Particularization of alpha contamination using CR-39 track detectors

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Abstract. Solid-state nuclear track detectors have found wide use in various domains of science and technology, e.g. in environmental experiments. The measurement of alpha activity on sources in an environment, such as air is not easy because of short penetration range of alpha particles. Furthermore, measurement of alpha activity by most gas ionization detectors suffers from high background induced by the accompanying gamma radiation. Solid state nuclear track detectors (SSNTDs) have been used successfully as detecting devices and as a passive system to detect alpha contamination on different surfaces. This work presents the response of CR-39 (for two types) to alpha particles from two sources, ^{238}Pu with energy 5 MeV and ^{241}Am with energy 5.4 MeV. The methods of etching and counting are investigated, along with the achievable linearity, efficiency and reproducibility. The sensitivity to low activity and energy resolution are studied.

1. Introduction

Work has been carried out for several years at Bristol University to optimize the formulation of CR-39 type plastic for uniform responsiveness to alpha particles, and to develop an automated technique to calculate track length (range) from geometric parameters of etched alpha tracks measured under the microscope [1–4].

Solid state nuclear track detectors (SSNTDs) have been the subject of immense commercial and scientific interest. Since their discovery, SSNTDs have been investigated by numerous researchers in a wide variety of applications [5]. On the other hand, the application of radiation in polymer technology is of great importance with a view to achieve some desired improvements in polymer properties [6].

SSNTDs have been used successfully as detecting devices for radon, neutrons and heavy ions [7–16]. A new possible use of plastics such as CR-39 as SSNTD's has been explored to monitor low-level alpha particle contamination of solid surfaces. SSNTDs can be placed in contact with the surfaces to be tested for contamination.

Since SSNTDs are completely insensitive to gamma radiation and beta particles, the detector, in the presence of mixed fields, registers only the alpha particles. This avoids the high background problem faced by other detectors. In addition, these plastic detectors can be cut into sizes and shapes according to the specific area that has to be monitored. In relatively short exposure times, monitoring of specific activities in the order of Becquerels or pico-Curies per gram is feasible. It is possible to identify isotopes of different alpha decay energies by employing a suitable chemical etching process.

The fundamental physical phenomenon is the deposition of alpha particle energy in a polycarbonate and the formation of a cone along the particle trajectory in the material after a specific chemical process. The cone thus formed is determined by the relationship between its geometrical size and the energy deposited. The surface of nuclear track materials observed under a microscope, after chemical etching for the same time and under the same conditions, shows elliptical conical cross sections of different axes. By measuring the number and diameters of the etched pits, a track diameter distribution can be obtained. Using this procedure, the alpha particle can be identified and consequently also the radioisotope. The influence of source characteristics on the alpha particle energy spectra has been reported in a number of papers. For example, resolution and peak tailing were recently investigated as a function of sample thickness [17]. In the present work we study the response of CR-39 (for two types) to alpha particles from two sources, ^{238}Pu with energy 5 MeV and ^{241}Am with energy 5.4 MeV to use this detector to monitor low-level alpha particle contamination of solid surfaces, and energy resolution is also investigated to identify the energies of alpha sources.

2. Experimental details

2.1 *Sample preparation*

If several nuclear track materials are available, a selection must be made taking into consideration the energy response of the material and the possible applications. For this experiment two types of CR-39 detector were used covered with 150 μm polyethylene. This polyethylene cover protects the detector material from interaction with radon during the transport and storage of the CR-39. This protection layer is removed the moment the process is started. For each detector material, even when it is of the same polymeric compound, the optimal etching conditions must be determined. For this work, we have selected the polymer commercially named CR-39 (poly allyl diglycol carbonate, PADC) from two different producers. One is from Tastrack (TT) with thickness 1.5 mm and the other from Intercast (IC) with thickness 1 mm.

2.2 *Alpha particles irradiation*

Irradiation was performed in air using two alpha sources, ^{238}Pu with energy 5 MeV and ^{241}Am with energy 5.4 MeV, with a 1 mm aluminium collimator placed between

the detectors and the sources. For reading we used only normal incidence alpha particles on the material detectors. In this work we propose a single step chemical etching. This process is optimized in such a way that it allows the observation of some of the characteristics of the particles that have interacted with the material originally via the signature left by the ionizing radiation. Using a 6.25 N NaOH solution at $70 \pm 0.1°$C, the chemical etching process for different time intervals was studied because of the different apparition times of alpha particles.

2.3 *Energy resolution*

To determine the energy resolution, this experiment was investigated in a separate study. In this part, we need to express the energy as a function of the diameter, $E = E(D)$. The displacement δE is carried out by means of the chain rule [18] of the calculus

$$\delta E = \frac{\delta E}{\delta D}\delta D. \tag{1}$$

Equation (1) can be written in a different form by using the approximation

$$\delta E/\delta D = \frac{(E_2 - E_1)}{(D_2 - D_1)}. \tag{2}$$

As a consequence of this approach, eq. (1) takes the form

$$\delta E/E = \frac{(E_2 - E_1)/(D_2 - D_1)}{0.5(E_2 + E_1)}\delta D, \tag{3}$$

where E_1 and E_2 are the energies of incident alpha particles, D_1 and D_2 are the mean track diameters corresponding to each energy and δD is the width of the diameter distribution. D_1, D_2 and δD are determined by fitting the data for the track size distribution.

In order to evaluate the response of the detector material (CR-39, TT), wide intervals of energies and resolution with the following energies 2.5, 3.25, 4.12, 4.86 and 5.2 MeV are examined. Irradiation was performed in air, with a 1 mm aluminum collimator placed between the detectors and the sources. For reading we used only normal incidence (90°) alpha particles on the material detectors.

The CR-39 chips were peeled and pre-etched in 6.25 M NaOH solution at 70°C for 2 h, prior to being exposed to the alpha particles. With this chemical treatment irregularities, contaminants and scratches are eliminated from the surface of the material. After the pre-etching, all detectors were washed in distilled water and dried in air. The detectors were irradiated and chemically etched in a 6.25 M NaOH solution at 70°C for periods of time ranging from 4 to 20 h. After the chemical etching, the detectors were again washed in clean running water for 10 min and each was sandwiched to be dried in desiccant paper.

To obtain the track diameter distribution and track density, the etched detectors were read and measured using an automatic digital image analysis system (LEICA), consisting of a DMRE optical microscope with 600× magnification, a
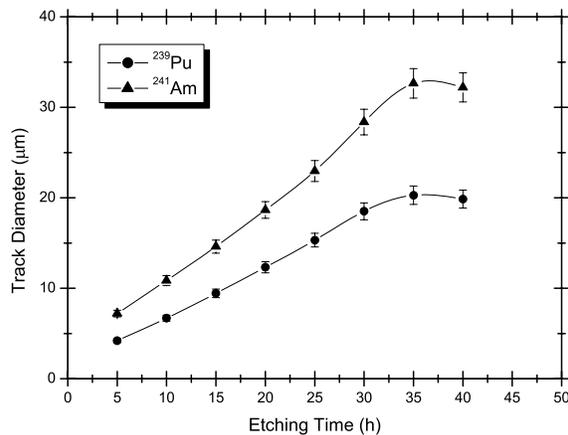
Figure 1. The track diameter as a function of chemical etching time for CR-39 (Intercast type) exposed to ^{239}Pu and ^{241}Am alpha particles.

charge-coupled device (CCD) camera and digital image electronic card in a PC as hardware and the LEICA WIN program as software. This system was equipped with motorized x–y stage and auto focus option controlled by special program operated under Windows 98. This system allows us to analyse the object structures with high spatial resolution. Two factors were considered: first, only circular tracks are measured, where the relationship of minor d and major D diameters d/D is between 0.9 and 1.0. Secondly, to obtain the track diameter distribution, only the major diameter was considered.

3. Results and discussion

Figure 1 shows the response of CR-39 Intercast to alpha particles from ^{239}Pu and ^{241}Am. It can be observed that the diameter of the tracks increases as a function of the chemical etching time. Another important feature to note is that the diameters of the tracks etched in this manner are of different sizes for different energies. Therefore, it is possible to define a chemical time for which the tracks can be distinguished by the counting system. For a selected etching time of 35 h the track diameters are 20.28 and 32.64 microns for ^{239}Pu and ^{241}Am, respectively. This difference is large enough for the measurement and counting systems to discriminate ^{239}Pu and ^{241}Am. The procedure may be extended to other isotopes that have differentiable alpha emission energies.

Diameters of the etched tracks were measured for all previously irradiated samples by using a microscope linked with the image analysing system LEICA DMRE. This system allows us to analyse the object structures with high spatial resolution. Figure 2 shows an image of the tracks of different sizes and diameters caused by alpha particles of different energies. Both plastic materials were exposed to same level activity of ^{239}Pu and ^{241}Am sources with same geometry. Figure 3 shows the responses of CR-39 samples from Tastrack (TT) and Intercast (IC), when exposed to the same level of ^{241}Am source. It can be seen that both CR-39 samples from
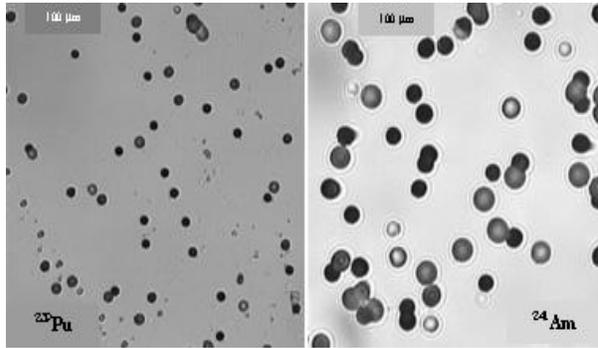
Figure 2. Microphotograph of track for two different alpha particles energies from one step chemical etching.
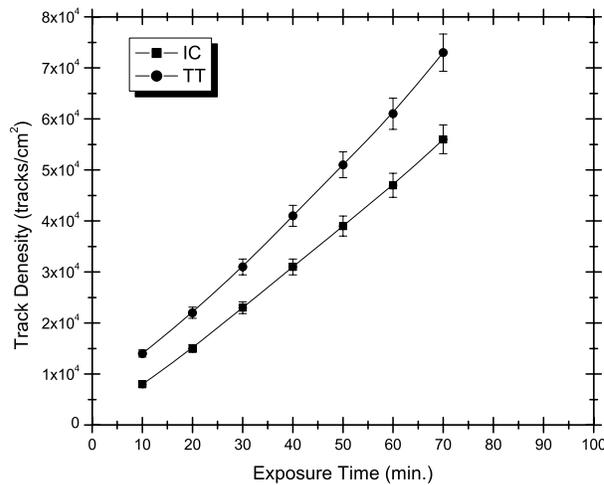

Figure 3. The dependence of responses of CR-39 (TT) and CR-39 (IC) to alpha particles with exposure time.

TT and IC show linear responses, but the response efficiencies are different for the materials, though exactly identical chemical etching and counting conditions are maintained. The unique response of each material does not allow the unconditional use of CR-39 from different makers and/or types.

The track etching efficiency is one of the most important criteria in the choice of a detector. The maximum value is found to be 85% for CR-39 (TT) and 79% for CR-39 (IC) when exposed to the same ^{241}Am source and processes for a 35 h time and read with same automated device (figure 4). Such efficiencies depend, in general, on the detector material, source energy, chemical etching time and reading system. The latent tracks with inclination less than cone angle, θ_c, (critical angle of etching) to a surface are not etched out, so in order to make the tracks visible by etching in a suitable detector surface, the tracks inclination should be greater than θ_c.
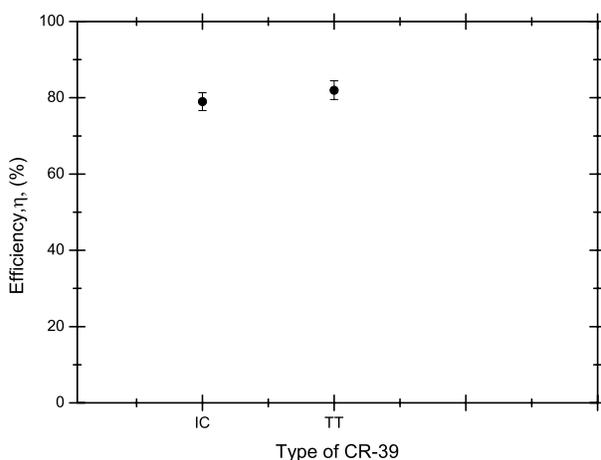
Figure 4. The efficiency of CR-39 (TT) and CR-39 (IC).

Table 1. Mean track diameter, D, for alpha particle energies after 12 h of etching time and the data for the energy resolution.

Energy (MeV)	Mean track diameter (μm)	Standard deviation	Area	$\Delta E/E$
2.5	17.869	0.44672	285.64	
3.25	16.776	0.4194	104.21	0.1794
4.12	15.459	0.38648	187.12	0.2808
4.86	14.578	0.36445	172.44	0.2405
5.2	13.647	0.34118	73.436	0.0459

In this study, it was found that the relative uncertainty in the efficiencies calculated from 150 measurements using CR-39 both from TT and IC under identical processing conditions was $\pm 3\%$. It must be pointed out that this is for laboratory conditions.

Experimentally it was observed that the track diameter distribution has a displacement along the x axis as a function of the etching time. For each etching time, from 4 to 20 h, a Lorentzian fit of the track distribution was made and the standard deviation calculated. The minimum value of the standard deviation was found for 12 h of chemical etching. Using this time as the optimum one, the Lorentzian fit to track diameter distribution for different energies are shown in figure 5.

The energies are identified as a function of the track diameter. To determine the response of the nuclear track detectors, the energies are plotted against the mean value of the respective track diameter. The response is linear, in this case from 2.5 to 5.2 MeV, as seen in figure 6. Using the track diameter distribution and the mean value and standard deviation as obtained from the Lorentzian fit, the energy resolution for different alpha-energies are calculated using eq. (3). The results are shown in table 1.
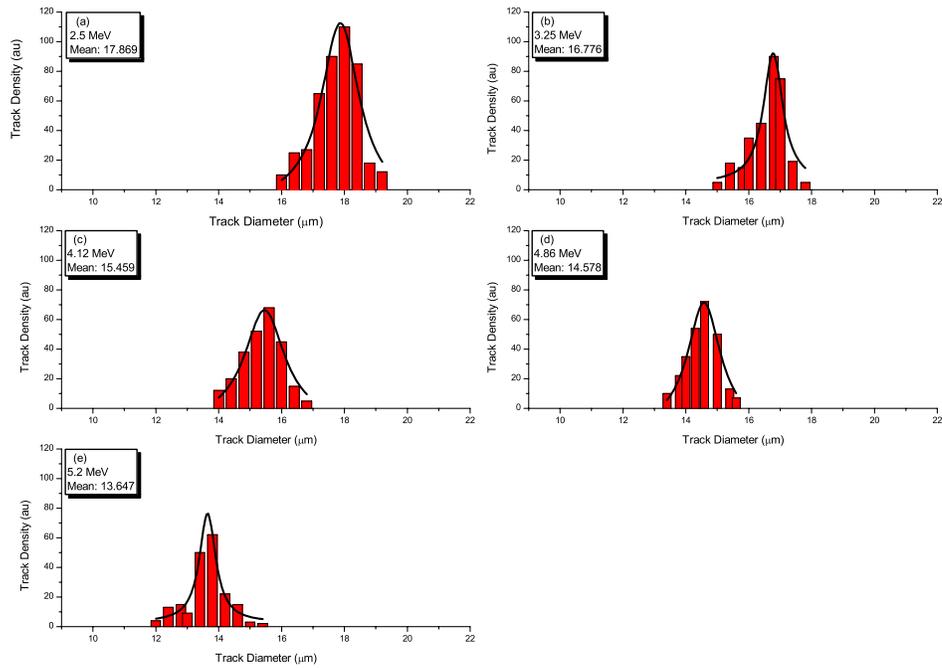
Figure 5. Track diameter distribution and Lorentzian fit for each energy of the ^{241}Am alpha source.
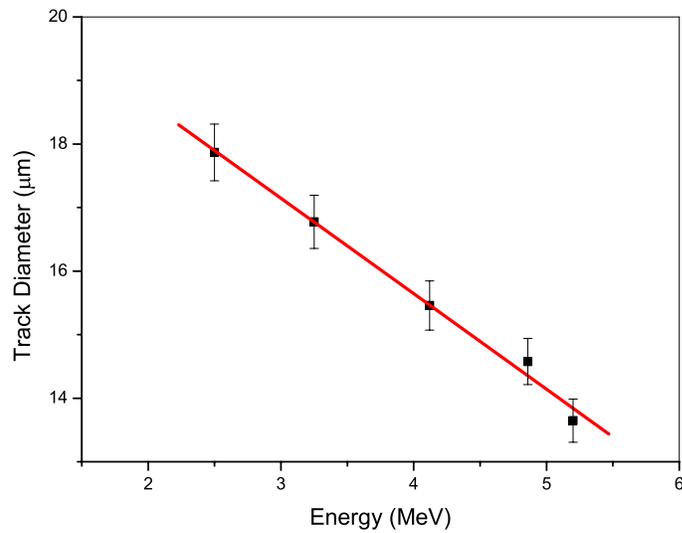

Figure 6. Particle energy and track diameter relationship.

4. Conclusions

Our study shows that SSNTD's are a new possibility for identification and measurement of alpha contaminants. Nuclear track methodology provides an alternative method for the analysis of alpha particle energy, especially in situations in which the measurements need to be *in situ* and/or in the open air. The selection of the detector material is very important and the chemical etching and reading parameters must be optimized for each type of material. The use of a single step chemical etching greatly simplifies the data handling and offers a great variety of alternatives for discrimination of alpha particle energies based on the track diameters.

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