CONTROLLED PRODUCTION OF NUCLEAR MICROFILTERS BY THE FISSION TRACK REGISTRATION TECHNIQUE

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INTRODUCTION

In the present experiment filters with holes in the 0.1 μm to 10.5 μm size range and pore density around 10^8 pores/cm^2 to 10^5 pores/cm^2 respectively have been developed by using the solid state nuclear track detection technique in Makrofol KG foils together with optical and scanning electron microscopes(SEM) for pore analysis. Some characteristics of these microfilters such as pore size and pore density fluctuation as well as the best chemical etching conditions were studied.

Filters with pore diameters in this size range (0.1 μm - 10.5 μm) are usually employed in the following application fields:
Air Pollution Analysis; Bacterial Analysis; Beverage and Food Quality Control; Blood Filtration; Cell Culture; Cytology; Water Microbiology; and in General Filtration such as in beverage stabilization, particulare removal, bacterial removal and in all general microporous filtration applications.

EXPERIMENTAL METHODS AND RESULTS

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The experimental method basically consists in exposing thin polycarbonate plastic films to a collimated beam of fragments produced by the U-235 fission with thermal neutrons. These fission fragments produce tracks with diameter around 0.008 μm across the entire thickness of the plastic foil which may be selectively attacked (etched) by an appropriate chemical solution producing a fine channel or hole with diameter up to 10.5 μm in the bombarded material. The hole diameter is thus controlled by the etching time and the density of holes is determined by the particle dose or irradiation time[1].

In Figure 1 is presented a sketch of a simple device named irradiation chamber we have used to produce the holes in the polycarbonate plastic film.[2]

The aluminium chamber containing the plastic foil and the fissile material was evacuated to 10^{-3} torr and then irradiated with thermal neutron at the beam-hole BH-10 of the IEA-R1, 2 MW pool type research reactor (Figure 2). The chamber evacuation is necessary in order to avoid absorption of the fission fragments by the air[3].

Inside this chamber the polycarbonate plastic Makrofol KG (47 mm diameter, 10 μm thickness) may be positioned at different distances from de fissile material depending on the required level of collimation for normal incidence of the fission fragments. The fissile target consisted of a disk of aluminium (1 mm thickness) onto which has been electrolytically deposited around 1 mg/cm^2 of U_3O_8 with 93.15 % enrichment in U-235, forming an active area with about 31 mm diameter. This layer of fissile material is much thinner than the range of fission fragments in U_3O_8

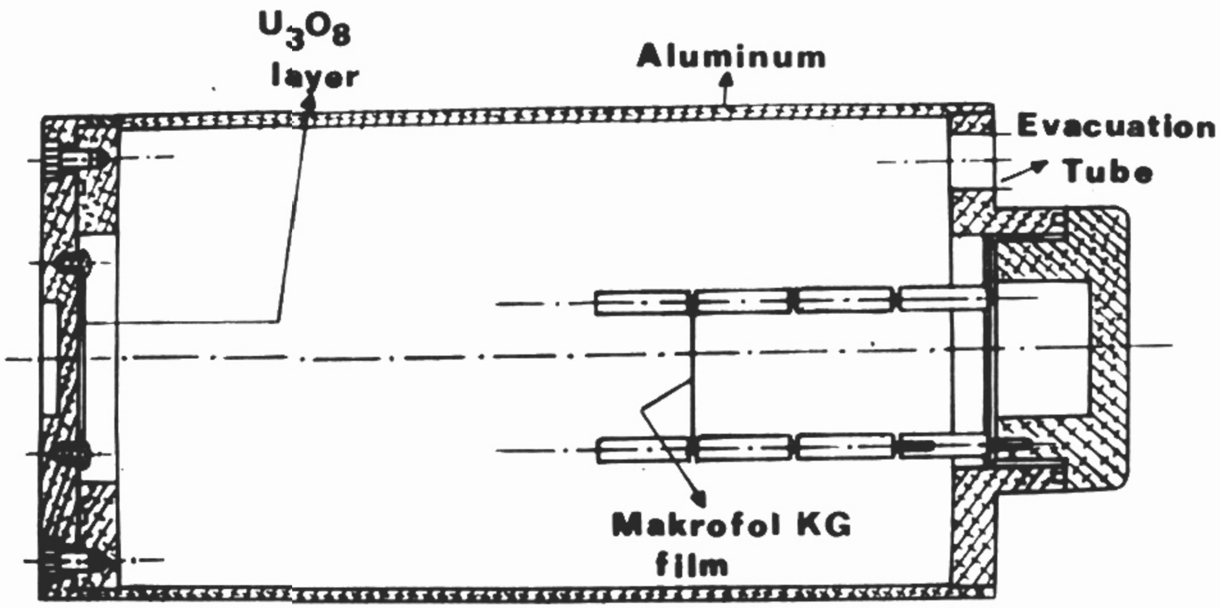


FIGURE 1: Irradiation Chamber

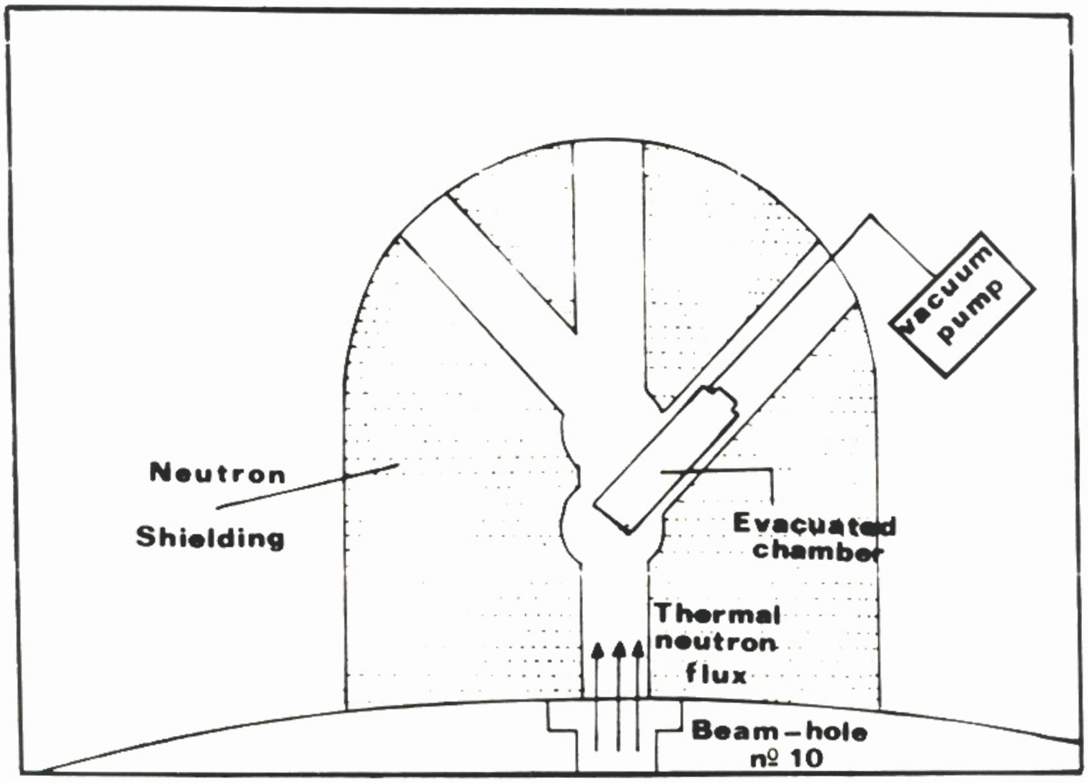


FIGURE 2 Experimental Arrangement

(10 mg/cm^2)[4] and thus one can say that practically all U-235 nuclei, emmiting fission fragments inside the solid angle of irradiation, contribute to the track production in the plastic film.

The thermal neutron flux density at the irradiation position of the Uranium sample has been measured by the activation technique with gold foils and the result obtained was 10^7 n/cm^2.s. The fragments produced by the nuclear fission reaction in U^{235} nuclei are isotropically emitted and they impinge perpendicularly on the plastic foil if it is positioned at an adequate distance from the U^{235} source.

In the present work we have obtained a good level of

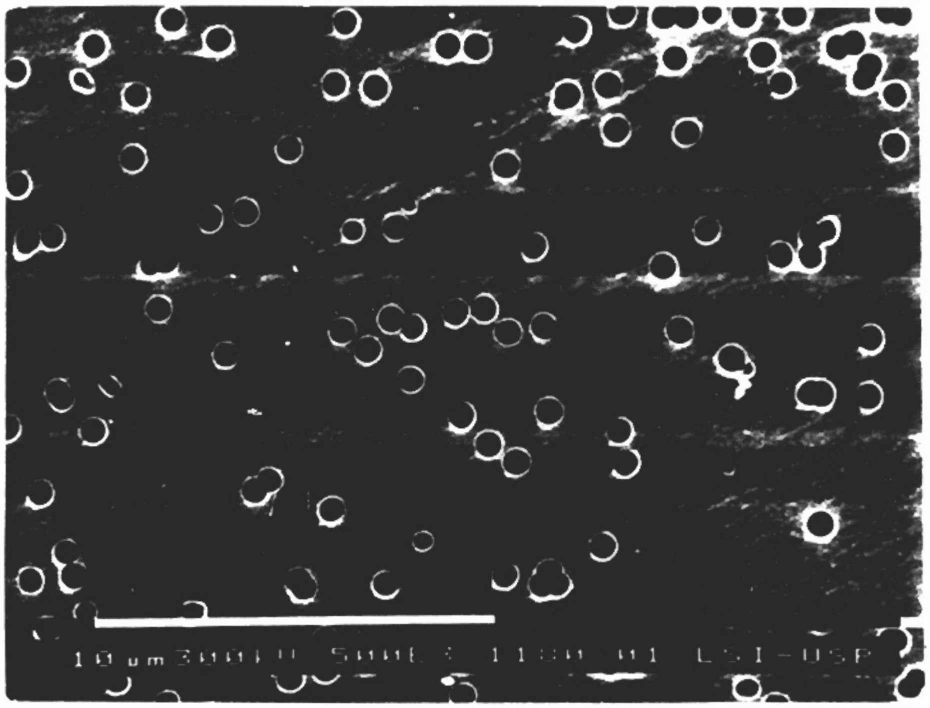


FIGURE 3 Pore size: 0.6 μm. Pore density: 2 10^7 pores/cm^2

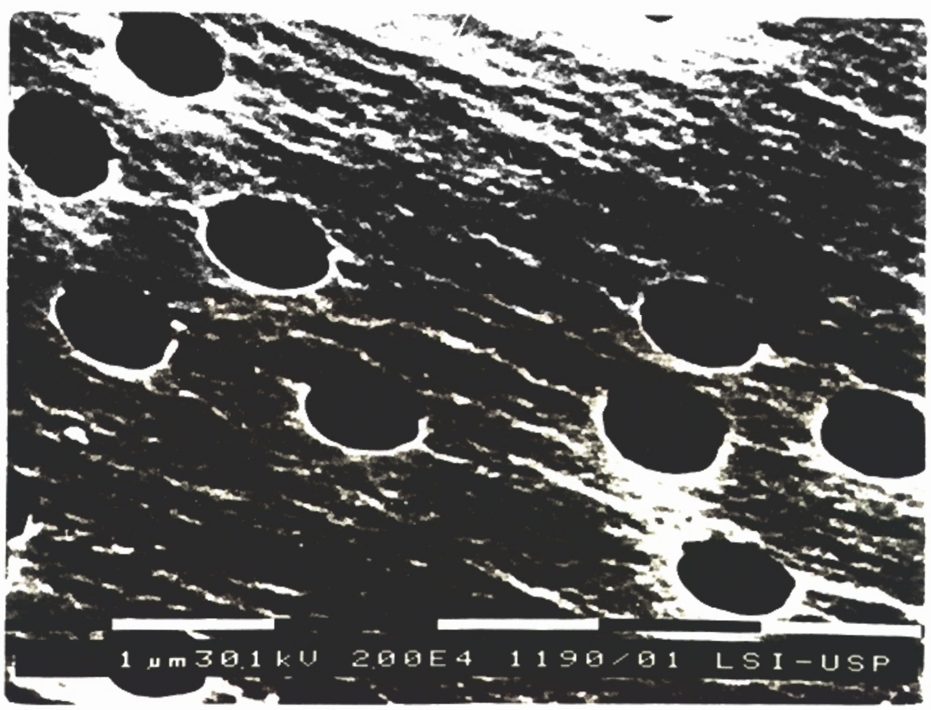


FIGURE 4 Pore size: 0.8 μm. Pore density: 2 10^7 pores/cm^2

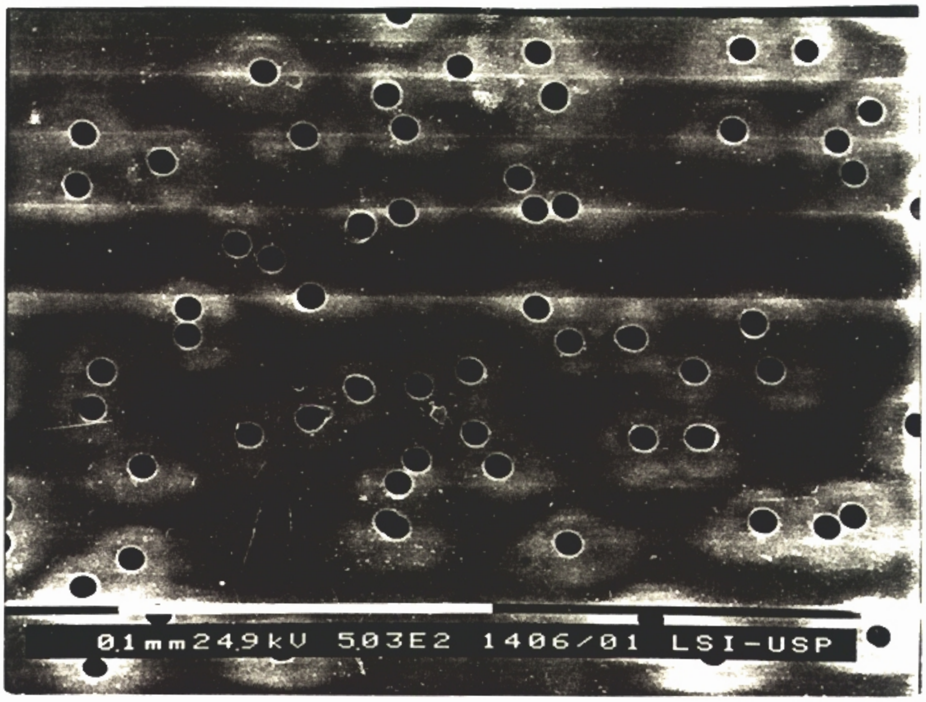


FIGURE 5 Pore size: 7.0 μm. Pore density: 10^5 pores/cm^2

FIGURE 6 Pore size: 5.5 μm. Pore density: 10^5 pores/cm^2

collimation (98% of the pores were practically circular) at the irradiation position 17.5 cm from the uranium source, using films with 4.2 cm active diameter. This result is in excellent agreement with the experimental studies carried out by Quinn et al[5].

For illustration, in Figure 3, 4, 5 and 6 are shown SEM microphotographes of four typical samples of Makrofol KG microfilters. As can be seen the produced pores are cylindrical and practically of the same size It is also apparent the smoothness on their surfaces and in the inner wall of the pores.

The pore size distribution in the film Makrofol KG has been verified, for a particular sample of microfilter, using an optical measuring microscope. A total of 500 pores diameters were analysed and the results obtained shown in Figure 7 are in good agreement with the histogram presented by Czilwa et al[6].

The mean diameter calculated for the distribution of Figure 7 was (6.5 ± 0.7) μm indicating a size pore fluctuation of about 11%. The distribution of holes was also verified performing several pore counting for several different areas of a particular sample of Makrofol KG microfilter. A fluctuation of about 10% has been obtained for the pore density after analysis of 47 different areas representing a total of 6000 holes. These fluctuations are in good agreement with those presented by commercially known track-etched membranes.

In order to obtain adequate times of filtration for microfilters with small pore size (lower than 1 μm) it is necessary to increase the pore density. However, for high pore densities the film gets damaged due to the high fluency of fission fragments that impinged on it

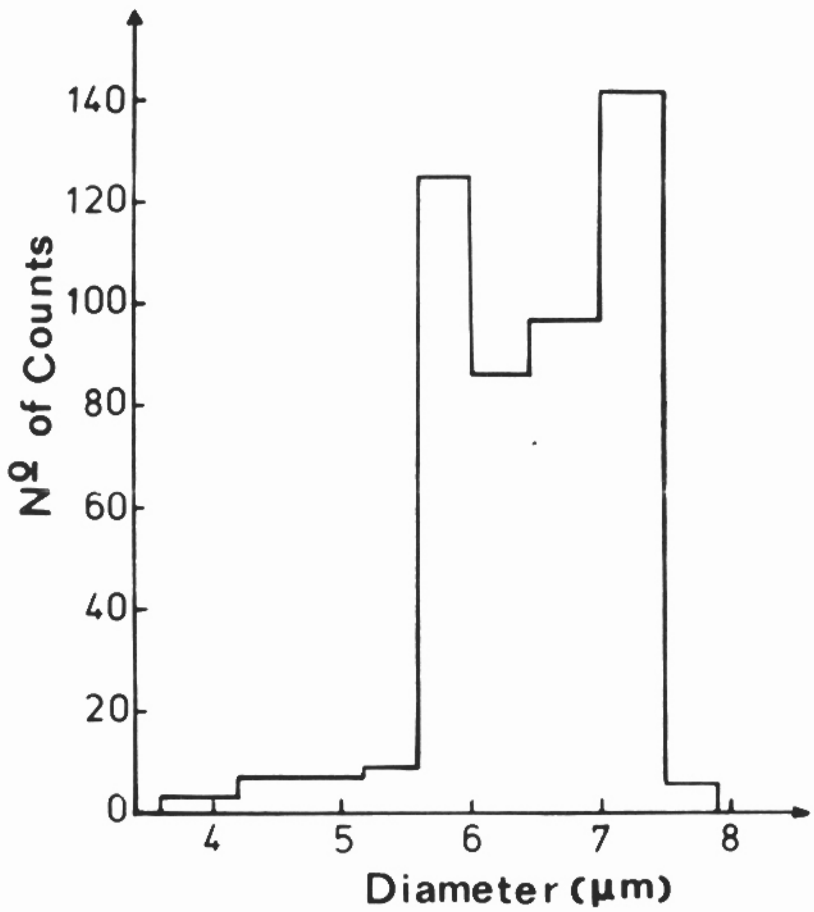


FIGURE 7 Pore size distribution

and consequently, the chemical attack on the films will be much stronger than for low pore densities[7,8]. Therefore different chemical etching conditions must be used depending on the particular pore size and pore density interval.

The best etching conditions obtained for the irradiated films of Makrofol in the pore size range of interest (0.1 μm to 10.5 μm) are presented in Table 1.

TABLE 1

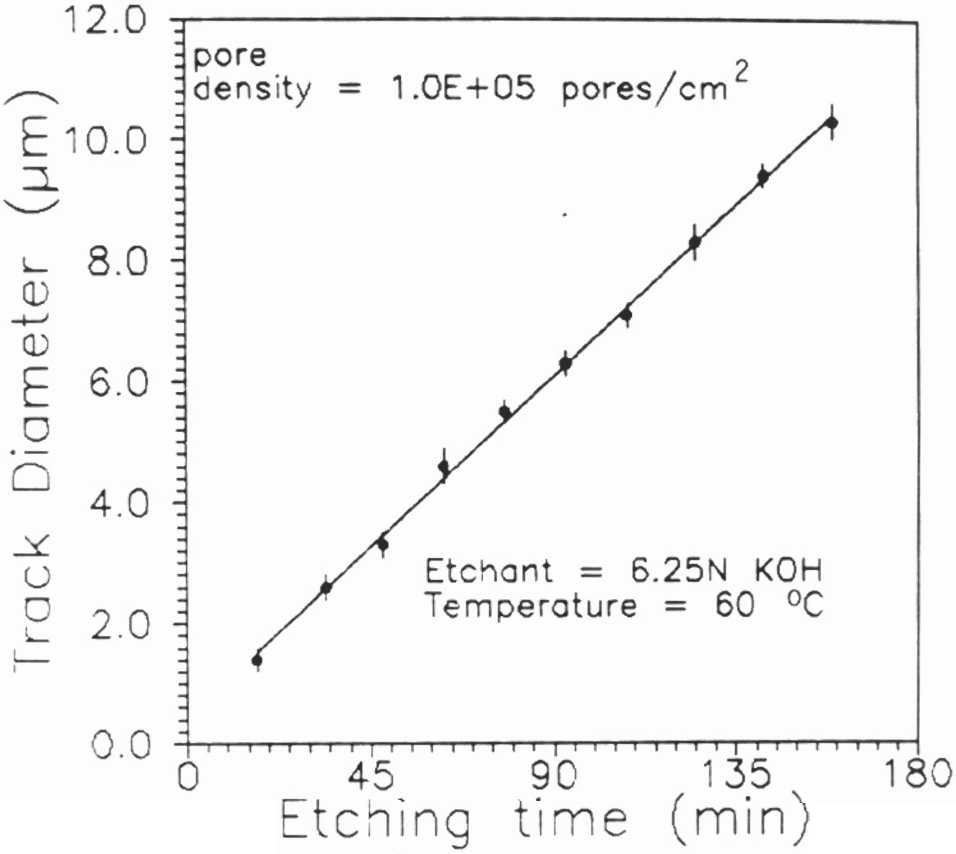
Chemical Solution	Temperature (^{o}C)	Pore Density ($pores/cm^2$)	Pore size (μm)
6.25 N KOH	60	10^5	1.40-10.5
6.25 N NaOH	45	$2\ 10^7$	0.30-1.10
5.00 N NaOH	35	10^8	0.15-0.47

In order to obtain the calibration curve track diameter versus etching time, around 10 irradiated films were etched in different time intervals for each etching condition listed in Table 1. The results obtained are shown in Figure 8, 9 and 10. The solid line in these figures represents the least squares fitting to the experimental data points.

For many applications fields it is important that the impurity or contamination level of the filters be the lowest possible. Special cares were taken during the handling of microfilters along all their steps of production. Table 2 shows the result obtained by neutron activation analysis technique for a particular sample of our filter. The present results are in excellent agreement with those reported by other commercially available filters[9].

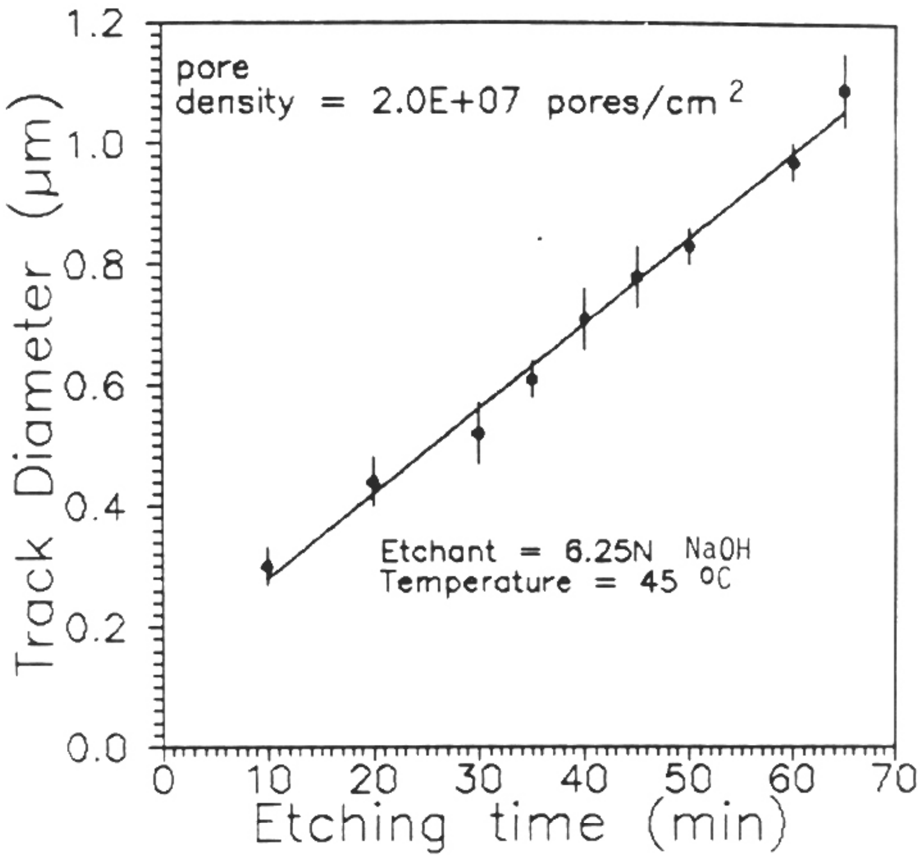
TABLE 2 Content of Trace Elements in Microfilter Material

METAL	ng/cm ^{2}	
Al	15.5	± 0.8
As	< 0.007	
Au	0.00046	± 0.00006
Ba	< 5.4	
Br	6.1	± 0.5
Ce	< 0.13	
Co	0.09	± 0.01
Cl	14	± 4
Cr	0.660	± 0.005
Fe	< 16	
La	< 0.005	
Mn	0.12	± 0.01
Mo	< 0.08	
Na	5.5	± 0.3
Rb	< 1.1	
Sc	< 0.0012	
Se	< 0.3	
Sb	0.005	± 0.001
Sm	< 0.0014	
Th	< 0.02	
V	< 0.03	
W	< 0.01	
Zn	< 2.1	



12.0
10.0
8.0
6.0
4.0
2.0
0.0
Track Diameter (µm)
pore
density = 1.0E+05 pores/cm2
Etchant = 6.25N KOH
Temperature = 60 °C
0
45
90
135
180
Etching time (min)

FIGURE 8



pore
density = 2.0E+07 pores/cm 2
Etchant = 6.25N NaOH
Temperature = 45 OC
Track Diameter (μm)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
0
10
20
30
40
50
60
70
Etching time (min)

FIGURE 9

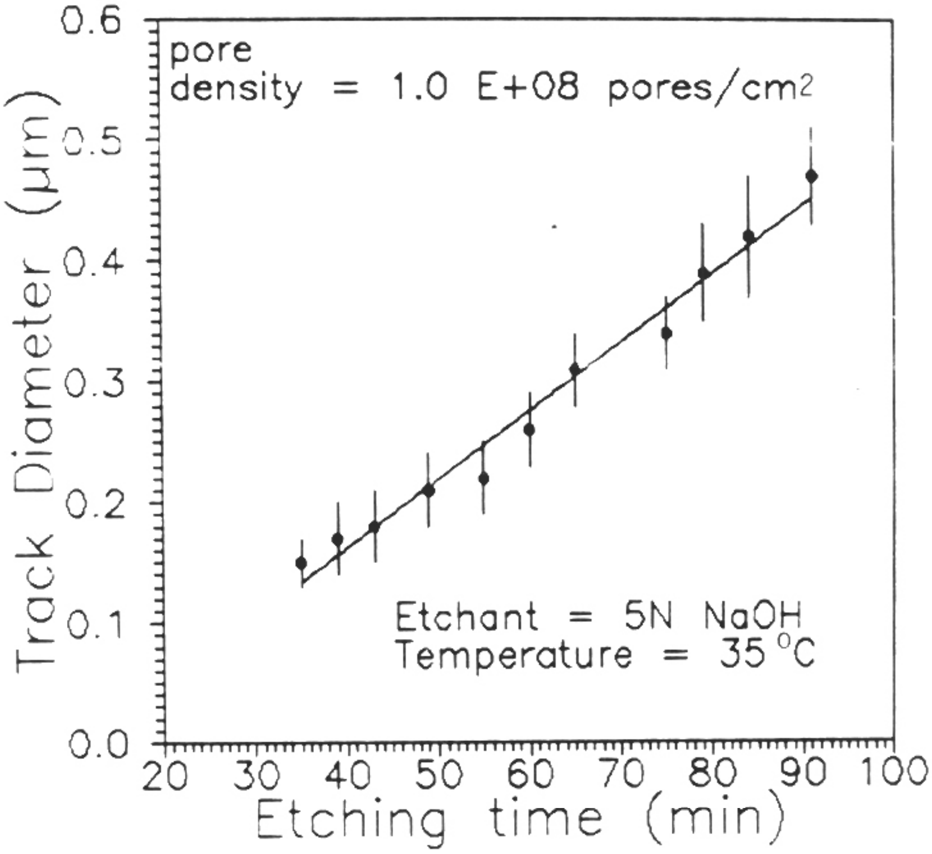

pore
density = 1.0 E+08 pores/cm2
Etchant = 5N NaOH
Temperature = 35 °C
Track Diameter (μm)
0.0
0.1
0.2
0.3
0.4
0.5
0.6
20
30
40
50
60
70
80
90
100
Etching time (min)

FIGURE 10

CONCLUSION

The track etched membranes (TEM) developed in this work have shown to be convenient and adequate for several applications in Industry and Biotechnology. The main characteristics of these microfiltration membranes were studied and the results obtained indicated comparable quality to commercially available TEMs.

The development of a new methodology to produce ultra-filtration membranes with pore size lower than 0.1 μm is in progress.

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