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Micro-collimator fabricated by alpha-particle irradiation of polyallyldiglycol carbonate polymer film and subsequent chemical etching

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ABSTRACT

In the present paper, we propose a method to fabricate a “micro-collimator” with a thickness of 15 μm . A commercially available PADC film with a thickness of 100 μm was first chemically etched with NaOH/ethanol solution to obtain a thin PADC film. This thin PADC film was then irradiated by 5 MeV α particles through a “macro-collimator” with a thickness of 5 mm and a hole diameter of 1 mm. This PADC film with latent tracks was then further etched in aqueous NaOH solution to form the micro-collimator with a thickness of 15 μm and etched-through air channels were all confined to a circular area with 1 mm diameter. Verifications were performed by checking the etched α -particle tracks formed on a test PADC film from α particles having traveled through a 20 μm support substrate attached to the 15 μm micro-collimator (both substrate and micro-collimator also made of PADC). The results are discussed.

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1. Introduction

Alpha-particle radiobiological experiments involve irradiating cells with α particles and require accurate positions where the α particles hit the cells, the latter being essential for dosimetric determination. In many of the α -particle radiobiological experiments, it is only feasible to control and quantify the α -particle energies incident on the cells if the α particles pass through the substrate to strike the cells which are in contact with the substrate, instead of passing through the fluid layer above the cells, which has a variable thickness (see Refs. [1–3]). As regards the determination of the number and energy of α particles actually incident on the embryo cells, there can be two approaches. The first one is to make use of a microbeam facility. Here the hit positions and the energies of the incident ions can be controlled. Another approach is to use a radioactive source to provide the α -particle irradiation and to use a solid-state nuclear track detector (SSNTD) as a support substrate to record the hit positions and the energies of the incident α particles. Although the procedures involved in the second approach are a little bit more tedious, they do not involve the expensive and sophisticated equipment associated with the microbeam facility. The SSNTD employed as the support substrate should be thin enough to allow passage of α particles with nominal energies (e.g., those from ^{241}Am source). A review on SSNTDs has been given by Nikezic and Yu [4].

Polyallyldiglycol carbonate (PADC) is a well-known SSNTD and is commercially available as the CR-39 detector. PADC films have gained their popularity as support substrates for cell culture in that they are transparent, biocompatible [5] and are not dissolved in the alcohol used for sterilizing the substrate. However, the thinnest commercially available PADC films are $\sim 100 \mu\text{m}$ thick and are thus not thin enough. Gaillard et al. [1] fabricated ultra-thin PADC films as support substrates for cell cultures but these films were not commercially available. Fabrication of these thin PADC films would require specialized expertise and dedicated equipment, which might not be easily available to all laboratories trying to perform α -particle radiobiological experiments. As a result, it is desirable if sufficiently thin PADC films can be fabricated from thicker commercially available PADC films, e.g., those with a thickness of 100 μm . Chan et al. [2] prepared thin PADC films from commercially available CR-39 SSNTDs with a thickness of 100 μm by etching them in 1 N NaOH/ethanol at 40 °C to about 20 μm .

For a more convenient and accurate dosimetric determination from the α -particle tracks developed on the support substrate upon chemical etching, the incident particles should all be preferably close to normal incidence with respect to the support substrate. For exceedingly oblique incidence, it will be difficult and tedious to determine precisely whether the alpha particles have actually hit the target areas. As such, it is desirable to collimate the α particles so that only those close to normal incidence can strike the support substrate. However, such a collimator should preferably be very thin to minimize the energy loss of the α particles when they travel through the air column in the collimator.

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Collimators commonly used in the laboratory might not be ideal for such purposes. For example, for these collimators, the minimum diameter of the mechanically drilled cylindrical bore is ~ 1 mm. To maintain its function to restrict α particles close to normal incidence, the collimator should have a minimum thickness of ~ 5 mm. The long air column involved will impose severe limitations in the thickness of the support substrate in such a way that the residual energy of α particles exiting the support substrate can still be useful for radiobiological experiments. For example, consider the case where we have a PADC support substrate with a thickness of $28 \mu\text{m}$. If the collimator has a thickness of 5 mm, after traveling an air column of 5 mm and passing through $28 \mu\text{m}$ of PADC, the energy of the α particles would have dropped to ~ 150 keV, which would be no longer suitable for α -particle radiobiological experiments. However, if a collimator has a thickness of $15 \mu\text{m}$, the energy of the α particles would only have dropped to 1.23 MeV, which would still be very useful for α -particle radiobiological experiments.

In the present paper, we propose a method to fabricate a “micro-collimator” with a thickness of $15 \mu\text{m}$ through α -particle irradiation of a PADC film and subsequent chemical etching. The feasibility of the fabricated micro-collimator was also tested, and the results are discussed.

2. Methodology

2.1. Fabrication of the micro-collimator

2.1.1. Preparation of thin PADC films

Thin PADC films were prepared from commercially available PADC films (from Page Mouldings (Persore) Limited, Worcestershire) with a thickness of $100 \mu\text{m}$ using the method proposed by Chan et al. [2]. The $100 \mu\text{m}$ PADC film were first cut into pieces with a size of $1.8 \text{ cm} \times 1.8 \text{ cm}$. These were then chemically etched in a 1 N NaOH/ethanol solution at 40°C , for which the bulk etch rate was about $10 \mu\text{m/h}$ [6]. During the chemical etching of PADC films in NaOH/ethanol , the byproduct sodium carbonate was accumulated on the surface of the films, which would slow down the etching rate [6]. Therefore, the films had to be rinsed with distilled water every two hours to ensure an efficient etching process. The PADC films were etched to a thickness of $41 \mu\text{m}$, which was monitored using a micrometer (Mitutoyo, Japan) with an accuracy of $\pm 1 \mu\text{m}$.

2.1.2. Alpha-particle irradiation of the thin PADC film through a macro-collimator

The $41 \mu\text{m}$ PADC film fabricated as described in Section 2.1.1 was irradiated by α particles with a collimator, which was a

rectangular block made of acrylic resin with a thickness of 5 mm and had a cylindrical bore with a diameter of 1 mm at the centre. We hereafter refer this 5 mm thick collimator as the “macro-collimator”, so that it can be differentiated from the collimator with $15 \mu\text{m}$ thickness to be fabricated in this work, which will be referred to as the “micro-collimator”. The $41 \mu\text{m}$ PADC films were irradiated by 5 MeV α particles through the macro-collimator for 15 min. The α -particle source employed in this work was a planar ^{241}Am source (with 5.49 MeV main energy under vacuum and an activity of $0.115 \mu\text{Ci}$). The irradiated PADC film was then mounted by epoxy (Araldite[®] Rapid, England) onto a plastic sheet with a $1.4 \text{ cm} \times 1.4 \text{ cm}$ rectangular opening at the centre.

2.1.3. Micro-collimator

Latent tracks were left in the PADC film after irradiation by α particles. Subsequent chemical etching could increase the track depth and enlarge the track opening (i.e., increase the major and minor axes of the diameter of the tracks) (see e.g., Ref. [4]). The mounted PADC film was etched on both sides in $6.25 \text{ N NaOH/water}$ with 5% alkylidiphenyloxide disulfonate (surfactant DOW-FAX 2A1, Dow Chemicals) under 70°C for 11 h, and the thickness of the PADC film became $15 \mu\text{m}$. The surfactant was added to increase the ratio between the track etch rate to the bulk etch rate [7], so that more cylindrical tracks can be formed (i.e., tracks with steeper walls). The etched PADC film with the thickness of $15 \mu\text{m}$ became our micro-collimator.

2.2. Testing the micro-collimator

To test the applicability of the micro-collimator, we employed it in an experimental setup as shown in Fig. 1(b), which mimicked a realistic one as shown in Fig. 1(a) used in the irradiation of cells or zebrafish embryos [2,3]. In the realistic irradiation setup shown in Fig. 1(a), the zebrafish embryo or cells are irradiated inside a Petri dish, with a hole at the bottom covered by a support substrate made by a PADC film. A micro-collimator is inserted between the α -particle source and the support substrate so that only those α particles close to normal incidence onto the micro-collimator can pass through. In the setup shown in Fig. 1(b) which mimicked the realistic one shown in Fig. 1(a), the zebrafish embryo or cells were replaced by another PADC film which was used to record the α particles after they passed through the support substrate, and we referred this PADC film as the α recorder. In real irradiation experiments, these α particles will strike the zebrafish embryo or cells.

In the present experiments, a thick PADC film with 1 mm thickness was employed as the α recorder, while a thin PADC film

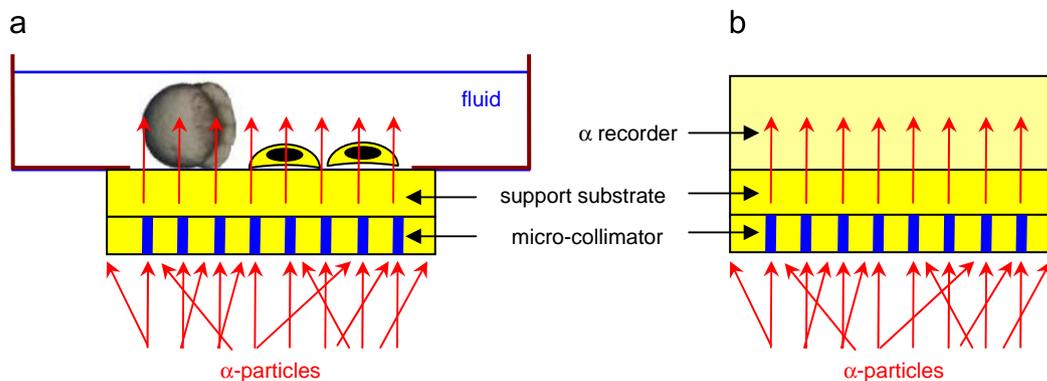


Fig. 1. (a) Schematic diagram showing the irradiation of a zebrafish embryo or cells inside a Petri dish, with a hole at the bottom covered by a support substrate made by a PADC film. A micro-collimator is inserted between the α -particle source and the support substrate so that only those α particles close to normal incidence onto the micro-collimator can pass through. (b) Same as (a) except that the zebrafish embryo or cells are replaced by a PADC film which is used to record the α particles after they have passed through the support substrate. In real irradiation experiments, these α particles will strike the zebrafish embryo or cells.

with 20 μm thickness was used as the support substrate. The number and incident angles of α particles passing through the micro-collimator were assessed by examining the tracks generated on the α recorder. The planar ^{241}Am source (with activity 0.115 μCi) was employed for α -particle irradiation.

2.2.1. Energy of α particles reaching the α recorder

Two α recorders denoted as A and B were used. Both were placed on top of a 20 μm support substrate. For irradiation of the α recorder A on the support substrate, the micro-collimator was inserted between the α -particle source and the support substrate, and irradiation was performed for 8 min. A parallel control experiment was carried out using α recorder B on the support substrate, but this time no micro-collimator was inserted between the α -particle source and the support substrate. For the control experiment, irradiation was performed for only 1 min to avoid too many tracks, which might make analyses difficult. Both α recorders were then etched in 6.25 N NaOH/water at 70 $^{\circ}\text{C}$ for 3 h to enlarge the tracks on the samples. Images of the enlarged tracks were captured from a camera attached on a microscope with magnification 200 \times . The diameters and the optical appearance of the tracks were examined.

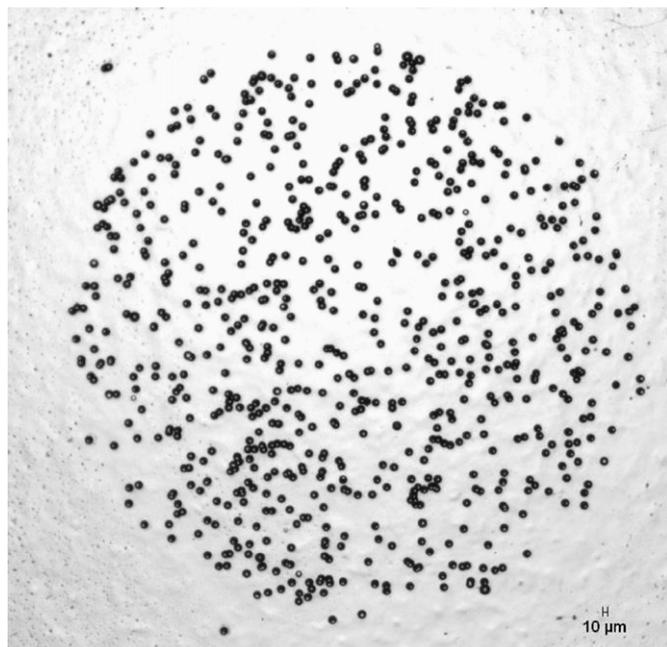


Fig. 2. Image of the micro-collimator.

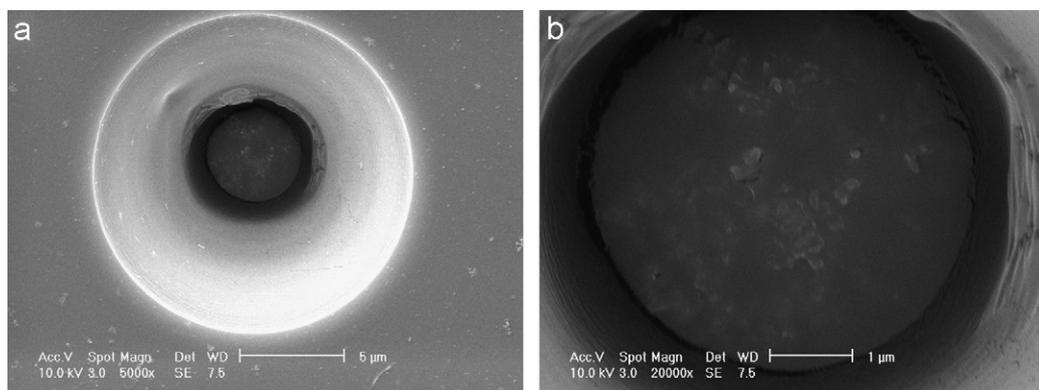


Fig. 3. Image of an enlarged pore in the micro-collimator captured by environmental scanning electron microscope under magnifications of (a) 5000 \times and (b) 20,000 \times .

3. Results

A micro-collimator with a thickness of 15 μm was fabricated through α -particle irradiation of a PADC film and subsequent chemical etching. Fig. 2 shows the image of such a micro-collimator after the enlargement of the latent tracks by 11 h chemical etching. By using the computer software ImageJ freely obtained from the website <http://rsb.info.nih.gov/ij/>, the number of the enlarged tracks located on this micro-collimator was determined to be 607.

Figs. 3(a) and (b) showed the images of the same enlarged etched track captured using the environmental scanning electron microscope (ESEM) (Philips XL30 Esem-FEG) under magnifications of 5000 \times and 20000 \times , respectively. The image in Fig. 3(b) clearly shows the pattern of an NEM tape (Nisshin EM Co. Ltd. Tokyo), which was a double-sided adhesive tape used to mount the micro-collimator onto a plate for imaging. This is a solid evidence to show that the track has been etched through and thus a channel has been formed in the PADC film after etching for 11 h.

The optical appearance of the tracks on α recorders A and B are shown in Fig. 4. Fig. 4(a) shows the image of α recorder A on which tracks were generated from 5.49 MeV α particles having passed through the micro-collimator and the 20 μm PADC support substrate. On the other hand, Fig. 4(b) shows the image of α recorder B on which tracks were generated from 5.49 MeV α particles having passed through only the 20 μm PADC support substrate, i.e., without going through the micro-collimator.

By comparing the images of the α recorders A and B, one can observe that most of the tracks on α recorder A were circular in shape. On the other hand, tracks generated by α particles with small incident angles (with respect to the PADC film surface) appeared on α recorder B but not on A. This clearly shows that, with the help of the micro-collimator, only α particles close to normal incidence can reach the target.

The shape of the tracks on the α recorder A can be divided into three categories, namely, (1) dark circular tracks, (2) bright circular tracks with dark circumference, and (3) dark elliptical tracks. In order to find out the energies of the α particles reaching the α recorder, the diameters of the tracks on α recorder A were measured and these were compared with those simulated using a computer program TRACK_VISION 1.0 [8] freely available on the website <http://www.cityu.edu.hk/ap/nru/vision.htm>. Fig. 5 shows sample results of these simulations.

The results indicated that the bright circular tracks with dark circumference with diameter 5.1 μm or above on α recorder A were generated by α particles with energies from ~ 0.9 to ~ 1.9 MeV with normal incidence. The dark circular tracks on α recorder A with the maximum diameter equal to 4.7 μm were generated by α particles with energy 2.85 MeV with normal incidence. By using the SRIM program (<http://www.srim.org/>), the

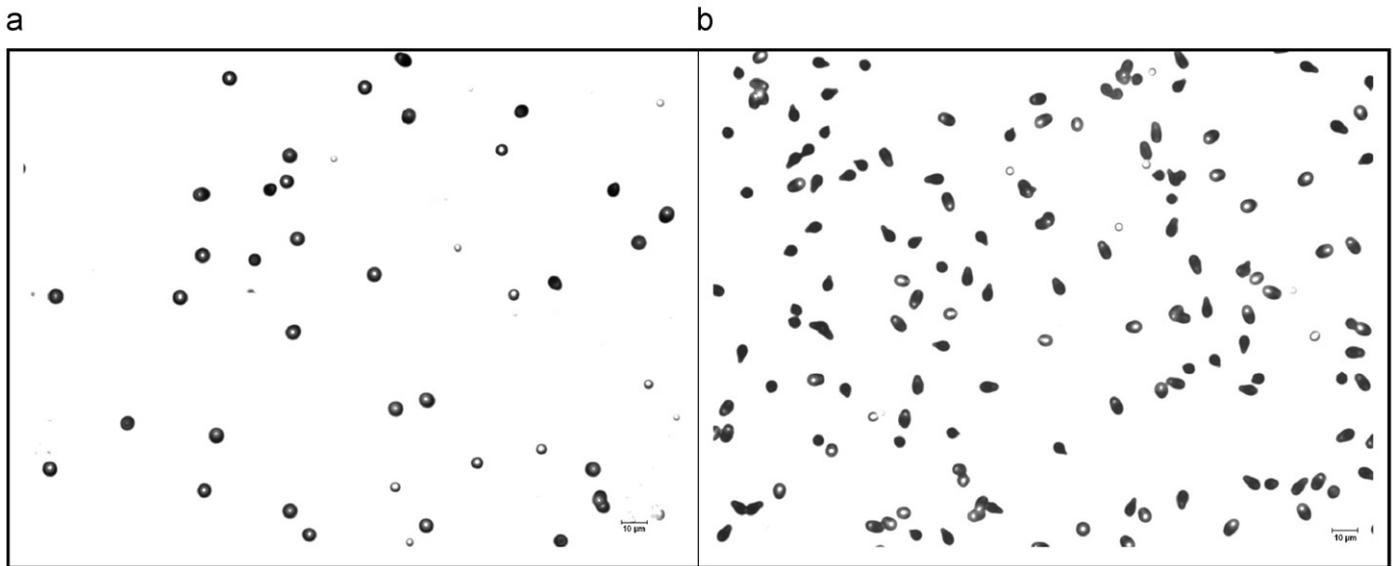


Fig. 4. (a) Image of α recorder A on which tracks were generated from α particles irradiated through the micro-collimator. (b) Image of α recorder B on which tracks were generated from α particles irradiated without the use of the micro-collimator.

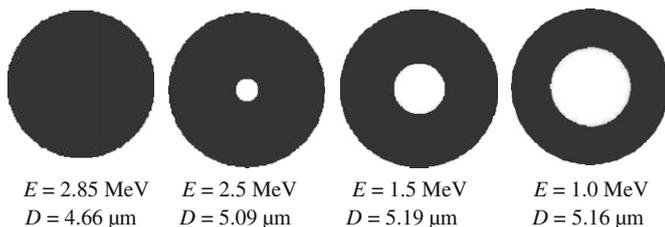


Fig. 5. Optical appearance of the tracks with different diameters D generated by α particles with different energies E , which are simulated using the computer program TRACK_VISION 1.0.

residual energy of α particles after passing through the air column in the micro-collimator and then a $20 \mu\text{m}$ support substrate was calculated to be 2.85 MeV. This agreed with the energy obtained by comparing the experimentally obtained and the simulated appearance of the track.

The energy distribution of the tracks was also measured by comparing the optical appearance of the tracks on the α recorder A with the results simulated using TRACK_VISION 1.0. From the result of the α recorder A, > 80% of the tracks were generated from α particles with energies $\sim 1 \text{ MeV}$ and with incident angles $\sim 65^\circ$ (and $< 90^\circ$), which encompassed our desired energies and incident angles.

4. Conclusions and discussion

A method was proposed to fabricate a micro-collimator with a thickness of $15 \mu\text{m}$ through α -particle irradiation of a PADC film and subsequent chemical etching. The feasibility of this fabricated micro-collimator was tested using PADC α recorders. The results showed that, with the use of the micro-collimator, the incident angles (with respect to the PADC film surface) of α particles could be restricted to $\sim 65^\circ$ to 90° , while α particles with small-angle incidence could not reach the sample. The dominance of α particles close to normal incidence would facilitate a more convenient and accurate dosimetric determination.

From the result of Fig. 4, > 80% of the tracks were generated from α particles with energies $> \sim 1 \text{ MeV}$ and with incident angles

$\sim 65^\circ$ (and $< 90^\circ$), which encompassed our desired energies and incident angles. The presence of the energy range was due to the fact that some α particles did pass through a small part of the collimator material instead of traveling the air column alone, which is a universal phenomenon for collimators.

In the present study, we used a PADC support substrate with a thickness of $20 \mu\text{m}$ to illustrate our results more clearly. In realistic experiments, however, although it is feasible to fabricate $20 \mu\text{m}$ PADC films, they are very fragile and it is not always successful to produce such thin PADC films intact and then to mount them onto the Petri dish using epoxy. Therefore, it is always desirable to be able to use thicker support substrates. In this way, the thickness of the collimator will be critical, as demonstrated by the example described in the introduction.

It would be even more desirable to fabricate micro-collimators with more cylindrical bores, so that the α particles passing through the collimators will be even more close to normal incidence. There can be two solutions. The first one is to use ions from accelerators instead of α particles. However, it is understandable that some laboratories cannot or do not want to involve expensive and sophisticated equipment associated with accelerators, so the following second solution might help. The second solution is to explore better etching conditions, particular with the use of surfactants. In the present work, we tried only the addition of 5% DOWFAX 2A1 into the etchant. With this surfactant, the slope of the track wall was modified from 23.4° to 19.8° . Further explorations of other surfactants as well as their concentrations will be made to further improve the slope of the etched track.

Acknowledgement

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