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Technical Note

Application of CR-39 Microfilm for Rapid Discrimination Between Alpha-Particle Sources

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ABSTRACT

This work presents a new technique for discriminating between alpha particles of different energy levels. In a first study, two groups of alpha particles emitted from radium-226 and americium-241 sources were successfully separated using a CR-39 microfilm of appropriate thickness. This thickness was adjusted by chemical etching before and after irradiation so that lower-energy particles were stopped within the detector, while higher-energy particles were revealed on the back side of the detector. The number of tracks on the front side of the microfilm represented all alpha particles incident on that side from the two sources. However, the number of tracks on the back side of the microfilm represented only the long-range alpha particles of higher energy that arrived at that side. Therefore, by subtracting the number of tracks on the back side from the number of tracks on the front side, one could easily determine the number of tracks for the short-range alpha particles of lower energy that remained embedded in the microfilm. Discrimination of the two energy levels is thus achieved in a simple, fast, and reliable process.

1. Introduction

Polymer-based solid-state nuclear track detectors are widely used for radiation detection in several important nuclear research applications, including cosmic ray measurements [1, 2], radon monitoring [3–5], particle identification, and neutron dosimetry [6–13]. At present, the most important type of detector is the poly allyl diglycol carbonate or CR-39 detector. Exposure of the CR-39 detector to heavy charged particles, such as alpha radiation, produces extensive ionization of the CR-39 material and dissociates the chemical bonds in the polymer, forming permanent tracks of the radiation path in the detector. The tracks vary in size, shape, and depth depending on radiation type, intensity, energy, and angle of incidence. For that reason, these tracks can be extensively investigated using different spectroscopic techniques such as ultraviolet–visible, Fourier transform infrared, and photoluminescence [14].

Spectroscopy using CR-39 to estimate the energy of incident alpha particles from the geometric measurements of the recorded tracks is an extremely challenging application. This is because alpha particles have a very short range in materials and can penetrate only a very thin layer of the CR-39 surface. For example, according to the Stopping and Range of Ions in

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Matter (SRIM) program [15], the range of 5-MeV alpha particles in CR-39 is 28.9 μm. In general, the range is highly correlated with the energy of the incident alpha particles [16]. Consequently, this generates a problem in the case of different alpha particles having very close energy levels. In that case, the ranges will be very similar, and the ability to discriminate between alpha particles will seem difficult to achieve.

Previous works on alpha spectroscopy have developed a matrix of energy equations as a function of the track diameter [17–20]. However, these approaches have used complicated geometric analyses of the track parameters, as well as calibration curves of the track diameter versus alpha energy. Another work on alpha particles from radon gas and radon daughters used two detectors [21]. The first was a CR-39 track detector to determine the incident fluence; the second was an LiF thermoluminescent detector to deduce the average energy of the alpha particles. However, that study was time consuming and required calibration of the two detectors. Therefore, it is important to search for a faster and less complicated method of alpha spectroscopy.

In this work, we present a new method using a CR-39 microfilm for the discrimination of the energy of alpha particles emitted from two different sources. The method is based on the experimental observation that the greater the energy of an alpha particle, the longer its range in the material. Therefore, by adjusting the thickness of a CR-39 microfilm to match the range of higher-energy alpha particles, low-energy particles will stop within the microfilm, whereas high-energy particles will pass the microfilm and can be revealed on the back side by chemical etching. It can readily be understood that, under these conditions, discrimination of the two energy levels is achieved accordingly. This work is a continuation of our previous work on improving radiation measurements using the CR-39 detector [22].

2. Materials and methods

2.1. Alpha-particle sources

We used two different alpha-particle sources from the commercially available reference standards. One source was ^{226}Ra, which emits alpha particles with a kinetic energy of 4.78 MeV; the other was ^{241}Am, which emits alpha particles with a kinetic energy of 5.49 MeV. Using the Bateman equation, we calculated the present activity of the two sources at the time of this study and found that both had the same activity of 150 nCi (5.55 kBq). In order to calculate the range of alpha particles in CR-39, we employed SRIM simulation software [15], available on the Internet. We chose the Transport of Ions in Matter (TRIM) section of the software to generate a list of stopping power and range values. The calculations were completed for 99,999 helium ions per simulation, a default used by the software. Fig. 1 is a plot of ionization, that is, the energy loss of the incident alpha particles to the target electrons as a function of the penetration depth in the CR-39 target. The dotted curve represents the 4.78-MeV alpha particles emitted from ^{226}Ra, and the solid curve represents the 5.49-MeV alpha particles emitted from ^{241}Am. End points of the curves represent the maximum penetration depth of the alpha particles in CR-39. The figure clearly shows that the greater the energy of the alpha particles, the longer their range. These range values were 33.3 μm for the 5.49-MeV particles and 27.0 μm for the 4.78-MeV particles.

2.2. CR-39 microfilm preparation and chemical etching process

Thin sheets of CR-39 microfilm (Fukuvi Chemical Industry Company, Tokyo, Japan) with $C_{12}H_{18}O_7$ molecular composition, 100 μm uniform thickness, and 1.32 g/cm^3 density were cut by a laser into pieces with dimensions of 1 × 1 cm^2. To determine the rate of the chemical etching process, five pristine CR-39 microfilms were etched under standard etching conditions in a 6.25N aqueous solution of NaOH maintained at 70°C by a water bath for 6 hours [22]. During the etching process, a magnetic stirrer was used to achieve uniform etching and to prevent accumulation of the etchant material on the surfaces of the microfilms. After etching, the microfilms were thoroughly rinsed with distilled water and dried in open air. The thickness of each microfilm before and after etching was measured using a sensitive micrometer; the average value of the bulk etching rate was found to be 1.06 μm/h, according to the following equation:

$$\text{Bulk etch rate} = \frac{\Delta d}{2t} \quad (1)$$

where Δd is the thickness reduction and t is the etching time. Our results for the etch rate agree exactly with those reported by Yamauchi et al [23]. In their work, it took about 40 hours to reduce the thickness of an unirradiated microfilm from 100 μm to 15 μm.

Next, a fresh set of six 100-μm-thick CR-39 microfilms was etched for 30.6 hours using the abovementioned etching conditions until the thickness of the residual active layer of each microfilm was reduced to 35 μm. This particular thickness is sufficient to prevent possible backscattering of alpha particles from a thick substrate at the back side of the detector. Indeed, alpha particles can penetrate the detector to the substrate, bounce from the substrate surface, and then enter a second time into the detector, which may contribute to the tracks at the back side of the detector. To ensure that alpha particles stop before reaching the substrate, the detector was etched to a thickness slightly larger than 33.3 μm (i.e., thicker than the range of the highly energetic 5.49-MeV alpha particles from ^{241}Am in CR-39).

Afterward, one blank microfilm was randomly selected and used as a control. The front and back sides of the control microfilm were scanned by a manual optical scanner to determine the existence of possible background tracks. Surface defects or high-density pits were not found in the control microfilm, and the background tracks were easily distinguished. The mean value of background track density was measured and found to be 4 ± 3 tracks/cm^2. This low count value indicated that the microfilm in hand had not been irradiated previously. At this point, the control microfilm underwent no further processing and was stored for future reference. It is worth noting that all the microfilms used in this work were kept away from the external environment in a clean room under controlled laboratory conditions. This

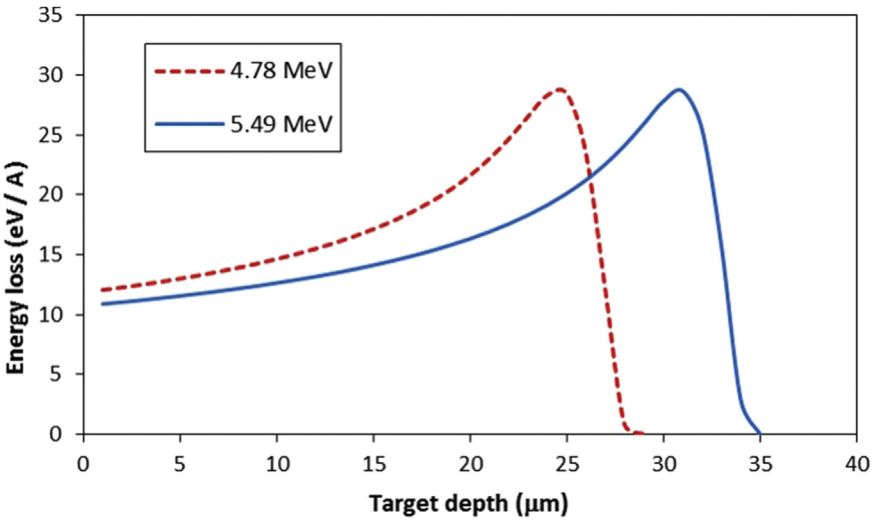

Fig. 1 – Stopping power of 4.78-MeV and 5.49-MeV alpha particles as a function of the penetrating depths in CR-39 target (calculated by SRIM-2013 software).

makes the collection of further background tracks from the environment or from some unaccounted-for source unlikely to happen. Nonetheless, we placed these microfilms on thick aluminum substrates to block any possible exposure to the environment occurring at the back side. In such a situation, there is no way for alpha particles to reach the back side except by coming through from the front side.

2.3. *Irradiation, counting, and energy identification of alpha particles*

Each microfilm was irradiated with alpha particles by placing the point sources (^{226}Ra and ^{241}Am) in close contact with the front side of the microfilm for 5 seconds. After irradiation, the irradiated CR-39 microfilms were etched again in a 6.25N NaOH solution at 70°C for a short time interval of 2 hours. After the etching, etched pits along the tracks of alpha particles in the microfilm became visible under an optical microscope and could be counted using an automated counting system. The system setting can positively identify the pits and ignore false positives. The characteristics of this system, and the procedure for track registration and analysis were described in detail in a previous publication [22]. The numbers of etch pits on the front and back sides of each microfilm were determined and verified by manual counting. Furthermore, background radiation was taken into account by subtracting the number of etch pits counted in the control (unexposed) microfilm from the number of etch pits counted in the irradiated microfilms. Discrimination of the alpha particles with two different energy levels is simply based on track counting on both sides of the microfilm, without need for calibration curves of the track diameter versus alpha energy.

3. Results and discussion

Initially, it was important to verify experimentally that the CR-39 microfilms prepared with 35-μm thickness were appropriate for discriminating between the two ranges of alpha particles used in this study. To do so, two microfilms were monoenergetically irradiated. One microfilm was exposed only to high-energy long-range particles from the ^{241}Am source, and the other microfilm was exposed only to low-energy short-range particles from the ^{226}Ra source. Fig. 2 shows a representative image of the etched tracks observed on both sides of the microfilm, the front side of which was in contact with the ^{241}Am source. As can be seen, the front and back sides of the microfilm have the same number of tracks. This observation clearly indicates that all alpha particles entering the front side penetrated the microfilm thickness and appeared at the back side. Therefore, it can be concluded that the microfilm thickness is, indeed, about the same as the range of these 5.49-MeV alpha particles.

Conversely, no tracks were observed at the back side of the microfilm whose front side was in contact with the ^{226}Ra source. This indicates that all alpha particles incident on the front side were stopped within the microfilm and remained embedded in it. Hence, the range of these 4.78-MeV alpha particles is shorter than the microfilm thickness. The above results confirm the possibility of using this very simple and practical procedure to discriminate completely and with certainty between alpha particles of two different energy levels.

For further quantitative analysis, Table 1 shows the number of tracks counted at the front and back sides of the three CR-39 microfilms after these films were irradiated by a combination of alpha particles emitted from the ^{226}Ra and ^{241}Am sources together.

The data in Table 1 clearly reveal that the alpha particles from both sources entering the microfilm produced nearly 4,800 visible tracks at the front side of the microfilm, whereas those arriving at the back side of the microfilm produced only approximately 2,400 visible tracks. Assuming that the number of alpha particles is determined by counting the visible tracks in the microfilm, it can be deduced that out of all the alpha particles from the two sources incident on the front side of the microfilm, only approximately half arrived at the back side of the microfilm. Most likely, these are the long-range 5.49-MeV alpha particles from the ^{241}Am source. By subtracting the

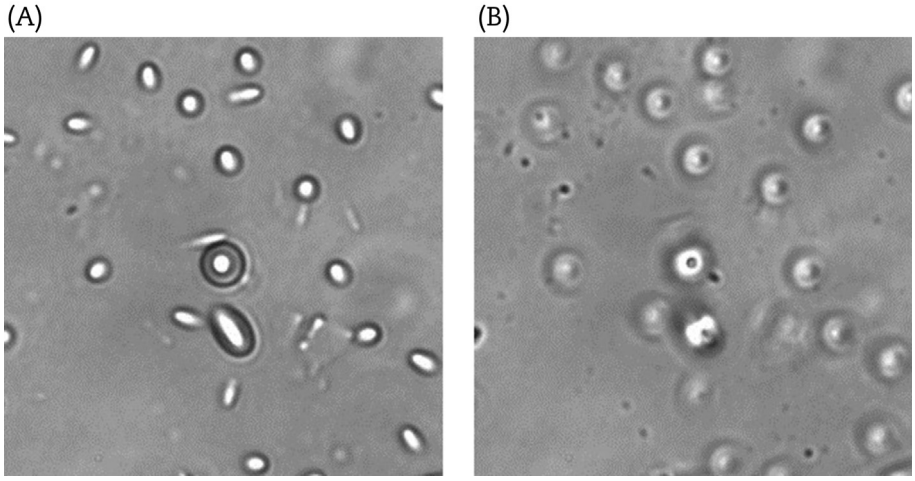

Fig. 2 – Recorded tracks for alpha particles with 5.49-MeV energy and normal incidence in CR-39 microfilm. The field of view taken at a specific position shows tracks on (A) the front side and (B) the back side.

Table 1 – Counts of the number of tracks of alpha particles in CR-39 microfilms.

	Counts on front side	Counts on back side
CR-39 microfilm #1	4,783	2,387
CR-39 microfilm #2	4,770	2,391
CR-39 microfilm #3	4,777	2,379
Average	$4{,}777 \pm 7$	$2{,}386 \pm 6$

track counts on the front and back sides of the microfilm, it can be found that nearly half of the incident alpha particles remained embedded in the microfilm. Most probably, these are the short-range 4.78-MeV alpha particles from the ^{226}Ra source. It is interesting to note that the numbers of alpha particles embedded in the microfilm and those that penetrated the microfilm are exactly the same. Essentially, this result confirms that the ^{226}Ra and ^{241}Am sources have equal activity, which is in agreement with our calculations of the present activity of the sources, as mentioned in Section 2.1.

The findings in this work provide new insight into using CR-39 microfilms to distinguish between alpha particles of different energy levels. The method is reliable, accurate, and suitable for environmental radiation measurements. It is relatively fast because of short etch times. In addition, it is simple because there is no need for calibration curves of the track diameter versus the energy of the incident alpha particles. Moreover, in the case of a mixed source emitting multiple alpha particles (n), the required number of microfilms for the identification of alpha particles must be $n - 1$. For instance, two CR-39 microfilms with different thickness are sufficient to distinguish between ^{222}Rn and its progenies ^{214}Po and ^{218}Po (under study).

Finally, it should be noted that this reported method requires prior knowledge of the alpha particles to be recorded and appropriate preparation of the CR-39 microfilm, in addition to suitable separation in energy of the alpha particles under investigation. In practical applications, the method can be used for environmental radiation monitoring. The method can also be used to discriminate between different types of natural and manmade radiation. However, further study is necessary to determine clear criteria and/or significant restrictions on the conditions for which the method is applicable. In particular, challenges may arise if the energy of the alpha particles is not known or if the alpha particles are comparable in energy, such that their separation in energy is too small. In addition, it may become increasingly difficult to quantify each population of particles if the separation between the high-energy cutoff of one particle is too close to that of the other due to broadening of the Bragg peak. At present, the method has been used for binary discrimination, not spectroscopy. With more development, there is certainly potential to move in the direction of spectroscopy.

4. Conclusion

In this work, we have developed a simple and rapid method of using CR-39 microfilms to discriminate between alpha particles of two different energy levels. The method proved effective in identifying alpha particles emitted from different sources with suitable different energy levels. This makes the method an appropriate option for nuclear science research and environmental radiation measurement. The method is in the first phase of experimentation, and future work will extend this study by further optimization of the microfilm and implementation of the method in advanced and complex applications.

Conflicts of interest

The authors have no conflicts of interest to declare.

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