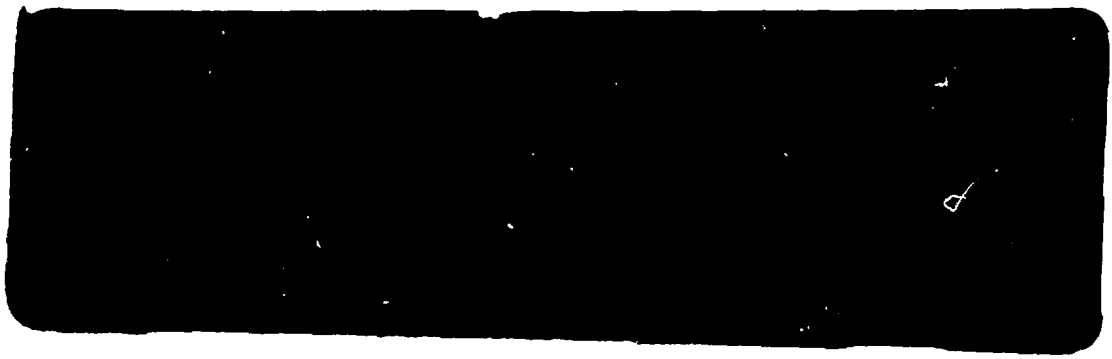

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# TRACK DETECTION METHODS OF RADIUM MEASUREMENTS*

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CONTENTS



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## 1. INTRODUCTION

Nowadays most of the measurements related to highly ionizing nuclei and based on the use of solid state track recorders may be considered as well-established ones. This type of nuclear detectors provide several advantages in wide-ranging fields of applications which may be attributed in part to their simplicity in handling and processing and to their insensitivity to beta- and gamma-rays. In addition to this attractive behaviour the wide region of their sensitivity is receiving great attention. Certain plastics are sensitive enough to permit the visualization of the tracks of alpha-particles emerging from both natural and man-made alpha-emitting sources . Therefore these are suitable for either direct or indirect measurements of the radium isotopes as well. In the following sections we will introduce the reader into the main principles and possible methods of such measurements with special emphasis on the recent developments in this field. We would like to emphasize, however, that up to now very few works have been reported on this topics and further studies are highly required to reveal all the essential details of such type of applications. Therefore the author's present survey should be considered as a first attempt in this direction which can't be complete.

In natural and man-made samples the radium occures either in equilibrium or in disequilibrium in the radioactive decay series. In most of the routine works it is convenient to apply the track method either to complete radioactive equilibrium or to complete disequilibrium. For both cases the technique of etch-track detectors is well-applicable. In doubtful "transition cases" one should apply additional nuclear or radiochemical methods to justify the assumed pattern of radio-equilibrium.

Another problem which sould also be considered here is the medium of the occurrence of radium. It is obvious that the method of monitoring alpha-activity concentrations depends on whether the medium of measurement is solid, liquid or gas. Most of the solid state nuclear track detectors are relatively insensitive to environmental effects during exposure but their registration efficiency may vary considerably on the phase and thickness of the sample and on the geometry of measurement. Therefore in the following we will devote some words to these aspects too.

## 2. TRACK PROCESSING

Ionizing nuclear particles can create latent tracks in the form of narrow cylindrical regions of intense radiation damage in most insulating solids. To our present knowledge such tracks may be revealed by four fundamental methods called chemical etching (Fleischer et al.,1975), electrochemical etching (Tommasino et al., 1981a, 1981b), track dyeing (Somogyi et al., 1979) and precipitation (Wendnagel et al.,1980).The quality of information gained from nuclear tracks depends on the way of track processing applied. To make the latent tracks visible, countable

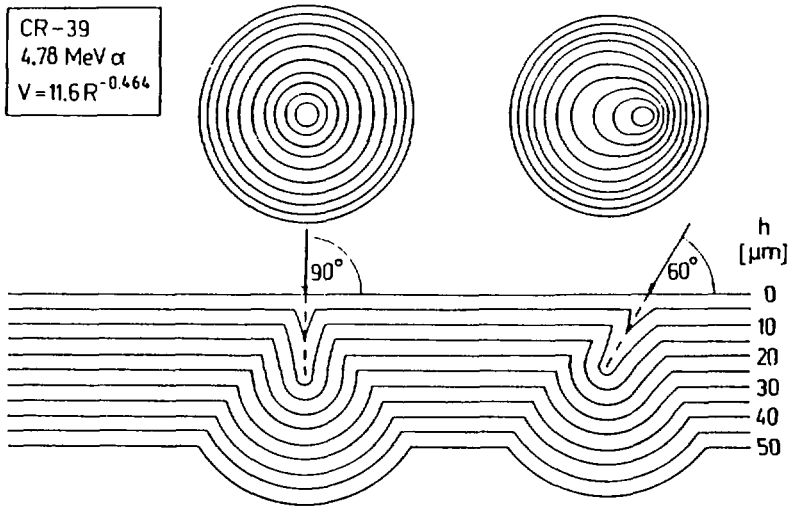


Fig.1. Illustration of the development of track contour and profile during the etching process for Ra-226 alpha-particles, entering the surface of a CR-39 type track detector at two different angles. The curves are calculated from the theory of etch-track formation (Somogyi,1980) using the indicated, empirically determined sensitivity function, V(R). Here and in all the figures shown in section 3.2.7 the range (R) is measured in μm.

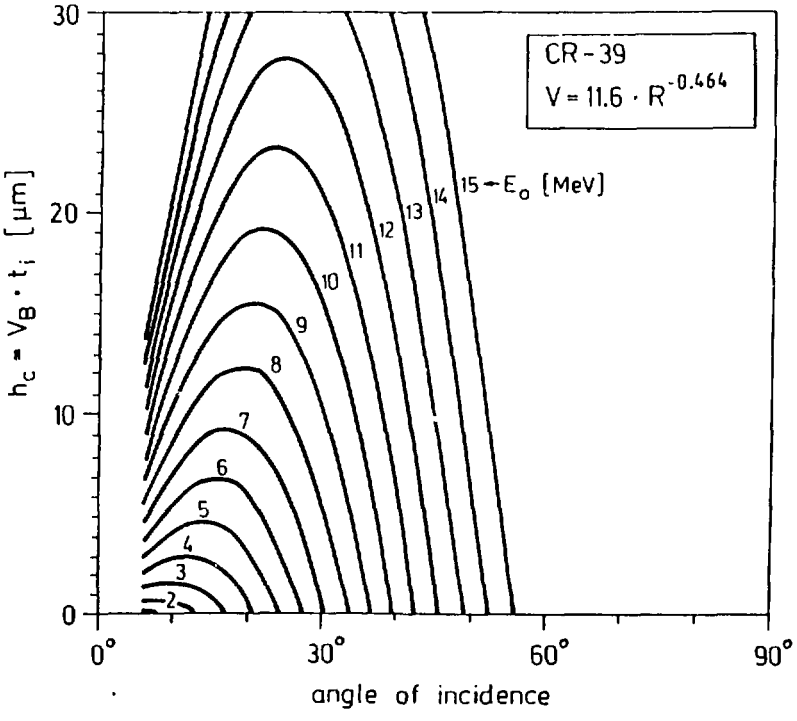


Fig.2. Illustration of the theoretically expected variation of the critical layer thickness removal, $h_c = V_B t_i$, on the incident angle and energy of alpha-particles in a CR-39 type track detector where $t_i$ indicates the incubation time of tracks prior to their appearance in etch-pit form during etching.

and measurable under optical microscope the most general and developed method is the preferential chemical etching along the damage trails. As this method seems to be the most useful, efficient and economical in both direct and indirect radium measurements, in the following this will be discussed in detail. In order to understand the proper use of etch-track method in radium measurements, one should have some pieces of information on the main rules governing the growth of etch-pits of alpha-particles entering the plastic track recorders, the essential steps of the track etching procedures and the registration characteristics of plastics most useful for alpha-activity measurements. In the next subsections we attempt to summarize the most essential information on these questions.

## 2.1. Etch-track formation in plastics

At present, the main principles describing the development of etch-tracks in plastic track detectors are well-known. By the theory of etch-track evolution one can calculate all the practically important parameters and trends of track growth during the etching process (Somogyi, 1980). One can predict for example the length, minor and major axes, contour and profile of etch-pits at arbitrary angles of incidence for any nuclear particles in a given plastic track detector as a function of the duration of etching. To accomplish such a task the knowledge of the variation of the so-called etch-rate ration, $V=V_T/V_B$, along the damage trail is required, the finding of which is still an empirical question. Here $V_T$ represents the rate of preferential chemical etching along the central track region and $V_B$ measures the dissolution rate of the undamaged bulk material of the detector attacked by the chemical etchant.

Restricting ourself only to the problem of alpha-track detection one can successfully use simple mathematical formulas to describe the etch-rate ratio (V) versus residual range (R) curves which indicate the etching sensitivity of plastics to nuclear particles. In case of the plastic track detectors suitable for natural alpha-activity measurements one may apply the relations $V=1+\exp(-AR+B)$ or $V=AR^{-B}$ for practical calculations, where A and B are fitting parameters. The first form is proposed for cellulose nitrate (e.g. LR-115 and CN-85 detectors produced by the Kodak-Pathé in France) and polycarbonate sheets (e.g. Makrofol-E, Bayer AG product). The second form of equation seems to to be applicable to CR-39 sheets (e.g. the Pershore, U.K., and the MA-ND, Hungary, track detectors).

To illustrate the typical phases of etch-track evolution and the practical problem we may meet in recognizing etched track pits, we have calculated the track profiles and contours characteristic of 4.78 MeV alphas emerging from Ra-226 and entering the surface of the presently known most sensitive track detector, CR-39, at two different angles. Fig.1 shows the growth of etch-pit contour and profile at different stages of the etching process.

One can see from Fig.1 that the users of alpha-sensitive plastics may meet complex geometrical forms of the tracks depending on the etching duration. In general one can differentiate the "incubation", "cone", "transition" and "sphere" phases of track

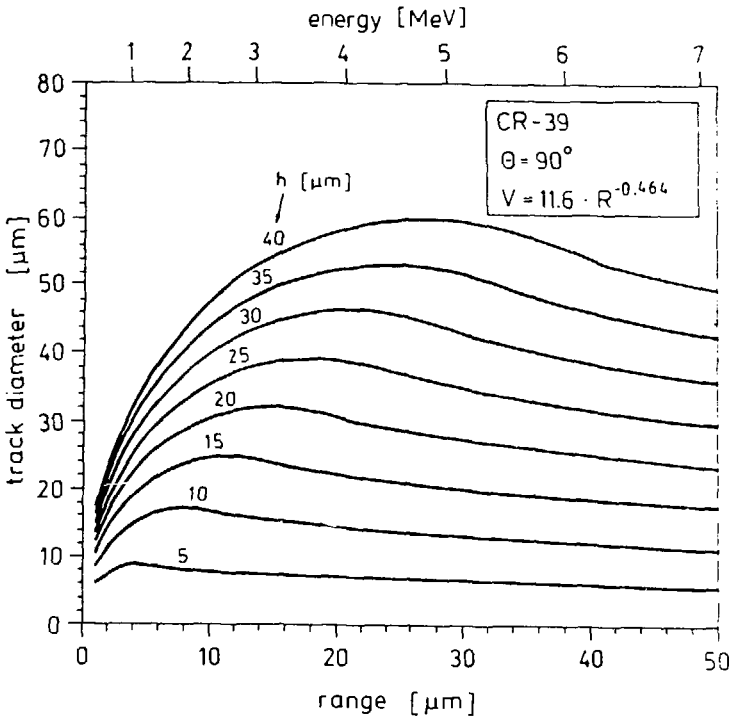


Fig.3. Curves calculated from the etch-track kinetics to illustrate the dependence of track size on the range (and energy) of alpha-particles in a CR-39 type track detector at different etching periods. The curves show that in a highly sensitive detector like CR-39 the track size is fairly insensitive for the variation of particle energy.

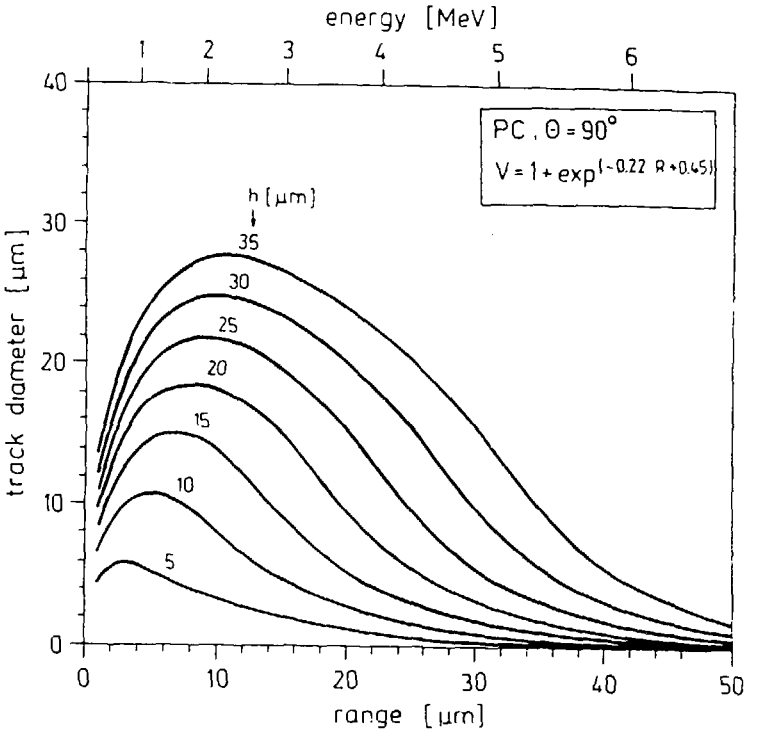


Fig.4. Curves calculated from the etch-track kinetics to illustrate the dependence of track size on the range (and energy) of alpha-particles in a PC type track detector at different etching periods. The curves show that in a lowly sensitive track detector like PC the track size is very sensitive to the variation of particle energy.

evolution. The knowledge of the dependence of the "incubation time" of tracks, $t_i$, (or the equivalent quantity $h_c$-$V_B t_i$, called critical layer thickness removal) on the particle parameters is of high practical importance as one can get visible tracks only after using longer etching times than $t_i$-$h_c/V_B$. The actual value of $h_c$ can be calculated from the solution of equation $V(R)\sin\theta=1$ after substituting the quantity $R$-$R_0$-($h_c/\sin\theta$), where $R_0$ is the "starting range" of a nuclear particle of $E_0$ energy, entering the track detector at $\theta$ angle to its surface. In Fig.2 we demonstrate the $h_c(\theta,E_0)$ curves calculated for alphas registered in CR-39 type nuclear track detector. The sudden delay in etch-pit appearance at given incident angles is a remarkable property indicated by these curves. Due to this, at exposures with broad-angle alpha beams, one meets a "track cut-off" effect which reduces the registration efficiency. With increasing etching time the "cut-off angle" (measured to the detector surface) decreases and one can get higher efficiency. This special behaviour of plastic track recorders should be taken into consideration in practice and an efficiency calibration has to be carried out as a function of the duration of etching.

Before performing a measurement it is also advisable to calculate expectable track sizesas a function of the etching time, t, (or the layer thickness removal, h=$V_B$t) by using the theory of track-etch kinetics (Somogyi, 1980). To select the proper length of etching period for a wider alpha-energy spectrum it is useful to calculate the track diameter at normal angle of incidence as a function of the range (or energy) of alphas at fixed layer thickness removals. The results of such a culculation are shown in Figs. 3 and 4 for the most and least sensitive detectors (CR-39 and PC) among plastics available for alpha-activity measurements.

## 2.2. Detector materials and etching procedures for alpha-tracks

The damage trails of heavier ionizing nuclei can be easily etched out and made visible under an optical microscope in a variety of insulating solids (minerals, glasses, plastics). The latent tracks of the lightly ionizing alpha-particles, however, are etchable only in certain organic polymers and only few of them are of practical importance. In this chapter we restrict ourself only to a brief description of the main characteristics of those detector materials and etching procedures which at present may be considered as the most advantageous ones for alpha-track registration. As concerns other alternatives and details we refer to more comprehensive communications (e.g. Fleischer et al.,1975; Monnin, 1980; Fischer and Spohr, 1983).

At present, one may consider the polymers called shortly PC, CN and CR-39 as the most frequently used alpha-particle sensitive track detectors. Some of their characteristics are summarized in Table 1. Recently several, new thermosetting resins were found which seem to have good optical properties and track etching behaviour (Fujii, 1985). Because of their capability to register nuclear tracks with different sensitivities, one may expect the development of a more extended family of polymers appropriate for alpha-activity measurements.

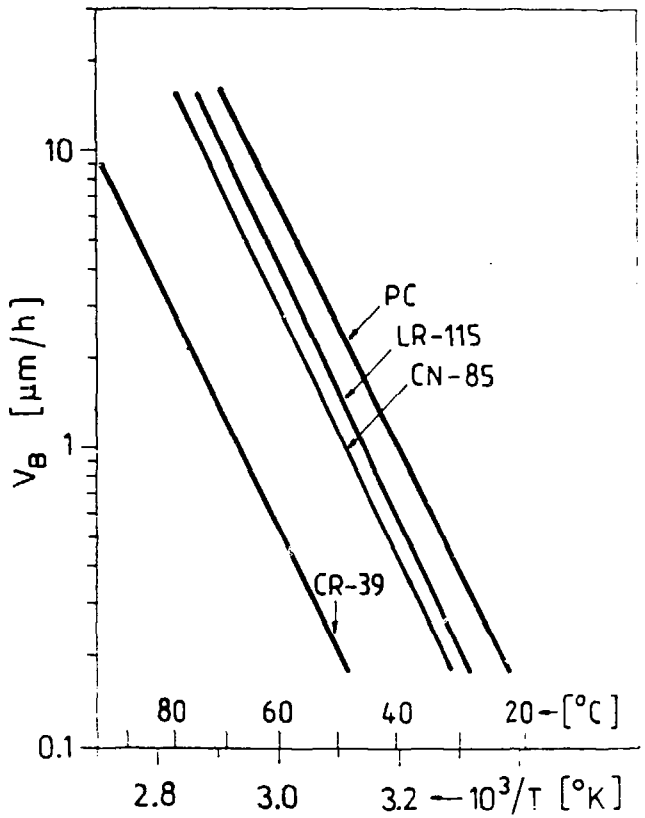


Fig.5. Dependence of the bulk etch rate of typical alpha-particle sensitive plastics on the temperature of track etching solutions indicated in Table I.

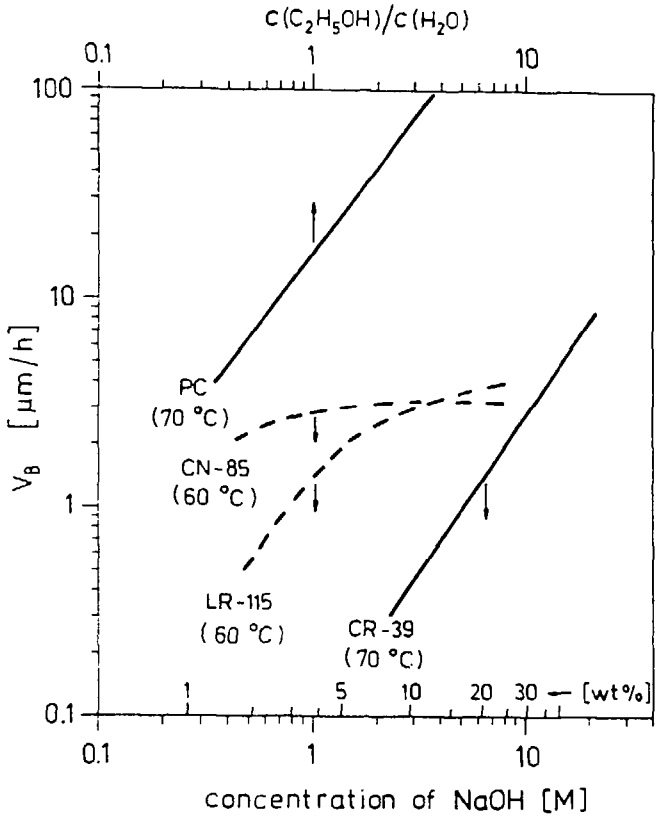


Fig.6. Dependence of the bulk etch rate of typical alpha-particle sensitive plastics on the etchant concentration: on the molarity of NaOH in water used for CN-85, LR-115 and CR-39 and on the alcohol/water ratio in the PEW etchant used for PC (see Table I).

Although there exist some useful guidelines in the literature the optimum condition of track etching, even at present, has to be determined mostly empirically for each track detector material. The etchants given in Table 1 for typical alpha-particle sensitive plastics may prove to be useful in most of the usual track measurements. By using these solutions for track revelation one can get the highest possible registration sensitivity and track contrast in the respective detectors considered. The actual optimum time of track etching depends on the aim of measurement, the energy spectrum of alpha-particles and the way of track counting and evaluation. For a first estimation of the optimum etching duration the following rough rule may appear to be useful. The track diameter related to alphas entering the plastics at right angle is nearly the same or less than the layer thickness ($h=V_B t$) removed from the single surface of detector during an etching time t (see Figs.3 and 4).

For revealing alpha-tracks the exposed detectors have to be immersed into an etching solution kept in a thermostat at a given temperature with an accuracy of about ± 0.1°C. An increase in both the temperature and concentration of etchant can speed up the chemical attack of the bulk of detector material considerable. This behaviour is illustrated in Figs.5 and 6 for PC, CN and CR-39 track detectors when using the etchants indicated in Table 1.

The bulk etch rate of plastics is an exponential function of the inverse temperature of etchant, 1/T(°K), and generally a power function of the etchant concentration, c(mol/dm³), according to the relation

$$V_B = f c^n \exp(-E/kT) \qquad (1)$$

where f and n are fitting parameters, k is the Boltzmann's constant and E is the activation energy for etching (Somogyi and Hunyadi, 1980). As a practical rule one may use that for PC and CR-39 a factor 2 and for CN a factor 2.5 change in $V_B$ is expected for each 10°C variation in the usual region of etching temperatures. With using NaOH in water solution in the 1-10 M range, the concentration dependence of $V_B$ can be well approximated by eq.(1) using the exponents n=3/2 for CR-39 and n=2 for PC detectors. From this point of view the CN sheets show unusual behaviour: their bulk etch rate is only slightly affected by the etchant concentration as seen in Fig.6 (Enge et al., 1974; Tanti-Wipawin, 1975; Hildebrand and Benton, 1980). Certain investigations have shown that such factors as stirring of the etchant, etching in interrupted periods or revolution of the CN sheet in the etchant can modify this tendency (Enge et al., 1974; Somogyi et al., 1978). To avoid temperature and concentration gradients, a controlled slow movement of the detector sheets in the etchant is recommended, but simultaneously a careful control of a possible change of $V_B$ is required.

During the etching period one should keep the quantity $h=V_B t$ constant to have reproducible registration efficiency. This requirement can be easily controlled by exposing a small area of the track detector with fission fragments from a Cf-252 source. After etching, the thickness of layer removed from the detector surface can be derived from the formula $h=d_f(90°)/2$ where $d_f(90°)$

Table 1. Characteristics of some plastic track recorders preferred to use in alpha-activity measurements

| material composition (density in $gcm^{-3}$) | short name | trade name (availability[X]) | etchant proposed for $\alpha$-tracks | $V_B$ ($\mu m/h$) |
|---|---|---|---|---|
| POLYCARBONATE $C_{16}H_{14}O_3$ (1.2) | PC | -Makrofol-E (Bayer AG, FRG) | - PEW solution (15g KOH + 40g $C_2H_5OH$ + 45g $H_2O$), 70°C | ~15 |
| CELLULOSE NITRATE $C_6H_8O_8N_2$ (1.52) | CN | -CN-85<br>-LR-115 type II (Kodak-Pathé, France) | - 10N.t% NaOH, 60°C<br>-the same, 60°C | ~3.2<br>~2.5-4.5 |
| ALLYL DIGLYCOL CARBONATE $C_{12}H_{18}O_7$ (1.31) | CR-39 | -CR-39 (Pershore Moulding Ltd, U.K.)<br>-MA-ND (MOM, Hungary)<br>-Tastrak (Bristol University, U.K.) | - 20wt% NaOH, 70°C<br>-the same, 70°C<br>-the same, 75°C | ~1.2-1.4<br>~1.23<br>~2.0 |

[X] All these sheets, except Makrofol-E, are manufactured as track detectors.

is the diameter of etched fission tracks at right incident angle.

The variation of the etch-rate ratio, $V(R) = V_T(R)/V_B$, describing the etching sensitivity of PC, CN and CR-39 sheets to alpha-tracks, as a function of the etching parameters is a complex and not completely understood phenomenon. In this respect we only refer to some adequate literatures (Tanti-Wipawin, 1975; Cartwright et al., 1978; Henshaw et al., 1979; Somogyi and Hunyadi, 1980; Amin and Henshaw, 1982; Durrani and Green, 1984; Chruścielewski et al., 1984).

Finally we will discuss here few smaller methodical questions which we should know before applying the PC, CN and CR-39 detectors in practice.

When using PEW solution, the only suitable etchant for alpha-tracks in PC, care should be taken to avoid the effect of causing brittleness of the sheet during etching. This effect can't come into action if the used piece of detector is not subjected to mechanical strain or deformation in the etchant (i.e. if the etching procedure is not carried out in too narrow test tubes as compared to the size of detector). It is also useful to know that the latent nuclear tracks in PC sheets are very stable against environmental effects, e.g. humidity and temperature (Urban et al., 1984), but an exposure to ultraviolet light (e.g. solar UV) may increase the etching sensitivity. The UV-sensitization, however, is effective only for the tracks of heavier nuclei and not for alpha-particles.

In case of an LR-115 film, which consists of about 12-13 μm thin red-dyed cellulose nitrate layer on a 100 μm transparent polyester support, a direct exposure to the UV-component of solar light should be avoided. The ultraviolet light can deteriorate the dyestuff and therefore the bulk etch rate may increase considerably. After a proper etching time (usually 2-3 hours when using the etchant given in Table 1) most of the alpha-tracks registered in LR-115 appear in the form of bright holes etched completely through the sensitive layer. The brightness of track holes is improved if immediately after etching the detector is immersed for few minutes in about 5% solution of ethanol in water to remove the colloidal etch-product layer formed on the surface and, in addition, the film is cleaned in water under ultrasonic agitation. Other useful advices to overcome smaller and greater disturbing effects in LR-115 type films are reported in a paper by Fantini and Renard (1982). It should also be noted that a special strippable version of the LR-115 film is manufactured and in this the etched-through tracks can be automatically counted under special condition by the so-called jumping spark technique (Tommasino, 1982), even after exposures to alpha-particles (Somogyi et al., 1978).

Up to now considerable efforts have been devoted to producing high sensitivity track detectors like CR-39 with isotrope and uniform response in different laboratories (see Table 1). In certain cases, however, a decrease in the registration sensitivity proves also to be very useful. It has been shown that this goal is achievable by adding certain organic solvents to alkaline etchants (Somogyi and Hunyadi, 1980; Amin and Henshaw,

1982). With certain alcohol and equeous hydroxide mixtures one can strongly de-sensitize the CR-39 sheets. In a mixture of 60cm$^3$ 6.25M NaOH and 40cm$^3$ pure methanol at 70°C for example the response to alphas can be completely "switched off", and at the same time the bulk etch rate can be increased considerably (one order and even more). One may utilize this effect to remove quickly larger surface layer thicknesses (around 100μm) prior the the use of a CR-39 sheet in low level alpha-activity measurements. Such a pre-etch treatment diminishes almost completely the background tracks originating from the environmental radon diffusing into the plastic sheet and exposing it during the storage period.

## 3. TRACK METHODS FOR RADIUM MEASUREMENTS

### 3.1. General considerations

First of all, it should be emphasized that - as far as the author is aware - published data on the use of alpha-particle sensitive solid state nuclear track detectors for direct radium measurements are very sparse in the scientific literature (Ellis and Jeffree, 1982; Pedraza et al., 1984). There are, however, sufficient data reported on the measurement of uranium (the parent nuclide of U-Ra decay series) on that of radon (the decay product of Ra) by using etch-track detectors. The techniques developed for these, in principle,can be used for radium analysis too. The usual aim of the application of track techniques to such tasks is to measure uranium concentrations in dating or related geological problems (Fleischer et al., 1975) and to carry out time-integrated radon exposures under different environmental conditions: e.g. in soils and natural waters for uranium prospection and radon transport studies (Alter and Price, 1972; Gingrich and Fisher, 1976; Fleischer and Mogro-Campero, 1978; Somogyi et al., 1978; Khan et al., 1980; Gingrich, 1984; Somogyi and Lénárt, 1986) and in dwellings, mines as well as in waste and construction materials for estimation of health hazards (Frank and Benton, 1977; Abu-Jarad et al., 1980; Gingrich et al.,1982; Fleischer et al.,1980; Langner et al.1983; Somogyi et al., 1986).

A way of the radium analysis can be based on the mapping of alpha-decaying elements in the complete U-Ra series by alpha-autoradiography or on the measurement of only the uranium itself by neutron-induced fissionography when the radioactive equilibrium in the sample is established. These methodical approaches will be shortly called in the forthcoming the uranium-alpha and uranium-fission methods of the radium anlysis.

The direct goal in the recent literatures published on radon measurements using some kind of track methods is not the determination of the radium content itself, from which the radon arises, but a quantitative or often only a qualitative estimation of the radon exhalation (emanation) from various media. A radium analysis by these track methods, however, may be practically performable if the radioactive equilibrium between the radon and radium is established and the boundary conditions of the measurements are clearly determined (e.g. the localization of the occurrence of radium producing the measured radon).

This type of indirect way of radium measurements via the alpha-decay of its first daughter element will be shortly called in the forthcoming the radon-alpha method of radium analysis. In all the practical application of this method one should take into consideration that an effective radioactive equilibrium (about 98%) for the radium-radon members of the decay series is reached in about 3 weeks, as the 1620 year half-life of Ra-226 can be considered infinitive when compared to the 3.8 day half-life of Rn-222. Finally we note that a direct radium measurement analysing alpha-tracks originating from the radium itself, which is separated from its parent nuclides seems also possible. This will be called the radium-alpha method of radium analysis.

In the end some words should be devoted to the estimation of the possible contribution of the members of Th-232 decay series to the above-mentioned measurements. This question, unfortunately, has received little attention in the literature, compared to the large number of studies on the U-238 and its decay products. The thorium in natural samples usually is more abundant than uranium by a factor of about 4. The determination of thorium content in a sample by track techniques is more difficult than that for the uranium itself. To solve this problem it is generally necessary to use double irradiation techniques: e.g. fissionography induced by both thermal and fast neutrons (or high energy charged particles). A useful survey of the methodical possibilities and problems concerning this question can be found in a book by Fleischer et al.(1975) and a paper written by Crozaz (1979). In addition to this, in the following, we will touch only on two questions: the response of plastic track detectors to the alphas of an equilibrium Th-232 series and the elimination of thoron effect (Ward et al., 1977), respectively.

## 3.2. The uranium-alpha method

Radium, along with uranium and thorium, can be found in natural materials (rock, soil, water, air, plants, etc.) and man-made products ( construction materials, industrial wastes, etc.) in very different levels of activity (Sheppard, 1980). The lowest limit of its detectability is obviously determined by the background "noise" and registration sensitivity of the method used. The actual value of both factors may greatly depend on the type of nuclear track detector and its storage and pre-treatment conditions. The lowest background track density (few alpha-tracks/cm$^2$) can be achieved by fresh CR-39 detector removing about 100 μm thick layer from its surface in a pre-etching process using alcohol/aqueous hydroxide etchants (Somogyi and Hunyadi,1980; Fews and Henshaw,1983). Without any pre-treatment few hundred alpha-tracks per cm$^2$ are generally found even in new CR-39 sheets and somewhat less than 50 etched-through track holes per cm$^2$ in LR-115 films etched to 6-5 μm residual thickness (which is optimum for alpha-activity measurements).

In the majority of natural materials the alpha-activity can reach an equilibrium concentration. Sometimes, however, certain elements my be selectively taken up by materials of biological origin ( root, grass, soil organism, algae, fish, bone, etc.) or by natural waters in a dissolution process. In radioactive

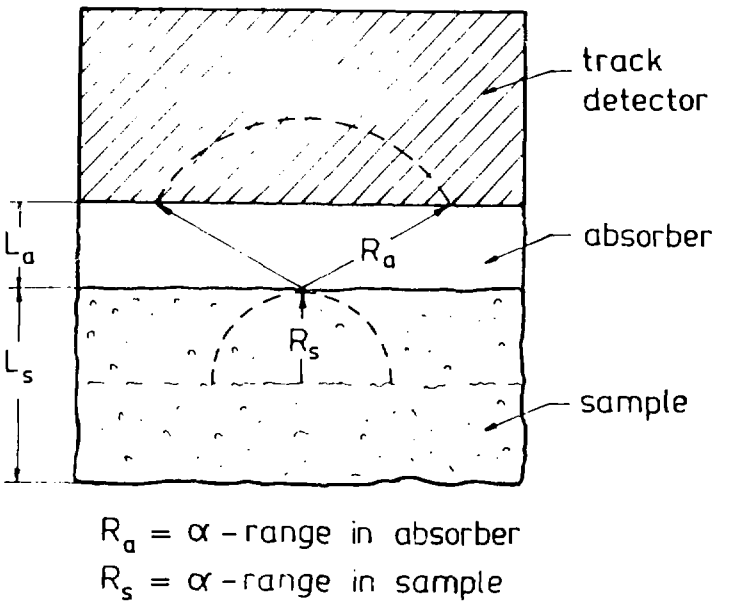


Fig.7. Schematic illustration of the geometry of measuring arrangement used to be applied to alpha-track and fission-track autoradiography. For attaining good spatial resolution a close contact ($L_a$ 0) and measuring average concentration an air gap or other absorber ($L_a$ /0) must be used between the sample and detector.

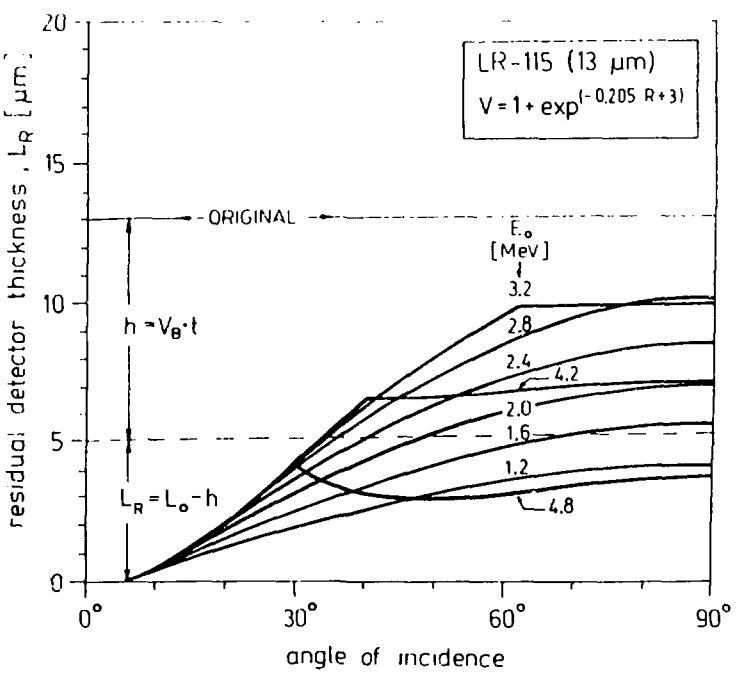


Fig.8. Calculated curves showing the variation of the residual thickness of an LR-115 foil in the moment of perforation of the red-coloured sensitive layer through the etched track holes as a function of the incident angle and energy of alphas.

equilibrium the alpha-decaying U-238 can give rise to seven additional alpha-emitting descendants, among them, the Ra-226 isotope too. (In case of the alpha-active Th-232 six subsequent alpha-emitting daughters appear in the decay series.) If the radioactive equilibrium is established in the series one may apply the method of alpha-track analysis for the estimation of the steady-state activity concentration of radium.

A schematic illustration of the possible, general geometrical arrangement of the above track measurement is shown in Fig.7. The registration sensitivity (defined by the track density obtained for unit alpha-activity concentration during unit exposure time) in this case obviously depends on the quantities $L_S$ and $L_a$ indicated in Fig.7, i.e. on the thickness of sample to to be masured and on the absorber (usually air gap) used, respectively. For a quantitative evaluation of this task, it is expedient to discuss separately the cases where $L_a=0$ (geometry of "contact autoradiography" ) and $L_a/0$ ( the case of "average alpha-activity measurement").

If the track detector and the sample are in close contact during an exposure time T, the registration sensitivity of the method ($S=\varrho/CT$) can be derived from the relationships

$$S_1=(L_S/2)\cdot(1-\sin\theta_c), \qquad \text{if } 0\leq L_S\leq R_S\sin\theta_c \tag{2}$$

$$S_2=(R_S/4)\cdot(\cos^2\theta_c-(1-(L_S/R_S))^2), \qquad \text{if } R_S\sin\theta_c\leq L_S\leq R_S \tag{3}$$

$$S_3=(R_S/4)\cos^2\theta_c \qquad \text{if } R_S\leq L_S \tag{4}$$

where $\varrho$ is the track density measured for a given alpha-decaying radionuclide occurring with C activity concentration in the sample and $\theta_c$ is the so-called critical (limit) angle for track registration. The frequent practical situations related to "thin" and "thick" samples are obviously represented by eqs.(2) and (4), respectively.

Another experimental geometry, when a "thick" sample covered with an absorber of finite thickness ($L_a\leq R_a$) is used, also frequently occures in practice. For this case a sensitivity calculation leads to the following equations

$$S_{31}=(R_S/4)\cos^2\theta_c-(L_aR_S/2R_a)\cdot(1-\sin\theta_c), \qquad \text{if } 0\leq L_a\leq R_a\sin\theta_c \tag{5}$$

$$S_{32}=(R_S/4)\cdot(1-(L_a/R_a))^2, \qquad \text{if } R_a\sin\theta_c\leq L_a\leq R_a \tag{6}$$

$$S_{33}=0, \qquad \text{if } R_a\leq L_a. \tag{7}$$

As concerns the use of eqs.(2) -(7) it is useful to know that for alpha-particles of $E_o$ (MeV) energy the range in normal air can be well approximated by the empirical formula

$$R_o(\text{cm})=0.322E_o^{1.5} \tag{8}$$

and in any solids by the Bragg-Kleeman's rule

$$R_S\gamma_S(\text{gcm}^{-2})=R_o\gamma_o\sqrt{\bar{A}_S/\bar{A}_o}=3.2\cdot10^{-4}R_o\sqrt{\bar{A}_S} \tag{9}$$

where $\gamma_o$ and $\gamma_S$ indicate the densities expressed in $\text{gcm}^{-3}$ as well as $\bar{A}_o$ and $\bar{A}_S$ the average atomic weights for air and the respective solid. For a material having $X_nY_m$ chemical composition and M molecular weight, $\bar{A}_S$ can be calculated from the relation

$$M\sqrt{\bar{A}_S}=nA_X\sqrt{A_X}+mA_Y\sqrt{A_Y}. \tag{10}$$

Now to get a formula for analysing radium with the "uranium-alpha method" one should take into account that in radioactive equilibrium the activity concentrations, $C_i(Bq/cm^3) = N_i(atom/cm^3)\lambda_i$ are equal for each radionuclide in the U-Ra series, that is

$$C_U = \ldots C_i = \ldots C_{Ra} = \ldots C_{Po} \qquad (11)$$

where $C_i$ denotes the activity of the i-type of radionuclide in unite volume of the sample. The track detector will obviously register the sum of tracks originating from every alpha-active nuclide. Therefore, in case of using "thick" sample and contact autoradiography for an exposure time T, from eq.(4) we have

$$\rho = T\sum C_i S_i = TC_{Ra}\sum S_i . \qquad (12)$$

Hence, after determining the alpha-track density, $\rho_{CR}$, by CR-39, the activity concentration of radium in Bq/kg can be derived from the equation

$$C_{Ra}(Bq/kg) = 4\rho_{CR}(T\cos^2\theta_c \sum R_{Si}\gamma_S)^{-1}, \qquad (13)$$

where $\rho_{CR}$, T and $\sum R_{Si}\gamma_S$ are expressed in alpha-tracks$\cdot$cm$^{-2}$, s and kg$\cdot$cm$^{-2}$, respectively. The radionuclide mass corresponding to the activity in unit mass of the sample can be calculated from

$$m_{Ra}(kg\ Ra/kg\ sample) = C_{Ra}A_{Ra}(n_A\lambda_{Ra})^{-1} = 1.7\cdot 10^{-17} C_{Ra}T_{1/2} \qquad (14)$$

where $T_{1/2}$ is the half-life of radium in years, $A_{Ra}$ is its atomic mass, $C_{Ra}$ is the activity concentration of radium in Bq/kg and $n_A$ is the Avogadro's number.

By means of the relationships (2)-(7) one can calculate the registration sensitivity of several track methods used for element concentration measurements. In Table 2 the results of some sensitivity calculations are given for alphas originating from equilibrium U-238 and Th-232 series and from non-equilibrium uranium, Th-232 and Ra-226 isotopes, occurring in various "thick" solid samples covered in close contact with an "ideal" track detector (i.e. when $\theta_c = 0°$). These data show the maximum alpha-track density expected from 1ppm parent nuclide in the sample.

When using CR-39 sheets the sensitivity data given in Table 2 should be multiplied by the factor $\cos^2\theta_c$ (see eq.(4)). The actual value of $\theta_c$, unfortunately, depends on the alpha-energy and the etching time used (see Fig.2 for CR-39). However, it cannot cause large error if we introduce an average critical angle around $\theta_c$(CR-39)=15° for all the alpha-active nuclides in the U-238 series (between 4.18 and 7.86 MeV). This assumption is quite satisfactory when we etch away about 10 μm thick layer from the surface of a CR-39 sheet to reveal the alpha-tracks (see Fig.2).

For LR-115 foils the situation is more complex. The residual thickness of an etched foil corresponding to the track holes appearance at different alpha-energies is shown in Fig.8. From this it is obvious that there exists an "energy window" in which one can only observe track holes. This is between about 1.9 and 4.2 MeV when the optimum residual thickness (5-6μm) of LR-115 foil is used. In this case the acceptance of the value of $\theta_c$(LR-115)=40°±5° as an average critical angle for track hole observation is advisable.

In addition we should note that , owing to the existence of the "energy window", the form of eqs.(2)-(7) must be modified for U-235 foils. For example, in case of a "thick" sample, instead of eq.(4) the relation

$$S_3=\frac{1}{4}(R_{Su}-R_{Sl})\cos^2\theta_c=\frac{1}{4}\,\Delta R_S\cos^2\theta_c \qquad (15)$$

has to be used where $R_{Su}$(4.2MeV) and $R_{Sl}$(1.9MeV) are the alpha-ranges in the sample , corresponding to the energies at the upper and lower ends of the "energy window".

By the alpha-track method consdired above one can evaluate very low-level alpha-contaminated environmental, industrial and biological samples and waste materials in nuclear and other industry where the experimental difficulties with using other methods are appreciable. Although the neutron-activation analysis has generally higher sensitivity, many elements in the sample can interfere with the measurements. The method based on alpha-track detection has usually lower accuracy, but its simplicity, low background and possibility of using long exposure times simultaneously for many samples, in most cases, compensate the drawbacks. A quantitative analysis of the alpha-activity in solids and liquids with the presented evaluation technique developed by the author can generally be carried out with an accuracy of 15-25%. A precise calibration (internal or external standard) method, however, can be adopted which may reduce the error limit. A comparison of the results obtained by the uranium-alpha method and the neutron activation analysis by Uda and Iba (1985) in quantitative estimation of uranium in metals has shown less than 15% deviation for samples of up to 100 pp U content. These authors have also derived experimental relationships between the absorber thickness and the alpha-track registration efficiency for thin surface sources. In addition, Few and Henshaw (1983) have proved that the track method using contact autoradiography can be successfully extended even to the evaluation of the energy spectrum of alpha-emitting nuclides (including Ra-226) in complex biological tissues (*Henshaw and Fews,1984*). The alpha-track method seems also to be applicable to liquids if suitable sample preparation techniques are available. Appropriate sample collection and preparation procedures for uranium and radium in spring waters and sediments have been reported recently by Mishra et al (1982).

### 3.3. The uranium-fission method

The uranium determination method based on fission-track analysis is well developed and known in the literature for samples made either from solids (see e.g. Fleischer et al., 1975; Matsuda et al.,1972; Crozaz,1979; Hamilton,1980; Koul et al.,1980; Igarashi et al.,1985) or liquids (e.g. Iyer et al.,1974; Fleischer and Delany,1976; Hamilton,1980). In great majority of old natural solid materials the radionuclides in the uranium decay series are generally in radioactive equilibrium and therefore the activity concentrations of U-238 and Ra-226 are equal. In this case the radium content in the sample can be obtained from the uranium concentration. Using appropriate nuclear track detectors one may also reveal the spatial distribution of radionuclides even on several micrometer scale.

Table 2. Calculated registration sensitivities of an ideal plastic track detector ($\theta_c = 0°$) exposed in close contact with various samples containing different combination of alpha-emitting radionuclides. The sensitivity data are expressed in $\alpha$-tracks $\cdot$ cm$^{-2}$/30 day exposure for 1 ppm parent nuclide concentration in the sample.

| SAMPLE PARAMETERS | | | TRACK REGISTRATION SENSITIVITY alpha-active radionuclides in the sample | | | | |
|---|---|---|---|---|---|---|---|
| material | density $\gamma_s$ (g$\cdot$cm$^{-3}$) | $\Sigma R_{si}\gamma_s$ for U-series (kg$\cdot$m$^{-2}$) | equil. U-238 series | equil. Th-232 series | only U-238+U-234 nuclei* | only Th-232 nuclei | only Ra-226 nuclei* |
| $SiO_2$(quartz) | 2.65 | 0.461 | 368 | 109 | 68.1 | 9.4 | $1.12\cdot10^8$ |
| $Na_2O\cdot CaO\cdot 6SiO_2$ | 2.5 | 0.472 | 376 | 112 | 70.1 | 9.7 | $1.15\cdot10^8$ |
| Biotite mica | 3.2 | 0.550 | 437 | 129 | 81.5 | 11.2 | $1.34\cdot10^8$ |
| $U_3O_8$ | 8.3 | 1.37 | 1090 | 324 | 203 | 28.1 | $3.35\cdot10^8$ |
| $ThO_2$ | 9.7 | 1.43 | 1133 | 337 | 211 | 29.2 | $3.49\cdot10^8$ |
| U(metal) | 18.7 | 1.54 | 1225 | 365 | 228 | 31.6 | $3.77\cdot10^8$ |

*1ppm Ra-226=$3.7\cdot10^7$ Bq/kg; 1ppm U-238=12.3 Bk/kg.

In the following we will consider only the problem of average uranium content determination in a homogeneous solid sample. In cases where nonuniform uranium distribution exists one may remove the heterogeneities either by dissolving the sample or by crushing it to a fine scale. Another simple solution is the use of a suitable air gap between the sample and track detector during the fission fragment registration as illustrated in Fig.7 for alpha-tracks. The relations (2)-(7) derived for the uranium-alpha method can also be applied to these methodical procedures. The effect of $\theta_c$, however, can usually be neglected as the critical angle for fission track registration in plastics and mica is only few degree.

The simplest technique for determining uranium by the fission-track method consists of placing an uranium-poor track detector (e.g. PC: Makrofol-E, Lexan or PET: Melinex-O, Hostaphan, Mylar) in contact between the sample to be studied and a calibrated standard of known uranium concentration (e.g. NBS glass wafers prepared by Carpenter and Reimer,1974). After irradiating the sample-detector-standard package with a thermal neutron fluence producing suitable fission-track density (more than 1000 tracks per $cm^2$), the detector should be etched and then scanned under randomly selected fields of view in an optical microscope at a magnification of about 500X. The concentration of uranium, C(U), in the sample and the equilibrium radium concentration, C(Ra), expressed in weight fractions can be given by the simple relation

$$C_x(U) \cdot C_x(Ra) \cdot (\lambda_{Ra} \cdot \lambda_U) \cdot C_o(\rho_x \cdot \rho_o) \cdot (R_o/R_x), \qquad (16)$$

where $\rho$ represents the density of neutron-induced fission tracks, R is the average range of fission fragments in $g \cdot cm^{-2}$, $\lambda$ is the alpha-decay constant and the subscripts x and o refer to the unknown and standard sample, respectively. We should note here that $R_o/R_x$ is nearly unit if the standard and unknown samples have similar elemental composition, but generally the fission fragment range increases with increasing atomic number.

In the uranium measurements considered, special care must be taken to avoid using any materials for treating, cleaning and mounting procedures with disturbing fissionable contaminations and causing track fading by overheating the plastic track detector during neutron irradiation in a reactor. Typical uranium contents found in different chemical reagents are published by Matshuda et al.(1972). The ordinary de-ionized water and the typical laboratory detergents (alcohol, aceton) are satisfactory agents as their uranium content is generally less than $10^{-9}$ g U in one g material. It is also useful to know that the gluing materials on the usual adhesive tapes may contain larger quantities of uranium, therefore their use must be avoided.

### 3.4 The radon-alpha method

For prediction of the level of radiation hazards originating from radium-containing materials, the determination of the rate of radon exhalation is of prime importance. For this purpose the most simplest and efficient method is to place the specimen to be measured in a cylindrical-shaped plastic or

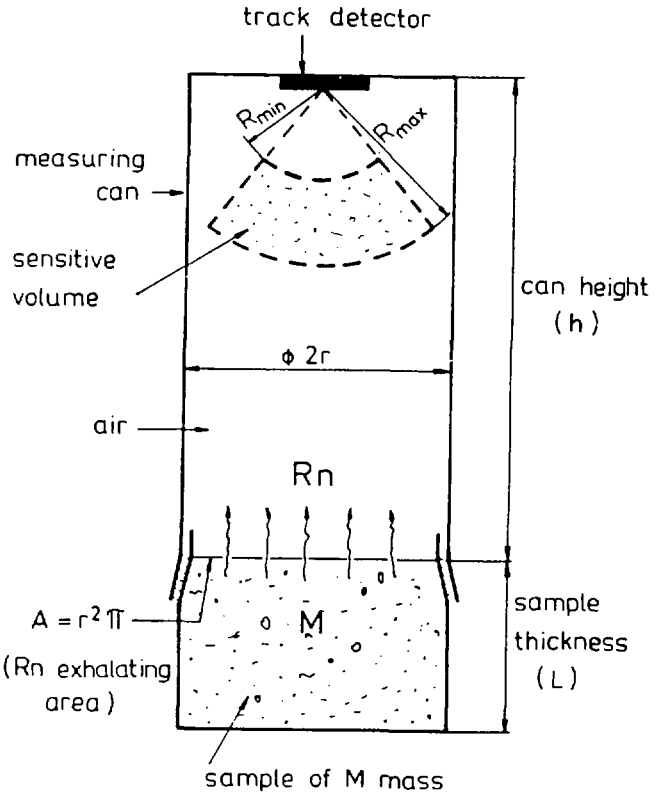


Fig.9. Cross-sectional view of a can-type arrangement for measuring radon exhalation from a radium-containing sample by means of alpha-particle sensitive track detector.

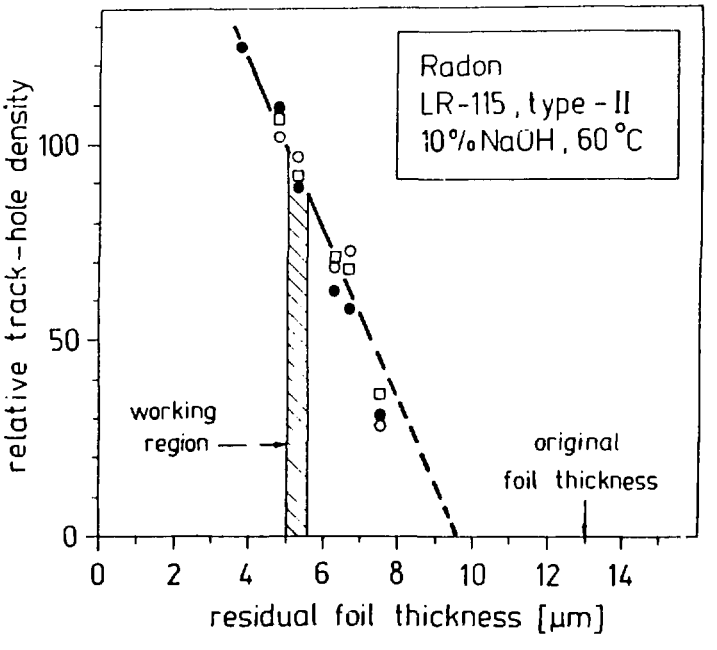


Fig.10. Calibration curve measured for LR-115 foils exposed to the same radon source and then etched to different residual thicknesses of the detector.

metallic can and to fasten a piece of track detector on its inside bottom to register the alpha-radiation from the radon entered the air volume of can. This type of measuring method, called shortly "can-technique", was proposed by Alter and Price (1972) for time-integrated radon monitoring and since that time it has been used extensively in numerous investigations concerning uranium prospection (Fleischer et al.,1975; Gingrich, 1984; etc.), environmental radon transport (Fleischer and Mogro-campero,1978; Somogyi and Lénárt,1986; etc.) as well as radon and daughter dosimetry (Frank and Benton,1977; Fleischer et al., 1980; Abu-Jarad et al.,1980; Gingrich et al.,1982; Somogyi et al.,1984; etc.).

Some attempts have also been made to examine critically the track-etch and other methods available for radon emanation measurements and to standardize the procedures in both field and laboratory measurements. In the papers reported on these questions (Gingrich et al.,1982; Langner et al.,1983; Cliff et et al.,1984) the reader can find various comparisons among the most common methods of radon (and sometimes thoron) concentration measurements based on scintillation counter, ionization chamber, solid-state alpha-detectors, gamma-spectrometers and track-etch techniques as well. In addition, some recommendations concerning the use, precision and evaluation of these methods have also been made.

In our present survey only the principle of the radon-alpha detection with special emphasis on the so-called effective radium content measurement is considered. With the "sealed can technique", however, used for the above purpose, all the important parameters of the radon transport in solids (areal and mass exhalation rates, $\emptyset_A$ and $\emptyset_M$; effective diffusion length, $z_d$; porosity of sample, p) can be determined with reasonable accuracy (Somogyi et al.,1986). A schematic drawing of the arrangement for measuring effective radium, i.e. radium in radioactive equilibrium with the measured radon released from the sample, is shown in Fig.9. After closing the can, the activity concentration of radon starts to increase with the time t elapsed according to the relation

$$C_{Rn} = C_{Ra}(1-e^{-\lambda_{Rn}t}) \qquad (17)$$

where $C_{Ra}$ is the effective radium content of the sample. This formula must be used when "pin-like" measurements in time are carried out , e.g. by emanometer, ionization chamber and scintillation counter (see e.g. Tóth and Fehér, 1976). A plastic track detector, however, measures the time-integral of eq.(17), i.e. the total number of alpha-disintegrations in unit volume of the can with S sensitivity, during the exposure time T. Therefore, the track density observed is given by the equation

$$\varrho = SC_{Ra}T_e = SC_{Ra}\left[T-\lambda_{Rn}^{-1}(1-e^{-\lambda_{Rn}T})\right] \qquad (18)$$

where $T_e$ denotes, by definition, the effective exposure time.

Now, we have to find S, the sensitivity of the radon measuring method when using a cylindrical measuring device equipped with plastic track-etch detector. The actual value of S can be derived from eq.(4) for CR-39 and from eq.(15) for LR-115 detector

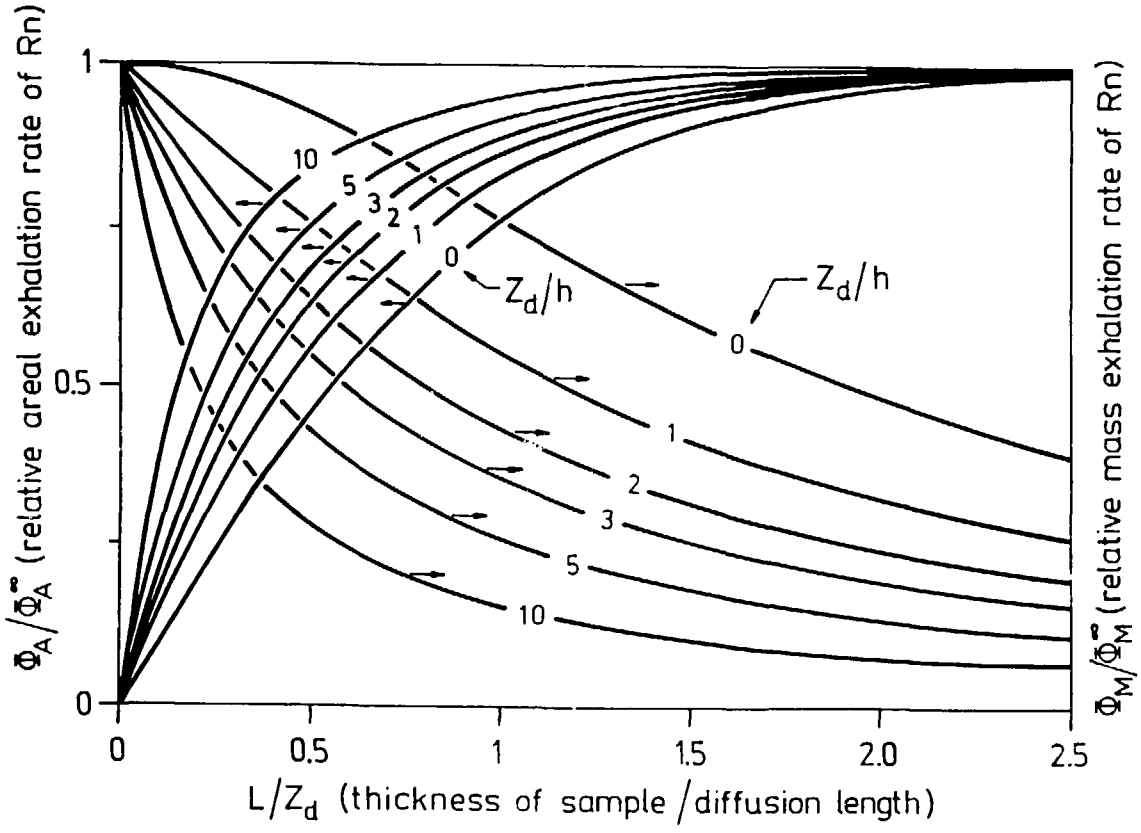


Fig.11. Variation of the normalized areal and mass exhalation rates of radon in a can-type measuring device shown in Fig.9, as a function of the thickness of sample when using different can heights, h, i.e. air gaps between the sample and detector. The curves are calculated at a porosity of p=0.5.

if the length and diameter of the can are "infinitive" compared with $R_S$ or $R_{Su}$, the meaning of which in the present case alpha-ranges in the can air, (i.e. in the radon-containing "sample"). At other can geometries the calculation is more complex due to the presence of alphas from Po-218 and Po-214 plated-out on the can wall and detector surface. However, using a can of 3.5cm radius and 10cm height the registration sensitivity of the LR-115 film to radon alphas becomes practically independent on the radon daughters concentration, and so on the plate-out effect and the degree of radioactive equilibrium as well (Somogyi et al.,1980). In this case the value of S is influenced only by the residual detector thickness obtained after completing the track etching procedure, as illustrated in Fig.10. Having etched the LR-115 film to the optimum, 5µm residual thickness, a value of S(LR)=(1/30)track-hole·cm$^{-2}$/day can be used for one Bq/m$^3$ radon in the can of 3.5cm radius,within about ±15% accuracy. For CR-39 this factor, assuming complete equilibrium, is about 9 times higher than S(LR), from which about 1/3 part comes from the airborne radon and 2/3 part arises from the plated-out Po daughters. According to the author's experiences in a completely sealed can practically the ratio S(CR)/S(LR)= 6± 20% is realized for solid specimens and the ratio of about 4 - 4.5 if one immerses an iverted cup under water for field radon measurement.

Finally , with using the "sealed can technique", the effective radium content of a solid sample, expressed in Bk/kg, can be obtained from the formula

$$C_{Ra}=(\varrho/ST_e)\cdot(hA/M). \qquad (19)$$

Here we should paid some additional words to the effect of can and sample dimensions on the radon (and effective radium) determinations. This question, in the author's opinion, has not been treated with the necessary carefulness in the relevant literature. A reduction in the radon exhalation rate, owing to a phenomenon sometimes referred to as "back diffusion", can be expected if the sample is placed into a closed container. A theoretical analysis of this problem (Somogyi et al.,1986) has shown that the "bound" areal exhalation of radon as a function of the geometrical dimensions (L and h, see in Fig.9) of a cylindrical can and sample can be described by the relationships

$$\emptyset_A=(\emptyset_{Am}/k)\cdot th(L/z_d) \quad \text{and} \quad k=1+(pz_d/h)\cdot th(L/z_d) \qquad (20)$$

where $z_d$ is the effective diffusion length of radon in a sample of p porosity and $\emptyset_{Am}$ is the maximum possible rate of radon exhalation from a "thick" sample (practically if $L>2z_d$).The trend of this effect is illustrated in Fig.11 at a porosity of 0.5. From eq.(20) it is obvious that the ratio of a "free" (if $h\to\infty$) and a "bound" (if can volume ≲ sample volume) areal exhalation rate of radon is determined by

$$\emptyset_A(free)/\emptyset_A(bound)=\varrho_f/\varrho_b=k. \qquad (21)$$

Therefore, in any measurements using the "sealed can technique", the appropriate formula involving the correction due to the "back diffusion" of radon is

$$C_{Ra}(Bq/kg)=(\varrho/ST_e)\cdot(hA/M)\cdot\left[1+(pz_d/h)\cdot th(L/z_d)\right]. \qquad (22)$$

To apply this formula in practice, the value of p and $z_d$ has to be known, which quantities can also be determined by using the "can technique" (see Somogyi et al.,1986).

Finally, we must note that the elimination of the thoron effect in effective radium measurements can be solved easily either by placing a thin, semipermeable plastic membrane between the sample and the sensitive volume of can (Ward et al.,1977) or by increasing the length of can up to about 30cm to permit the decaying-out of the short half-life (56s) thoron along its diffusion path (Somogyi et al.,1984).

3.5. The radium-alpha method

Most of the current techniques for direct radium measurements involve a saparation of radium from the major sample constituents by ion exhange or co-precipitation with barium or lead sulphate. If the radium can be well separated from other elements and it can come to equilibrium with its daughters, the sample can be analyzed by the alpha-track method. In this case the Ra-226 itself and the Rn-222, Po-218 and Po-214 isotopes contribute to the total alpha-track density observed. The effect from the last alpha-emitter, Po-210, in the decay series can be neglected as it originates after the decay of long-lived Pb--210 ( 22 year half-life).

We should note that the sample preparation technique suitable for barium is generally applicable to radium, as well. A typical method is the co-precipitation of the usually trace quantities of radium with $BaSO_4$ carrier. The radon in the freshly prepared sample is allowed to come to equilibrium in about 3 weeks, then an alpha-autoradiogram is taken with an alpha-sensitive plastic track detector in contact with the sample. For quantitative measurements a standard reference sample or the knowledge of the registration sensitivity of the detector for radium and its alpha-active daughters is required. The later quantity may be estimated from the relationships (2) and (12). However, we should take into account that in certain cases,when using thinly deposited radium sources, not all the newly generated Rn-222 atoms are retained on the source surface. According to Hashimoto et al.(1979), if the Ra-226 containing sample is prepared by an electroplating procedure from isopropanol solution on stainless steel or platinum plates, about 50% or respectively 15% of the equilibrium Rn-22 activity is retained on the samples.

Another simple technique to prepare radium sources for measuring low activities (<100 $Bq/m^3$) extracted from large volumes of natural waters is reported by Bland (1982). He prepared discs of commercial nylon and cellulose impregnated with hot, saturated $KMnO_4$ solution. The radium dissolved in the water is adsorbed by such discs simply by leaving them for several hours in the natural water.

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