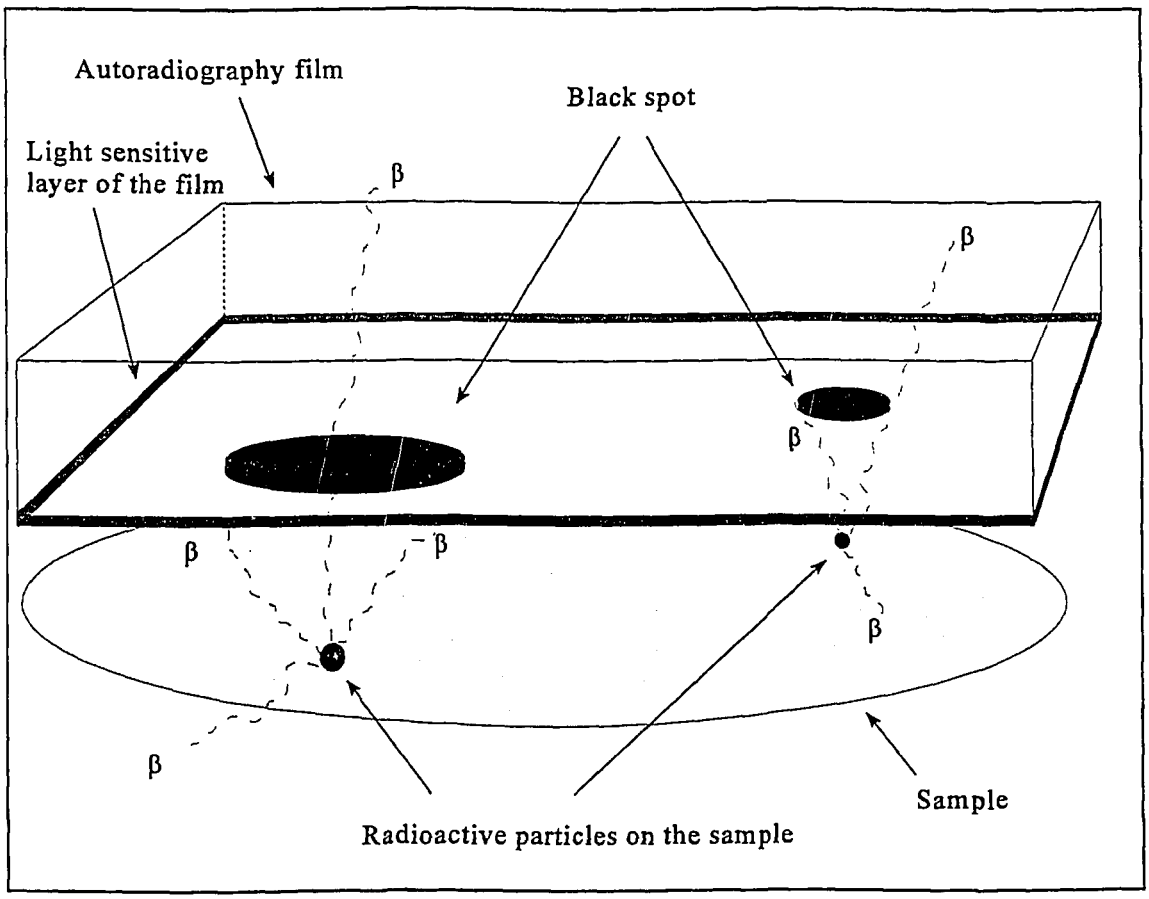

Figure 1. *Autoradiography using film that has only one sensitive layer. Beta active particles on the sample (swipe sample, air filter etc.) cause blackening of the light sensitive layer of the film. Size of the black spot depends on the exposure time, activity of the particles, nuclides in the particle and exposure geometry. In reality, the black spot does not have sharp edges.*

2 EQUIPMENT IN THE FILM EXPOSURE AND CHEMICAL PROCESSING

2.1 Film exposure

High-quality emulsion-coated film and a light proof exposure cassette are needed for the exposure (Fig. 2). The film used in the present study is coated on one side only. This light sensitive layer and the sample with radioactive particles are put in close contact (Fig. 3).

After a certain period of time the film can be processed either manually or using commercially available automatic processor. To estimate the activity of individual particles the size of the black spots must be evaluated. Then the exposure time must be known.

The distance between the sensitive layer of the film and the particle, as well as materials between them, has an influence on the formation of the black spot and, consequently, on activity estimation of the particle. This is true particularly for low-energy beta emitters. The particles may be located deep in a sample matrix or on the surface.

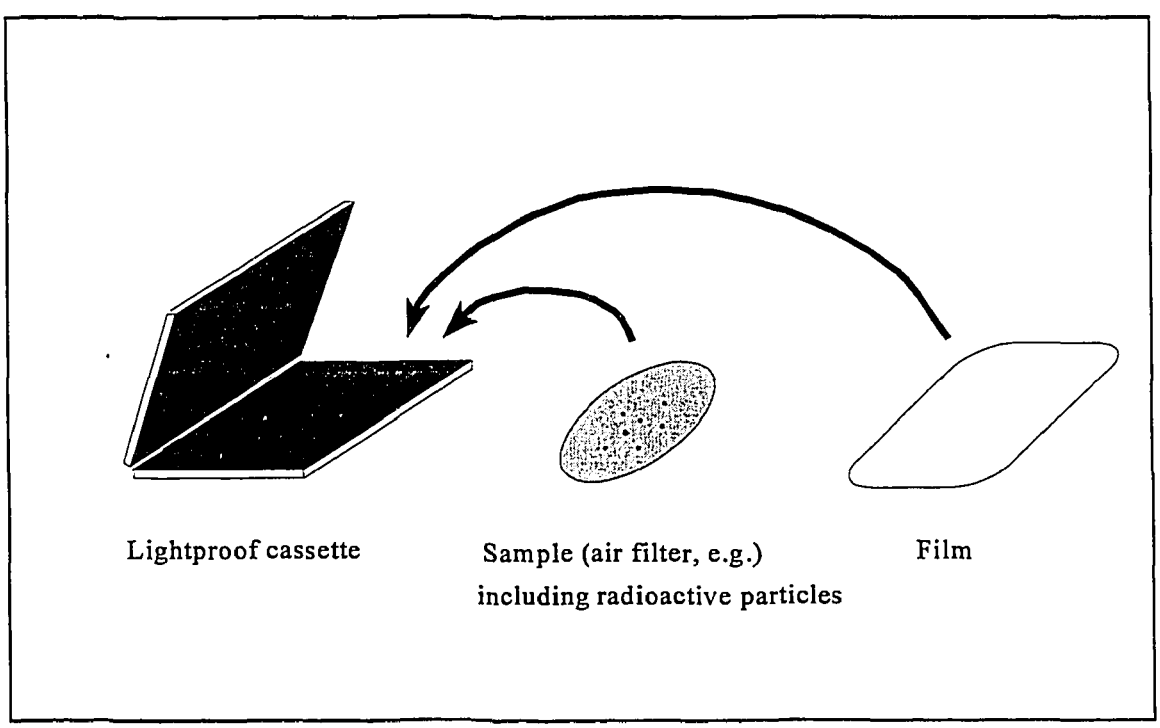

Figure 2. *Equipment needed for the film exposure. The sample and the film are placed in a lightproof cassette.*

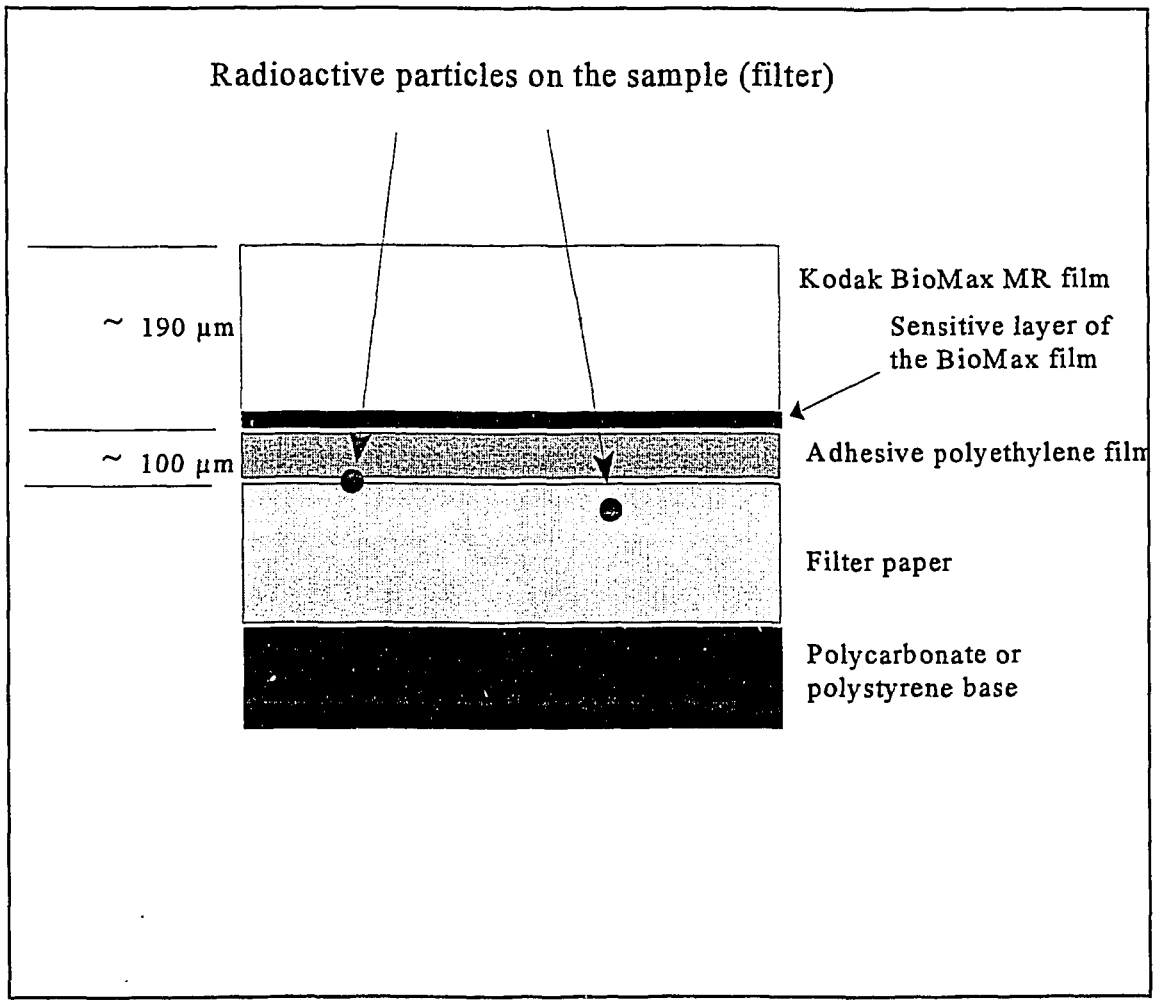

Figure 3. *An example of the cross section of the irradiation geometry. Particles on the sample and sensitive layer of the film are on close contact provided that adhesive polyethylene film is not used (this geometry enables detection of pure alpha emitters). Then the particles should be fixed carefully to the filter using spray-on-laquor, e.g. Only high-energy betas may arrive to the sensitive layer of the film if more absorbing material is put between the film and the particle.*

2.2 Chemical processing of the films

The equipment needed for the film processing are similar to those used in a standard X-ray film development process. Developing and fixing procedures must be performed in a dark room condition with running water. The chemical processing and rinsing can be performed in three sectional racks which save a lot of space (Fig. 4). Each of the chemicals in rack 1 are in the separate sections. Rack 2 contains only rinsing water. Automatic film processing machines are also commercially available.

After the exposure, the film is put in a film holder designed for the racks. The film in a cassette is set to the rack 1 in the section that contains developing chemicals. After 3—5 minutes (the time depends on the temperature of the liquid bath) the film is put in a stop bath for 30 seconds. The fixing bath takes 10 minutes. After that the lights can be turned on for the final rinsing with water (about 2 min in each section of rack 2). The film must be dried for subsequent analyses.

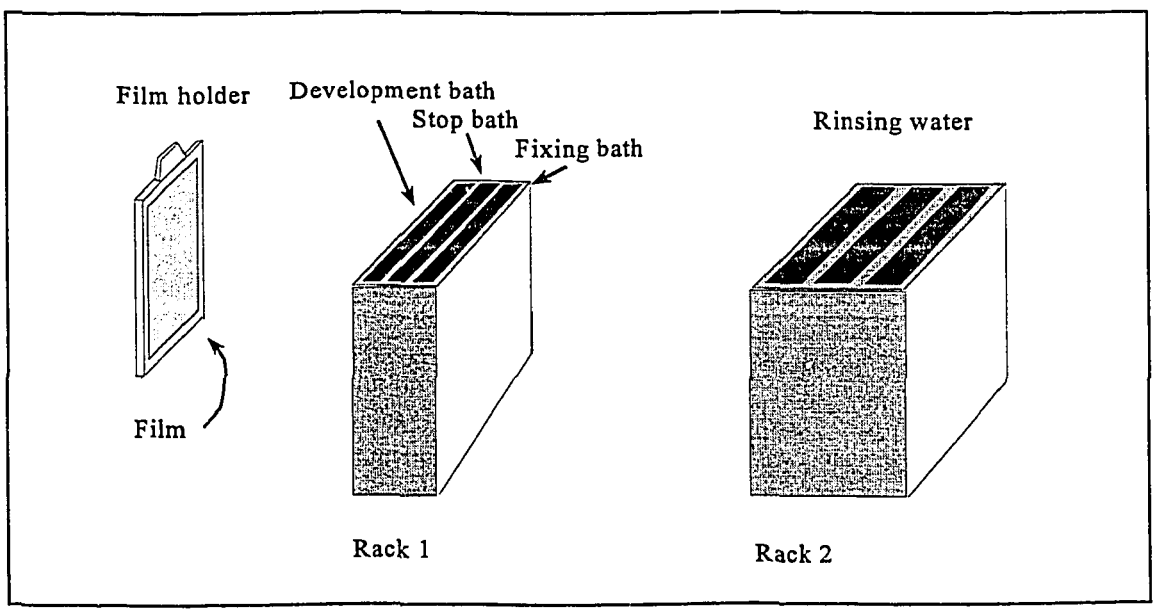

Figure 4. Equipment needed for the film processing in the field.

3 ANALYSIS OF THE BLACK SPOT

3.1 Formation of the black spot

Blackening of the film is a function of beta dose. The higher the dose, the greater the number of blackened silver halogenide crystals and, therefore, the darker the colour. The centre of the spot receives the highest beta dose and, thus, has the darkest colour. Along the radius of the spot the beta dose decreases and the colour gets gradually lighter. When the beta dose inside a small area of the film rises over a certain level all the silver halogenide crystals are exposed and the area comes fully blackened. Increasing the number of beta disintegrations raises the beta dose gradually and the saturated black centre begins to grow.

The beta dose that the film receives depends on the measuring geometry (properties of the film and material between the film and the particle), on particle properties (particle activity and self-absorption of beta particles) and on the energy of beta particles (nuclides). If the particle is in a homogeneous media the radius of the black spot cannot be larger than the maximum range of the beta particles. The air gap between the film and the particle may have a significant influence on the size of the black spot.

For particle of unit activity locating as shown in Fig. 3 the radial beta dose rate in the sensitive layer of the film behaves as shown in Fig. 5. In this calculation it was assumed that particle is in the infinite water medium. The distance between the particle and the sensitive layer of the film is 100 μm. Let us assume that a dose of 10 mGy is needed to produce a 'black' film (c.f. Fig. 10). It is now possible to calculate the diameter of the black spot as a function of the number of disintegrations (Fig. 6) for each nuclide separately.

The distance between the film and the beta active particle is so large that the most low-energetic betas (< 0.1 MeV) do not arrive to the sensitive layer of the film. Low-energy beta emitters, such as ^{95}Nb, may cause a black spot with a diameter less than 0.5 mm whereas the spot size for high-energy beta emitters, such as ^{106}Rh and ^{144}Pr, may be more than 1 cm provided that the exposure time is long enough. In practice, the diameter of the spot is considerably smaller than the maximum diameter.

Fig. 6 shows that it is theoretically possible to evaluate the activity of the particle from the size of the black spot. However, the particles often contain a mixture of nuclides. In practice, high-energy beta emitters or those nuclides which have the highest activity have a major influence on the size of the spot. Both theoretical calculations and test measurements are needed to find out the relationship between particle activity and spot size.

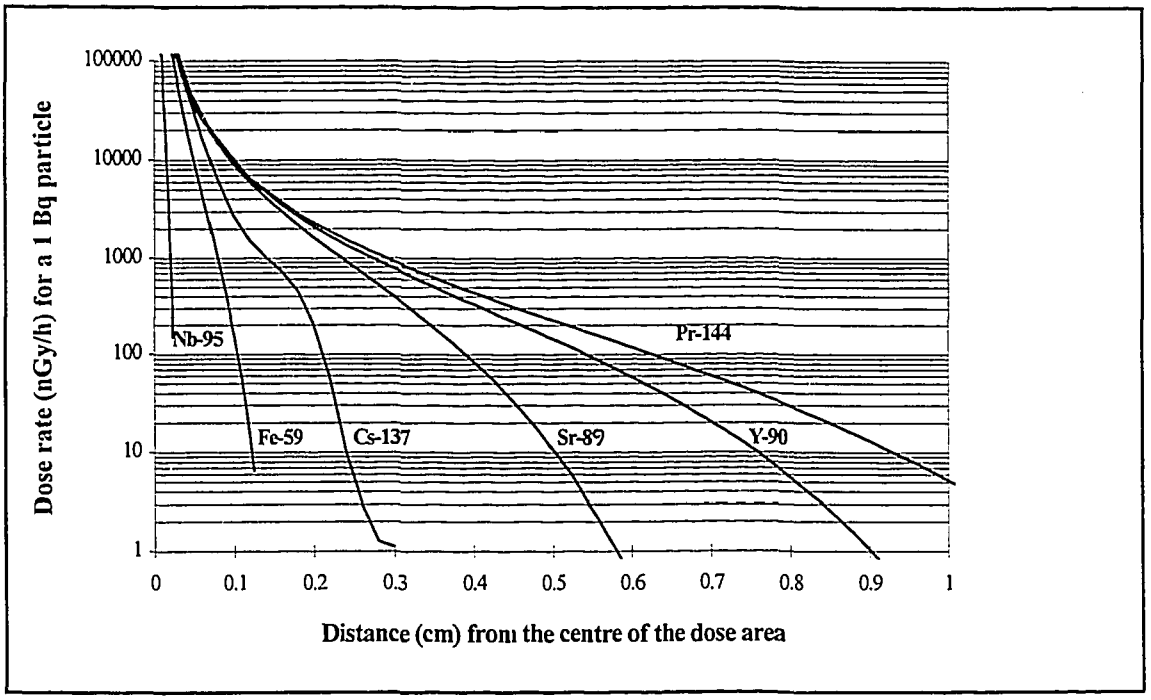

Figure 5. Radial dose rate (nGy h^{-1}) caused by different beta emitters in the sensitive layer of the film (depth 100 μm). The activity of point isotropic nuclides is 1 Bq. Beta dose rates calculated by Cross et al. (1992) are used for the estimation.

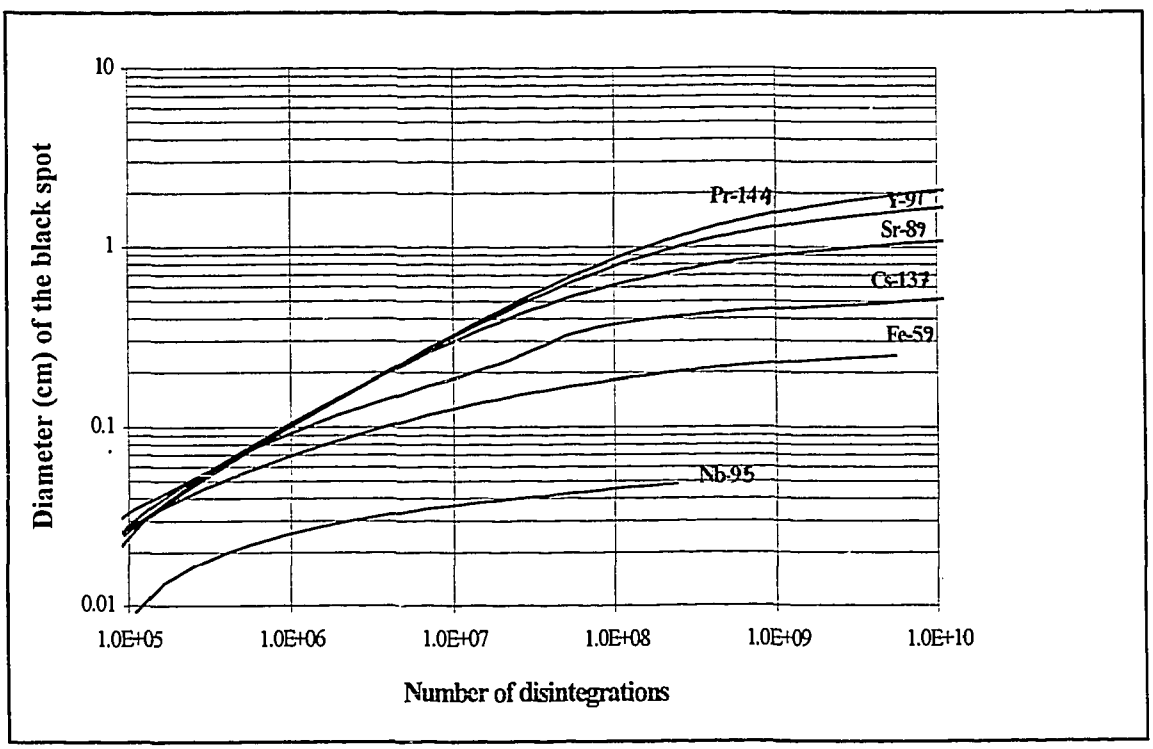

Figure 6. Diameter of the black spot (10 mGy isocurves) versus number of disintegrations (depth of the dose area 100 μm) for different beta emitters.

3.2 Measurement of the spot size

A simple method to measure the size of the spots is to make a digital image (Fig. 7) with a scanner, store it using a certain image file format and analyse the contents with suitable software. A digital image is easy to produce with a scanner (Hewlett Packard ScanJet IIc) and Windows software. The image of a single spot was scanned in black and white with resolution of 300 dots per inch (dpi). The information was stored in a Windows bitmap file with indexed 8 bit colours which in this case corresponds to 256 shades of grey.

Several computer codes are available for image analysis but they are too complicated and expensive for this application. Therefore, a simple code was written to cope with the image and activity calculations (appendix 1). Each byte in a bitmap file is read as a character and its numeric value is returned (ANSI code of a 8 bit character). This value corresponds to the colour index of the pixel. In the area selected, our program counts the number of pixels representing each colour.

The colour 'black' has an index 0 and the colour 'white' an index 255. In practice, light reflection from the surface of the film changes the darkest colour index to 10 or more. The area, a_{pix}, of one square-shaped pixel is 0.0072 mm^2 (side 0.084 µm). The pixel size determines the minimumsize of the black spot. The area of the black spot is obtained by calculating the number, n, of the black pixels. The calculation of the diameter of the spot is straightforward:

$$d_{spot} = (4\ n\ a_{pix}/\pi)^{1/2}.$$

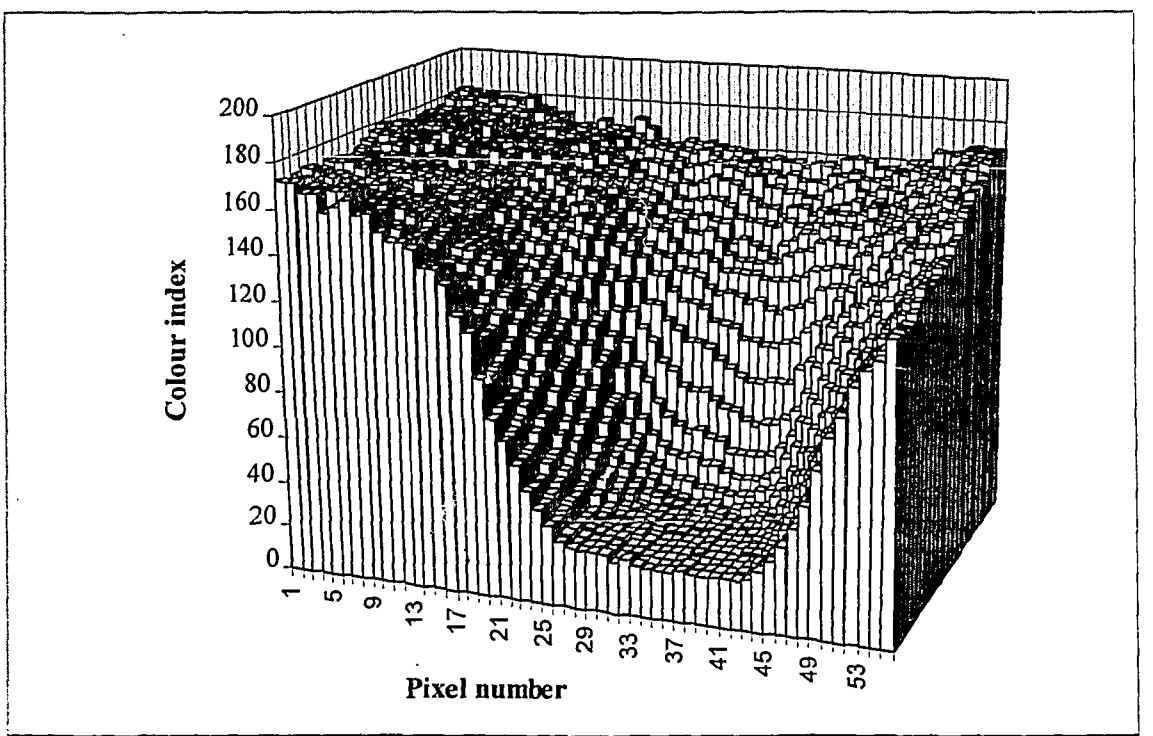

Figure 7. Cross section of a digital image of a black spot on an autoradiography film. Each column is a square-shaped pixel. Colour index (in this case < 200) refers to the shade of grey.

Selection of the colour 'black' has to be performed carefully. Systematic error follows if only the darkest colours are used for the spot size estimation (Fig.8). In addition, the spot size may have an influence on colours the scanner can detect.

Resolution of the scanner determines the absolute minimum detectable size of the black spot. It is not possible to detect a black spot smaller than a_{pix} using a digital scanner. In practice, a multiple of a_{pix} is the true minimum detectable size. It must be determined experimentally. According to the test measurements, the diameter of the black spot should be greater than about 0.3 mm (~10 pixels).

Let us assume that we have two ideal black spots of different sizes, i.e. the spots are assumed to be sharp-edged and fully blackened whereas the backgroud is white. Let one spot be large (several mm in diameter) and the other small (like the fullstop at the end of this sentence). After several experiments it was found that, according to the digital scanner, the centre of the large spot is more 'black' than the centre of the small spot (Fig. 9). Thus, selection of the colour 'black', known here as cut-off colour, must be performed separately for each spot and in such a way that comparison of spots of different sizes is possible.

Several tests were performed for ideal spots. The cut-off colour index must be selected according to the derivative of the cross-section curve. Extreme values of the derivative curve give appropriate colour. In practice, the spots on the autoradiography film are not sharp-edged. However, it was found that selection rules for defining the cut-off colour are similar to those used for the ideal spots.

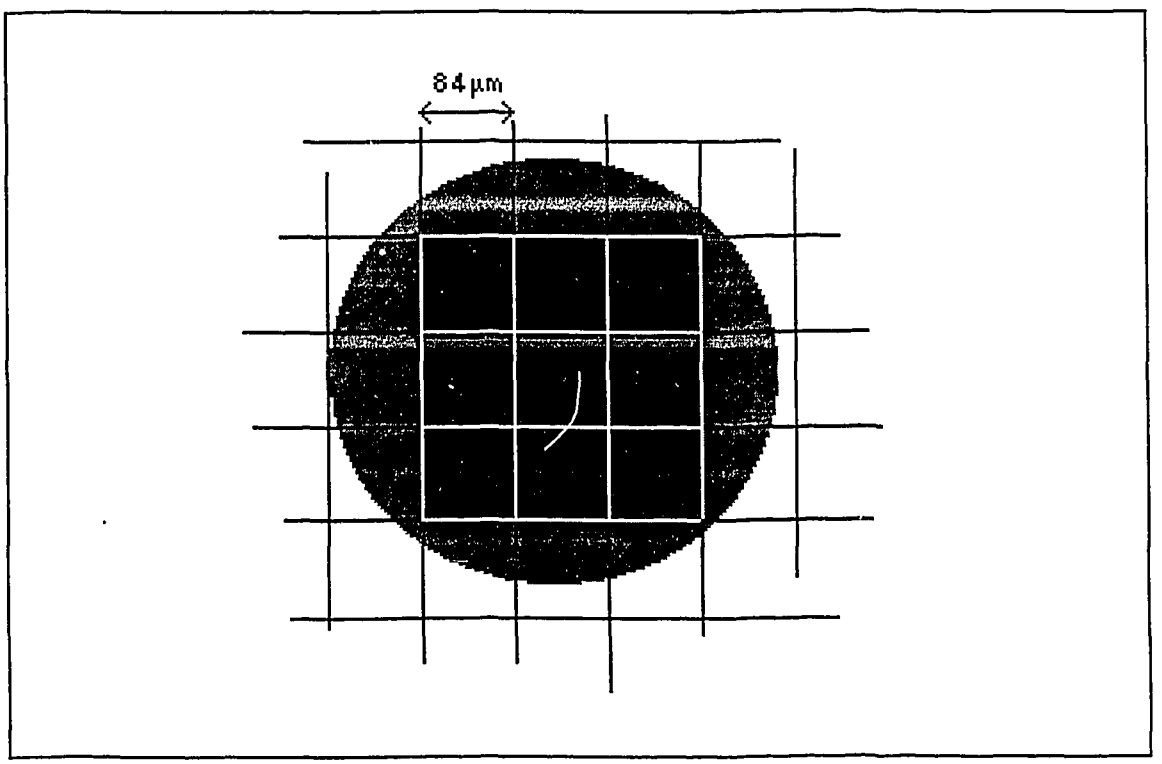

*Figure 8. An ideal black spot detected by a scanner. Each pixel is shown as a square-shaped area. Only pixels located at the centre of the spot are considered 'black'. Other pixels contain partly black from the spot and partly white from the background. Thus their colour index is between black and white. According to the black pixels the spot size is 9*a_{pix} which is considerably smaller than the true spot size. To avoid systematic errors also these 'grey' pixels must be taken into account in the size estimation.*

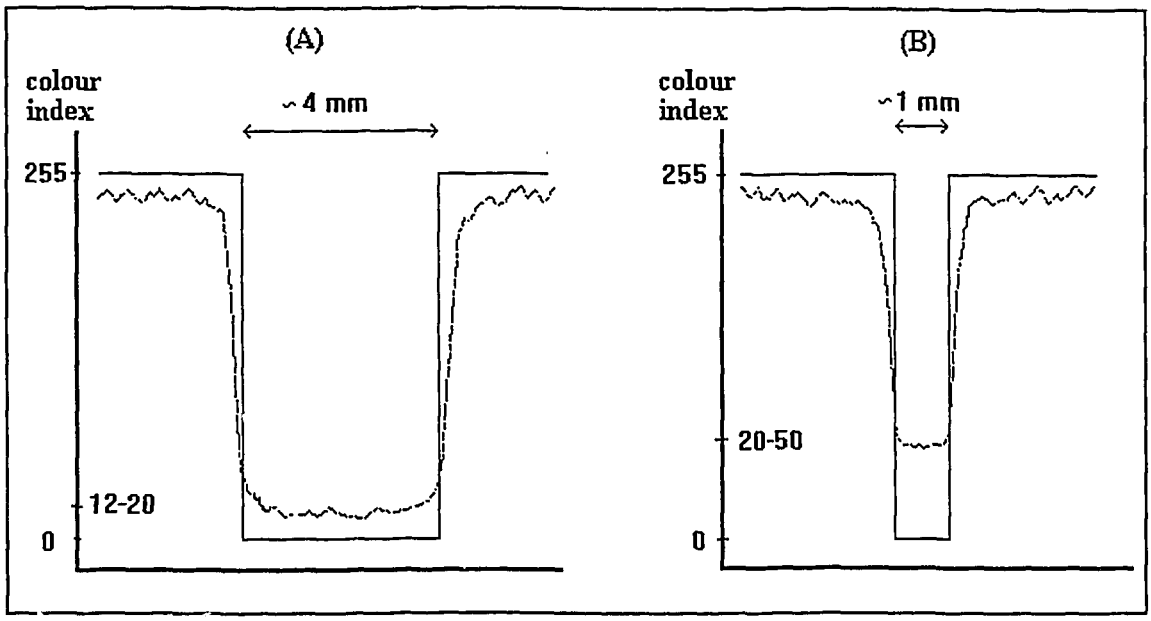

Figure 9. Cross section (solid line) of an ideal large black spot (A) and an ideal small black spot (B). The scanner identifies the spots as smooth-edged (dashed line). Colour index for the ideal background (= 'white') is 255 and colour index for the ideal spot (= 'black') is 0. For large spots the minimum index for the colour black is 12—20 because of the light reflection from the surface of the film. For small spots the minimum value of the index may be up to 50. Similarly, colour indices 'white' are smaller than 255.

3.3 Calibration

Activity estimation from the size of the black spot is based on the assumption that each colour index represents a certain dose. Unfortunately, blackening of the film is not directly proportional to exposure. Diffuculties may arise if the amount of blackening is evaluated for different doses.

However, the problem is simplified if only the cut-off dose (i.e., the cut-off colour) is considered. The main question then is what is the minimum dose needed to blacken the film? The edge of the black spot receives this dose. Theoretical calculations compared to measured spot sizes, i.e. calibration, are needed to evaluate the cut-off dose. This limit is the basis for the estimation of the activity of particles.

Two different approaches are possible for the calibration purposes: (1) Exposure geometry of the films is designed in such a way that dose rate coefficients for water-water boundary can be used in the calibration (i.e. the radioactive particle is in infinite water medium). These coefficients are well known and they can be easily used in the calculations. The difficulty is that all materials near the radioactive particle should be similar (with respect to beta particle interactions) to water. The properties of autoradiography film itself, for example, differ from the properties of water. (2) An other possibility is to use a fixed geometry different to water-water boundary and calculate beta dose rates for this specified geometry. Although this approach may be better than method 1 used in this study, it is beyond the scope of the present study.

Several test particles of known activity were used for the calibration. Some of the particles contained only high-energy beta emitters such as ^{106}Rh (^{106}Ru) and ^{144}Pr (^{144}Ce) whereas some of them contained only low- and medium-energy beta emitters such as ^{58}Co, ^{59}Fe and ^{60}Co. Some particles contained a mixture of low-, medium- and high-energy beta emitters.

Autoradiography film is a detector that reacts to the total absorbed dose. It does not distinguish whether the dose comes from high-energy beta emitters or low-energy beta emitters. All that is needed is to calculate the 'theoretical' curve of radial beta dose as a function of the number of disintegrations and compare the detected spot size to the 'theoretical' curve. This comparison, performed for particles of known activity, gives a possibility to estimate the activity of unknown beta active particles.

Calculations and experiments with a test particle are shown in Fig. 10. This particle contains ^{106}Ru/Rh (1.9 Bq), ^{137}Cs/Ba (1.8 Bq), and ^{144}Ce/Pr (36 Bq). The dose of approximately 10 mGy refers to the cut-off colour (Fig. 10 A). 'Theoretical' diameter of the black spot versus number of disintegrations is shown in Fig 10 B. The diameters are calculated by assuming that the cut-off doses are 5, 10 and 20 mGy, respectively. The diameters are calculated using point source beta dose rates for different beta emitters in water-water boundary (Cross et al., 1992). These 'theoretical' dose rates are based on Monte Carlo calculations. Detected diameters for different cut-off colour indices are inserted in Fig. 10 B. The effect of changing cut-off colour indices for black spots of different sizes is prominent. Because the high-energy beta emitters ^{106}Ru/Rh and ^{144} Ce/Pr produce much larger black spots than the medium-energy beta emitter ^{137}Cs/Ba (their total activity is also much higher) only the activity of the high-energy beta emitters is included in the number of total disintegrations.

Let us assume that the measured diameters in Fig. 10 B are for a particle of unknown activity. If the activity calculation is based on 'theoretical' estimation of beta dose curves ranging from 5 to 20 mGy then the estimated activity has an uncertainty by a factor of about 3—4 (the difference of the number of disintegrations for a specified diameter of the black spot). Because the real dose range is smaller in this case (about 7—13 mGy, Fig. 10 B) the uncertainty of estimated activity is smaller.

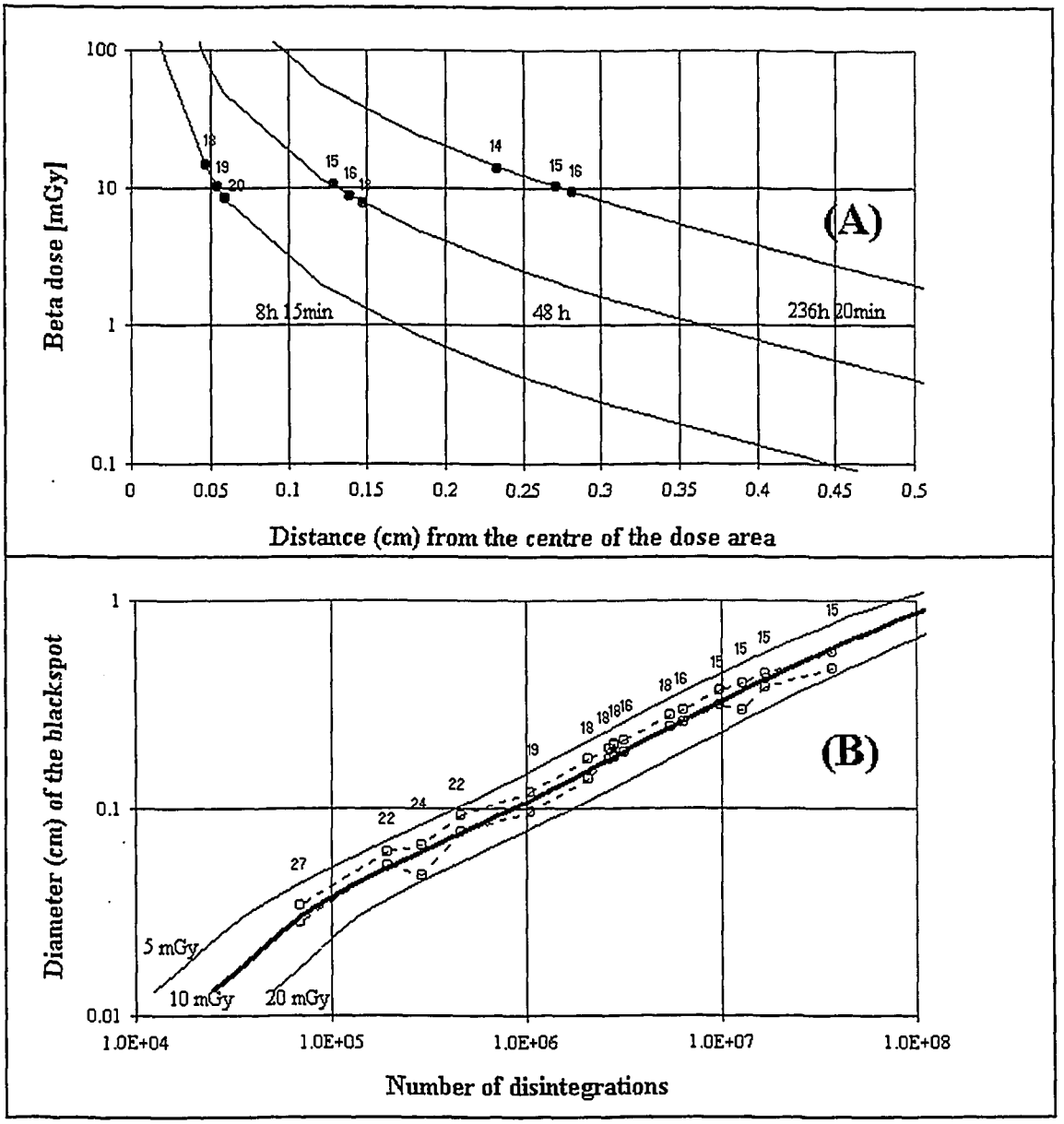

Figure 10. (A) *Beta dose caused by a test particle as a function of radial distance from the centre of the dose area (depth 100 μm) for three different exposure times. The points and the numbers above each curve represent the cut-off indices for colour 'black' used for the size estimation. The effects of small changes of the cut-off indices are clearly seen.* (B) *Diameter of the black spot versus number of disintegrations for the same test particle. The uppermost curve (thin solid line) represents the cut-off dose of 5 mGy whereas the other two curves are for the dose of 10 (thick solid line) and 20 mGy (thin solid line). Cut-off colour index for each size measurement is marked above each pair of points. The two points that represent each measured diameter refer to the 'most probable cut-off value +1' (upper point) and to the 'most probable cut-off value — 1'(lower point). The dashed lines that join the measured sizes are only to guide the eyes. Fifteen autoradiograms with different exposure times ranging from 30 min to 10 days were needed to produce the data.*

3.4 Estimation of activity from the size of the black spot

The dose of ~10 mGy that represents the cut-off colour index 'black' is the basis for the activity estimation. Combining this result to the detected size of the black spot gives a possibility to estimate the activity of particles.

The size of the black spot is proportional to the exposure time and activity of the particle. In addition, the spot size depends strongly on the nuclide in question. Nuclides with high beta energy may produce considerably larger spots than those with low beta energy. Autoradiography film reacts to the total absorbed dose. It does not distinquish whether the dose comes from high-energy beta emitters or from low-energy beta emitters. The high-energy beta emitters may mask the effects of low-energy and medium-energy beta emitters. Then one may ask which nuclides and, correspondingly, how few disintegrations can be detected in autoradiography.

Activity estimation is basically a simple task if the particle contains only one specified radioactive isotope. If the particle contains several radioactive nuclides with different beta energies the situation is more complex. Thus, some information about the nuclides in a radioactive particle must be available. The problem is illustrated by the following example.

Let us assume that a particle contains two radioactive isotopes. Let the isotopes be ^{137}Cs that can produce a spot smaller than 5 mm in diameter, and ^{144}Pr that can produce a spot a few centimeters in diameter (Fig. 6). Three different cases are considered here:

(A) Activity of ^{137}Cs is ten times higher than the activity of ^{144}Pr.
(B) Activity of ^{137}Cs and ^{144}Pr are equal.
(C) Activity of ^{144}Pr is ten times higher than the activity of ^{137}Cs.

Calculated diameter of the black spot as a function of the number of disintegrations is presented in Fig. 11 A—C for these cases. Three curves are present in each of the figures. In curve (1) both nuclides are taken into account for the number of disintegrations. Only ^{144}Pr affects the size of the black spot if the spot is larger than 5 mm in diameter. In curve (2) only ^{144}Pr is taken into account for the number of disintegrations but ^{137}Cs is included in the spot size estimation. ^{137}Cs may have an influence on the spot size when the number of disintegrations is small. In curve (3) ^{137}Cs is completely omitted in the number of disintegrations and in the spot size estimation.

The following conclusions can be made:

- If the activity of high-energy beta emitters is higher than the activity of medium- and low-energy beta emitters the spot size is determined only by the high-energy beta emitters (Fig. 11 C). For such particles it is not possible to estimate the activity of medium- and low-energy beta emitters with autoradiography.

- If the activity of low- and medium-energy beta emitters equals with high-energy beta emitters the spot size is determined mainly by the high-energy beta emitters (Fig. 11 B).

- If the activity of high-energy beta emitters is considerably lower than the activity of medium- and low-energy beta emitters the spot size is determined by their combined effects (Fig. 11 A). Low- and medium-energy beta emitters determine the spot size for a small number of disintegrations (spot diameter smaller than a few mm) whereas high-energy beta emitters determine the size of the large spots.

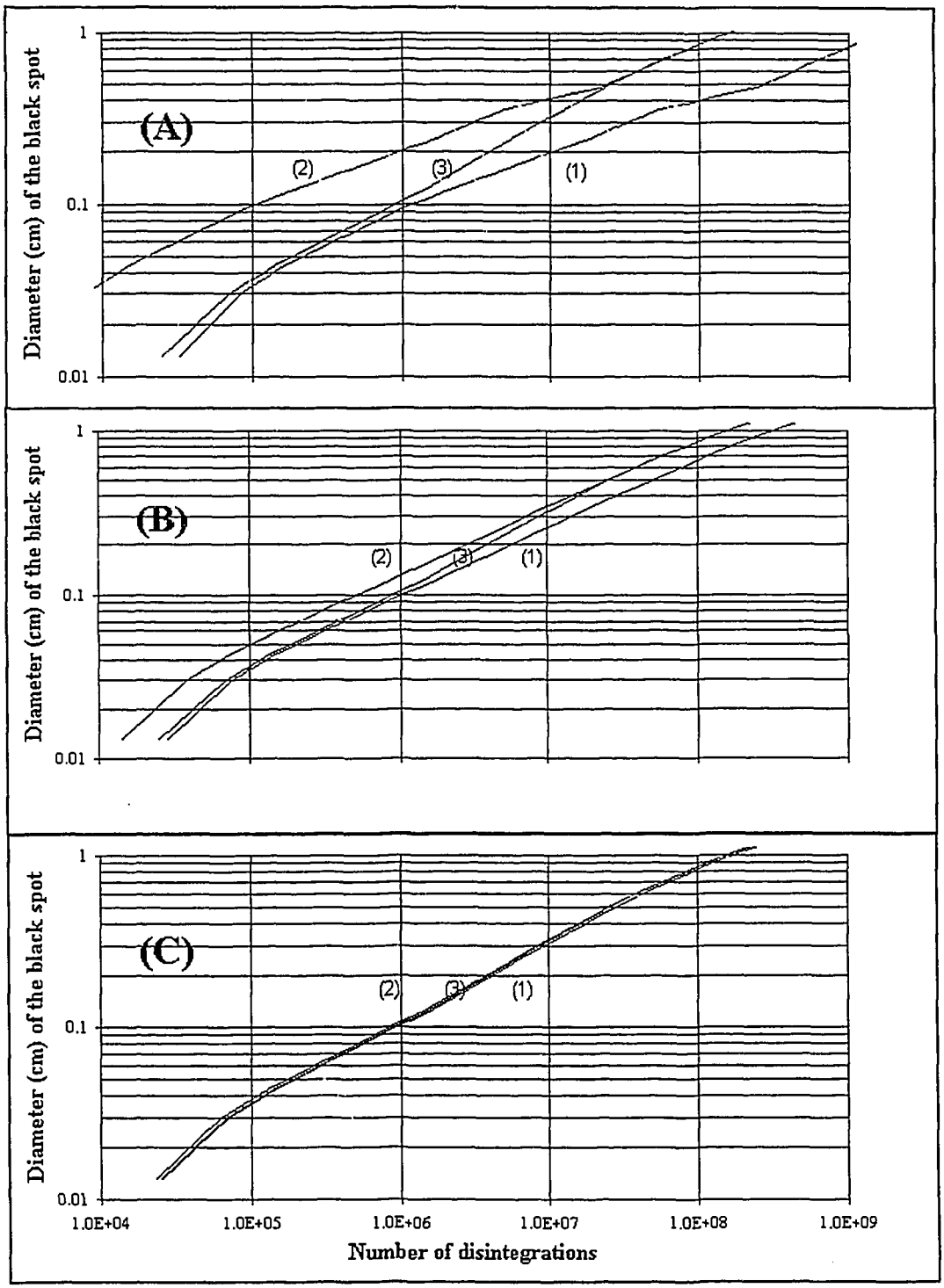

Figure 11. *Diameter of the black spot versus number of disintegrations. Nuclides in this hypothetical particle are ^{137}Cs and ^{144}Pr.* **(A)** *activity of ^{137}Cs is ten times higher than the activity of ^{144}Pr.* **(B)** *activity of ^{137}Cs and ^{144}Pr are equal.* **(C)** *activity of ^{144}Pr is ten times higher than the activity of ^{137}Cs. In curves (1) both ^{137}Cs and ^{144}Pr are included in the number of disintegrations whereas curves (3) include only the disintegrations of ^{144}Pr. In curves (2) ^{137}Cs is taken into account in the size estimation, not in the number of disintegrations.*

3.4.1 Estimation of particle activity from an unknown sample

Here it is assumed that the nuclide composition in a sample is unknown. Let us also assume that radioactive material is in a particulate form rather than distributed evenly in a sample. Localization of the radioactive particles is the main target of the autoradiographic analysis. However, some activity estimation of the particles can also be performed.

Several autoradiograms with different exposure times help to improve the quality of the results. If the measured spot diameters are compared to the calculated curves (c.f. Fig. 6) it is, in principle, possible to evaluate the nuclides and their activity in a particle. In practice this is impossible partly because of the tedious and slow procedures of exposure, film development and analysis.

However, even a single autoradiogram may be valuable. Activity of high-energy beta emitters may be estimated if the diameter of the black spot is larger than about 5 mm. In this case the activity must be estimated using only the number of disintegrations of the high-energy beta emitters (see Fig. 11). If another autoradiogram is produced for the same particle using an exposure time ten or hundred times longer, it might be possible to identify ('an educated guess') the nuclide with the highest beta energy. The same procedure may be useful also for smaller spots.

As an example, let us assume that we have a radioactive particle of unknown activity and that we have detected a black spot of 6 mm in diameter (solid horizontal line in Fig. 12, c.f. Fig. 6). The exposure time has been 10 h (= 36000 s). The horizontal axis is now the number of disintegrations divided by the exposure time, i.e. the activity. In this case the particle must contain high-energy beta emitters because the spot is large. Good candidates are ^{144}Pr, ^{90}Y, ^{89}Sr, e.g. If we assume that the particle contains mainly ^{144}Pr then the activity is close to 1 kBq. If the particle contains mainly ^{89}Sr then the activity is approximately 3 kBq. The activity of low- and medium-energy beta emitters remains unknown.

Additional information may be obtained using a longer exposure time. Let us assume that another autoradiogram with an exposure time of 100 h (~ 4 d) is produced for the same particle and let the diameter of the black spot now be 12 mm (solid horizonal line in Fig. 12). Now it is clear that ^{89}Sr cannot explain the detected size of the spot. The total activity of nuclides such as ^{144}Pr, ^{90}Y, ^{106}Rh is then close to 1 kBq.

Additional information may also be obtained using a shorter exposure time. The majority of the relevant beta active nuclides produce a black spot of a diameter of 0.8—1.0 mm when the number of disintegrations is 10^6 (note that Fig. 6 presents only a few beta emitters). When the diameter is close to 0.3 mm the number of disintegrations is 10^5. Thus, it is possible to evaluate the total activity of the particle, not only the activity of high-energy beta emitters. However, the activity of very low-energy beta emitters cannot be estimated in the presence of a mixture of nuclides. ^{95}Nb is such a nuclide ($E_{\beta,max}$ = 160 keV for the most probable decay branch). If, for example, the diameter of the black spot is 1 mm for an exposure time of 10 h, the total activity of the particle would be approximately 30—40 Bq (Fig.12).

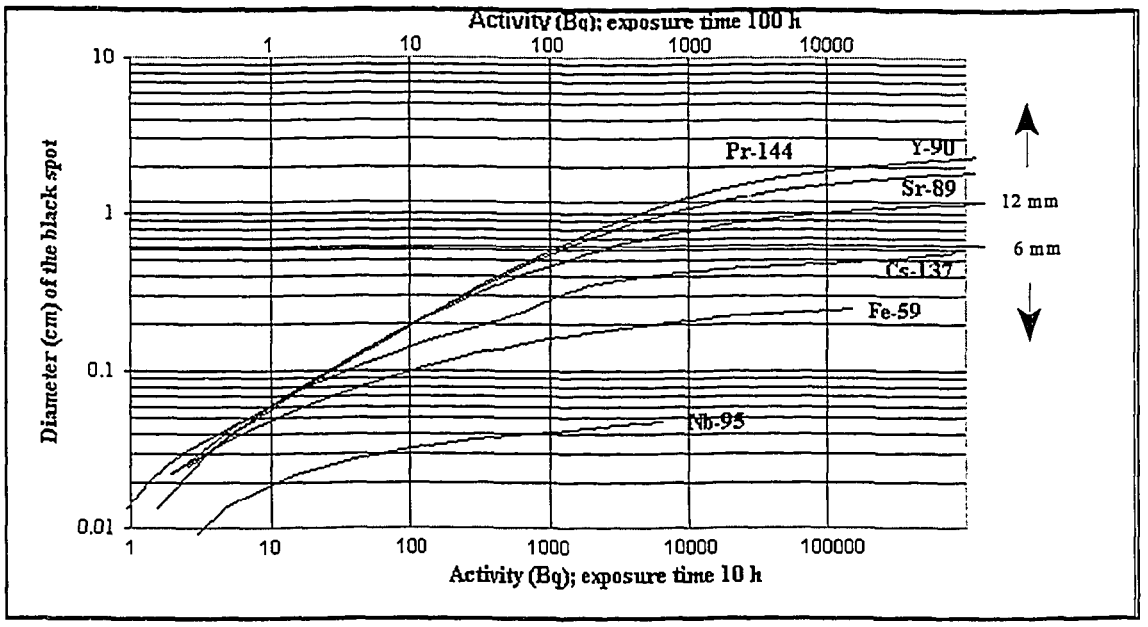

Figure 12. Graphical activity estimation from the diameter (6 mm and 12 mm) of the black spot. Exposure time is 10 h or 100 h. The activities are calculated separately for each nuclide. The curves represent cut-off dose of 10 mGy (depth 100 μm).

3.4.2 Sample purification

Sometimes the sample may consist of large amounts of non-interesting bulk material. A typical example is a swipe sample that contains inactive material and only a few radioactive particles. Alternatively, the sample may contain a large number of radioactive particles and, among them, a few radioactive or otherwise interesting particles. It may be useful to isolate the most interesting part of the sample to avoid disturbing effects in the possible subsequent analyses.

Autoradiography can be used, as was shown in the previous section, for the activity analysis of particles although the nuclide composition of the sample is unknown. Much more information about the sample and particles within it is available if the activity of the whole sample is measured beforehand using a gamma-ray spectrometer.

As an example, let us analyze the activity of single particles in an air filter (Fig. 13). The most relevant nuclides from the point of view of spot formation are ^{58}Co, ^{60}Co, ^{59}Fe, ^{95}Zr, ^{110m}Ag, ^{124}Sb, and ^{181}Hf (Table I). ^{51}Cr and ^{54}Mn cannot be fairly detected in autoradiography. ^{131}I is probably in small particles evenly distributed in a filter (grey background of the filter, Fig. 13).

The total activity of the filter (~0.5 kBq) was measured by a gamma-ray spectrometer. On the basis of Fig. 13 it is evident that the activity of the most radioactive particle is below a few tens of Bq's. The maximum number of disintegrations per particle must be considerably smaller than 10^8 because the exposure time was 10 days.

Let us first assume that the relevant nuclides are evenly distributed among the particles. The diameter of the black spot (1) in Fig. 13 is approximately 2.8 mm. Nuclides ^{58}Co, ^{60}Co, ^{59}Fe, ^{95}Zr, ^{110m}Ag and ^{181}Hf cannot produce a spot of this size (see Fig. 6). Thus, particle (1) must contain ^{124}Sb or non-gamma emitting high-energy beta emitters such as ^{90}Y. Figure 12 shows that spot diameter of 2.8 mm may be explained by assuming that the activity of ^{124}Sb is ~10 Bq (250 Bq must be divided by 24 because exposure time is now 24 times the

exposure time of 10 h used in Fig. 12). The estimated activity seems to be too high. Either pure beta emitters such as Sr-90/Y-90 are present in the particle or the image consists of several adjacent spots located less than 1 mm apart. The slightly irregular shape of the spot support this conclusion. Further analyses for this particle are needed (Table I).

The activities of medium- and low-energy beta emitters in particle (1) cannot be evaluated by autoradiography. The diameter of the black spot is not a sensitive function of the number of distintegrations. Activities cannot be determined accurately (perhaps within a factor of two).

When the size of the black spot is smaller than about 1 mm only total activity can be calculated. The diameter of the black spot (2) in Fig. 13 is 0.9 mm. Thus, it must contain medium-energy beta emitters such as ^{59}Fe. A significant amount of high-energy beta emitters cannot be present. The number of disintegrations of all nuclides is about $4*10^6$ (Fig. 6) which gives for the total beta activity of medium-energy beta emitters close to 5 Bq. The filter contains more than thirty particles with an activity above 5 Bq.

Diameter of spot (3) is 0.4 mm which gives for the total activity of medium-energy beta emitters 0.4 Bq. In the filter there are hundreds of particles with an activity higher than 0.4 Bq.

Particles (1), (2) and (3) were detached from the filter material and analyzed using a gamma-ray spectrometer (Table I). Nuclides are typical activation products. The estimated activities of particles (2) and (3) agree (by a factor of about 2—3) with these measurements. Nuclides in particle (1) cannot produce observed black spot. Further purification and analysis for this particle are still needed.

Table I. *Nuclide-specific activity (and its error estimate (%)) of an air filter. Particles 1, 2 and 3 were isolated from the filter using autoradiography (see Fig. 13). Measurements were performed by a gamma-ray spectrometer. Only beta emitting nuclides ^{58}Co, ^{60}Co, ^{59}Fe, ^{95}Zr, ^{110m}Ag and ^{181}Hf are included for the total beta activity.*

Nuclides	Activity of the air filter (Bq)	Activity of particle 1 (Bq)	Activity of particle 2 (Bq)	Activity of particle 3 (Bq)
^{51}Cr	77 4 %	- -	- -	- -
^{54}Mn	89 5 %	0.35 17 %	6.5 7 %	0.25 9 %
^{58}Co	160 4 %	0.27 21 %	5.7 8 %	0.28 9 %
^{59}Fe	90 2 %	- -	3.9 11 %	0.12 27 %
^{60}Co	53 3 %	0.1 37 %	2.5 9 %	0.10 13 %
^{95}Zr	30 3 %	22 3 %	- -	0.18 13 %
^{110m}Ag	11 3 %	- -	- -	- -
^{124}Sb	1.7 10 %	- -	- -	- -
^{131}I	47 2 %	- -	- -	- -
^{144}Ce	3.0 17 %	2.8 12 %	- -	- -
Total beta activity	350	25	12	0.7

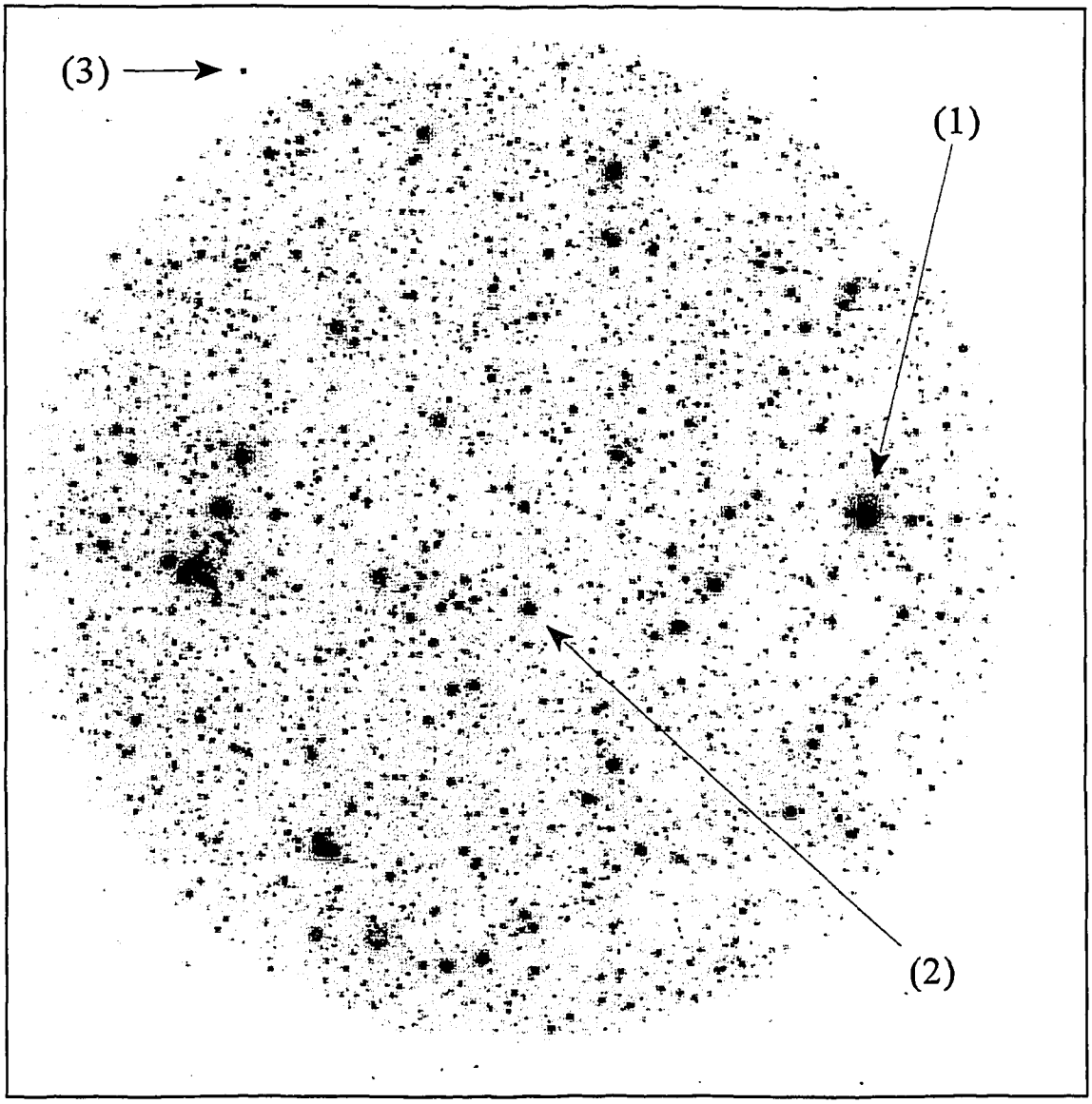

Figure 13. An autoradiogram of an a filter through which 25 m³ air has passed. The aerosol sample has been collected in a nuclear power plant. Film exposure time is 10 days. Total activity of the filter is close to 0.5 kBq (nuclide-specific activities are in Table I). Hundreds of particles with activity up to a few tens of Bq's were found in the filter. Particles (1—3) were used for more detailed analyses.

3.5 Detection limits

The detection limit of beta active materials depends on exposure geometry, type of autoradiography film and exposure time. Distribution of radioactive materials (particulate or evenly distributed materials) has a crucial effect on the detection limit. However, the type of beta active nuclide in a sample has only a minor influence on these limits in a fixed exposure geometry.

Let us suppose that the film type is the one used in this study and let the exposure geometry be as shown in Fig. 3. Let the particles be distributed in the sample such as the particles in Fig. 13. Test measurements showed that

(1) Dark spots cannot be identified on a film if the number of disintegrations in a particle is considerably below 50000. In terms of activity, a particle with activity of 1 Bq can be detected using an exposure time of 0.5 d. Then, rather a grey spot than a black spot appears on the film.

(2) More than 10^5 disintegrations are needed for a digital scanner to detect the spot as 'black'. The diameter of the black spot is then larger than 0.3 mm.

(3) Detection limits are lower if absorbing material is removed between the sensitive layer of the film and the particle. The limits may be considerably higher if a significant amount of absorbing material is present between the film and the particle. Especially low-energy beta emitters are difficult to detect.

(4) A light multiplying emulsion layer can be inserted above the film. The detection limit is then better than without it. However, the analysis of spot size as a function of the number of disintegrations is beyond the scope of the present study.

(5) If evenly distributed radioactive background material exists on the film, the detection limit for individual particles may be much higher.

(6) The simplest way to detect low active materials is to use long exposure times (10 d).

4 DETECTION OF ALPHA ACTIVE PARTICLES

The range of alpha particles in water is below 70 μm. Thus, the absorbing medium between the sensitive layer of the film and the radioactive material must be removed (see Fig. 3). Also self-absorption of alpha particles in the source material may greatly affect the detection limits. Quantitative activity analysis for large (tens of micrometers) alpha-active particles cannot be performed. However, qualitative analysis for surface activities is possible.

The detection limit (Bq cm^{-2}) of autoradiography was estimated using massless sample (surface area 4 cm^2) prepared initially for drinking water analysis (Salonen, 1993). The sample was analyzed using a low-background liquid scintillation spectrometer (QUANTULUS ™). The sample contains only isotopes of uranium and its daughters (Table II).

Two autoradiograms were performed. They showed that radioactive material was evenly distributed on the sample plate. Exposure time of one day could not produce a good image. Three days of exposure (more than 20000 α-disintegrations per square centimeter) are needed to produce an image that can be distinquished from the film background. However, the colour of the image was not 'black'.

Two additional massless samples were used to evaluate the number of disintegrations for a fully black spot. These samples are normally used for calibration of alpha spectrometers. Total activity (^{241}Am, ^{239}Pu, ^{244}Cm) of the samples per square centimeter was 1400 Bq and 14000 Bq, respectively. More than approximately 10^7 disintegrations per square centimeter are needed for a black spot.

The limits mentioned above may change considerably if the samples are not massless. Alpha particles that enter the film come mainly from the part of the sample which is closest to the film. In addition, it may be difficult to exclude the presence of beta active nuclides in the sample. However, the effects of alpha particles can be removed by adding absorbing material between the film and the source (as shown in Fig. 3).

Table II. *α-activity of uranium isotopes in the massless sample. Total alpha activity is 0.1 Bq cm^{-2}. The daughters of ^{238}U (^{234}U, ^{234}Pa) are in equilibrium. Their total activity is 0.045 Bq cm^{-2}. Because of their low activity, as compared to alpha active nuclides, they do not influence spot formation markedly.*

Nuclide	Surface activity [Bq cm^{-2}]
^{232}U	0.038
^{234}U	0.038
^{238}U	0.025

5 IAEA TEST SAMPLES

IAEA gave four swipe samples for test analyses in autoradiography (Table III). A simple beta counting was performed first to get an idea of the level of activity in the samples. The nuclide contents in each sample were obtained with gammaspectrometric analysis. Then, autoradiography was used. Finally, some of the identified particles were used in further analysis.

5.1 Gamma spectrometric analysis

The samples were counted in Williams measurement geometry without any special preparation. Therefore, the quantitative results may be slightly inaccurate (error estimation is not performed). Only sample 8856-02-24 contained species of post-irradiation origin (Table IV).

5.2 Autoradiography

The samples were attached to a piece of paper to make sure that they do not move during the exposure and subsequent handling. Samples 8856-04-04 and 8856-02-24 contained a lot of loose material which was fixed with spray-on-laquor. Three autoradiograms were produced (two samples at the time):

(1) Samples 8856-04-04 and 8856-02-24. Exposure time 52 h 37 min.

(2) Samples 8856-04-04 and 8856-02-24. Exposure time 21 h 09 min.

(3) Samples 8856-01-03 and 8856-07-03. Exposure time 143 h 53 min.

Both samples in autoradiograms (1) and (2) produced clear images (Fig. 14). Sample 8856-01-03 in autoradiogram (3) revealed no beta active material. Only a small amount of beta active materials was found in sample 8856-07-03.

Radioactive material in samples 8856-04-04 and 8856-07-03 is almost evenly distributed. Particles in sample 8856-04-04 broke down easily in a process to detach them from the carrier matrix. They were more like clusters of the sample material rather than single radioactive particles. Sample 8856-02-24, however, contained single beta active particles. They were small and surrounded by inactive sample material. Thus it is impossible to detect them by eye. Some of these particles were detached for further analyses (see Fig. 14).

Table III. IAEA test samples for autoradiography.

Number	IAEA code	IAEA Remarks
1	8856-01-03	Cloth swipe from an office area. Contains traces of low-enriched U.
2	8856-02-24	Cloth swipe from a facility for post-irradiation examination of fuel. Contains fission and activation products (^{60}Co and ^{137}Cs).
3	8856-04-04	Cloth swipe from a fuel-fabrication facility. Contains up to several milligrams of natural U.
4	8856-07-03	Cloth swipe from nuclear material storage facility. Contains traces of natural U and picogram amounts of Pu.

Table IV. Gammaspectrometric results of IAEA swipe samples (Bq per sample). Note that the daughters of ^{238}U (^{234}Th and ^{234m}Pa) emit high energy betas. In equilibrium their activity is close to the activity of the parent nuclide. The daughter of ^{235}U (^{231}Th) emits low energy betas.

	8856-01-03	8856-02-24	8856-04-04	8856-07-03
^{238}U	-	-	400 Bq	20 Bq
^{235}U	0.05 Bq	-	20 Bq	0.7 Bq
^{60}Co	-	4 Bq	-	-
^{134}Cs	-	1.2 Bq	-	-
^{137}Cs	-	23 Bq	-	-
^{125}Sb	-	0.1 Bq (?)	-	-
^{241}Am	-	0.2 Bq (?)	-	-
^{154}Eu	-	0.3 Bq	-	-
^{144}Ce	-	0.3 Bq	-	-

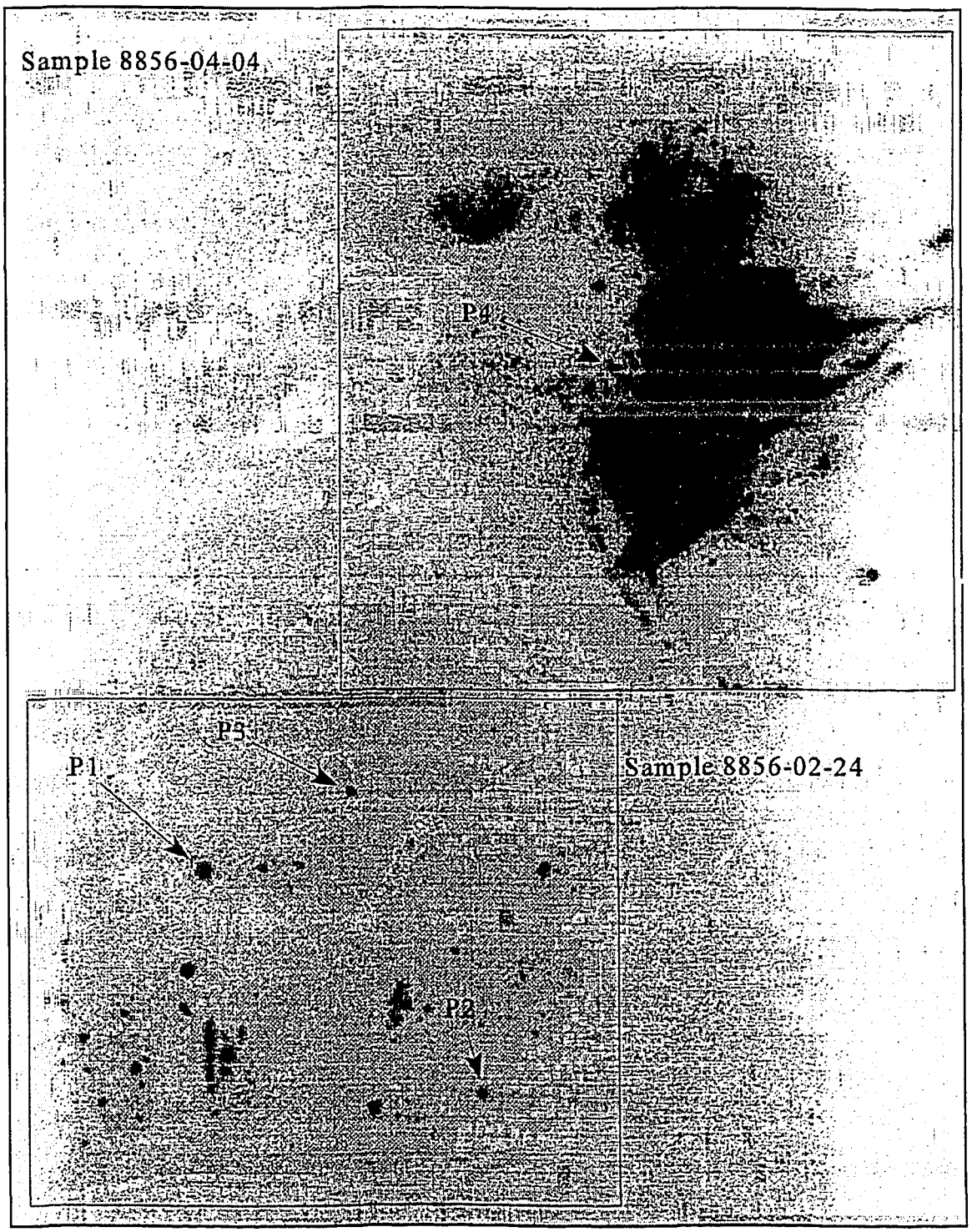

Figure 14. Scanned image of an autoradiogram (1). Particles P1—P4 were detached for further analyses.

5.3 Analysis of detached particles

Four detached particles were analyzed in gamma-ray spectrometry (Table V). The activities of particles P1 and P4 are 1.5 Bq and 5 Bq, respectively. These particles were not further analyzed although the sizes of the black spots do not have positive correlation with the activities.

The activity of particle P3, measured by gamma-ray spectrometer, is nearly ten times larger than the activity of particle P2. However, the autoradiograms show that their activities should be almost equal because the spot sizes are equal. The difference may be explained by assuming that particle P2 contains also pure beta emitters.

Particles P2 and P3 were analyzed using a liquid scintillation counter (Salonen, 1993). The beta spectrum from particle P2 shows the presence of $^{90}Sr/^{90}Y$ (Fig. 15). The beta spectrum from particle P3 shows also the presence of pure beta emitters (Fig. 16). According to the beta spectra the total activities of particles P2 and P3 are equal (Table V).

Alpha spectum of particle P3 shows small amounts of ^{241}Am (0.013 Bq). This is also supported by the amount of low energy betas (Fig. 16). Liquid scintillation counting is sensitive for ^{241}Am. The detection limit is lower than the detection limit in a long gamma spectrometric analysis.

Table V. Activities of particles P1—P4 and their error estimates (%) measured with a gamma-ray spectrometer and scintillation counter (particles P2 and P3).

Particle	Nuclide	Activity (γ-spectrometer)		Total β-activity (β-spectrometer)	
P1	^{137}Cs	1.5	5 %	-	
	^{134}Cs	0.04	20 %		
P2	^{137}Cs	0.03	27 %	0.58	5 %
P3	^{137}Cs	0.2	15 %	0.62	5 %
P4	$^{234}Th/^{234m}Pa$	5.4	30 %	-	
	$^{234}U/^{231}Th$	0.03	30 %		

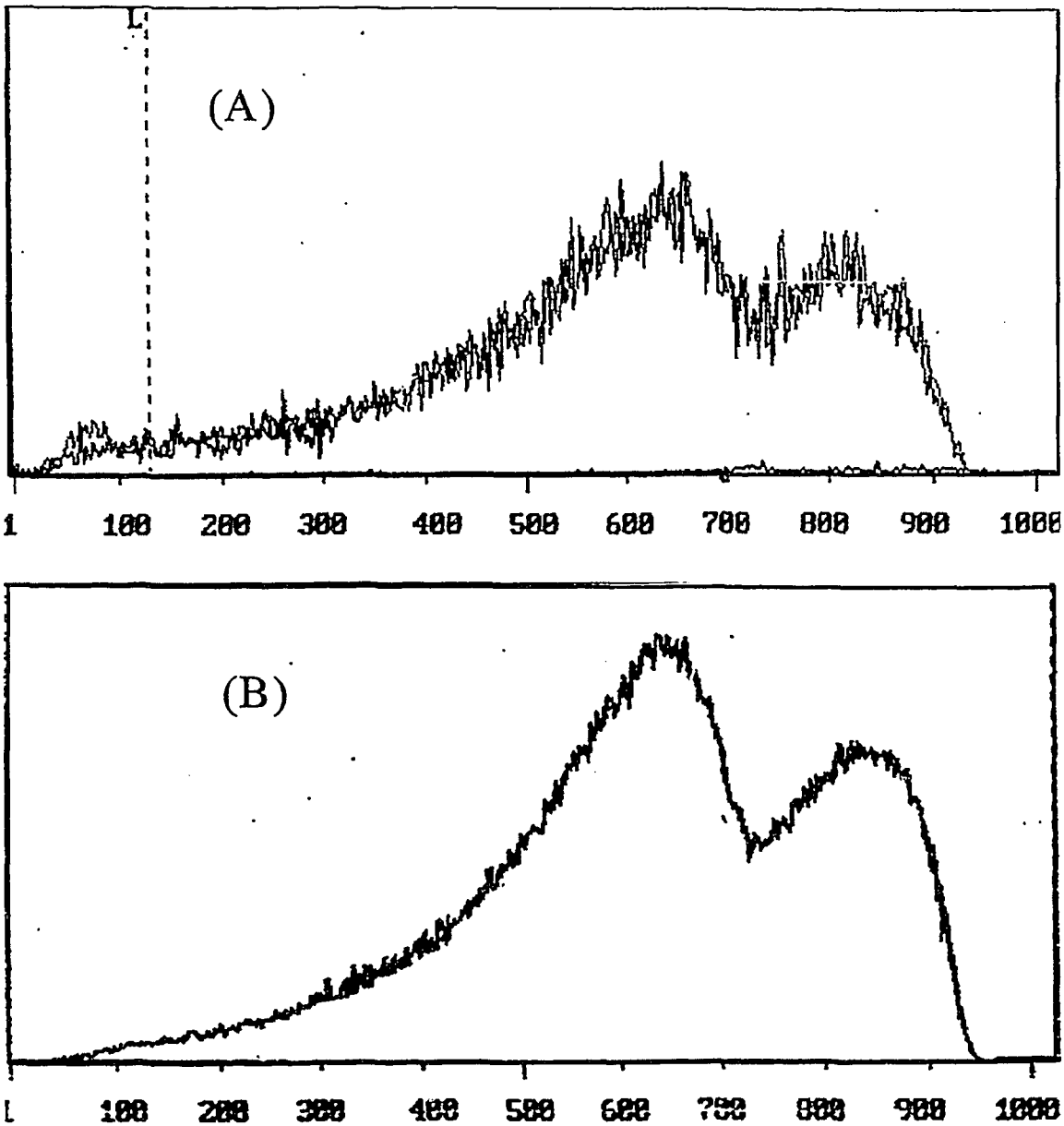

Figure 15. (A) *Beta spectrum of particle P2 and* (B) *reference beta spectrum of pure* $^{90}Sr/^{90}Y$ *(Laina Salonen, STUK).*

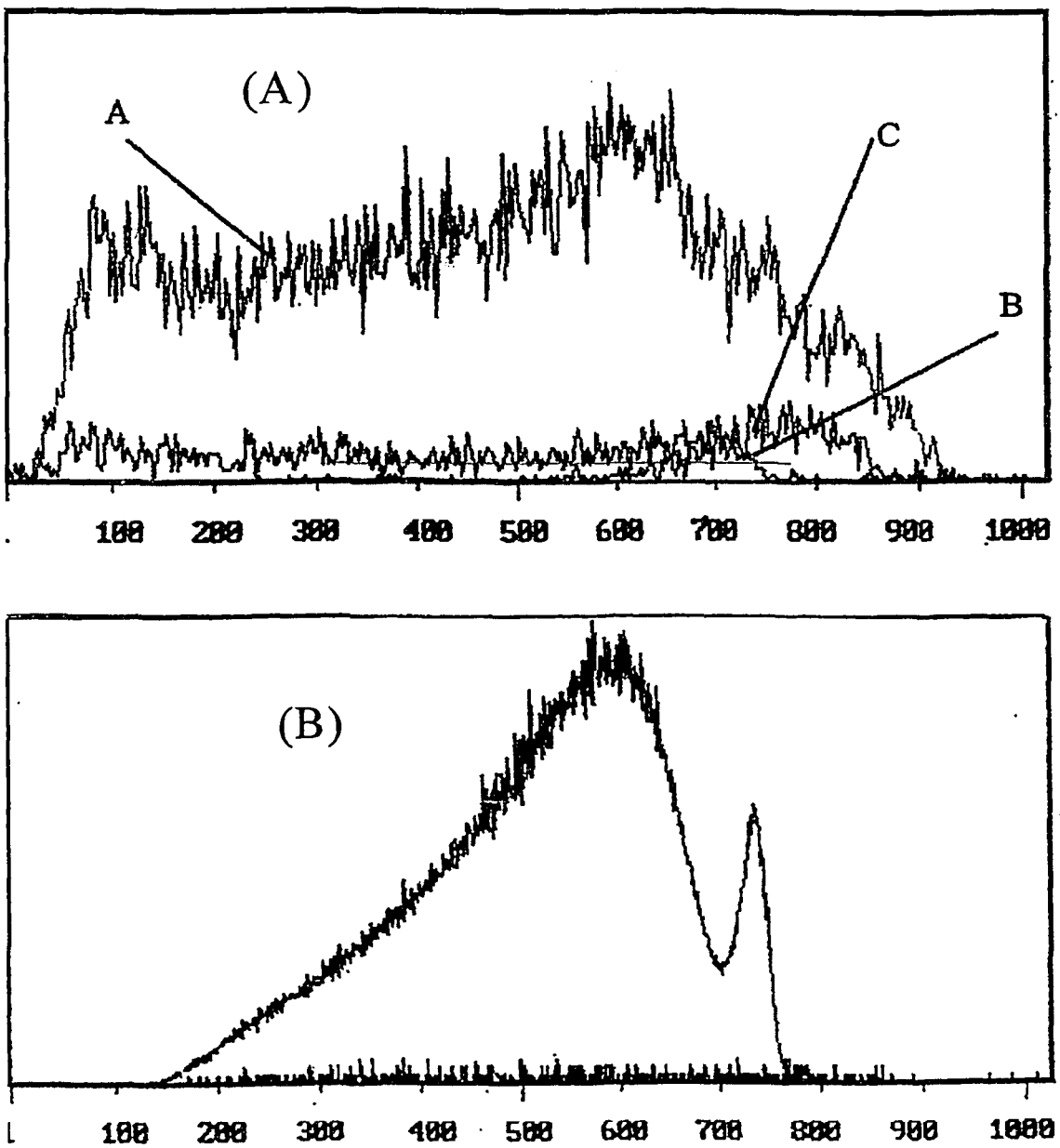

Figure 16. (A) *Beta and alpha spectra of particle P3. A = beta spectrum, B = alpha spectrum, C = beta background.* (B) *Reference beta spectrum of* ^{137}Cs *(Laina Salonen, STUK).*

6 AUTORADIOGRAPHY IN THE FIELD

Autoradiography is widely used in laboratory conditions for the environmental monitoring of radioactive particles. So far, its suitability for in-field conditions has been limited. Although the amount of chemicals and water needed in the film processing is small, their transportation and use in the field may be difficult. A darkroom and water must be available. However, sample preparation and exposure of the films can be easily performed.

A portable autoradiography unit was constructed (Fig. 17). It contains, among other things, chemicals and equipment presented in Figs. 2 and 4.

Let us assume that a set of swipe samples has to be examined in the field using autoradiography. The procedure is as follows:

(1) Particles in a sample have to be fixed using spray-on laquor or adhesive film (Fig. 3) to avoid contamination of the exposure cassettes.

(2) The sample and the autoradiography film are put in a lightproof cassette (a dark room is needed). Several samples can be used at the same time if the area of the swipe samples is considerably smaller than the area of the autoradiography film. This is recommended because sampling is then more representative. The exposure time must be recorded for the activity analysis of particles. Location of the samples and the film must be fixed if the particles are detached from the sample for further analyses.

(3) The chemicals of development bath, stop bath and fixing bath are diluted by water and substituted to the developing rack. Rinsing rack contains water.

(4) The procedure of film development (section 2.2) must be performed in a darkroom.

Activity analysis of particles cannot be easily performed without a computer. Long exposure times (needed for samples containing low amounts of radioactive material) can be used if phases 1 and 2 are performed in the field and the other phases in a laboratory.

In the field conditions it is possible to get the answers to the following questions, provided that the film development process can be performed:

(1) Does radioactive material exist in the sample? Is the radioactive material evenly distributed in the sample or located in the particles?

(2) Does expected radioactive material exist in the sample? For example, radioactive materials found in fuel-fabrication facilities are different from those found in spent fuel storage facilities.

(3) Do other than expected radioactive materials exist in the sample? For example, if a swipe sample from the fuel fabrication facility contains high-energy beta emitters (i.e., large black spots in autoradiography) it is a signal of post-irradiation material. Further examination in a laboratory is then definitely needed for this sample.

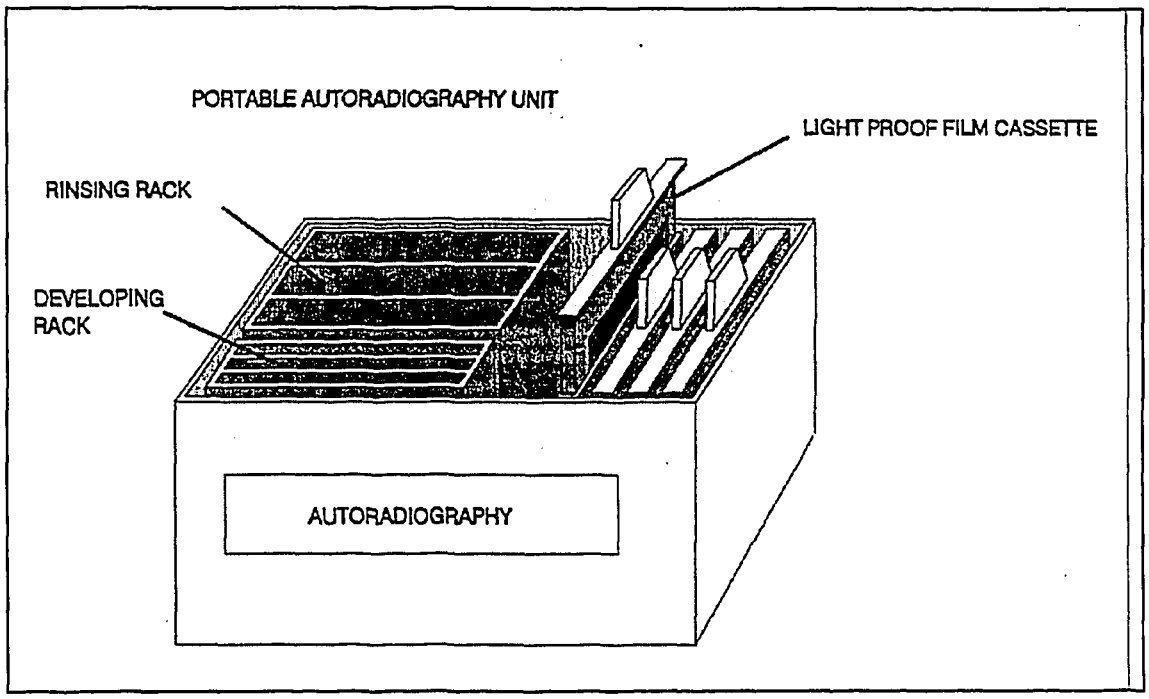

Figure 17. *A portable autoradiography unit. All equipments and chemicals (excluding water) needed for in-field autoradiography are within the unit. Weight of the system is below 20 kg.*

7 CONCLUSIONS

(1) Autoradiography is a simple method that can be easily used to detect radioactive materials and to localize beta active particles. Activity of the particles may be below 0.1 Bq provided that the exposure time is several days.

(2) Sampling and film exposure can be easily performed in the field. Film processing and data analysis can also be performed in the field. However, there is no commercial product available. The equipment used in the present study are not intended for routine use.

(3) Radioactive particles can be easily detached from the sample. This can be performed also in the field. Detached particles as such are appropriate for gamma spectrometric or other analyses. This minimizes the amount of non-interesting bulk material that may considerably affect the accuracy of these subsequent methods.

(4) Autoradiography can be used for pre-screening of samples. Further examination should be performed in a laboratory if other than tracer nuclides are suspected.

(5) For the exposure geometry and film type used in this study, more than 50000 beta emissions per particle are needed before a dark spot can be identified on the film. For example, a particle with an activity of 0.1 Bq can be detected if the exposure time is 5 days.

(6) A digital desktop scanner and suitable data analysis software enable to estimate the activity of single particles. More than 10^5 disintegrations are needed before a spot can be identified as 'black'. The diameter of the black spot is then larger than about 0.3 mm.

(7) Pure beta emitters, such as $^{90}Sr/^{90}Y$ that cannot be identified in gamma-ray spectrometry, can be detected in autoradiography.

(8) In autoradiography nuclide identification and, thus, activity estimation is possible only in certain situations. However, the nuclides can be grouped into high-, medium- and low-energy beta emitters for activity estimation. The total activity of beta active nuclides can be evaluated if the number of disintegrations is smaller than 10^6.

(9) Detection of alpha active nuclides is possible if no absorbing material exists between the sensitive layer of the film and the sample. The size of a black spot caused by a pure alpha active particle is then smaller than 0.1 mm^2. More than 20000 α-disintegrations per square centimeter are needed to produce an image that can be distinguished from the film backround. In practice, beta particles may mask the effects of alpha particles.

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APPENDIX 1

IMAGE ANALYSIS OF AUTORADIOGRAMS

An image analysis code, known as SPOT, for autoradiography was developed using Visual Basic application development system for Windows. The code is intended to analyze images created by a digital scanner. Before analysis, a single spot on an autoradiography film is scanned (300 dpi) and stored in a directory specified by the user. To avoid long loading times and to save disk space the bitmap file (*.bmp) should be as small as possible, typically smaller than a few hundred kilobytes (or in terms of area, smaller than a few square centimetres). Each image is composed of square-shaped pixels that have a colour index (shade of grey) between 0 to 255. The cut-off colour index that represents the colour 'black' determines the size of the black spot and, consequently, enables activity analysis of the radioactive particle.

The main window of the code is shown in Fig. A1. The specified image must be opened before the analyses.The image appears below the Open image button.

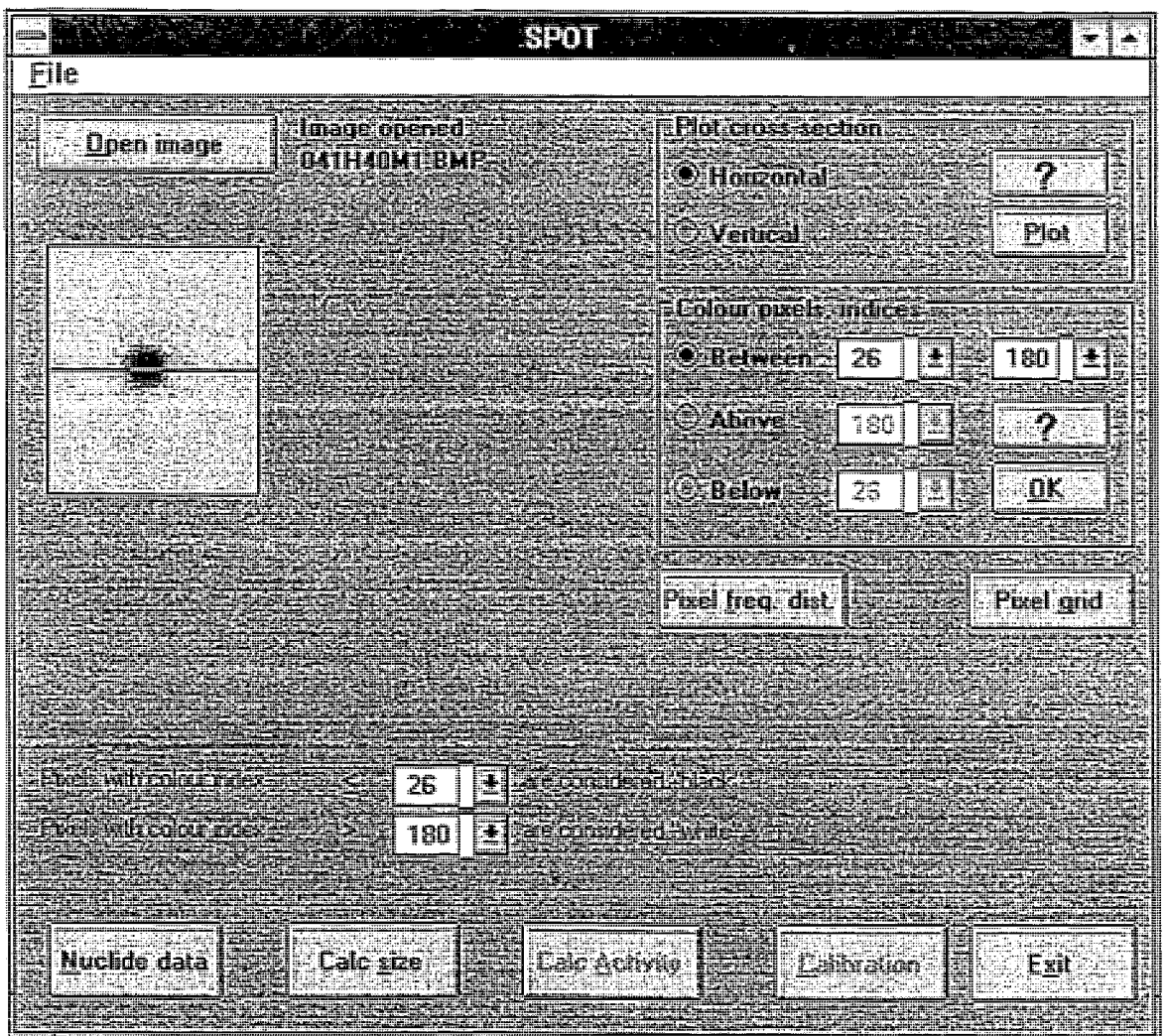


Figure A1. Main window of the SPOT code. The upper part of the window contains the tools for image analysis (plot and colour pixels) whereas the buttons in the lower part can be used for size and activity analysis and calibration. A data base of beta active nuclides has been constructed to facilitate dose rate calculations and other analyses.

A **Horizontal** or alternatively a **Vertical** cross-section of the spot can be plotted (c.f. Fig. 9). A cross section of the spot gives a possibility to estimate an appropriate cut-off colour index for the colour 'black'. **Colour pixels**-tool can be used as an aid for estimating the cut-off index. Pixels above or below a specified colour index (or between a specified range) are then coloured red. The frequency distribution of pixels can be plotted using the Pixel freq. dist. button. The colour index of each pixel can be seen in a table format using the Pixel grid button. The user specifies the colour indices that represent 'black' and 'white' ('white' is not used in the present code). They are NOT automatically selected in the present version of the code.

Nuclide data enables to plot figures such as Fig. 5 and Fig. 6, e.g. The database includes the nuclide-specific beta dose rate at different distances from a point source of unit activity in an infinite water medium (uppermost selection in Fig. A2). The other two selections are 1) exposure time needed to produce a dose D in the sensitive layer of the film (the distance between the film and the particle is d mm) as a function of radial distance and 2) diameter of the black spot (the edge of the black spot receives D mGy) vs. number of disintegrations. These quantities can be calculated either for several nuclides or for a single nuclide which make sensitivity analyses possible. Then, for example, diameter vs. number of disintegrations can be plotted for different doses D (see Fig. 10B).

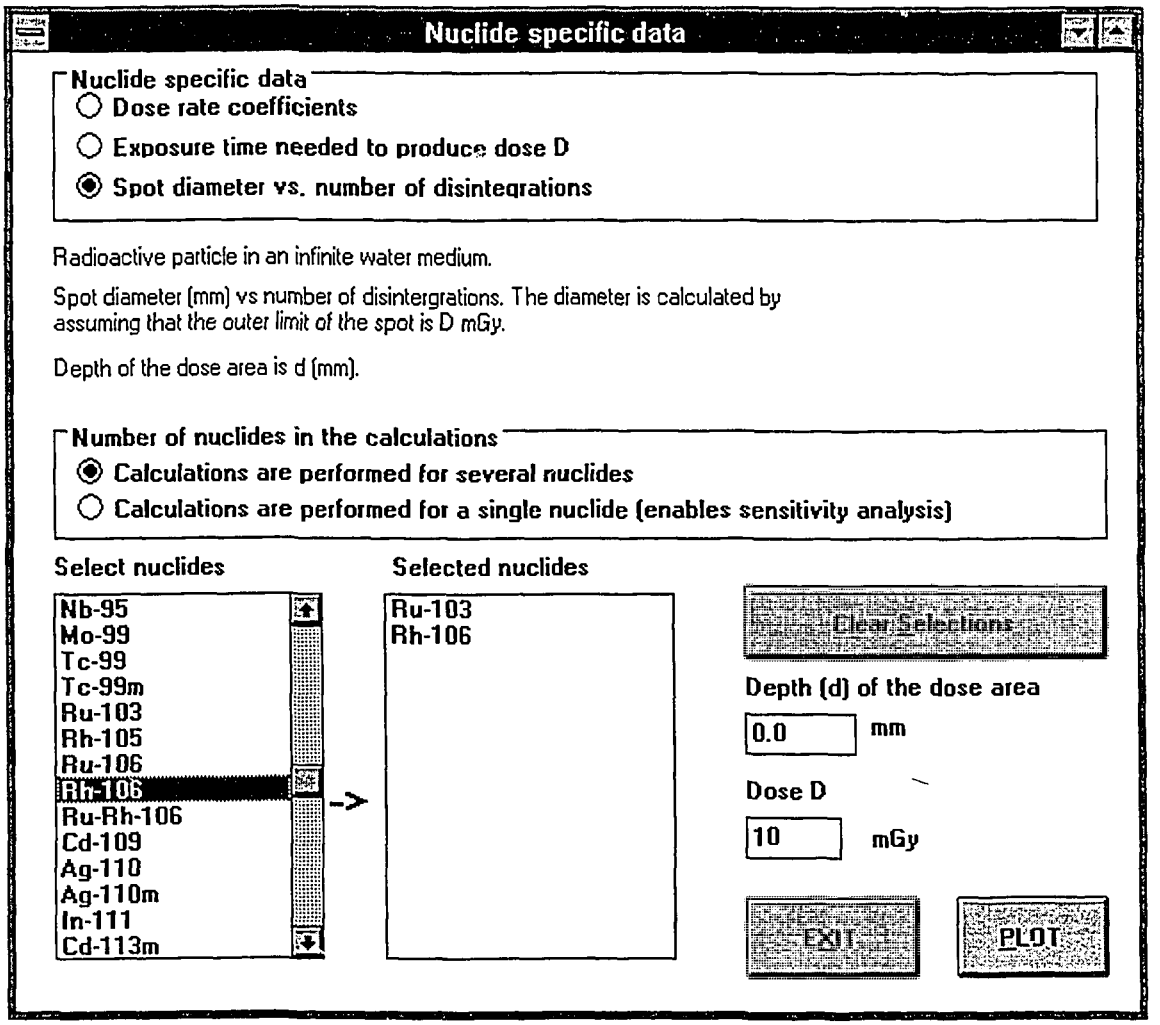
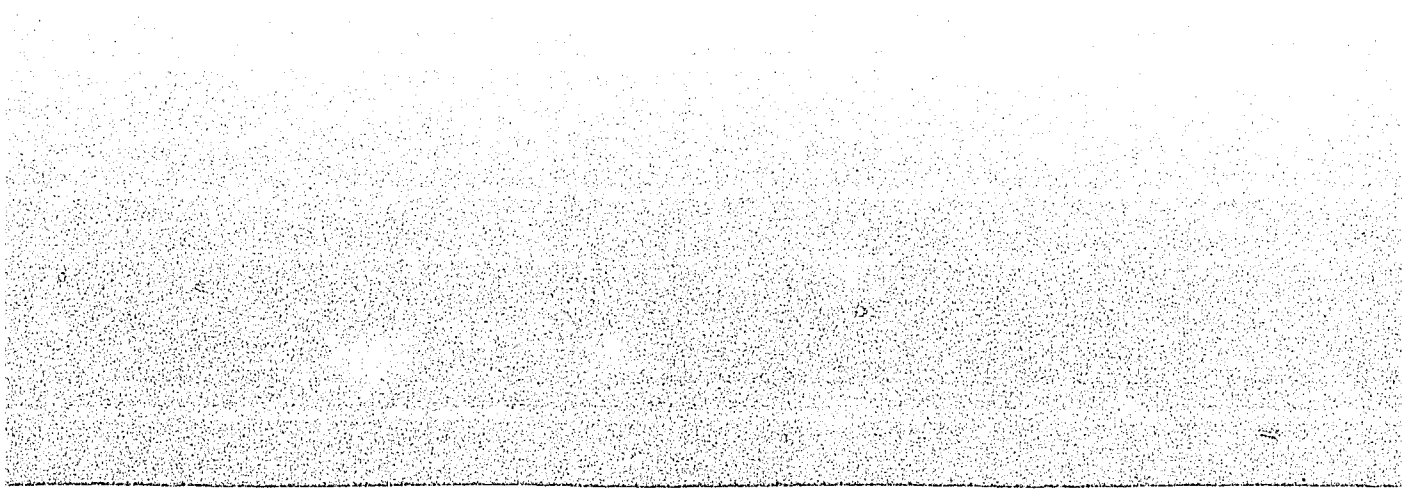


Figure A2. Plotting nuclide-specific data (spot size vs. number of disintegrations) for ^{103}Ru and ^{106}Rh.



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