BULK ETCHING RATE ASSESSMENT FOR POLYMER TRACK RADIATION DETECTORS

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Poly(Allyl Diglycol Carbonate) (PADC) detectors are widely used in nuclear particles detection due to their high sensitivity. Particles impinging the polymer form latent tracks which are enlarged through a chemical etching process. This study presents a quantitative assessment of the bulk etching rate obtained by two approaches: (i) mass difference and (ii) fission fragments method . Bulk etching rate is the main parameter reflecting the etching procedure's efficiency and tracks development. The obtained values of this parameter are furthermore important for the tracks reading and counting processes relevant for dosimetry applications. We present the results obtained through the above-mentioned methods applied on detectors that have been exposed to Th-228 and Cf-252 radioactive sources.

Keywords: PADC, SSNTD, nuclear track detectors, chemical etching, CR-39, bulk etching rate

1. Introduction

Poly(Allyl Diglycol Carbonate) (PADC) detectors are Solid State Nuclear Track Detectors (SSNTD) used in the nuclear particles detection. They are highly sensitive at energetic charged particles (protons, light and heavy ions) [1,2]. Via recoil protons, they are also used in neutrons detection. Charged particles ionize molecules close to their path in material, generating a volume enriched with free chemical radicals called "latent tracks" [2,3]. Using these detectors for radiological or dosimetry purposes implies a chemical treatment to conveniently enlarge the tracks developed by the impinging radiation. Through the immersion of PADC detectors in a basic aqueous solution (NaOH) in controlled conditions, the detector layers will be etched and the tracks enlarged. Status, challenges, best practices, and perspectives of the research in the field are recently reviewed by e.g. Bolzonella et al [3].

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The evaluation of the etching method is an indispensable process to characterize the behavior of a PADC material following the etching treatment under specific conditions. Establishing the etching method parameters is the first requirement in building a particle dosimetry system based on solid-state nuclear track detectors [3,4]. Additionally, to harmonize the individual monitoring systems (IMS), precise values of the bulk etch rate related to the types of used detectors are required. Differences among batches of detectors provided by different suppliers have been reported [3]. With the purpose of assessing the PADC detector quality, the European Radiation Dosimetry Group (EURADOS) has created a dedicated Task Group.[5].

In the present study, two different sizes (10 mm x 10 mm x 1 mm and 20 mm x 25 mm x 1.5 mm) of film-like PADC detectors have been used. This paper reports the bulk etching rates obtained for detectors of two different geometries manufactured by Track Analysis System Ltd [6]. A graded approach has been employed to assess the etching process with different methods. Detectors were repeatedly etched and measured. Our work has been focused on the evaluation of bulk etching rate by two methods: (i) mass difference and (ii) fission fragments one [2,3].

In sections 2 and 3 of this paper we detail the experimental procedure and respectively the measuring methods. The obtained results are reported in Section 4. Finally, in Section 5 conclusions and perspectives are given.

2. Experimental procedures

2.1 Irradiation

As a general approach, for each irradiation, we have exposed two detectors placed at contact with the radioactive source to ensure the capture of alpha particles and fission fragments. Therefore, the exposure is considered to be performed at normal incidence. Four batches of detectors have been irradiated as follows: (i) Batch #1 consisting of detectors with sizes of 10 mm x 10 mm x 1 mm has been exposed for minutes (1 min to 10 min) to a Th-228 source (Λ = 20 kBq) to emphasize the tracks corresponding to alpha particles and (ii) Batch #2 consisting of detectors (with sizes of 20 mm x 50 mm x 1.5 mm) has also been irradiated for minutes (1 min to 10 min) with fission fragments and alpha particles emitted by a Cf-252 source (Λ = 20 kBq). In case of the above presented irradiations an over-exposure of the detectors has been noticed. The over-exposure is observed when the detectors cannot provide distinguishable but overlapped particle tracks after the etching procedure. A representative example can macroscopically be observed in Fig. 2.

The last two batches (batch #3 with 10 mm X 10 mm x 1 mm detectors and batch #4 with 20 mm x 25 mm x 1.5 mm detectors) have been exposed to the same Cf-252 radioactive source for several seconds (between 5 and 30), to ensure that the fission fragment tracks do not overlap and are distinguishable and therefore the fission fragments method can be applied, as presented in section 3.1.

2.2 Etching

Two of the most important etching parameters are V_T (track etching rate) and V_B (bulk etching rate) as they are defined e.g. in Ref.[2,3,7]. Their values are strongly dependent on the detector material, immersion time, etching solution molarity, and temperature. We have chosen to etch all the detectors in a 6.25 N NaOH solution as suggested in Ref. [8,9] at a constant temperature of 85 ^oC. In accordance with the literature [6,10,11,12], around 5 μm/h has been expected as value for the bulk etch rate. The etchant concentration has been checked with a hydrometer or by assessing the pH of the aqueous solution and then adjusting the basic component of the enchant.

The chemical etching process was performed in two hour long successive immersions; after every step, the detectors have been washed in distilled water for 10 minutes, then immersed in 2% CH_3-COOH solution for 30 minutes (for neutralization), washed again in distilled water for 15 minutes, and finally let for natural drying. All detectors were immersed in more than 20 l of solution to ensure that the dispersion of small plastic pieces of detectors has a quite negligible influence. To ensure a uniform concentration in the whole volume of the solution, an automatic stirrer has been permanently functioning during the chemical etching. Additional efficiency upgrades of the chemical process can be induced by increasing molar concentration, temperature or adding ethanol [6].

The detectors developed fission fragments tracks which were measured after the first two hours etching time through an optical microscope. Each batch contained two blank detectors with the scope of monitoring the tracks formed due to natural radioactivity background (see Fig. 2).

3. Measuring methods

3.1 Fission fragments technique

The fission fragments technique is based on a geometrical assessment of the tracks developed throughout the etching process. The proper developing of the tracks takes place when the etch rate along the track (V_T) is higher than the bulk etch rate V_B, a valid assumption for angles of incidence smaller than the critical angle, as it is presented in Ref. [2,12]. Assuming a constant track etch rate (V_T) as

considered in Ref. [2] for normal incident particles, the track opening (D) can be estimated according to Eq.1, where t is the etching time. The opening has been measured for the tracks formed by normal-incident fragments by using an optical microscope of 50X magnitude and the adjacent software.

$$D = 2\,t\,V_B \tag{1}$$

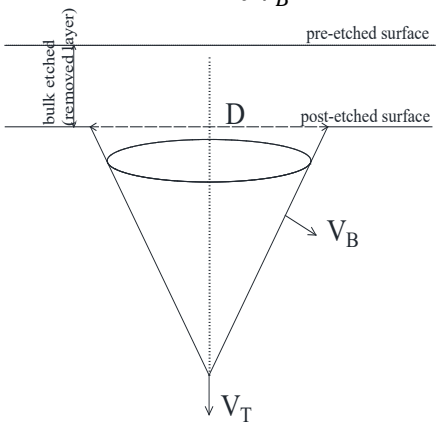

Fig. 1. Geometry of track development, where D is the track opening, V_B is the bulk etch rate, and V_T is the etching rate along the track.

The method is applied for round shaped tracks of fission fragments obtained when the radiation exposure is performed at normal incidence, accordingly to the geometrical approximations based on Fig. 1. The shape of the track is an important parameter for the image obtained in two dimensions throughout the optical microscopic analysis.

3.2. Mass difference technique

Each detector has been weighted using a 5 micrograms precision balance before starting the chemical process and after every two hour long etching step. Using the change in mass of each detector after removing layers in the etching process and assuming a specific density for each detector and a specific measured area, the bulk etch rate can be calculated as specified in the Equation 2, according to Ref. [2].

$$V_B = \frac{\Delta m}{2A\rho t} \tag{2}$$

In Eq.2, A represents the area of detector, ρ is the density of the detector and t is the etching time. Geometrical sizes of the detectors were necessary to calculate the density value for each detector. In our study, we use the obtained

values of density instead of the ones provided by the manufacturer when evaluating the bulk etching rate by the mass difference method. The devices we used for measuring the geometrical sizes consists of micrometer, caliper and optical microscope. In the optical microscope measurement, detectors thickness has been measured by cross-sectional view and by step height method. Step height method implies two different focuses of the objective lens – first on the inferior surface and the second one on the upper surface of the detector. It has been found proper to use the values of the low uncertainties measurements, therefore the values obtained from the optical microscope measurements.

The mass difference method provided accurate results only for non-irradiated detectors; in case of using this technique for irradiated detectors, the etchant interacts with a larger surface of the detector due to the new latent tracks formed by the impinging radiation. This enhanced chemical reaction of the etching process led to overestimated results for bulk etching rate.

4. Experimental results and their interpretation

4.1 Results obtained by mass difference assessment

All detectors have been weighted after every etching step with the scope of determining the mass etched during the chemical process. Mass difference method has been applied to all batches, even for the detectors that have been over-exposed (Fig. 2) which cannot be assessed through the fission fragments technique. This method was applied to provide proper values for blank detectors and to confirm the increase of etched mass as the damaged area increases too in the case of irradiated samples.

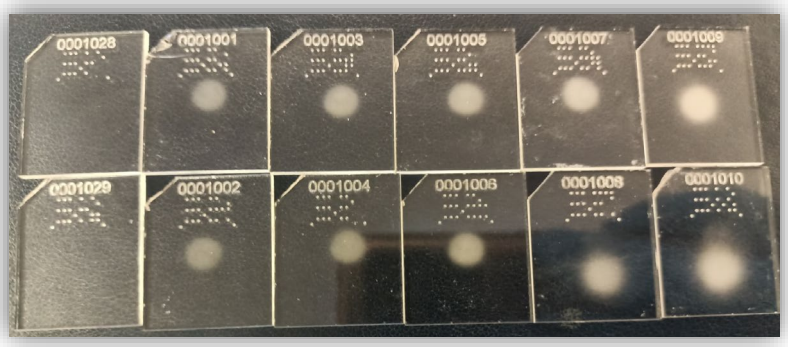

Fig. 2. Progressively over-exposed detectors (from left to the right: blank detectors follower by detectors exposed for 1 min, 2 min, 3 min, 5 min and 10 min). Picture captured after the first two hours of the etching process. As mentioned in the section 2.1, for each irradiation case, we used 2 detectors.

Table 1

Average bulk etching rate obtained throughout the mass difference method after each two hour long etching time

Batch number (as it is defined in Chapter 2.1)	Average V_B after the **first 2h** etching step [µm/h]	Average V_B after the **second 2h** etching step [µm/h]	Average V_B after the **last 2h** etching step [µm/h]
#1	6.51 ± 0.37	7.73 ± 0.81	8.24 ± 0.7
#2	4.87 ± 0.21	5.80 ± 0.17	7.18 ± 0.21
#3	4.25 ± 0.44	6.58 ± 0.57	7.82 ± 0.57
#4	2.85 ± 0.20	5.41 ± 0.14	7.08 ± 0.25

As it is presented in the Table 1, the samples have been etched in three consecutive steps, each of them of two hour long. Repeated measurements of the detectors masses have been performed before and after each etching step. An increasing trend for bulk etching rate values has been observed due to the increasing damaged area that the etchant is interacting with. Accordingly, a constant bulk etching rate could not be assumed over the entire process due to the higher quantity of material what is etched from the developing tracks.

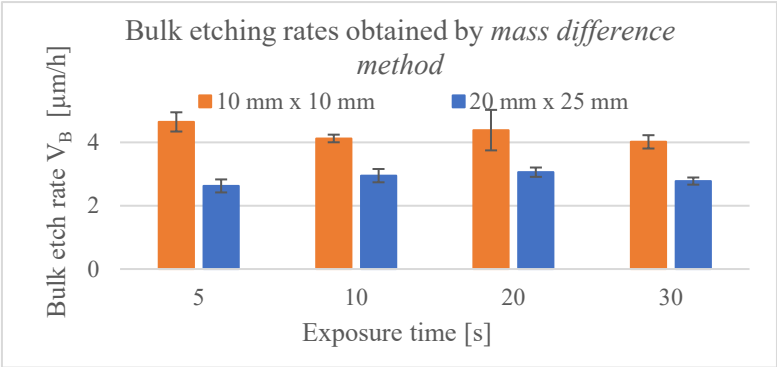

Fig. 4. Comparison between the bulk etch rates obtained by mass difference method for small (10 mm x 10 mm) and larger (20 mm x 25 mm) detectors, after a total of 6h etching time

In the Fig. 4 the bulk etching rates obtained by using the mass difference method for the smaller and bigger samples are given. The big difference in terms of etched layers during the same etching process for detectors provided by the same manufacturer is an additional argument to not use this technique for irradiated samples. On the other hand, there is a quasi-constant value among the batches of detectors with the same shape and lengths.

4.2 Fission fragments technique

The diameters of fission fragments tracks have been measured at the optical microscope at 50X magnitude. Lateral diameters of the tracks were measured for a minimum of 20 tracks uniformly selected from the etched surface. Fission fragments technique has been applied to lower exposed batches (for irradiation times between 5 and 30 seconds). For the over-irradiated detectors, we were not able to discriminate between fission fragments tracks in the central part of the irradiation spot due to the overlapping phenomena. This can macroscopically be observed in Fig. 2. The fission fragments technique was not proven to be time efficient due to the large amount of particle tracks needed to be measured.

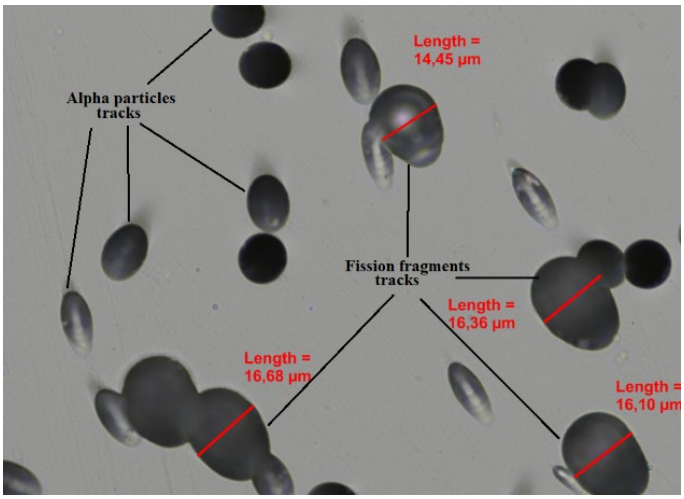

Fig. 3. Fission fragments and alpha particles tracks after two hours of etching. The measured tracks with the value written next to them correspond to the fission fragments

Clear pictures of fission fragments have been obtained as presented in Fig.3 Diameters measurement implies a small uncertainty too due to the optical approximation of tracks edges. Central fission fragments tracks are described by a well-defined oval shape and are easily distinguishable from the alpha particles tracks due to their larger size. Representative alpha particles and fission fragments tracks can be observed in Fig. 3. The darker and round shaped ones correspond to the particles impinging the material at normal incidence, while the tilted ones form brighter, thicker oval shaped tracks.

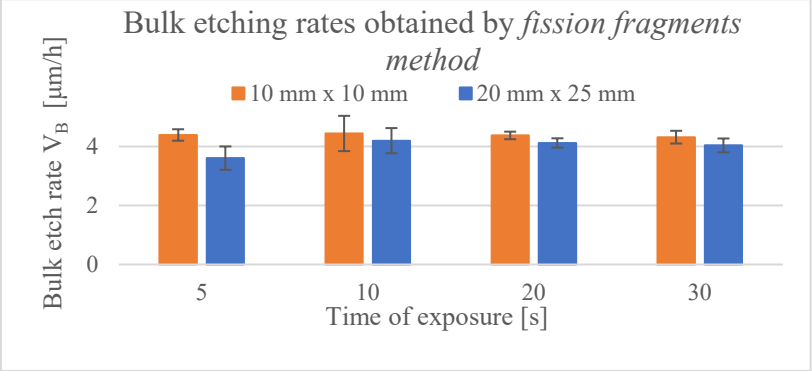

Fig. 5. Comparison between the bulk etch rates obtained by fission fragments method for small (10 mm x 10 mm) and larger (20 mm x 25 mm) detectors, after a total of 6h etching time

Small differences within the uncertainties between bulk etch rates obtained by microscope analysis could be explained by the two different optical microscopes we used. The tracks developed in the areas towards the edges of the detector have lower diameters and depths as it could be observed through the optical microscope analysis. Fission fragments range in PADC material is around 10-15 micrometers, therefore the optical analyses could be performed only after the first two hours etching step for which the removed layer is below the mentioned values. In Fig. 5, the fission fragments method results are presented, confirming the method stability independent of the detector dimensions (in opposition with the results from Fig. 4). Through this method, similar bulk etching rate values in the range of 4 µm/h have been obtained, independently to the size of the detector or exposure level.

4.3 Comparison between the results obtained by the two methods

Table 2

Bulk etching rates obtained by using mass difference and fission fragments methods

Detectors dimensions	**Bulk etch rate V_B by *mass difference method*** [µm/h]	**Bulk etch rate V_B by *fission fragments method*** [µm/h]
1 cm x 1 cm	4.29 ± 0.37	4.38 ± 0.31
2 cm x 2.5 cm	2.85 ± 0.22	4.01 ± 0.39

In Tab. 2 we present the results of the bulk etching rates obtained by using the mentioned methods for both (10 mm x 10 mm x 1 mm and 20 mm x 25 x 1.5 mm) types of detectors. Higher bulk etch rate values have been obtained for small detectors (10 mm x 10 mm x 1 mm) by the two measurement techniques. As it can

be noticed, a significant difference has been obtained between the two bulk etch rates for larger detectors, proving that mass difference method cannot be used for irradiated detectors. It is found to be inappropriate due to the amount of latent tracks formed by the impinging radiation causing larger area of interaction with the etchant. This observation is essential for dosimetry applications in order to perform the specific etching process related to the type of PADC dosemeters used in the monitoring routine. In addition, the dosimetry software used in particle tracks reading should be customized to identify and count the tracks of specific dimensions.

5. Conclusions and perspectives

In this work, we have assessed the bulk etching rate for PADC detectors by two different measuring methods: (i) mass difference and (ii) fission fragments. We have found that the fission fragments one is not time-efficient due to the numerous tracks required to be measured for good statistics. At the same time, this method requires qualitatively developed tracks, with clear edges. The mass difference method is well suited to estimate the bulk etch rate for non-irradiated detectors. In case when this method is applied for irradiated detectors, higher bulk etching rates are obtained due the latent tracks development and the increased surface that the etchant in interacting with. The results obtained through the fission fragments method provided uniformity among the studied batches, independent of their size. In contrast, by using the mass difference method we have obtained different bulk etch rates for the two types of studied detectors.

The etching procedure has a strong influence on the latent tracks development, a feature important for the dosimetry applications. The results presented in Table 2 will be considered as an input information in the future etching procedures performed within the passive dosimetry system foreseen to be operated in the Dosimetry Laboratory at Extreme Light Infrastructure – Nuclear Physics. Further studies will be performed in testing and validating the passive dosimetry method used in the laboratory.

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