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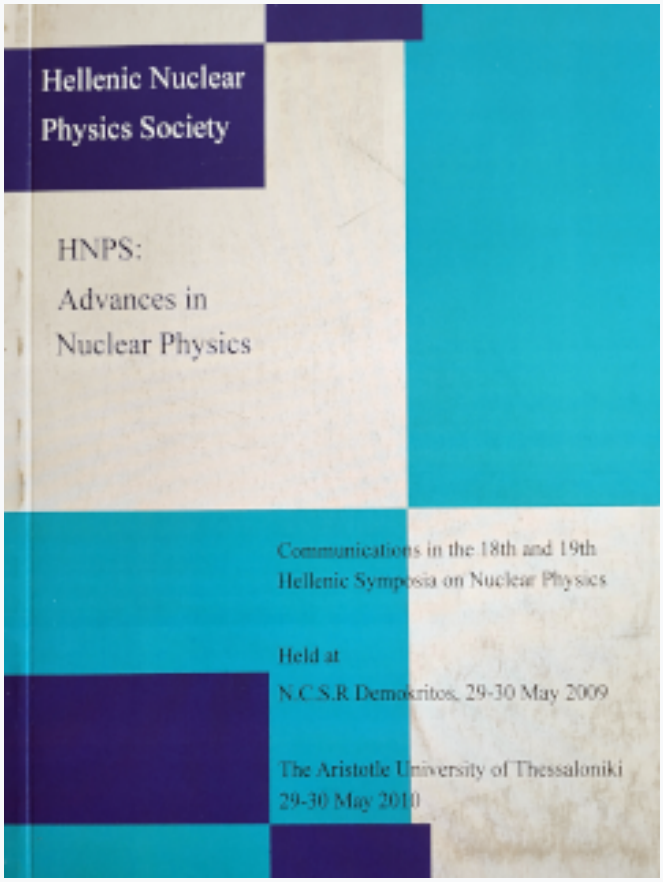

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Detection of plated-out radon progenies on CR-39 detectors

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Abstract

A spectrometric study of alpha particles using CR-39 solid state nuclear track detectors is described. It is based on the application of software imposed selection criteria, concerning the geometrical and optical properties of the tracks which are created by alpha particles of specific energy falling on the detector at given angles of incidence. These selection criteria are based on a preliminary study of tracks' parameters (major and minor axes and mean value of brightness), using the TRIAC II code. Since no linear relation was found between the energy and the geometric characteristics of the tracks (major and minor axes), we resorted to the use of an additional parameter in order to classify the tracks according to the particles' energy. Since the brightness of tracks is associated with the tracks' depth, the mean value of brightness was chosen as the third parameter. This method could be applied as a low cost technique in studies concerning the radon's daughters ^{218}Po and ^{214}Po behavior in air.

Key words: Radon, polonium, radio-ecology, SSNTD, TRIACII.

1. Introduction

Radon and its polonium daughters (^{218}Po, ^{214}Po and ^{210}Po) are alpha particle emitters. Solid state nuclear track detectors are widely [1] used in radon measurements through the detection of the emitted alpha particles. The number of tracks recorded per unit area provides information on the radon radioactivity concentration of the investigated environment. Although this information is valuable for radiological studies, counting only the total number of tracks is not useful to differentiate these radioisotopes and thus study their behaviour. However, the tracks' geometrical and optical characteristics convey information on the particles' energy and angle of incidence. Following a standard chemical etching procedure, the shape of the created tracks in the special case of vertical particle incidence is circular, whilst it is elliptical in the general case. The size of the tracks depends both upon the energy and the angle of incidence. Geometrical models concerning the growth of the tracks' length predict that there is no linearity between the axis (major and minor) of elliptical tracks

and the particles' energy. As a result, the identification of radon daughters with an alpha spectrometric method, based on the use of SSNTDs is not a trivial task [2–9].

In the present work, using the TRIAC II code [10], a comprehensive study of the major axis, the minor axis and the mean value of tracks' brightness is presented. The results verify in agreement with previous research [11–14], the geometrical models' predictions, showing the absence of a linear relation between the tracks' openings with energy. The behaviour of those parameters was investigated for a wide range of particles' incidence angles and for different chemical etching durations, ranging from 6 up to 14 h. As it will be elucidated below, increasing the etching time, the elliptical tracks' shape gradually converts to circular and a linear relation between the mean values of brightness with energy can be established. Although this is an important finding, which can be applied for spectrometry, etching for 14 h or more outgrows tracks and the resulting overlapping hinder the schematic differentiation of the tracks. Even when an image analysis program, which can analyze overlapping tracks, is used, the overlap of tracks sets an upper limit to the measurement of the surface concentration of the tracks, approximately 250 tracks/mm2. Outgrown overlapped tracks create a continuous band, thus an accurate estimation of the geometrical parameters is impractical. Finally, using the results of tracks' parameters analysis, a method to separate the tracks of deposited on the detectors ^{218}Po and ^{214}Po is proposed. Deposition is one of the main removal mechanisms of airborne aerosol particles in addition to ventilation and decay for radioactive aerosols. While ventilation and decay are well understood and usually easy to quantify, the understanding of the deposition process is still not adequate. Moreover, deposition depends on the properties of the aerosol particles such as size, charge, shape and condition of internal surfaces and also on air flow. The method described here is a method for the direct detection and differentiation of deposited radon's progeny and it could be applied as an inexpensive tool to the understanding of the deposition processes and the behaviour of radioactive aerosols.

2. Experimental methods.

The CR-39 detectors used in the present study were supplied by Pershore Mouldings Ltd. (Worcestershire, UK), with a nominal thickness of 1000 μm. The cut detectors (1x10 cm^2) were systematically irradiated inside a vacuum chamber (10^{-2} torr) with alpha particles and energies ranging from 1.5 MeV to 5.5 MeV. An americium (^{241}Am) source (main energy 5.48 MeV) was employed in combination with number of energy absorbers. The detector irradiation setup consisted of a source mounting, an absorber holder and a narrow cylindrical tube made of Plexiglas acrylic material, acting as a beam collimator. The collimator was placed on a rotator, with the exit a few millimetres close to CR-39 detector. Rotating the collimator, the angle of incidence to the detector's surface was varied from 90^o (vertical incidence) up to 35^o, in steps of 5^o. As it was found after some test runs, no tracks were recorded for angles smaller than 45^o and chemical etching times up to 8 h. Angles of incidence smaller than 45–40^o are close to the critical (or registration) angle. This is the minimum angle of incidence with respect to the detector's surface in which tracks can be formed by the chemical etching

of the detectors. The initial energy of the alpha particles was reduced by absorbers. Three different materials (kapton, polyethylene, aluminium) were used in the form of thin foils. The energy of the particles falling on the detector's surface was determined by an alpha spectroscopy system based on a surface barrier silicon detector. The detectors were etched in a 6 N aqueous NaOH solution, maintained at 75° C in a water bath with a temperature control better than 1° C. Following the first 6h of etching, the detectors were removed from the etchant, were rinsed with de-ionized water and finally they were dried in an oven at 50° C overnight. Then a number of images of the detectors' surfaces were captured with the use of a microscope – video camera – frame grabber – computer recording arrangement. For each combination of energy-angle-chemical etching duration, a number of 200–400 tracks were analyzed from three CR-39 detectors, thus achieving adequate statistics. Following the completion of the image acquisition step, the same detectors were etched for another hour and their tracks were again recorded and analyzed. This procedure was repeated until the detectors' etching time totaled 14 h.

3. Results and discussion.

3.1. Spectrometric study.

The association between the geometrical characteristics (major axis indicatively) of the tracks and the energy of the alpha particles for different etching times is presented in figure 1. The parameters of major and minor axis did not correlate with the energy of the particles for all angles of incidence. It is notable that after the first steps of etching (6–7 h), wider tracks are created by particles of lower energies. However, this feature is not sufficient for classifying tracks according to their energy. For example, particles with the energy of 5.5 MeV, which impact vertically onto the detector's surface, produce tracks with the same value of major (or minor) axis as the particles of lower energy do, when they collide with the detector at smaller angles of incidence. Moreover, as the

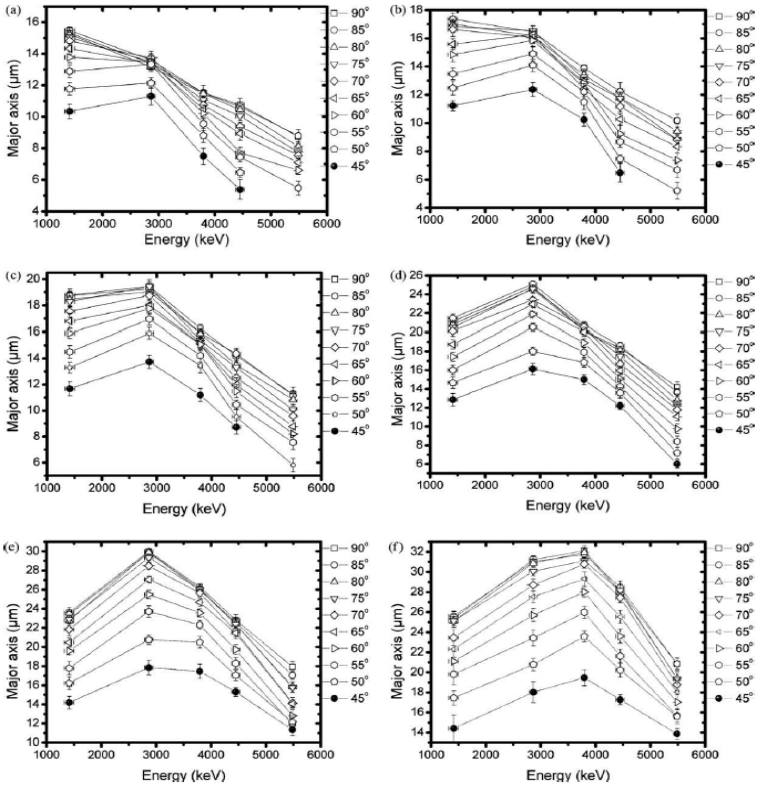

Figure 1: The relation between the major axis and the particles' energy for etching duration: (a) 6 h, (b) 7 h, (c) 8 h, (d) 10 h, (e) 12 h, (f) 14 h.

duration of the etching process is increased, particles with greater energies gradually produce wider tracks' sizes, however more particles with different energies and (or) angles of incidence concurrently register into tracks with the same value of major and minor axis. The visualization of tracks is carried out by placing the detector between the source of light and the microscope's objective lens. A ray of light transverses the detector following a straight line trajectory, which is redirected, when it is refracted out in air. The change in direction is due to the change of the optical medium and depends on the angle of refraction. Since there is not a preferred angle, the total contribution to the brightness of the optical field is almost uniform. However if before exiting, a light ray enters a region where a track was developed, a series of optical phenomena like refractions and reflections affect its final deflection. This is the reason explaining why the particles' tracks seem darker from the detector's surface and can be recognized from the background of the image. To study the brightness of the tracks, the mean value of tracks' brightness was estimated by the TRIAC II program for all the combinations of incident particle energies-angles and chemical etching durations. The association between the mean brightness of the tracks and the alpha particles' energy for different angles of incidence and etching durations is presented graphically in figure 2. Following digitalization, brightness was expressed by a value in the range 0–255. A brightness value of 0 corresponded to total black, while a value of 255 represented maximum illumination of the optical field. Then, as the duration of etching is increased, the tracks from particles with greater energies register darker. After 14 h of etching, a close to linear relation between brightness and particles' energy, referred to the same angle of incidence was found. Taking into account the evolution of this parameter with etching, additional etching was found to improve the linearity. Although this finding is important for spectrometric purposes, the outgrowth of tracks sets an upper limit to the tracks' surface concentration. For our experimental setup, this limit was 250 tracks mm^{-2}. More tracks would lead to an extensive overlapping of the tracks, thus affecting the accuracy of the measurements.

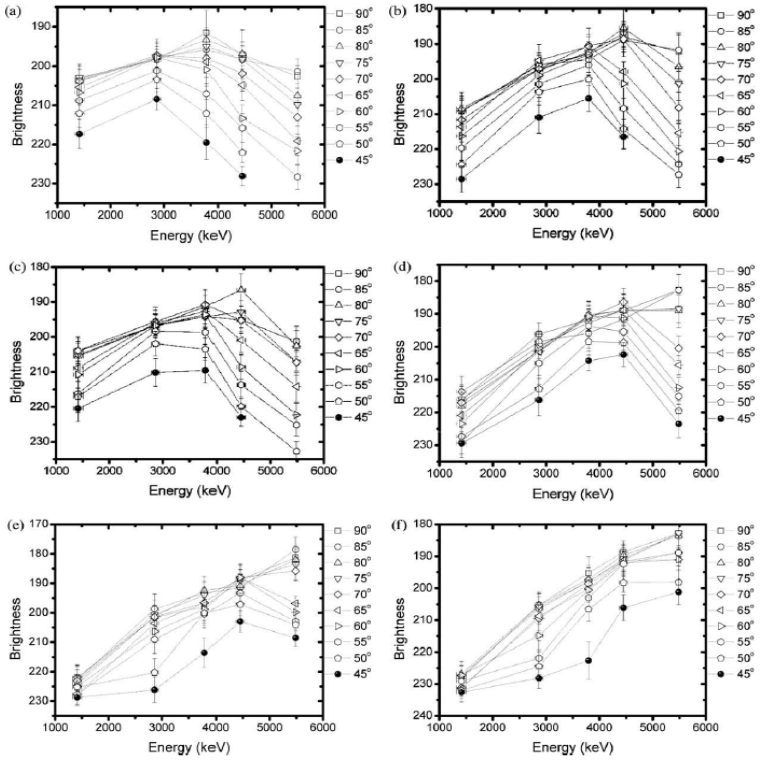

Figure 2: The relation between the brightness and the particles' energy for etching duration: (a) 6 h, (b) 7 h, (c) 8 h, (d) 10 h, (e) 12 h, (f) 14 h.

3.2. The discrimination of deposited ^{218}Po and ^{214}Po, as an application of the spectrometric study.

As an application of the described spectrometric study, a method of direct detection and separation of the deposited radon's progeny ^{218}Po and ^{214}Po was tested. After the generation of radon's daughters in air, these isotopes are found at three states, as attached to atmospheric aerosols, as unattached (free) and as deposited nuclei on available. Their deposition is an important removal process and many studies concerning the estimation of the portion of the deposited progeny have been proposed. The method, which is proposed in the present study, depends on the direct detection of deposited isotopes ^{218}Po and ^{214}Po, considering as deposition surface, the surface of CR-39 detectors and using the results of the spectrometric analysis. The radon progeny, deposited on the detector's surface emit alpha particles, which impact on the detector with their entire energy; in particular, ^{218}Po emits alpha particles with initial energy of 6.0 MeV and ^{214}Po particles with initial energy of 7.7 MeV. A number of experimental problems had to be solved since our experimental data were ranging between 1.5 MeV and 5.5 MeV. An approach to overcome these complications was to shield the detectors with an appropriate absorber so to reduce the particles energy. A number of calculations were performed using the code SRIM to determine the kind and the thickness of the absorber material. An aluminium foil of 15 μm thickness was found optimal for our purpose. Close enough; the thickness of the commercially available aluminium foils was estimated equal to 14.2 μm. This absorber reduces the energy of the emitted particles from ^{218}Po to 3.6 MeV and from ^{214}Po to 5.8 MeV.

Two CR-39 detectors, one of them covered with commercial aluminium foil, were placed for 60 h inside a non ventilated chamber, with a volume of 67 L, together with a ^{226}Ra radioactive source of 4000 Bq activity. To monitor the exposure of CR-39 detectors, a Continuous Air Monitoring (CAM) PIPS detector was placed inside the chamber, capable to operate under environmental conditions. The spectra from this detector, acquired using standard electronics were analysed to validate the results of the proposed method. Following the exposure in the radon rich environment, the CR-39 detectors were etched in a 6 N aqueous NaOH solution, maintained at 75^{o} C for 8 h. A number of images containing a total of 3500 tracks from each detector was scrutinised by the TRIAC II program. The results of the analysis are presented in figure 3. The energy spectrum of the detected alpha particles accumulated using the CAM-PIPS detector is presented in Fig. 3(a). This is

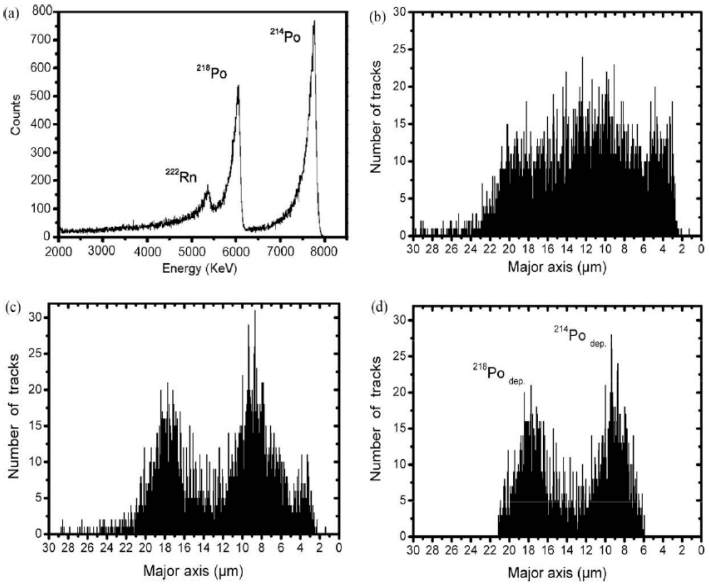

Figure 3: The results of the exposure to a radon rich air environment.

a typical radon environment spectrum, where the deposited progeny onto the detector's surface produce two well separated peaks, while radon, remaining free in air, emits alpha particles from a region around the detector, producing a continuous spectrum up to 5.5 MeV. Histograms of the measured major axis values from the registered CR-39 detector tracks are also presented for the uncovered detector (Fig. 3(b)) and for the covered with the absorber detector, before (Fig. 3(c)) and after (Fig. 3(d)) the application of the energy selection criteria. Comparing the histograms derived from the uncovered and the covered detectors, two groups of values for the major axis are recognized from the uncovered detector tracks. This is due to the deposition of the polonium isotopes onto the absorber surface in combination with the appropriate kind and thickness of the absorber, which allow the formation and recognition of tracks from particles of those isotopes under the chosen chemical etching conditions. The histogram of Fig. 3(c) was obtained by all the recognised tracks and neither selection criteria nor other discriminative procedure was applied. After imposing the selection criteria, the noticeable regions remain, providing an evidence of the radon's progeny differentiation.

4. Conclusions

The method described here for the detection and differentiation of the alpha emitter radon's progeny (^{218}Po and ^{214}Po) was based on the spectrometric analysis of the geometrical and optical characteristics of the particles' tracks. This study focused on the CR-39 solid state nuclear track detector has shown the absence of a linear relation between both major and minor axis of the tracks with the particles' energy. Instead, a linear relation between the mean value of brightness and energy is established after a prolonged chemical etching (greater than 14 h). The usage of this finding is limited by the maximum surface concentration of the tracks, due to tracks overlapping.

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