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Detection of α-Particles and Neutrons in Electrochemically Etched Polycarbonate Track Detectors

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Abstract

The application of polycarbonate, in particular of Makrofol DE, has been investigated with respect to the detection of α-particles in radon diffusion chambers and the detection of fast neutrons via recoils and α-particles as well as thermal neutrons using (n, α) radiators.

The comparison of the dosimetric properties of different types of Makrofol DE materials has shown for nearly all materials a similar registration efficiency for α-particles and fast neutrons, however a better background behaviour for Makrofol DE 1-4 previously used at KfK.

The same two-step electrochemical etching (ECE) technique at an electric field strength of 26.7 $kV \cdot cm^{-1}$ applied for α-particle and neutron detection resulted for Makrofol DE 1-4 in a background track density and standard deviation of 5 ± 2 cm^{-2}. In the neutron energy range above the threshold of about 2 MeV the fast neutron response of $40 \cdot cm^{-2} \cdot mSv^{-1}$ allows to indicate already 0.1 mSv.

The two-step (ECE) technique allows, on the other hand, the detection of α-particles in the energy range 0.5 MeV to 4 MeV.

Nachweis von α-Teilchen und Neutronen in elektrochemisch geätzten Polykarbonat-Kernspurdetektoren

Zusammenfassung

Die Einsatzmöglichkeiten von Polykarbonat insbesondere von Makrofol DE als Kernspurätzdetektor wurden hinsichtlich eines Nachweises von α-Teilchen in Radondiffusionskammern und eines Nachweises von neutroneninduzierten Rückstoßkernen und α-Teilchen sowie von thermischen Neutronen unter Verwendung von (n,α)-Radiatoren untersucht.

Der Vergleich der dosimetrischen Eigenschaften verschiedener Materialsorten von Makrofol DE zeigte für alle Materialien eine vergleichbare Nachweisempfindlichkeit gegenüber α-Teilchen und schnellen Neutronen, jedoch ein besseres Backgroundverhalten für das im KfK bereits benutzte Makrofol DE 1-4.

Die Anwendung desselben zweistufigen elektrochemischen Ätzverfahrens bei einer elektrischen Feldstärke von 26.7 $kV \cdot cm^{-1}$ zum Nachweis von α-Teilchen und schnellen Neutronen führte bei Makrofol DE 1-4 zu einer Background-Spurendichte und Standardabweichung von $5 \pm 2 \cdot cm^{-2}$. Das Neutronenansprechvermögen von 40 $Spuren \cdot cm^{-2} \cdot mSv^{-1}$ ermöglicht im Neutronenenergiebereich oberhalb der bei 2 MeV gefundenen Energieschwelle den Nachweis schon von 0,1 mSv.

Das zweistufige elektrochemische Ätzverfahren gestattet andererseits den Nachweis von α-Teilchen im Energiebereich 0,2 MeV bis 4 MeV.

Content

1 Introduction

Polycarbonate, in particular Makrofol foil of about 300 µm thickness has been found to be a sensitive track etch detector for the registration of α-particles and has been successfully applied in the past in radon diffusion chambers to measure within countrywide surveys the mean radon concentration in dwellings and free in air (1). Makrofol combined with (n, α) radiators such as BN1 from Kodak-Pathé is a γ-insensitive thermal neutron detector which has been applied in albedo neutron dosemeters (2) as well as in moderator spheres for the low level measurement of the neutron dose equivalent in mixed neutron/photon radiation fields such as the natural radiation background (3). In comparison with CR-39, however, the registration efficiency for neutron induced recoils is much lower because recoil protons are not detected (4).

The two-step electrochemical etching technique, recently adopted for CR-39 to improve the detection of low energy recoil protons has been also applied for polycarbonate detectors resulting in an increase of the neutron response by a factor of about 3 in comparison with the combined pre-etching and electrochemical etching technique previously applied for Makrofol (4,5).

In order to establish an optimized electrochemical etching (ECE) technique the main purposes of this work were:

- The application of the same two-step ECE conditions for the detection of thermal neutrons using (n, α) radiators as well as fast neutrons via neutron induced recoils and α-particles and, above all, the registration of α-particles in radon diffusion chambers.
- The comparison of various types and batches of Makrofol DE manufactured by Bayer AG with respect to background track density and registration efficiency for α-particles and neutrons.
- The study of the energy dependence of α-particle registration in Makrofol using different etching techniques and α-particle absorbers in front of the detectors.
- The study of the energy dependence for fast neutron induced recoils and α-particles.

2 Detector material

The detector material examined was Makrofol DE, manufactured by Bayer AG, Leverkusen, which replaced the former Makrofol E. Both materials are of the same composition, differ, however, in the way of production. "E" and "DE" stand for extruded material. Because different types of materials are now available on the market we examined the dosimetric properties of the following types:

- Makrofol DE 1-4, 300 µm thick, glossy on one surface only, without protection cover, namely:
  (a) samples from the 50 kg stock at KfK/HS, for routine use,
  (b) samples from Bayer AG, batch marked 18.05.88,
  (c) sample No. 1. from Bayer, batch marked 26.05.88,
  (d) sample No 2 from Bayer, batch marked 26.05.88.

- Makrofol DE 1-1, transparent, i. e. glossy on both sides, namely:
  (a) DE 1-1 C, 250 µm thick, batch marked 18.05.88, one side covered by a polyethylene foil,
  (b) DE 1-1 SC, 350 µm thick, batch marked 18.05.88, one side covered by a thicker green polyethylene foil, the other by a thinner colourless polyethylene foil.

The protective covers were removed before irradiation and/or etching of the detectors. Colourless polyethylene covers have been found to be some kind sticky.

For all the experiments the detectors were cut into a detector size of 24 mm x 24 mm. The transparent Makrofol DE 1-1 detectors were carefully marked in order to distinguish between the sides, Makrofol DE 1-4 detectors were marked with pencil on the mat side.

3 Irradiations

3.1 Thermalized neutrons

Detectors from all types of Makrofol DE were mounted together with boron radiators using a distance of 0.3 mm between radiator and the etched detector surface. We irradiated the (n, α) radiator-detector combinations in the KfK calibration hall with thermalized neutrons produced by a Cf-252 neutron source inside a cylindrical moderator of 4.25 cm thickness. The detector-radiator sandwiches were positioned at a distance of 50 cm from the source centre, the detectors nearer the source, the radiators behind the detectors, and rotated around the source. An irradiation time was chosen to give a reading similar to a dose equivalent of 0.25 mSv of Cf-252 neutrons.

3.2 Fast neutrons

The irradiation in the KfK calibration hall was performed using the Cf-252 neutron source producing spontanous fission neutrons with a mean energy of about 2.1 MeV. The detectors of each type, mounted together with Makrofol DE 1-4 radiators were placed in a distance of 50 cm from the source centre and rotated during the irradiation with a neutron dose equivalent of about 19 mSv.

Makrofol DE 1-4 was also irradiated in a similar way, using an Am-Be neutron source with a mean neutron energy of about 4.5 MeV and an irradiation with a neutron dose equivalent of 10.35 mSv.

3.3 α-particles

In order to compare the registration efficiency for 1.5 MeV α-particles the detectors from all types of Makrofol were irradiated with about 880 α-particles per cm^2. Moreover, routinely used Makrofol DE 1-4 detectors were applied to study the energy dependence of α-track registration using monoenergetic α-particles produced in the energy range of 0.5 to 4.5 MeV. For this purpose a thin, electro-deposited Am-241 source of 5.6 kBq was mounted, in front of collimator, at a distance of about 4 cm from the detectors. By changing the air pressure in the irradiation chamber it was possible to obtain nearly monoenergetic α-particle beams in the above mentioned energy range. An α-particle spectrometer with a semiconductor silicon surface barrier detector has been used to determine α-energy. The α-particle fluence was also equal to about 880 cm^{-2} in all irradiations.

4 Etching and track counting

The standard KfK etching chambers and high voltage/high frequency generator manufactured by Physikalisch-Technische Werkstätten (PTW), Freiburg, have been used. The two-step ECE conditions, optimized in previous experiments (5), were as follows:

- The etching solution was a mixture of 80 vol.% of a 30 weight % KOH solution in H_2O and 20 vol.% of ethanol.
- For both etching steps an etching temperature of 35° C and an electrical field strength of 26.7 $kV \cdot cm^{-1}$ was used, taking into account the detector thickness.
- During the first ECE step the frequency of 100 Hz was applied for 5 hours, in the second step 2 kHz for 1 hour.
- Only one side of the detector was etched, except for Makrofol DE 1-1 SC, 350 μm thick, which has been etched on both sides.

For the registration of tracks a microfiche reader was used with magnification of about 35 times. The track density was counted by eye using photo reprints and the marked quarter of a circle of known area. Background tracks were counted directly on the screen of the microfiche reader from the area within the whole marked circle of 1.15 cm^2. In the case of Makrofol detectors etched on both sides it was difficult to count the tracks because the focus of the microfiche reader did not separate them sufficiently.

For α-particles of various energies the mean track diameter was estimated approximately on the basis of microfiche reprints.

5 Comparison of different types of Makrofol material

As a result of our study the background track density and the track registration efficiency for α-particles and neutron induced recoils are presented in Table 1 for the various types of Makrofol DE. For routinely used Makrofol DE 1-4, the response for Am-Be neutrons is added.

The difference in the background track density is probably connected with the thickness of the protection foil and the storage conditions of the detector material before use. The smallest background value has been found for Makrofol DE 1-4 SC, 375 μm thick, on the side which was covered by a thicker polyethylene foil. Also routinely used Makrofol DE 1-4 cut from the big, tightly wound roll has rather small background. (Any storage of the detectors in paper envelopes should be avoided, even for short periods of time, because the intrinsic radioactivity of the paper may result in a remarkable increase of the background which may double the background, for instance, already after storage periods of some days.)

Within the limits of error the efficiency of α-particle registration both for the detection of thermal neutrons using the (n, α) reaction with boron radiators and for 1.5 MeV α-particles has been found to be the same for all kinds of Makrofol DE.

For Cf-252 neutrons, the efficiency for the registration of neutron induced recoil tracks is also similar for all kinds of Makrofol, but that of Makrofol DE 1-1 SC, 375 μm thick, is somehow smaller. The reason of this discrepancy is not clear, but, in general, the application of this type of Makrofol does not offer special advantages, so that no further investigation has been made.

The investigation shows that the routinely used material Makrofol DE 1-4 of 300 μm thickness is the detector material with a low value of background track density and its standard deviation. Fig. 1 shows the frequency distribution of the background track density found on the basis of 68 unirradiated detector foils etched at different periods of time. For routinely used Makrofol DE 1-4 the mean background track density and the standard deviation was found to be 6 ± 2 tracks per cm^2.

Material	Background (cm-2)	1.5 MeV α-particles [1] (cm-2)	Cf-252 neutron response: moderated, BN1 radiator [2] (cm-2)	Cf-252 neutron response: fast recoils, Makrofol radiator ($cm^{-2} \cdot mSv^{-1}$)
Makrofol DE 1-4 (300 μm):				
routine material	5.3 ± 2.1	875 ± 60	2190 ± 120	25.9 ± 1.5
test sample of 18.05.1988	26.7 ± 6.9	885 ± 66		
test sample of 26.05.1988, batch I	14.8 ± 8.6	914 ± 39		
test sample of 26.05.1988, batch II	9.7 ± 4.6	943 ± 25		
Makrofol DE 1-1 (375 μm):				
green covered side	2.6 ± 1.7	881 ± 14	2080 ± 420	10.7 ± 0.7
white covered side	10.4 ± 2.1	814 ± 54	2300 ± 240	16.9 ± 1.8
Makrofol DE 1-1 (250 μm): non-covered side	17.7 ± 3.6	-	2220 ± 270	27.3 ± 1.0
covered side	8.7 ± 3.6	840 ± 42	2200 ± 260	26.1 ± 1.2

1) α-irradiation with a fluence of 880 cm^{-2}

2) Track density related to neutron dose equivalent of 1 mSv for the unmoderated Cf-252 neutrons

Table 1: Comparison of the background and registration efficiencies of various types and batches of Makrofol DE polycarbonate (Bayer)

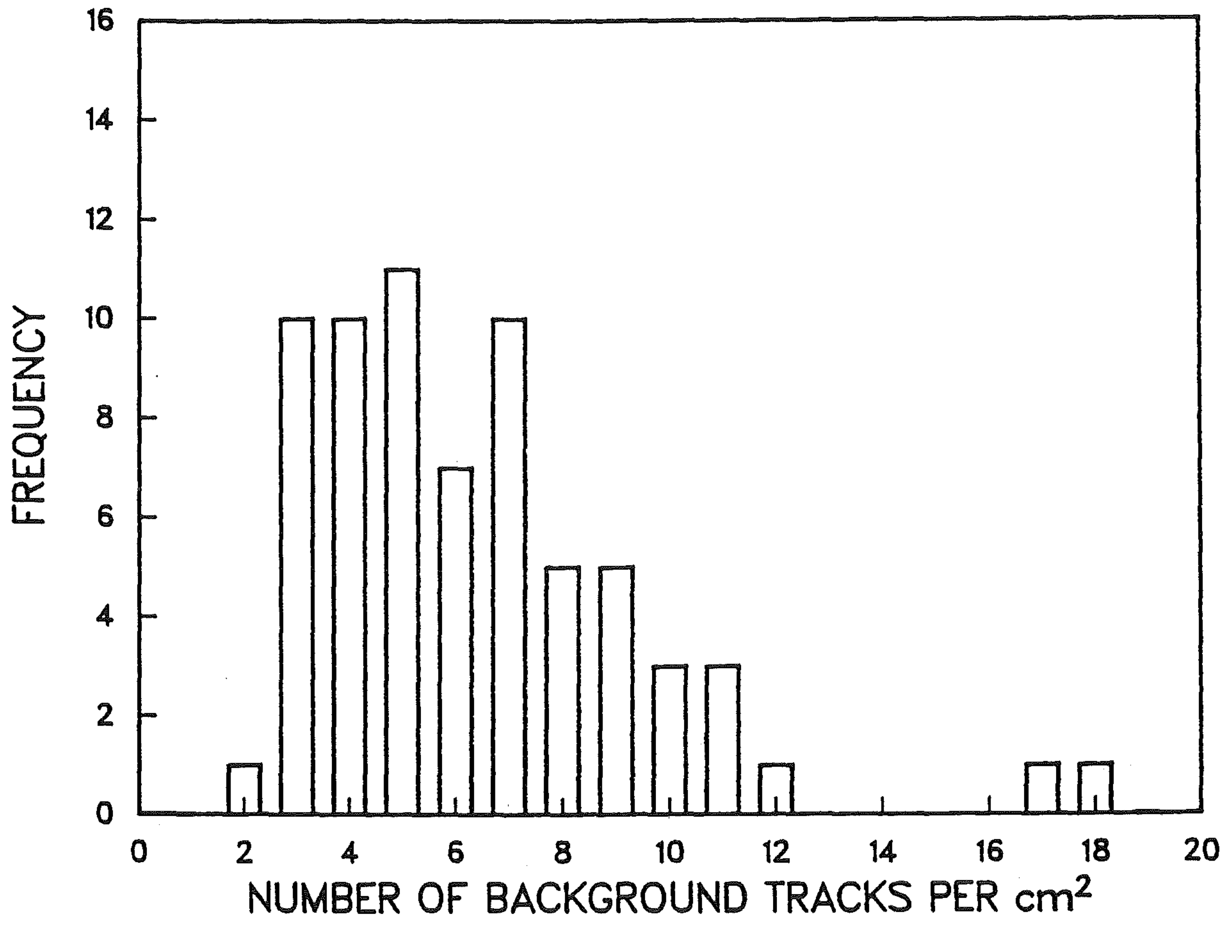

Fig. 1: Frequency distribution of the background track density of Makrofol DE 1-4 after two-step eletrochemical etching.

6 Energy dependence of α-particle registration

For Makrofol DE 1-4 and the two-step ECE technique, the registration efficiency for α-particles and the most probable track diameter vs. α-energy are given in Table 2 and Figure 2. The two-step ECE technique at 26.7 $kV \cdot cm^{-1}$ results in an upper energy threshold at about 3.5 MeV; α-energies as low as 0.5 MeV are registered. Previously investigated etching conditions which make use of another etchant and an electrical field of 23 $kV\ cm^{-1}$ at 50 °C (5) extend the α-energy range up to 5 MeV. Because of the higher etching rate at 50 °C instead of 35 °C, however, etching through effects cannot be avoided. As a consequence this etching conditions cannot be applied routinely.

α-energy (MeV)	Track density (cm^{-2})	Efficiency[1)]	$\pm \sigma_n$	Track diameter (μm)
0.5	787	0.89	0.05	170 ± 15
0.7	839	0.95	0.02	165 ± 15
1.0	815	0.93	0.05	160 ± 15
1.5	824	0.94	0.09	160 ± 15
2.0	886	1.01	0.02	150 ± 15
2.5	785	0.89	0.02	135 ± 15
3.0	801	0.91	0.03	120 ± 15
3.2	812	0.92	0.05	110 ± 15
3.4	887	1.01	0.08	95 ± 15
3.6	805	0.91	0.05	85 ± 15
3.8	530	0.60	0.29	65 ± 15
4.0	35	0.04	0.02	40 ± 15

1) quotient of tracks per cm^2 and α-particles per cm^2 with the standard deviation for four detectors

Table 2: α-particle registration efficiency and most probable track diameter vs. α-energy for Makrofol DE 1-4 and the two-step ECE technique.

Within the radon diffusion chamber radon decay products are deposited on the hemispherical surface in a distance of about 1.5 cm in front of the detector. After the decay of Rn-222 with an α-particle of 5.5 MeV, there is a decay of Po-218 with an α-particle of 6.0 MeV and of Po-214 with an α-particle of 8.78 MeV. For the application in the small diffusion chamber the detector should therefore record high energy α-particles above 4 MeV.

The routinely used etching conditions for α-particles detection, namely conventional pre-etching and ECE at 33.3 $kV \cdot cm^{-1}$ and 35 °C, were adopted in order to record α-particles of high energies within the radon diffusion chamber. The corresponding results are presented in Figure 3 for the bare as well as the Mylar covered Makrofol DE 1-4.

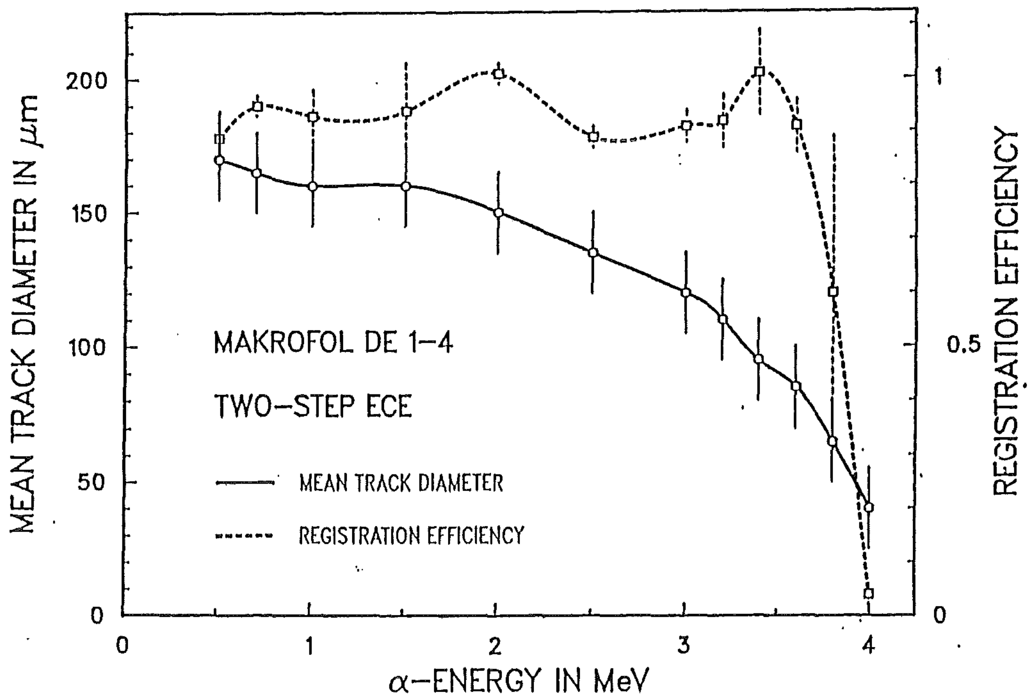

Fig. 2: α-particle registration efficiency and mean track diameter vs. α-energy for Makrofol DE 1-4 after two-step ECE at 26.7 $kV \cdot cm^{-1}$.

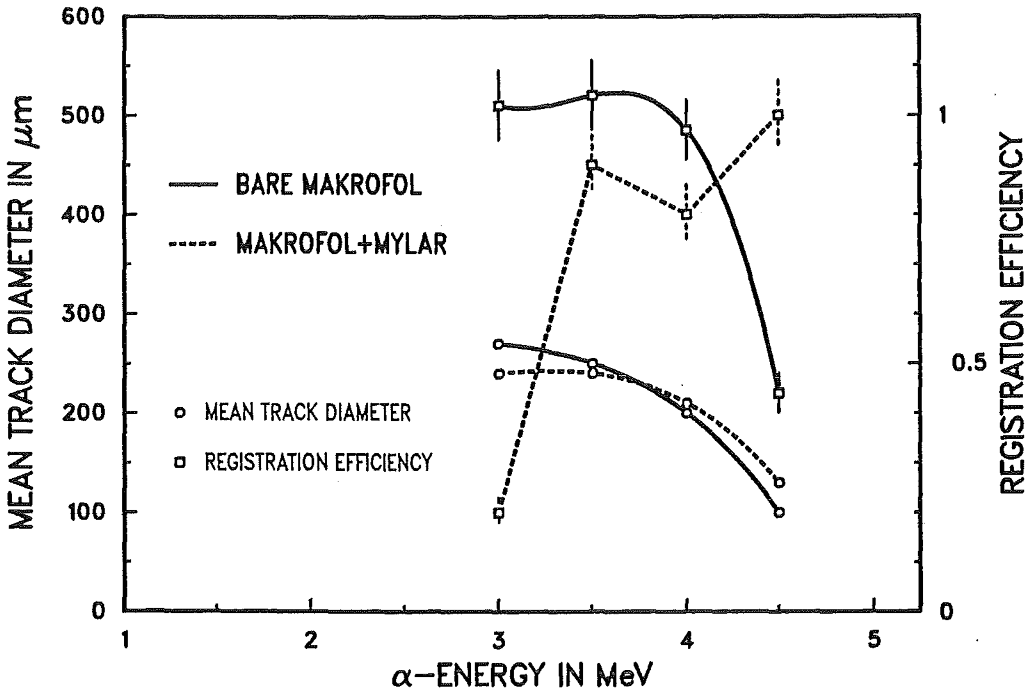

Fig. 3: α-particle registration efficiency and mean track diameter vs. α-energy forbare and Mylar covered Makrofol DE 1-4 after pre-etching and electrochemical etching at 33.3 $kV \cdot cm^{-1}$.

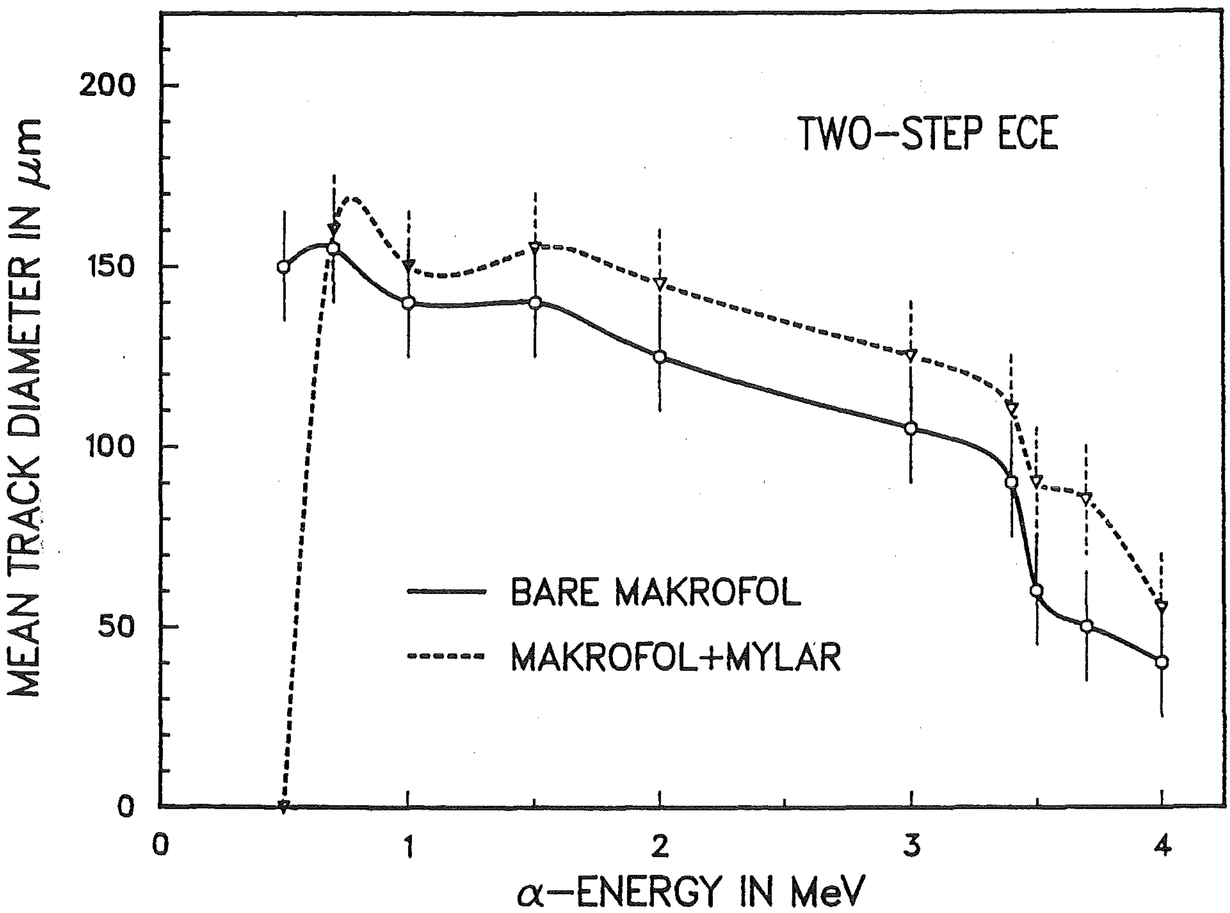

Fig. 4: Mean track diameter vs. α-energy for bare and Mylar covered Makrofol DE 1-4 after two-step electrochemical etching.

Using the two step ECE technique, the effect of the Mylar cover in front of the α-detector is shown in Figure 4 and Figure 5. The results indicate that after the penetration of α-particles through the Mylar cover the track diameter is about 20 μm larger. The Mylar foil, on the other hand, reduces the registration efficiency of low energy α-particles below 1.2 MeV and increases the α-particle registration in the energy range 3.5 MeV to 4 MeV.

For the use in the small sized KfK radon dosemeter, the Makrofol detector must be covered therefore by an absorber foil in order to detect high energy α-particles expected from the decay of Po-218 and Po-214.

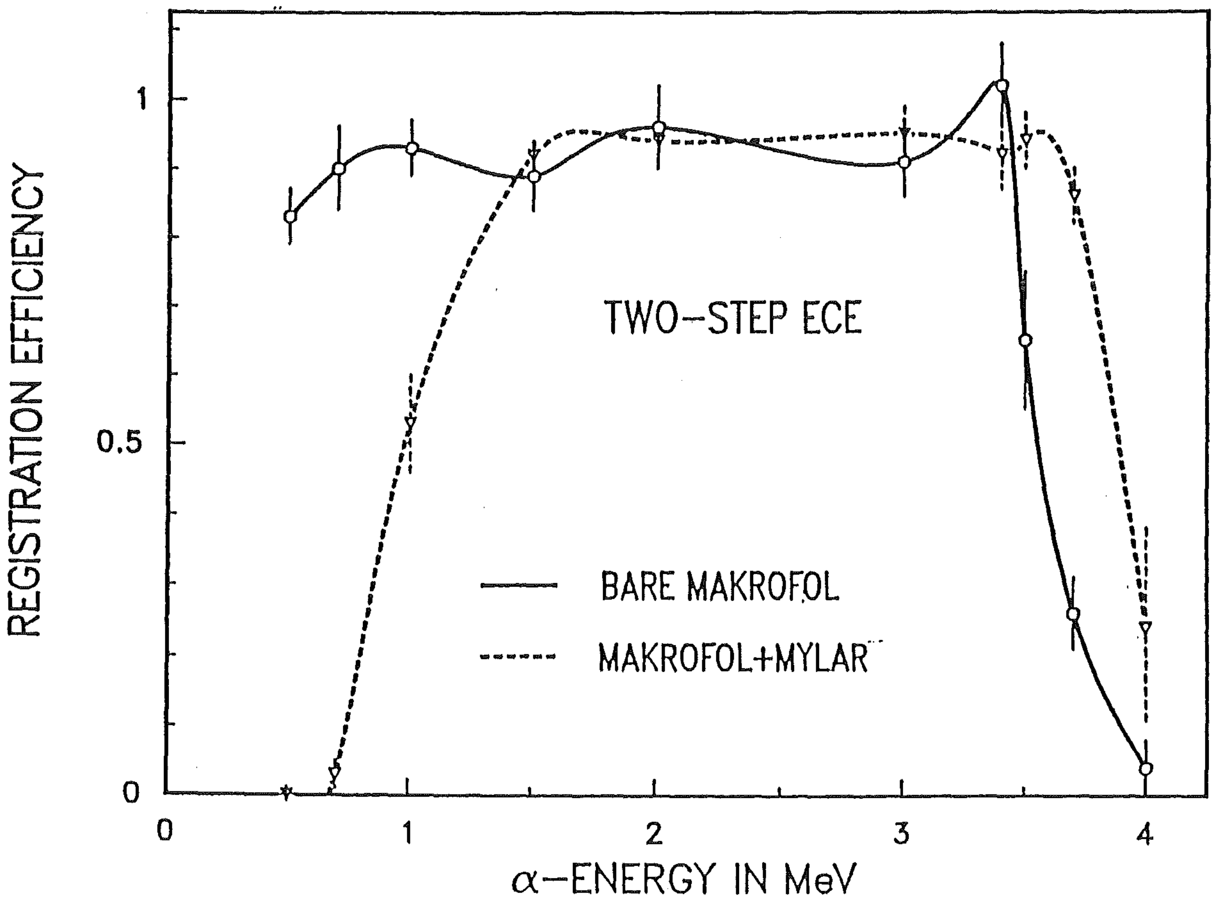

Fig. 5: α-registration efficiency vs. α-energy for bare and Mylar covered Makrofol DE 1-4 after two-step electrochemical etching.

7 Energy dependence for the detection of fast neutrons

The detection of neutron induced recoils and α-particles in Makrofol DE 1-4 was investigated in the energy range 0.575 MeV to 14 MeV. The irradiations with monoenergetic neutrons of 0.575 MeV, 1.2 MeV, 1.6 MeV, 2.0 MeV, 2.5 MeV, 5 MeV and 14 MeV have been performed at the Physikalisch-Technische Bundesanstalt, Braunschweig. The results of this study are presented in Figure 6 and Table 3.

Using the two-step ECE technique and one Makrofol foil as a radiator in front of the detector foil, the energy threshold was found at about 2 MeV. For 0.575 MeV neutrons and irradiations with 500 mSv, the neutron induced recoil tracks were found within the standard deviation of the background track density. Above the energy threshold a neutron response of about 42 $cm^{-2} \cdot mSv$ was found. Figure 6 shows that the neutron response significantly increases by a factor of 2 for 14 MeV. This may be explained mainly by the registration of α-particles due to the $^{12}C(n, n', 3\,\alpha)$ reaction at neutron energies above 12 MeV. The α-particles contribute to the carbon recoils resulting in an increase of the response by a factor of about 2 at 14 MeV.

Neutron energy (MeV)	Dose equivalent (mSv)	Track density[1] ± σ_n (cm^{-2})	Response ($cm^{-2} \cdot mSv^{-1}$)
1.2	406.4	590 ± 36	1.5 ± 0.1
1.2	112.3	133 ± 25	1.2 ± 0.3
1.6	178.5	1054 ± 46	5.9 ± 0.3
2.0	51.4	868 ± 57	16.9 ± 1.1
^{252}Cf[2]	19.3	500 ± 28	26 ± 2
2.5	21.44	720 ± 25	34 ± 4
Am-Be[2]	10.3	493 ± 42	48 ± 4
5	15.24	595 ± 41	39 ± 3
14.8	11.02	880 ± 50	80 ± 5

[1] After subtraction of the actual background track density of (6 ± 2) cm^{-2}; standard deviation of five detectors.
[2] Irradiations with KfK sources.

Table 3: Neutron energy response of Makrofol DE 1-4 for the detection of neutron induced recoils and α-particles; irradiated at PTB (January 1989), two-step ECE technique.

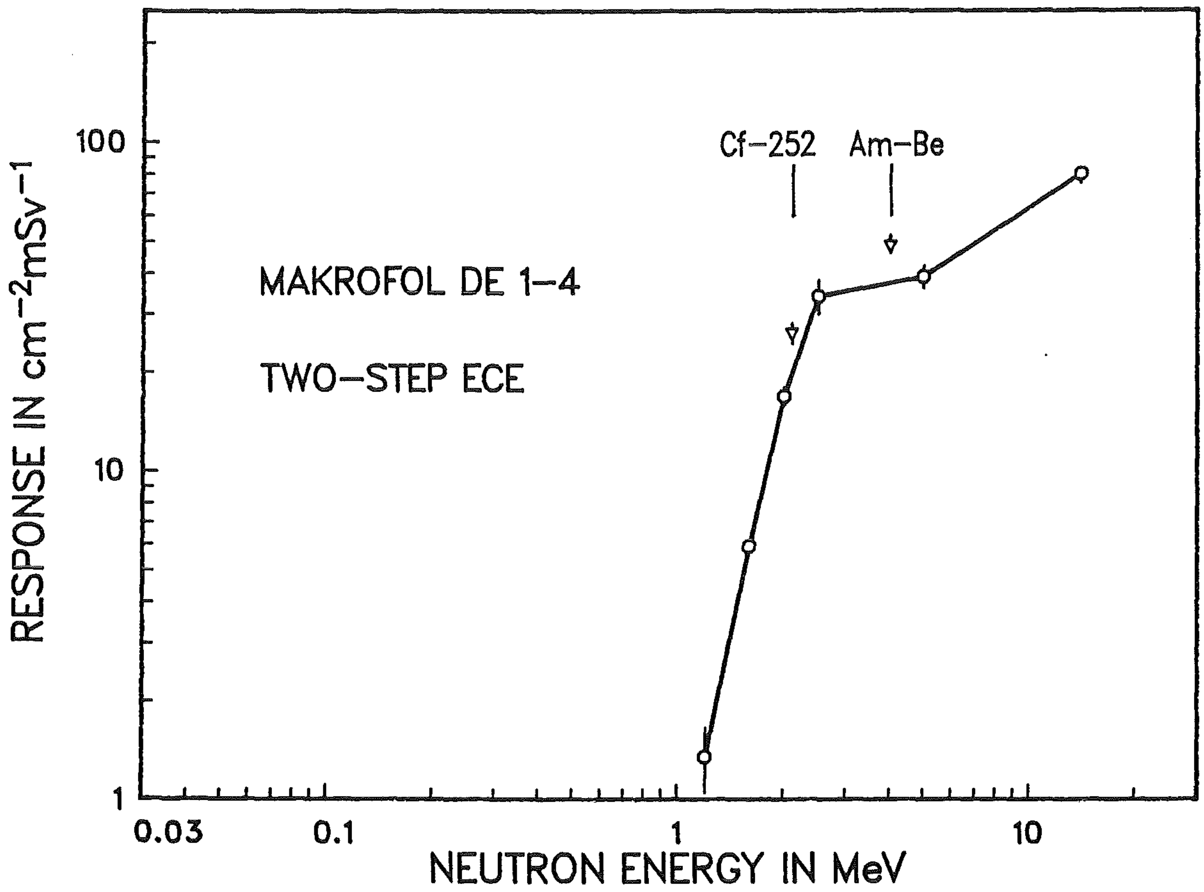

Fig. 6: Response for the registration of neutron induced recoils and α-particles in Makrofol DE 1-4 vs. neutron energy after two-step electrochemical etching.

These results nearly agree with those found previously after chemical etching (6) and after applying pre-etching and elechtrochemical etching (4). With respect to the energy threshold the corresponding values were found to be about 1.2 MeV and 3.5 MeV, respectively. The data for 14 MeV, however, was more or less equal to that for Am-Be neutrons.

In comparison with the neutron detection in CR-39 (8) which is shown in Figure 7 for the two-step ECE technique, Makrofol DE 1-4 cannot register recoil protons. The neutron response of Makrofol DE is therefore lower by a factor of about 10 and neutrons below 1 MeV cannot be detected.

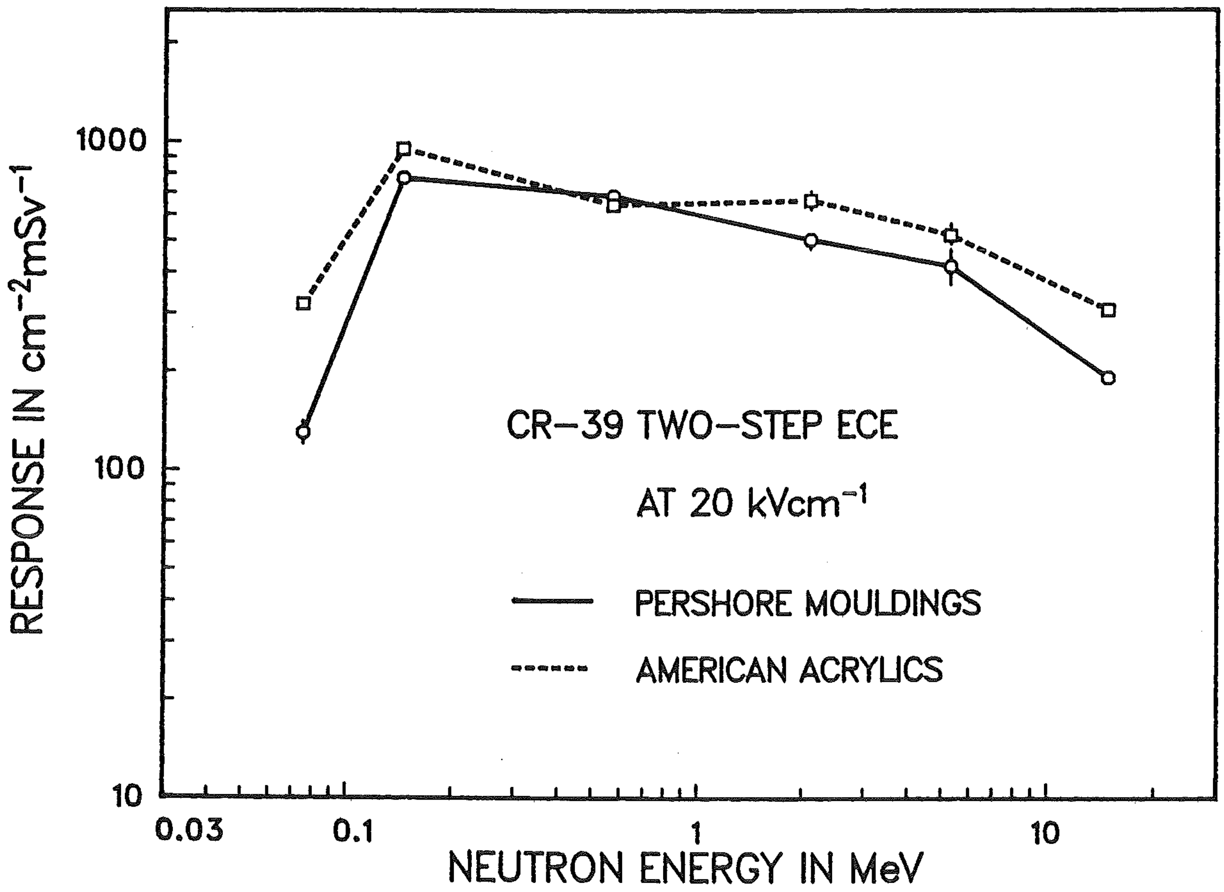

Fig. 7: Response for the registration of neutron induced recoils and α-particles in CR-39 vs. neutron energy after two-step electrochemical etching at 20 kV·cm^{-1}.

8 Conclusions

The investigations of different types of Makrofol DE and samples of various productions have shown that significant differences were found mainly in the background track density and its standard deviation. The previously used Makrofol DE 1-4 commercially available in amounts of about 50 kg is still the best material for a routine application having a background track density of about 6 ± 2 cm^{-2}.

The use of transparent Makrofol DE 1-1 would slightly improve in particular the automatic track counting but makes the handling of foils more difficult because of the need to indicate the side for particle registration and to mark the detectors. Before detector preparation, on the other hand, the detector side with the lower background must be found experimentally.

The application of the same two-step electrochemical etching at 26.7 kV cm^{-1} for the detection of thermal neutrons using (n, α) radiators together with Makrofol detectors as well as the detection of fast neutrons via neutron induced recoils and α-particles resulted in a neutron response of Makrofol DE 1-4 which was found to be a factor of 3 higher compared with the previously applied etching conditions, namely pre-etching followed by an electrochemical etching. With respect to the relative low background track density of 6 ± 2 tracks per cm^2, the lowest detectable neutron dose equivalent is about 0.1 mSv. The neutron response of Makrofol was found to be 42 $cm^{-2} \cdot mSv^{-1}$ and shows an energy dependence of about +100% at 14 MeV and about -50% at the energy threshold of about 2 MeV. Combined with the routinely used KfK albedo dosemeter the Makrofol neutron recoil detector may indicate the dose equivalent of fast neutrons above 2 MeV separately with a high sensitivity, in particular in the application area of research accelerators.

With respect to the detection of α-particles in Makrofol DE 1-4, the two-step electrochemical etching technique allows, in general, the registration of α-particles in the energy range 0.2 MeV to 4 MeV. These etching conditions are applicable for radon diffusion chambers of larger size. For smaller diffusion chambers an additional absorber in front of the detector is needed in order to indicate α-particles in the energy range of about 6 MeV and 8 MeV. In comparison with CR-39, Makrofol 1-4 shows practically the same registration efficiency with a similar track diameter distribution and α-energy range.

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