Turkish Journal of Physics

Volume 37 | Number 3 Article 10

1-1-2013

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TÜBİTAK

Turkish Journal of Physics

http://journals.tubitak.gov.tr/physics/

Research Article

Turk J Phys
(2013) 37: 356 – 362

doi:10.3906/fiz-1210-1

Identification of spontaneous fission fragments by using thermally annealed PADC films

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Received: 01.10.2012 • **Accepted:** 13.02.2013 • **Published Online:** 13.09.2013 • **Printed:** 07.10.2013

Abstract: One of main requirements in applied nuclear physics is the need of a detector capable of detecting and differentiating different particles. Thus, the fission products from a ^{252}Cf radioactive source, after about 12 half-life periods of alpha decay, are registered using virgin and annealed CR-39 polyallyldiglycol carbonate (PADC) films. In the case of virgin CR-39, a single peak appears corresponding to the heavy group of these products, while the other peak related to the light group is obscured. The annealed CR-39 detectors, on the other hand, were demonstrated to be highly sensitive to the fission fragments, and as a result we have observed the characteristic twin peaks of fission fragments from that radioactive source.

Key words: ^{252}Cf radioactive source, spontaneous fission, mass distribution, nuclear track detectors, annealed CR-39 detectors

PACS: 25.85.Ca, 29.40.Wk, 29.40.Gx, 65.60.+a

1. Introduction

Spontaneous fission is a significant process for a wide range of transuranic nuclides of extremely large mass number [1]. Petrzhak and Flerov [2] in the 1940s discovered the phenomenon of spontaneous fission and this was later confirmed [3]. Soon afterwards, several groups spent considerable time on further studies of this phenomenon [4–6]. This was followed by the discovery of damage trails, or tracks, as they became known, in LiF crystals [7] previously in contact with a uranium foil. The tracks were induced by thermal neutrons and developed with a chemical solution. The most sensitive solid-state nuclear track detector (SSNTD) for recording tracks from low-energy protons to fission fragments is known as CR-39 and is preferred over other nuclear detectors as it shares some good properties of other track registering detectors (such as nuclear emulsions and cloud chambers) but requires no electronics or dark-room processing. Later, in the United States, a SSNTD pioneer group [8] used track techniques to study the detection and registration efficiency of the signature of fission fragments in the presence of a very high alpha background. Over 10 years ago, another research group from India studied the probable influence of alpha activity on the mass distribution of ^{252}Cf fission fragments and analyzed spontaneous fission data from a ^{252}Cf-deposited source after about 1 half-life for alpha emission, and also at the end of almost 4 half-lives when the alpha activity (half-life: 2.65 years) was reduced significantly while the spontaneous fission activity (half-life: 85 years) was reduced slightly [9]. In addition, the branching

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ratios of alpha/spontaneous fission of ^{252}Cf on these 2 dates were 18.242 ± 0.421 and 2.398 ± 0.031, respectively [10]. There is a similar concern that mass distribution of spontaneous fission fragments from samples of lime was observed [11]. It should also be noted that nuclear track detectors (NTDs) of polymer, pristine or thermally treated, or glass and mica have already been applied to many studies of the detection of the nuclear fission products and the registration properties of these fission products or lighter particles, e.g., alpha particles in those detectors, as reported by several research groups [12–20].

The objective of the present paper was to study the influence of thermal annealing on the mass distribution of ^{252}Cf fission fragments using virgin and annealed CR-39 detectors when the alpha activity is reduced by a highly significant amount while the spontaneous fission activity is reduced slightly. In addition, we introduce a novel method for identifying the light and heavy fission fragment groups by using thermally treated polyallyldiglycol carbonate (PADC)-based NTDs.

2. Experimental

CR-39 detectors, 660 μm thick, were purchased from Intercast Europe S.P.A. of Parma, Italy. Three sets of detectors were prepared for this experiment, and each was exposed to a deposited source of ^{252}Cf that had an alpha activity of 1 μCi measured in October 1977. Irradiation was done in air using 2π geometry. These detectors were annealed in a temperature-controlled oven for different time intervals (4–10 h) and temperatures (90, 120, and 150 °C). The first set of CR-39 detectors was exposed, but not annealed, in order to compare results with the results from the annealed detectors (for the control experiment). The second set of detectors was first irradiated with fission fragments and alpha particles, followed by annealing in the oven (exposure before annealing). The third set was first annealed in the oven and then exposed to the ^{252}Cf source (exposure after annealing).

After exposure and annealing or vice versa, the detectors were etched in a 6.25 N standard aqueous solution of NaOH maintained at 70 °C by a water bath; these are the most commonly used etching conditions for CR-39 NTDs. The temperature was kept constant with maximum uncertainty of ±1 °C. The etching time was appropriately chosen to ensure that the fission fragment tracks were easily observed using an optical microscope with appropriate magnification, while the alpha tracks were almost invisible. However, the etching time was 4 h for virgin CR-39 NTDs and 4 h, 1 h, and 4 min for pre- and postannealing exposure at 3 different temperatures of 90, 120, and 150 °C, respectively. The etch pit diameters were used to measure the mass of the particles [21].

3. Results and discussion

We measured track diameters to estimate the atomic mass ratios of the fission fragments emitted from a ^{252}Cf radioactive source by using nonannealed and thermally annealed high polymer film composed of PADC in the form of CR-39-based NTDs. For the virgin detector exposed to a ^{252}Cf source, we measured the diameters of the tracks along the minor axis for 4 h of etching time. In this case, a plot of the number of tracks versus track diameter shows that the fission fragments are clustered into a "heavy group" showing only a single peak, while the other "light group" was unclear, as shown in Figure 1. There was a significant reduction of alpha emission after about 12 half-life periods of alpha activity, and the fission fragments tend to have a mass distribution indicated by a single peak.

When the thermally treated CR-39 detector was used, representing the preannealing exposure case, the fission fragments recorded also exhibited a mass distribution with a single peak, as previously recorded using

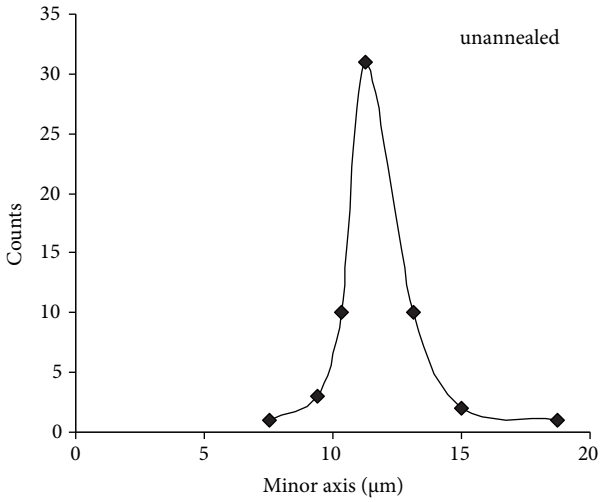

Figure 1. Mass distribution of tracks along the minor axis for the fission fragments from ^{252}Cf by using virgin CR-39 NTDs etched for 4 h.

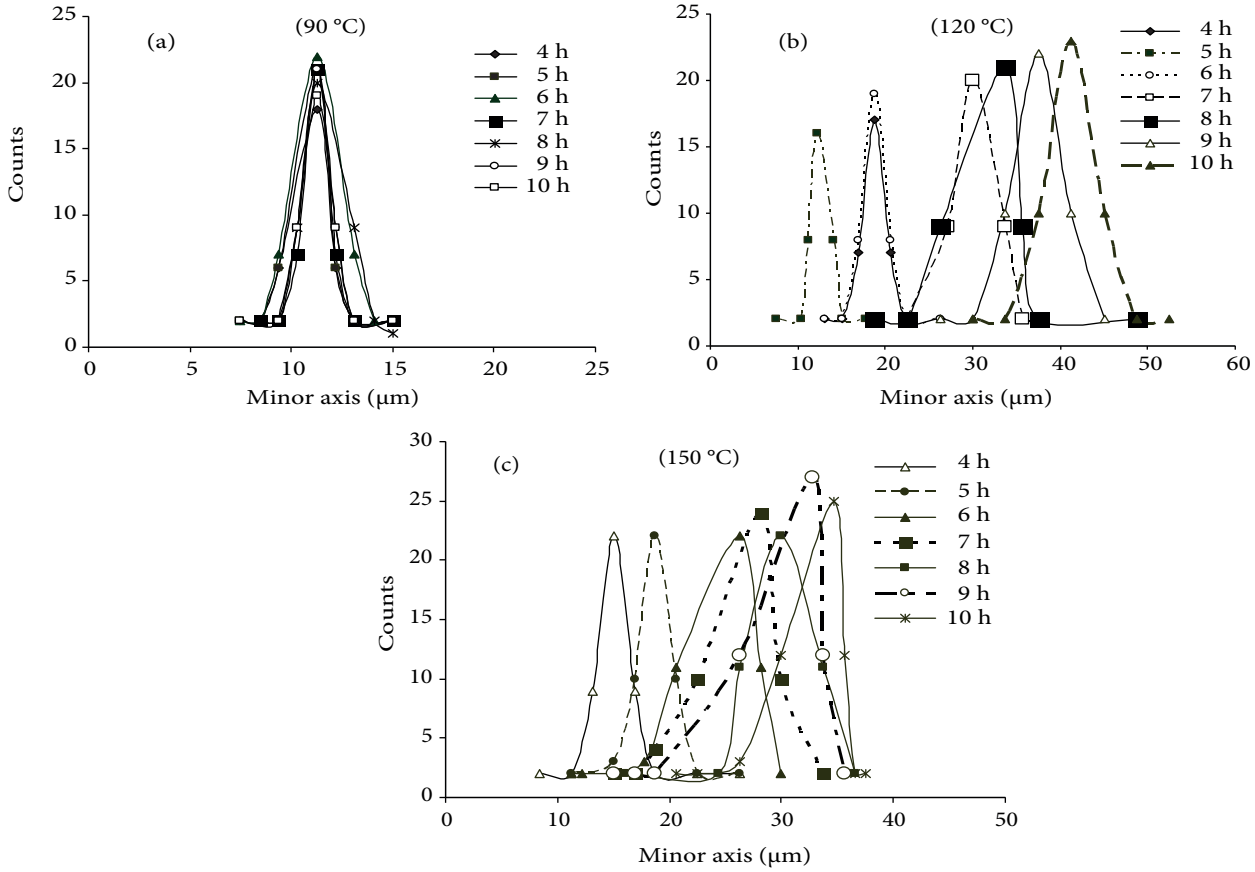

Figure 2. Mass distribution of tracks along the minor axis for the fission fragments from ^{252}Cf by using annealed CR-39 NTDs as a function of annealing time at 3 different temperatures, 90, 120, and 150 °C, with preannealing exposure etched for 4 h, 1 h, and 4 min, respectively.

a virgin detector. At an annealing temperature of 90 °C, the upper mass peak remains fixed at its typical position, as seen in Figure 2a, while at the higher temperatures of 120 and 150 °C, this peak moves towards larger diameters with the increase of annealing time at the given temperature, as shown in Figures 2b and 2c. It is interesting to note that the mass distribution that produces a single peak is the result of fission fragments already registered in the material before the effect of the thermal annealing process.

In principle, nuclear fission, resulting from either spontaneous or neutron-induced fission, is normally asymmetric. Thus, the fragments tend to cluster into a "light group" and a "heavy group". In the case of spontaneous fission from ^{252}Cf, both groups show 2 peaks with average mass numbers of 108 and 143, which gives a ratio of R = 1.32 ± 0.01, as reported in [22]. However, such distinct clustering for fission fragments is discernible in the plots of a number of tracks versus track diameter using annealed detectors (postannealing exposure) at annealing temperatures of 90, 120, and 150 °C. The track diameters were measured along the minor axis for each experiment with etching times of 4 h, 1 h, and 4 min. In all cases of postannealing exposure, the plot of the number of tracks shows that the fission fragments are clustered into 2 groups; the light group is again observed and the heavy group is still present, showing 2 peaks, as seen in Figure 3. It should be noted that the reappearance of these 2 peaks is attributed to the modification of the registration properties of heavy ions in a CR-39 polymeric track detector induced by thermal annealing. This observation proved that the nature of fission decay of this nuclide does not depend on the age of the source, but rather on the nature of the changes in the registration properties of charged particles in that polymeric detector induced by thermal annealing as we reported previously [23].

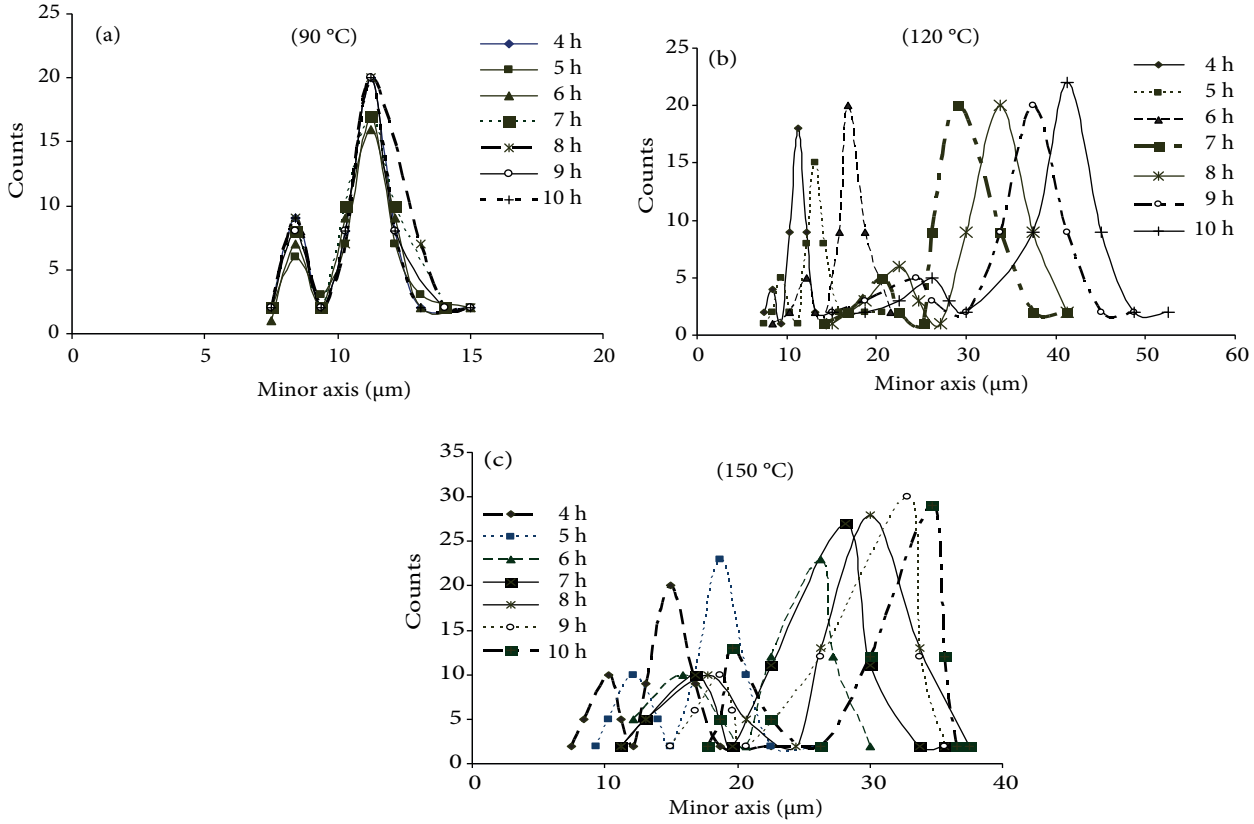

Figure 3. Mass distribution of tracks along the minor axis for the fission fragments from ^{252}Cf by using annealed CR-39

NTDs as a function of annealing time at 3 different temperatures, 90, 120, and 150 °C, with postannealing exposure etched for 4 h, 1 h, and 4 min, respectively.

Our results are summarized in the Table. At an annealing temperature of 90 °C, the mean ratio of R = 1.33 ± 0.08, based on an average of track diameters at the peak positions, is closely related to the ratio of the average values of the light and heavy groups, R = 1.32 ± 0.01, as previously reported [22]. Thus, we can state that the measured ratio R for the thermally treated CR-39 detector, at relatively low temperature, is in excellent agreement with the ratio reported in the literature. However, at the higher annealing temperatures of 120 and 150 °C, this ratio is shifted towards higher values, with the increase of the annealing time for the given temperature reported in the Table. The results show that the thermal annealing treatment leads to an increase in the mean track diameter of the fission fragment tracks, as given in the Table, whereas α-particles show the opposite behavior, leading to the complete erasing of the tracks [23]. The value of the increase in the diameter of the fission tracks depends on the annealing time and temperature. From the Table, it can be seen that the ratio R at an annealing temperature of 120 °C has a similar dependence on annealing time as that for 150 °C, while at 90 °C this ratio remained constant.

Table. Measured track diameters along the minor axis of the ^{252}Cf fission fragment tracks, observed fission fragment (FF) groups, and ratio R in a CR-39 polymeric track detector as a function of time of thermal annealing (exposure after annealing) at 4 different temperatures: room temperature (RT) and 90, 120, and 150 °C. D_h and D_l stand for the average track diameter along the minor axis of the fission fragments corresponding to heavy and light groups, respectively.

Annealing temperature (°C)	Annealing time (h)	(μm)	(μm)	Observed FF groups	$R = \frac{D_{\mathrm{h}}}{D_{\ell}}$
23 (RT)	0	obscured	11.25 ± 0.60	1	—
90	4	8.44 ± 0.40	11.25 ± 0.70	2	1.33 ± 0.08
	5	8.44 ± 0.45	11.25 ± 0.55	2	1.33 ± 0.07
	6	8.44 ± 0.45	11.25 ± 0.65	2	1.33 ± 0.08
	7	8.44 ± 0.50	11.25 ± 1.00	2	1.33 ± 0.11
	8	8.44 ± 0.45	11.25 ± 0.70	2	1.33 ± 0.08
	9	8.44 ± 0.45	11.25 ± 0.50	2	1.33 ± 0.07
	10	8.44 ± 0.45	11.25 ± 0.50	2	1.33 ± 0.07
120	4	8.44 ± 0.45	11.25 ± 0.60	2	1.33 ± 0.08
	5	9.80 ± 0.45	13.13 ± 0.65	2	1.40 ± 0.07
	6	12.19 ± 0.55	16.88 ± 0.70	2	1.38 ± 0.06
	7	20.63 ± 0.50	29.06 ± 0.85	2	1.41 ± 0.06
	8	22.50 ± 0.50	33.75 ± 0.85	2	1.50 ± 0.04
	9	24.38 ± 0.65	37.50 ± 0.75	2	1.54 ± 0.03
	10	26.25 ± 0.45	41.25 ± 0.70	2	1.57 ± 0.02
150	4	10.31 ± 0.80	15.00 ± 0.75	2	1.45 ± 0.09
	5	12.19 ± 0.85	18.75 ± 0.65	2	1.54 ± 0.08
	6	15.94 ± 1.15	26.25 ± 0.80	2	1.65 ± 0.08
	7	16.88 ± 0.90	28.13 ± 1.00	2	1.67 ± 0.06
	8	17.81 ± 1.15	30.00 ± 1.20	2	1.68 ± 0.08
	9	18.75 ± 0.55	32.81 ± 1.00	2	1.75 ± 0.04
	10	19.69 ± 0.45	34.69 ± 0.55	2	1.76 ± 0.03

Finally, it is easy to explain why the annealed detector (in the case of exposure after annealing) under typical conditions registered more counts of fission fragments than the unannealed detector, allowing the light

group of fission fragments to be observed once again. Figure 4 shows that the fission fragment track density increased linearly with annealing time at 3 different temperatures of 90, 120, and 150 °C for both processes with exposure before and after annealing, respectively. It should be noted that, in the case of postannealing exposure, the track density is fairly greater than those in the other case. This is because the annealed CR-39 track recording films are sensitive to heavily ionizing particles, such as fission fragments, but not to alpha particles. The annealing process presumes that the annealing of damage trails occurs as a result of the diffusion of atomic defects through the crystal lattice or movement of molecular fragments within NTDs [24]. The annealing of latent fission tracks and their etching, as well as the fission fragment track formation in CR-39, have been discussed in detail using the crucial analysis of measured tracks [25,26]. The detection efficiency, η % $= [1 - (V_B / V_T)]$, of thermally annealed CR-39 polymeric detectors, where V_B and V_T are the bulk and track etch rates, respectively, is enhanced significantly for fission fragments, whereas it is decreased greatly for alpha particles [12,14,20,23]. Additionally, fission tracks formed at an angle less than the critical angle are not revealed because the normal surface advances faster than the etch rate along the track. When the fission fragment enters the annealed surface at an angle equal to or greater than the critical angle, it will be enhanced and visible after etching. The details of the registration properties of thermally treated CR-39 polymeric detectors (transient to glass-like track detector) were already fully explained in a separate paper [23]. Therefore, we have recorded 2 groups of fragments, which tend to have a mass distribution with the most probable values indicated by twin peaks in the plot of the number of tracks as a function of track diameter.

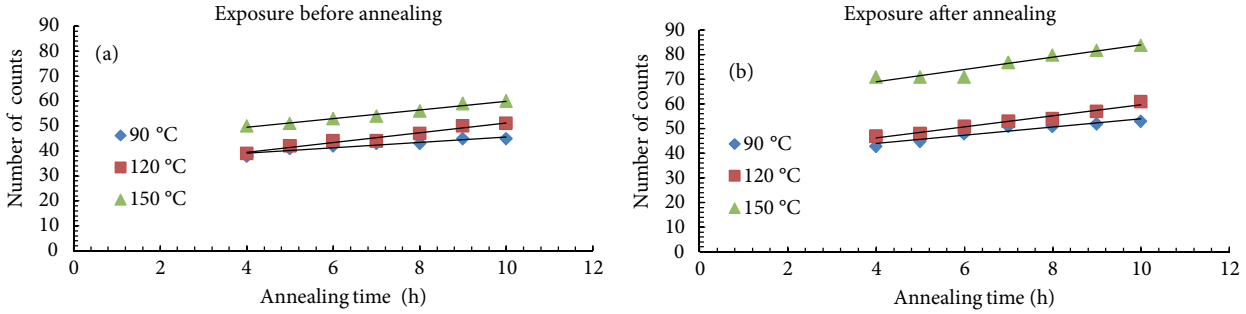

Figure 4. The variation of the number of counts of the fission fragment tracks as a function of annealing time at 3 different temperatures, 90, 120, and 150 °C, with a) exposure before annealing and b) exposure after annealing.

4. Conclusion

This study describes a method for detecting the mass distribution of fission fragment tracks from the spontaneous fission of a ^{252}Cf-deposited source at the end of almost 12 half-lives (in the absence of significant alpha activity) using an annealed CR-39 detector. We have determined the ratio, R, of the track diameters at the peak positions corresponding to the average mass distribution of the fission fragment clusters. This method is based on the thermal treatment of the CR-39 SSNTDs, which causes a significant change in the registration properties of the detectors: sensitive to the highly ionizing particles such as fission fragments of heavy nuclides, but not to alpha particles, as reported recently [23]. This annealing method has been successfully used in our attempts to separate light and heavy fission fragment groups in CR-39 SSNTDs leading to the reappearance of the twin peaks, which is a main characteristic of the fission process. Reliable R-values of 1.33 ± 0.08 were obtained with this method, particularly at lower temperatures and shorter periods of annealing. We therefore conclude that the R-values determined by the annealed SSNTDs are consistent with the recommended value of 1.32 ± 0.01 as reported [22].

Acknowledgments

We would like to thank the Benghazi University Zoology Department of the Faculty of Science for providing an optical microscope during the track scanning work and give special thanks to Prof Mahmoud Fadiel, the former dean of our faculty, for his encouragement and assistance. We are also indebted to Dr Dief-Allah Tawati, the former chairman of the Physics Department, for invaluable help and facilitating during this work. We also acknowledge Mr Mohammed El-Mshatay for his technical assistance.

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