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Passive α-particles spectrometry by polycarbonate SSNTD using new etching conditions

S.M. Hosseini Pooya [a,b,c,*], H. Afarideh [a], M. Taheri [b], M.R. Kardan [b,c]

[a] Faculty of Nuclear Engineering & Physics, Amirkabir University of Technology, Tehran, Iran
[b] National Radiation Protection Department, Iranian Nuclear Regulatory Authority, PO Box 14155-4494, Tehran, Iran
[c] Applied Radiation Research School, Nuclear Science & Technology Research Institute (NSTRI), Tehran, Iran

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ABSTRACT

By adding ethylenediamine to the common etchant composition (potassium hydroxide, water, methanol and/or ethanol) in electrochemical etching of lexan polycarbonate detectors, a passive α-particle spectrometry is resulted in the energy range of 0.8–5.2 MeV. This spectrometry is based on the track size distributions, which have a tailed normal shape and FWHM of less than 0.5 MeV. Also the energies of α-particles emitted by a ^{226}Ra micro-precipitated source and its α-emitter daughters, which were detected by this method, were in accord with that of a surface barrier detector. Consequently, this method can be developed for alpha spectrometry such as radon/thoron and their α emitter daughters' detection, separation and measurements.

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1. Introduction

Most of the alpha spectrometry methods have been performed by active methods such as surface barrier detectors. However, the solid state nuclear track detectors (SSNTDs) have a wide application in α-particle detection and measurement. Several attempts have been made to achieve α-particle spectrometry by means of SSNTDs. The first report on the fact that track diameter evolution during the etching process depends on the energy and type of the particles was made by Somogyi (1966). The phenomenon was illustrated for alpha tracks in cellulose nitrate and for fission tracks in soda glass, and was proposed for the identification of nuclear reaction products. Detailed experimental data were later published on the energy dependence of track diameters for α-particles in different kinds of SSNTDs including Makrofol E PC detector (Somogyi and Schienk, 1970). Among different kinds of SSNTDs, lexan polycarbonate (PC) has good characteristics in ECE method (Tommasinio, 2004). Discrimination of ^{241}Am, ^{239}Pu and ^{244}Cm by ECE and PC films shows been reported (Wong and Tommasino, 1982). Also several steps of ECE and CE with a nearly long duration of etching time (7 h) have been applied to extend the detection range of Makrofol PC (Al-Najjar et al., 1989). Discrimination of radon, thoron and their daughters by applying various ECE and CE processes on two PC films have been reported (Doi et al., 1992). In order to extend the range of α-particle detection and spectrometry by means of lexan PC, pre-soaking by methanol (Dadvand and Sohrabi, 1997), etching by PEMW (Sohrabi et al., 1998) and pre-etching by ethylenediamine (Dadvand and Sohrabi, 1999) have been used.

All of the mentioned researches have improved alpha spectrometry via ECE on lexan PC. But a simple method which can separate alpha energies in a wide energy range with a short etching time without any CE, pre-soaking or pre-etching processes had not been introduced. Recently Taheri and Hosseini Toudeshki (2005) introduced the new etchant composition, PEdMW (potassium hydroxide, ethylenediamine, methanol and water), to enhance the alpha particle detection ranging 0.5–4.7 MeV in lexan PC nuclear track detector. The etching time was 70 min, but the efficiency response was not flat and the responses did not show stable behavior for the lowest and highest energies of the detection range (i.e. 0.5 and 4.7 MeV). As well, the spectrometry characteristics of the new etchant were not studied in their research work.

The present work first introduces new etching conditions to improve the repeatability and energy response in tracks detection, especially in the lowest and highest energies of the extended range of 0.8–5.2 MeV. Second, it is demonstrated that considering the new etching conditions as well as tracks diameter distribution, acceptable energy separation is obtained in lexan PC nuclear track detector, which has not been reported by other investigators.

* Corresponding author at: Faculty of Nuclear Engineering & Physics, Amirkabir University of Technology, Tehran, Iran. Fax: +98 21 88009502.
E-mail address: mhosseini@aeoi.org.ir (S.M.H. Pooya).

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2. Materials and methods

Lexan PC sheets (manufactured by General Electric, USA) with 250 μm thickness (with two masked layers on both sides) were cut into 2 cm diameter pieces and used as detectors. In order to have approximately monoenergetic α-particles, a source of ^{241}Am was placed at the end of a cylindrical collimator with 0.5 cm^2 surface area (see Fig. 1). The alpha particles emitted from the source are passed through the air column in the collimator and reach the PC film surface so that its degraded energy will depend on the collimator length. Considering the air pressure of 640 mmHg and 27 °C temperature in the laboratory, the collimator length was selected as 6 mm for 5.2 MeV up to 44 mm for 0.8 MeV. The alpha energy calibration was performed with the same geometry of Fig. 1 via a surface barrier detector (Canbera, USA).

The masked layer of one side of each film was removed and irradiated by the monoenergetic alpha energies by means of the irradiation setting which is presented in Fig. 1. Then the irradiated films (without removing the other masked layer) were etched at 27 ± 1 °C temperature by the new etchant compositions. PEMEW contained 15% KOH (potash), 5% C_2H_5OH (ethanol), 30% CH_3OH (methanol), 20% $NH_2(CH_2)_2NH_2$ (ethylenediamine) and 30% H_2O (water) by weight. Three interval etching times, step 1: 1500 kV–15 min, step 2: 1300 kV–30 min, step 3: 1100 kV–55 min, were applied by a programmable PTW-FG5 AC HV power supply (PTW GmbH, Germany) at 2000 Hz. These optimum values were chosen based on both the maximum detection for tracks with the smallest size (i.e. for 5.2 MeV α-particles) and the minimum overlaps for tracks with the largest size (i.e. for 0.8 MeV α-particles) during the etching processes (Taheri, 2005).

A sample of micro-precipitated ^{226}Ra on a filter was used as an α-particle source with mixed α-particle energies. Also, the surface barrier detector was used for comparison studies.

The etched PC films images were taken by a high-resolution scanner with transparency adapter (Umax Scanner Power Look III) .The diameter for each track was calculated as the average of track size in vertical and horizontal dimensions and distribution of track diameters in each energy determined by a software (Taheri, 2005).

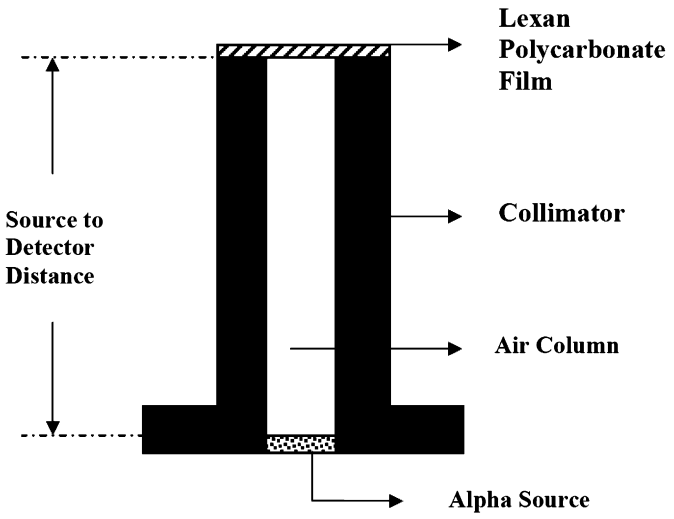

Fig. 1. The used system irradiating the PC films with monoenergetic α-particles by different source to detector distances (SDDs).

3. Results and discussion

3.1. Etching conditions improvement

The common etchant in ECE is a simple alkali (e.g. KOH or NaOH); however, the sensitivity of the etchant can be enhanced considerably by adding alcohol (methanol or ethanol) to the aqueous dissolution due to its property to increase the mobility of etchant ions, solve the etched molecule fragments of the polymer detector and remove the chemical reaction zone (Tommasino and Harrison, 1985). Taheri and Hosseini Toudeshki (2005) showed that the ethylenediamine in the etchant enhances the alpha detection energy range to 0.5–4.7 MeV. The role of both ethylenediamine and etching conditions can be explained as follows:

- Ethylenediamine increases the bulk etching rate (V_B) so that the latent tracks which are formed by α-particles with higher energies can be etched (Taheri, 2005).
- Higher electric field strength (i.e. 1500 kV in the first interval time) allows starting the treeing process immediately after applying the HV for superficial latent tracks which are formed by α-particles with lower energies (e.g. ~1 MeV).
- The latent tracks of the more energetic alphas are etched more slowly so that it takes a longer period to develop its related pits to a size. Since the etching time is constant (100 min), more number of 'treeing processes' may take place for the latent tracks which are formed by alpha particles with lower energies, and it leads to considerable differences in track sizes in various energies.

The etching time was suggested as 15 and 40 min for step 2 and 3, respectively, by Taheri and Hosseini Toudeshki (2005). If this regime is used, the maximum detectable energy would be 4.7 MeV. The detection efficiency is reduced by 40% at this energy and a lot of over-loadings (i.e. creating a hole on the film surfaces and occurring of shortcut circuit) may happen in the polymer. If the time intervals are rather increased, certainly the overloading will happen and etching process cannot be continued. So in this study the PC films were applied with its resistive masked layer to avoid any over-loading during all experiments. Consequently, it is possible to increase the duration of the second and third steps to 30 and 55 min, respectively, so that the detection range is enhanced up to 5.2 MeV. Also, applying longer etching time in interval 3 leads to better appearing of the latent tracks which are formed by alpha energies up to 5.2 MeV. On the other hand, decreasing 5% methanol concentration and adding 5% ethanol in the etchant composition leads to a stable response in 5.2 MeV. The role of 5% ethanol is that it decreases the etching velocity slightly (without any decrease in total concentration of alcohol in the etchant), so that the efficiency in the lowest and highest energies of detection range (i.e. 0.8 and 5.2 MeV) is not changed by a little change in peripheral conditions (e.g. temperature).

Fig. 2 shows the images of tracks in different energies ranging from 0.8 to 5.2 MeV and the differences between track sizes, which can be clearly seen. Track diameters are 60–330 μm corresponding to the α-particle energies ranging 5.2–0.8 MeV. As it can be seen, the majority of tracks in the given energies are isodiametric. Fig. 3 shows the diameters of the majority of tracks versus alpha energy. The standard deviation of track diameters is less than 10 μm.

In order to study the energy response of this method, the relative responses (the ratio of the number of appeared tracks in a given energy to that of 4.5 MeV as an optional reference value) were measured for the given energies of 0.8, 1.5, 2, 2.6, 3, 3.6, 4,

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