

4. Measurement techniques

The measurement of ^{222}Rn , ^{220}Rn and their decay products is always based on the detection of the radiation they emit. As it can be clearly seen in Figures 2.1 and 2.2, these radionuclides are α -, β - and γ -emitters. All these emissions, in their interaction with matter, give rise to various signals such as local defects, electrical currents or pulses and light intensities. Thus, practically any of the available detection instrument or system able to utilise, independently or in combination, the registration of these signals, see Knoll (2000), is potentially appropriate for the measurement of the above radionuclides. The only conditions to account for are the corresponding adaptation of the system used to the gaseous nature of ^{222}Rn and ^{220}Rn , and/or to the complex gas-particle behaviour of their airborne daughters.

The most common detection techniques used in ^{222}Rn , ^{220}Rn and their progeny monitoring are pulse ionisation chambers, charged electrets, semi-conductor diodes, solid and liquid scintillation phosphors, and activated carbon collectors. Other methods employing Nuclear Track Detectors (NTDs) or ThermoLuminescent Detectors (TLDs) have also a wide applicability. The choice of these techniques or methods depends on the specific application, information needed, sampling duration, and sensitivity requirement. An extensive and complete evaluation of the performance characteristics as well as the state-of-the-art of the above measurement techniques can be found in OECD/NEA (1985), Urban and Schmitz (1993), Harley (1992), George (1996), and in Monnin and Siedel (1997).

In this chapter, a brief description of the measurement principles of ^{222}Rn , ^{220}Rn and their progeny is given in conjunction with a comparison of the main methods with respect to their ability to provide long-term exposure levels in private homes and workplaces. Special attention is paid to the NTDs, as this is the type of detectors used in this work.

4.1. Principles of measurement

In contrast to their short-lived daughters, which are — as it has been seen in the previous chapter — chemically reacting solid minerals, ^{222}Rn and ^{220}Rn are radioactive

noble gases. Therefore, it is useful to distinguish between the measurement principles of ^{222}Rn and ^{220}Rn and that of their decay products.

4.1.1. Detection of ^{222}Rn and ^{220}Rn gases

It must be borne in mind that the measurement of ^{222}Rn and ^{220}Rn concentrations can provide only information on the upper limit for the potential α -energy exposure due to the presence of their daughters in air. The concentration of ^{222}Rn and/or ^{220}Rn is generally easier to measure than the concentration of any associated individual daughter. The instrument has to be designed in a way that only the noble gas and not its airborne progeny (neither in airborne-unattached state nor in aerosol-attached form), is permitted to enter the detector sensitive volume¹. In this case, the gas concentration is measured by the registration of the radiation (primarily α -particles) emitted by its atoms present in the sensitive volume and/or by the subsequent decay products. Another possibility could be done by collecting the gas on activated charcoal and measuring the contents of its progeny after absorption by means of γ -spectrometry technique.

^{220}Rn gas may in principle be detected using the same methods as are applicable for ^{222}Rn . However, the relative short half life of ^{220}Rn makes the direct determination of its concentration very difficult unless it can be measured immediately after it is formed. In most cases, this concentration is inferred from the measurements of ^{220}Rn daughters. In the case of ^{222}Rn measurement only, the elimination of ^{220}Rn contribution is possible thanks to the fact that the diffusion length of this last in a given medium is very small with regard to that of ^{222}Rn (see Chapter 2). When the purpose is to measure ^{220}Rn itself, the problem of avoiding the contribution of ^{222}Rn is much difficult to solve, since any method used for this discrimination will eliminate the ^{220}Rn concentration as well. To date, the methods usually used to measure ^{220}Rn are based on i) a combination of different filters in a multi-detector (two) system, in such a way that the ^{220}Rn concentration is obtained from the difference of the readings of the detectors used, as it is the case of the dosimeter developed in the present work, ii) delayed coincidences between the α -emissions of ^{220}Rn and ^{216}Po . Owing to the short half-life of the ^{216}Po (145 ms), it is expected to be in radioactive equilibrium with its precursor (^{220}Rn), so that, it is possible to distinguish between $^{220}\text{Rn}/^{216}\text{Po}$ pair counts and random coincidence pulses. An example of the application of this method is given by Limoto and Kurosawa (1996), in which they use a scintillation cell facing a photomultiplier tube connected to three time-gates pulse-coincidence circuit.

¹The volume in front of the detector in which the radiation emitted has a non zero probability to be detected.

4.1.2. Detection of ^{222}Rn and ^{220}Rn daughters

The measurement of ^{222}Rn and ^{220}Rn daughters is of a great importance because it lets the direct determination of the potential α -energy concentration (see Section 2.3). Almost all the detection systems used for the measurement of ^{222}Rn and ^{220}Rn daughters are based on their deposition properties. Having these decay products deposited on surfaces, filters or detectors, makes the handling more convenient, allowing moving of the sample, application of vacuum, etc. A simple method consists of drawing a given volume of air through a filter during a preset period of time. Radioactive decays from the filter can be counted during the sampling, after sampling, or both. By counting two, three or even more times the activity of this filter after the sampling at optimised time intervals, concentration ratios of ^{222}Rn or ^{220}Rn progeny in the sampled air are able to be estimated with quite acceptable accuracy. Particularly, separation between ^{222}Rn and ^{220}Rn daughter concentrations could be performed at counting times longer than 30 minutes. Another method that determines precisely the airborne individual daughter concentration is by performing α -spectrometry on the filter.

A separate measurement of the airborne-unattached and aerosol-attached ^{222}Rn and/or ^{220}Rn decay products, could be performed by passing the sampled air previously to the filter through long tubes, parallel plates or fine-meshed wire-screens. In this situation, the airborne-unattached daughters, because of their high diffusivity, deposit to a large extent on the available surfaces (tube, plates or wire-screen) before reaching the back-up filter. The aerosol-attached progeny pass more or less unhindered through these surfaces and are mostly collected by the filter. In principle, the resulting activities of the tubes (eventually on the parallel plates or the wire-screens) as well as of the back-up filter, permit to determine the size distribution, between airborne-unattached and aerosol-attached states, of each individual ^{222}Rn or ^{220}Rn daughter. This parameter is of great interest, with regard to human lung dosimetry, because it affects the deposition pattern of ^{222}Rn and ^{220}Rn progeny in the respiratory tract.

4.2. Detector classification

The fact that there are a lot of types of measurement systems for ^{222}Rn , ^{220}Rn and their progeny make difficult to classify them. Certainly, there is not a unique criterion for this classification. In addition to the type of radiation that is detected, there are several other characteristics to distinguish between the measurement techniques. We have already mentioned one in the previous section and it is whether the technique is able to measure ^{222}Rn and ^{220}Rn gases, or their decay products. Another major characteristic is the time

resolution of the considered technique. Accounting for this criterion, we can consider three broad types of techniques: grab-sampling, continuous measurement and time-integrated measurement.

Grab-sampling: In this technique, the radioactive content of discrete air samples, usually collected at a single point over a short time, is analysed. The result obtained is representative for the instant when the sample was collected. This technique is useful for a quick estimation of the radionuclide concentration, and can be used also for tracking the time evolution in a given site while, for example, a remedial measure is being installed.

Continuous measurement: This technique is designed to provide information on the time evolution of the radionuclide concentration. The result of the counting is known continuously and is normally registered during the whole measurement period. This technique is basically used for research studies, where the effect of a given parameter (for instance ventilation rate) is explored, and for testing remedial techniques.

Time-integrated measurement: In this case, the technique provides a single concentration measurement averaged over the exposure period, which may span from a few days to several months or even to a year. It is used normally for carrying out wide surveys over a given region, nation or state to determine the annual average of the radionuclide concentration with the aim of estimating the annual dose.

There is another way of classifying the detection systems that has not a clear definition but that is widely used in the ^{222}Rn research community. Detection systems can be *active* or *passive*. Unfortunately there is no generally agreed definition of this classification and, therefore, some instruments might be called active or passive, depending on the definition applied. Perhaps the most common definition is the following:

- A detection system is active if it requires for operation some kind of external or internal power supply, including those that have battery packs.
- A detection system is passive when it does not require any external or internal power supply for operation.

This definition implies that all instrumentation having running parts such pumps and electronics (battery supplied) are active, independently on the type of detector used. One can also make another differentiation between active and passive detectors: active detectors are those that provide a pulse signal when the radiation interacts with their active volume as, for instance, a liquid scintillator; while, passive detectors are those that

register the interaction of radiation in a permanent way at least during the exposure such is the case of NTDs and TLDs. However, according to the common definition we have adapted for the active and passive detection systems, an instrumentation that uses NTDs together with pumps to collect a certain volume of air is an active system.

4.3. Detector comparison

Each system has its own characteristics. Calling them advantages or disadvantages depends on the goal of their use. The main purpose of this work is to develop a dosimeter for long-term measurement of ^{222}Rn , ^{222}Rn progeny and ^{220}Rn concentrations in private homes and workplaces to better obtain the annual equilibrium factor, and therefore, the associated effective dose at which population are exposed to.

The indoor ^{222}Rn , ^{220}Rn and their decay products concentrations present high time-fluctuations on different time-scale cycles (from hours to days, months and even years). Therefore, to obtain the annual radiation dose due to the presence of a given radionuclide in indoor environments, the best option would be to measure its concentration during the whole year period. In general, for practical reasons, this is not possible and, therefore, it is of interest to know to which extent a given result is representative of the annual average radionuclide concentration. Swedjemark (1984) carried out an interesting study, based on measurements of ^{222}Rn and its decay products in various Swedish houses, to estimate the statistical uncertainty entailed in sampling periods shorter than one year. The results of his study are summarised in Table 4.1.

The uncertainties in the estimation of annual average dose from exposure to indoor ^{222}Rn progeny may vary from 33% for one-hour sampling to 13% for three-month measurement. The uncertainty in this estimation from a single grab-sampling with a time duration less than one hour may be even higher than 33%. In conclusion, in order to achieve a good accuracy of the indoor annual exposure due to the presence of ^{222}Rn and ^{220}Rn decay products, it is essential to use instruments integrating over periods of several months for their measurement. Obviously, it is possible to use continuous measurement systems or short-time grab sampling systems (for instance 24 hours) during the whole year to obtain the required annual average concentration. However, continuous measurement systems are normally active devices, which are mostly expensive, noisy, large-sized, unportable and they can only be handled by technical staff. Grab sampling methods also require the presence of technical staff as well as a logistic support to take samples at a sufficient frequency to be the mean value accurate enough. In contrast, passive systems using time-integrating detectors, such as NTDs or TLDs, may provide good estimations of annual averaged dose.

In fact, the Commission of the European Union suggested the use of passive systems integrating over several months for long-term measurement of ^{222}Rn , ^{220}Rn and their progeny concentrations in private homes and workplaces (Bochicchio, et al., 1995).

Table 4.1. Relative Standard Deviation (RSD) from the *true* annual average of different sampling periods (Swedjemark, 1984).

Sampling period	RSD	Sampling method
Grab sampling with restrictions	22%	Scintillation cell
Grab sampling without restrictions	33%	Scintillation cell
24 hours	25%	Activated carbon
3 days	22%	Activated carbon
1 week	20%	Activated carbon
2 weeks	18%	TLDs
1 month	18%	TLDs
3 months	13%	NTDs

An advantage of the TLDs is the possibility of performing direct measurement of the absorbed dose, while their main disadvantage is the fact that they are normally sensitive to photons making necessary the subsequent discrimination of the TL signals induced by ^{222}Rn , ^{220}Rn and their decay products from those produced owing to the presence of the environmental external radiation. The main advantages of the NTDs are that they are sensitive to α -particles but not to β -particles nor to γ -rays, they are mostly unaffected by the environmental conditions (humidity, low temperatures, moderate heating and light) and, in principle, they do not require neither expensive nor complicated instrumentation for their reading. In addition, the registration property of NTDs is an intrinsic quality of the material they are made of and the signal produced by the radiation is relatively permanent, which makes the reading not to destroy the signal as is the case of TLDs. Another important feature is that they have a selective response, so that they can only detect those particles able to produce a damage in their structure higher than a given threshold. This fact allows to select which α -energies are to be measured. All these reasons have made the use of NTDs in ^{222}Rn studies very attractive and the number of laboratories and institutions using them is in continuous growing with time. A recent review of the NTD applications in ^{222}Rn monitoring is given by Nikolaev and Ilic (1999). In the next section, some fundamentals of NTDs are given together with a description of the electrochemical etching technique applied to polycarbonate detectors.

4.4. Nuclear track detectors (NTDs)

Nuclear track detectors are basically electrical insulating solid materials — including minerals, crystals and plastics — in where the passage of heavily charged particles (from protons upwards) create, along of their paths, damage zones on an atomic scale called *latent tracks*. An eventual treatment of these latent tracks using chemical or electrochemical etching allows their visualisation under optical microscopes or by a naked eye, respectively. Polymeric plastics are best suited to measure α -particles because they are durable, simple, stable, inexpensive, easy-to-use and high sensitive. The polymer detectors commonly used in ^{222}Rn , ^{220}Rn and their progeny measurements are cellulose nitrates (LR-115)², polyallyl di-glycol carbonate (CR-39)³, and bisphenol-A polycarbonates (Makrofol)⁴. The former two detectors are more sensitive than the Makrofol detector. However, the LR-115 has a limited layer of $\sim 13\ \mu\text{m}$, which makes the measurement of ^{222}Rn and ^{220}Rn decay products difficult since it detects only α -particles with energy below 4 MeV (Bonetti et al., 1991; Planinic, 1992). Also, it has the inconvenient of a complicate track counting and presents a relative high fading when the exposure duration is more than 4 weeks. On the other hand, the sensitivity and the background track densities of CR-39 vary with each batch of detector material. In turn, the Makrofol detector presents a constant product quality of its batches, a reproductive energy response, a low background track density, and a negligible fading. For all these reasons, the Makrofol detector is chosen as to be the most appropriate in this study.

4.4.1. Track formation mechanism

When an ionising particle interacts with matter, it transfers a part or the totality of its energy to the electrons of the medium with a linear rate of energy loss, $\frac{dE}{dx}$, that is function of the particle characteristics (mass, charge and energy) and of the detector material used (see Appendix B). When the energy loss is above a critical value, $(\frac{dE}{dx})_{\text{min}}$, called *registration threshold*, which is specific for the detector used, local structure transformations of this last are produced. These transformations, in the case of polymers, can be explained by ruptures in the molecular chains and/or formations of new components chemically very reactive along the particle trajectory. The latent track then formed has a typical diameter from 1 to 10 nm (Fleischer et al., 1975). Figure 4.1 gives a schematic illustration of a chain scission in polymers caused by the passage of heavily charged particles.

Due to their size, latent tracks can only be visualised under extremely high mag-

²Manufactured by Kodak-Pathé, France.

³Manufactured by Pershore Mouldings Ltd., England.

⁴Manufactured by Bayer-AG, Germany.

nification by means of transmission electron microscopes or equivalent. However, if the detector is etched with the help of a suitable hot and concentrated chemical reagent or *etchant* — often a simple alkali (e.g., NaOH or KOH) or acid (e.g., HF or HNO₃) — they can be enlarged up to a diameter of few μm . In the next section, the principles of chemical etching are given. However, before continuing with this section, an important fact has to be taken into account and it is worthwhile to remark: it is well-known that, after exposure, the latent track in polycarbonate detectors may grow *a posteriori* with time reaching stability only after a given period (Domingo et al., 1990). Then, if the detector is etched when the latent tracks are still growing, these lasts will be seen as produced by lower ionising particles. This difference can be avoided if waiting a given elapsed time between the exposure of the detectors and their subsequent etching. This fact leads to the importance of the storage of the detectors, as they must be in a place under well-controlled conditions so that any parameter affecting the evolution of the latent track may not act. Some of the factors that might affect the evolution of the latent track are the presence of ultraviolet radiation, the presence or absence of oxygen as well as humidity in the detector environment, the exposure to gamma radiation, and the temperature of the detector (Khan et al., 1993). Normally the use of a refrigerator to storage the detectors at around 4 °C, during at least one week in heat-sealed radon-proof bags (i.e., thick aluminised plastic bags available), is enough to overcome this problem.

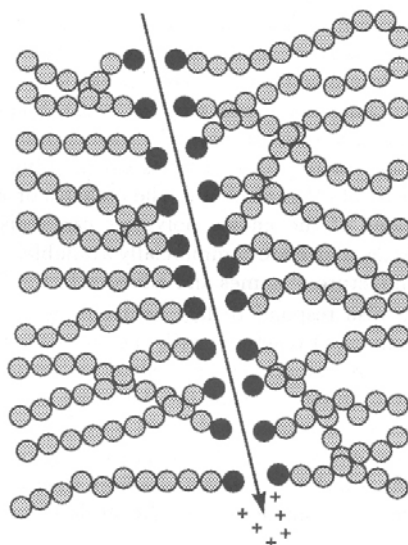


Figure 4.1. Schematic illustration of a chain scission in polymers caused by the passage of heavily charged particles — adapted from Durrani (1997).

4.4.2. Chemical etching (CE)

The chemical etching consists of introducing the detector foils into a thermostatically controlled bath at temperatures ranging from 25 °C to 70 °C, being the commonest etchant an aqueous dissolution of NaOH or KOH of molarities ranging from 2 to 8 M. Typical etching times range from 2 to 6 hours. In certain applications, ethyl alcohol (C_2H_5OH) may be added to the etchant in a certain proportion to enhance the registration sensitivity of polycarbonate plastics. The main advantage of the CE is that it is possible to obtain relevant information of the incident particle such as its energy, charge, mass and incident angle just from the geometrical analysis of the track (see for example Domingo et al., 1996).

The chemical etching is based on the fact that the chemical reactivity of the latent track is higher than the surrounding undamaged detector material. Certainly, when a polymer is subjected to an etchant, the surface of the polymer is removed with a bulk etch rate, v_B , that is lower than the etching rate through the damage trail (latent track) known as track etch rate, v_T . As it can be seen in Figure 4.2, the formation of the track can be easily explained by applying the Huygens principle, assuming both the track and bulk etch rates to be constant and isotropic. After a given etching time, t , the new surface of the plastic will be determined by the surrounding of all the elemental hemispheres (of a radius $v_B t$) centred in the original surface of the plastic, while the preferred chemical etching along the particle trajectory generates a conic etch pit profile.

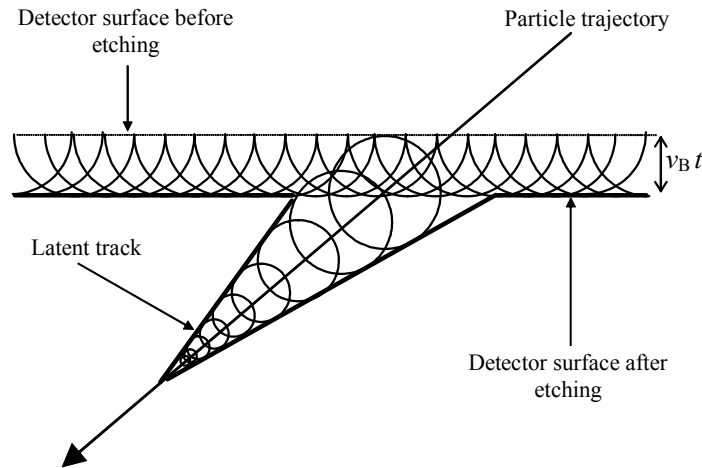


Figure 4.2. Application of the Huygens principle to explain the latent track formation within polymer detectors.

For any charged particle incident at a dip angle, ϕ , which is the angle between the particle trajectory and the detector normal, the component of the track etch rate (v_T) perpendicular to the detector surface must exceed bulk etch rate (v_B) in order to produce a track, so the following condition must be fulfilled

$$v_T \cos \phi > v_B \quad (4.1)$$

Hence there exists a critical angle of registration given by $\phi_c = \arccos(v_B/v_T)$ above which the damage trail is not developed into a track. The bulk etch rate depends only on the plastic detector used and on the etching conditions (type of etchant, % of alcohol mixture, temperature, etc), while the track etch rate also depends on the incident particle characteristics (charge, mass and the rate of energy loss). The ratio v_T/v_B is known indistinctly as intensity of the registered signal or the detector response.

The shape of the track changes drastically with prolonged etching, going through three phases: conical, of transition, and spherical (see Figure 4.3). The final geometry of the etch pit profile depends, therefore, on the instant when the chemical etching is stopped. The conical phase lasts until the etching reaches the end of the damage trail. In this phase, the shape of the track is conical and its open surface is elliptical. When this phase finishes, the transition phase starts in which the bottom of the track is being rounded whereas the open surface is still elliptical. The etching rate is, then equal to the bulk etch rate in all directions. The spherical phase is reached when the shape of the track is spherical and its open surface circular.

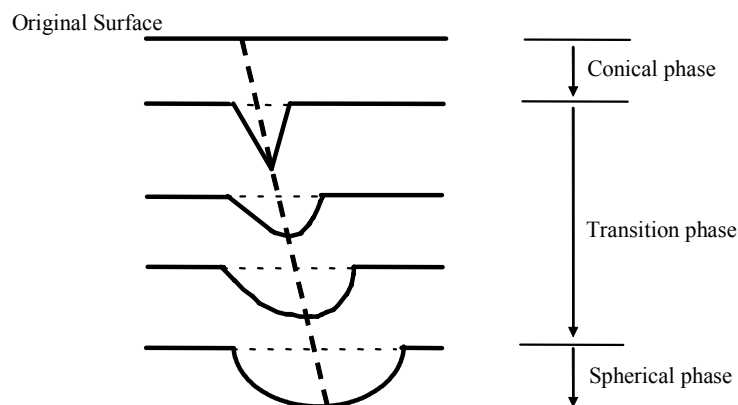


Figure 4.3. The evolution of an etch pit profile with prolonged chemical etching — adapted from Durrani and Bull (1987).

4.4.3. Electrochemical etching (ECE)

The electrochemical etching is an etching technique that increases the size of the tracks up to two orders of magnitude, so that they can be counted by means of low-magnification devices such as microfiche readers, slide projectors, or low-magnification ($\sim 10\times$) microscopes. It is specially used when the incident particles are not so heavy, as for instance protons and α -particles, since they produce small tracks that are difficult to be observed. This size-magnification means a great advantage when the track density is low (say below 10^3 cm^{-2} as it is normally the case of ^{222}Rn measurements in houses) and makes the track counting much easier and faster. The main disadvantages are that the specific information of the track is lost and that the method is not suitable for high track densities (above few 10^3 cm^{-2}) due to the track overlapping effect. The presence of imperfections and/or contaminants within the NTD can produce tracks similar to those produced by the radiation, so that the ECE can increase the background reading.

The ECE consists of applying a high AC electric field across the plastic detector during the chemical etching (Tommasino, 1980). The irradiated plastic foil is sandwiched between two separate volumes of electrolyte, of which one is an etchant. An alternating high voltage is applied across the plastic foil by means of two electrodes that are immersed in the electrolyte dissolutions. In this manner, the NTD detector behaves as an insulator between the two dissolutions. The electrochemical etching conditions most often used are electric field strengths of several tens of kV cm^{-1} at frequencies of few kHz (Durrani and Bull, 1987). The high AC electric field produces thermal phenomena that may increase the etch rate as well as electrical phenomena that lead to electromechanical stresses in the polymer and even to electric breakdowns. In the first stage of the ECE, the most important process is the formation of the initial cone according to the CE mechanisms described in the previous section. Once the conical track has been formed, the electric field at the tip of the conductive needle can be many times greater than that applied to the remaining detector material. The very high localised stress gradients can initiate electrical phenomena, which, when combined with the chemical action of the etchant, produce tree-shaped damage at the track tip. Figure 4.4 shows one example of these tree-types of damage tracks induced by the plutonium α -emissions in CR-39 (Tommasino, 1997). There have been many attempts to understand the basic processes that drive the ECE, but all of them are simplified qualitative models (Al-Najjar et al., 1979; Tommasino, 1980; Lück, 1986). To date, there is not any theoretical quantitative study that allows its comparison with experimental results. This is consequence of the fact that a detailed knowledge of the mechanisms that drive the processes occurring in an ECE is not available: effect of electric breakdowns in dielectrics due to small penetrations of conductor fluids inside the dielectric medium. However, due to the huge

number of experimental studies already carried out around the world, there is a detailed knowledge of the ECE properties. The most relevant parameters of this technique and their effects are described in the next section.

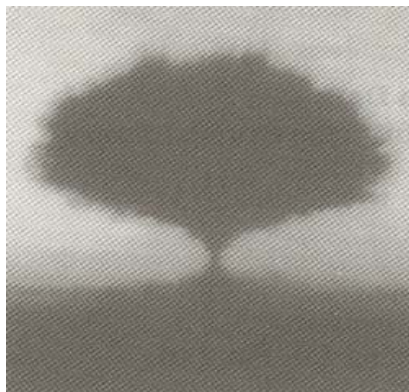


Figure 4.4. Example of the tree-type damage tracks induced by the plutonium α -emissions in CR-39 — adapted from Tommasino (1997).

4.4.4. Relevant parameters

Electric field strength

As we have said above, the presence of the electric field is crucial to magnify the tracks to the desired size. The increase of the applied voltage means a stronger etch and, therefore, can improve the detection efficiency (Sohrabi, 1987). However, there are some limitations to the value of the applied voltage, since a too electric field strength can make perforations in the detector foil producing short-circuits. Another inconvenient of applying a higher voltage is that it normally increases the background track density. Commonly, the ECE electric field strength ranges from 20 to 40 kV cm⁻¹.

Type of plastic detector

The detector plastic constitutes the dielectric in the etching process and, hence, is a determinant element. An important characteristic is its thickness, since, the wider is the foil, the stronger is the etching that it can resist without being perforated. It must be taken into account that the use of a wider plastic reduces the electric field strength unless the effective voltage is increased.

A good plastic detector should have a good response or sensitivity, i.e., the track etch rate (v_T) should be clearly higher than the bulk etch rate (v_B) and its energy threshold appropriate for the purpose of the particular study being carried out. It is also important

for the plastic to be as homogeneous as possible, since the presence of inhomogeneities will be manifested as an increase of the background tracks. Obviously, the more transparent is the plastic detector, the very easier will be the track counting.

Frequency

The effect of the ECE frequency applied is to magnify the tree-type damage tracks (Wong and Tommasino, 1982; Lück, 1986). An increase of the ECE frequency means an increase of the tree diameter and of the track density, while the tracks produced by low ionising particles are not affected. Typical values of ECE frequencies are of the order of kHz.

Etching temperature

Increasing the etching temperature augments the kinetic energy of the ion components within the etchant and, therefore, their mobility making the etching stronger. As said above, the common etching temperatures are in the range between 25 and 70 °C, due to the technical difficulties for keeping low temperature and to the plastic deformations that may happen in a too hot environment.

Type and concentration of the etchant

The effect of increasing the etchant concentration is to increase the bulk etch rate. The most common etchants are alkaline or acids, being the alkaline perhaps more widely used. In the case of Makrofol detectors, the most common etchant used is KOH aqueous dissolution, since a dissolution of NaOH has the inconvenient of the sal sodium precipitation on the detector surface that limits the uniform etching at high NaOH concentrations (Torrelles et al., 1988). This effect may occur also when using high KOH concentrations. The difference is that the precipitates of the sal potassium is more soluble in water than that of the sal sodium. Likewise, the sal potassium precipitation effect appears only for even more higher KOH concentrations.

Ethyl alcohol concentration

The reactivity of the etchant can be enhanced considerably by adding alcohol to the aqueous dissolution because it increases the mobility of the etchant ions, solves the etched molecule fragments of the polymer detector and removes them from the chemical reaction zone (Tommasino and Harrison, 1985). Normally, high bulk etch rates are achieved for intermediate ethyl alcohol concentrations. A small percentage of alcohol (below 10% of the total etchant volume) in the etching dissolution has almost no effect on the chemical

etching. On the other hand, if the alcohol concentration is too high (above 90% of the total etchant volume), the absolute quantity of etchant ions is too small to etch the plastic detector.

Bulk etch rate

In general, it is of interest to choose a bulk etch rate as high as possible to reduce the required etching time, provided that the response of the detector is kept optimum. The bulk etch rate is approximately independent on the applied voltage and frequency; while depends strongly on the etching temperature, on the percentage of alcohol, and on the type and concentration of the etchant.

Etching duration

The importance of the etching duration is given by the fact that it determines in conjunction with the bulk etch rate the removed layer. Normally, five, six or even more hours are necessary and the optimisation of the etching conditions is required to limit the etching duration to a working day. If the etching duration is long enough, then the percentage of alcohol may be reduced in a significant way by the evaporation process, so that, the bulk etch rate may not longer be considered as constant.

Removed layer

This is a key parameter to be taken into account when optimising the etching conditions. The range of the ionising particles in a given medium is a function of their mass, energy and charge. When we are interested on a given type of particles (as α -particles in our case) penetrating a given material, all the parameters are fixed but the incident energy. Hence the range will be function of the particle incident energy. Since α -particles deposit most of their energy at the end of their trajectory, the main damage will be located there. Then, if we want to register α -particles of a given energy, it is necessary for the removed layer to be of the order of their range within the detector medium. Consequently, the removed layer determines a lower energy threshold of the α -particles that are detectable in the case of using the ECE technique.

4.4.5. Different ECE techniques

The ECE etching often consists of two or even three sequential steps. In this section, we describe the different etching techniques commonly used to electrochemical etch the NTDs.

Pre-etching

It consists of removing a certain NTD layer by means of a standard CE for a given period before applying the AC electric field (Hassib, 1979; Al-Najjar et al., 1989; Sohrabi and Sadeghi, 1991). The pre-etching technique cleans up scratches on the detector surface, reduces the background track density and, perhaps the most important advantage, allows the discrimination of tracks developed by lower penetrating α -particles. These tracks are erased because, when the ECE starts, they are already in the transition or spherical phase and the electric field does not reach the high values as in the conical phase. The size of these tracks does not increase by the electrical treeing phenomenon and remains unchanged with microscopic dimensions. As we will see in Section 8.2.2, this technique offers the possibility of doing α -spectrometry using electrochemically etched NTDs, being the lower α -energy threshold regulated by the duration pre-etching and the higher α -energy threshold by the total removed layer. These threshold levels define the range of detectable α -energies, which is commonly known as the detector α -energy *window* response. In general, the response of electrochemically etched Makrofol detectors can be produced with different α -energy ranges and widths, if the CE and ECE are properly regulated.

Two-step ECE

This technique consists of two sequential electrochemical etchings: a first low frequency ECE (of the order of few hundred Hz) acting as a *memory effect* followed by a high frequency ECE (typically of several kHz) (Piesch et al., 1991). In this case, the main objective is opposite to the pre-etching, since the low energy tracks, which are normally lost during the CE, now will be visualised. The low frequency during the first step produces small trees in these tracks that prevent them to be rounded enough to be erased in the second step ECE. Therefore, we will be interested on this technique when we look for a wide energy window and for a high detection efficiency.

Post-etching

This technique consists of applying again a chemical etching to the NTDs, once the electrochemical etching is finished. By keeping the electrochemically etched detectors in the etchant for a given time, the tracks become more circular and a small spot appears in their centre. This spot is of great utility when it is necessary to distinguish between the overlapped tracks.

