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ON THE ELECTROCHEMICAL ETCHING OF NEUTRON-INDUCED TRACKS IN PLASTICS AND ITS APPLICATION TO PERSONNEL NEUTRON DOSIMETRY

G.M. Hassib*), J.W.N. Tuyn and J. Dutrannois
CERN, Geneva
Switzerland

ABSTRACT

The recently developed electrochemical etching method seems to be a promising technique to reveal neutron-induced recoil tracks in plastics. However, very little work has been done and the data presented are preliminary.

The purpose of the work presented here was to investigate in some detail the most important parameters affecting the electrochemical etching kinetics of recoil tracks in Makrofol polycarbonate ($\sim 500\ \mu$) and to find the optimum conditions for applying this technique to personnel neutron dosimetry.

The electric track diameter and the neutron sensitivity were studied as a function of frequency, applied voltage, etching condition, neutron dose, neutron spectrum, and the directional dependence.

Special cells were designed in such a way that only one surface of the plastic foils was etched electrochemically while the other surface was made conductive and then connected with the earth terminal of a high voltage function generator. With this technique the electric tracks appear as dark stars on a quite clear background, which permits a more reliable evaluation of the track density, using any automatic or semi-automatic scanning equipment.

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*) Present address : Atomic Energy Establishment, Cairo, Egypt.

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INTRODUCTION

During the last few years extensive efforts have been devoted to the search for a personnel neutron dosimeter with better characteristics than those of the conventional photographic nuclear track emulsions. Dielectric track detectors have been proposed as a valuable alternative method because of several advantages. Combinations of fissile materials and thin plastic foils to be counted by a spark counter have been used in routine and accident personnel neutron dosimetry in different laboratories.

However, radiation hazard and contamination risk inherent in fissile materials have not encouraged a large-scale application of this method. The detection of fast neutrons by registration of induced recoil nuclei and/or alpha particles produced in (n,α) reactions in plastic track detectors may overcome most of these limitations. The application of this technique in practice has encountered some problems due to the fact that the resulting tracks are characterized by a wide spectrum of diameters and ranges which make the visual counting difficult, unreliable and time-consuming.

The recently developed electrochemical etching technique introduced by Tommasino(1) makes it possible to overcome the difficulties of using non-fissile radiators. Employing the electrochemical etching method the recoil nuclei and alpha particle tracks can be enlarged to such a size that they can be seen by the naked eye or displayed on a screen by using a slide projector (Fig. 1).

The method is based on the fact that the particle damage trails in plastics are characterized by a high conductivity compared with that of the undamaged region. On applying a pulsed high voltage during the etching process an electrical breakdown is induced at the end of the damage trails in the bulk of the plastic foils.

In this technique the plastic foil to be etched electrochemically is placed between two insulated containers filled with the etchant in such a way that a good contact between the plastic detector and the containers maintains the electrical insulation of the two solutions. Platinum electrodes in the containers are connected with a high-voltage function generator.

In preliminary experiments using this technique (2), the application of the electrochemical etching method for dosimetry purposes proved to have a number of advantages over other photographic or non-photographic nuclear track registration methods.

To use this technique in the optimal way it is essential to have a well established fundamental knowledge of the influence of several parameters of the method. Among the essential parameters are the type of detector and the thickness of the foil, the applied voltage and frequency, the nature, temperature and concentration of the etchant, the neutron energy and cell geometry. So far these parameters have never been studied systematically.

It is the aim of this work to investigate in detail the influence of the most important parameters affecting the registration of neutron-induced tracks in polycarbonate plastic detector. The study was restricted to the nature and concentration of the etchant, etching time, frequency, applied voltage, neutron energy, and directional dependence. Special cells were designed in such a way that only one surface of the plastic foils was etched electrochemically while the other surface was made conductive and then connected with the earth terminal of a high-voltage function generator. With this technique the electric tracks appear only on one surface of the plastic foil while the other surface remains clear without any disturbing background. All measurements were done using 500 μ thick Makrofol*) E sheets after being irradiated with 14 MeV and PuBe neutrons.

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*) Trade name Bayer, Germany.

CONCENTRATION AND NATURE OF ETCHANT

An extensive effort was made to study the effect of concentration and nature of etching solution on the track diameter and registration efficiency of Makrofol E. For these experiments a special cell with six separate chambers and a common electrode was designed such that six foils could be etched simultaneously using different etchant concentrations or different etching solutions.

KOH was chosen to study the effect of etchant concentration on the track diameter in a given electrochemical etching condition. Figure 2 shows the results of these measurements for solutions containing 15% - 40% KOH. In this figure the electric track diameter increases with increasing etchant concentration to a maximum at about 25% and then decreases to a minimum at about 35% KOH, after which the diameter increases again. This tendency follows approximately the electrical conductivity curve at the region between 15% and 35% as it may be expected from electrochemical kinetics. It means that for a given etchant there is an optimum concentration at which the electrical conductivity reaches its maximum value. In the case of KOH the optimum concentration was found to be around 25% at room temperature.

Studies have shown that the addition of ethyl alcohol to the etching solution increases the bulk etching rate of the plastic detectors(3). In this work we have tested different mixtures of ethyl alcohol and 25% KOH solution. The highest track diameter was found at a mixture containing 1:1 volume ratio of alcohol and 25% KOH solution as shown in Fig. 3. At a given etching condition the rate of track diameter growth using alcoholic solution was found to be more than three times higher than that for 25% KOH.

EFFECT OF ETCHING TIME

Compared with the conventional etching method, the electrochemical etching technique is controlled by additional parameters affecting the rate of track growth. Figure 4 shows the electric track diameter as a function of etching time at different time intervals. In this figure the rate of track growth increases with increasing time interval. This tendency may be attributed to the continuous generation of heat due to the electric current in the etching solution. The induced heating accelerates the rate of track diameter growth up to a point where the rate of heat generation is equal to the rate of heat loss by radiation. In this condition the growth of the tracks behaves in the same way as in the case of conventional etching.

ELECTRICAL PARAMETERS

The efficiency of the electrochemical etching is strongly dependent on the frequency and the applied voltage. Figure 5 shows the effect of the frequency on the maximum track diameter and the neutron sensitivity at frequencies between 1 and 10 kHz. The track diameter increases with increasing frequency up to a point where it levels off, while the change in the neutron sensitivity is negligible within ± 20%.

Figure 6 shows the increase in the neutron sensitivity as a function of applied voltage using two etching solutions with different etching rates. From these experiments one can conclude that there exists a threshold value of the applied voltage for the appearance of the electric tracks. Above this threshold and for a given neutron dose the number of electric tracks increase with increasing applied voltage up to about 3 kV, where the track density levels off. Moreover, one can see from Fig. 6 how the nature of the etching solution is strongly affecting the registration efficiency of the detector. It shows that the higher the etching rate of the solution, the higher the registration efficiency. The alcoholic solution gives an increase of a factor of two for the neutron sensitivity compared to 25% KOH in a given condition.

These results show that for maximum efficiency and reproducible quantitative measurements using 500 μ thick foils, one should carry out the electrochemical etching process using alcoholic solutions at more than 3 kV and more than 5 kHz at room temperature. Of course, optimum etching conditions have to be re-established whenever a different foil type or thickness, etchant or etching temperature is used.

NEUTRON ENERGY AND DIRECTIONAL DEPENDENCE

For the application to neutron dosimetry of the electrochemical etching technique three main points have been briefly investigated. The first was the energy dependence on the neutron sensitivity, the second was the directional dependence, and the third was the lower detectable limit in relation to the background tracks. Foils were irradiated with PuBe and 14 MeV neutrons at different angles. The foils were etched electrochemically under identical conditions. The neutron doses were derived from measurements with a Rem counter.

Table 1 shows the results of these measurements. It can be seen that the neutron sensitivity varies by a factor of about 3 between PuBe and 14 MeV neutrons for normal incidence. This factor increases as the angle of neutron incidence decreases. These results agree well with those of previous studies(4) using visual hole counting of cellulose nitrate LR-115.

It can be seen also from Table 1 that the detector sensitivity is strongly dependent on the direction of neutron incidence. It is of interest to point out that, unlike conventional etching, the directional dependence is less pronounced with 14 MeV neutrons than with PuBe neutrons. This tendency may be due to the differences in the critical particle detection angle.

BACKGROUND TRACKS AND LOWER DETECTION LIMIT

One of the important problems we have faced is the background tracks in the plastic foils. The background track density depends on the age of the plastic foils, the condition of storage, and the etching condition and determines the reliability of the method for measurement of low neutron doses. After several attempts to solve this problem we reached the following conclusion.

The plastic foils have to be etched electrochemically to reveal the background tracks using the same etching condition which will be applied later for etching the exposed foils. After irradiation, a short conventional etching before applying the high voltage gives the background tracks a quite different configuration than the newly induced neutron tracks as shown in Fig. 7. By using this process the lower detection limit, within ± 10% reproducibility, becomes lower than 500 mrem in the present condition. This limit could be lowered by using new thinner plastic foils.

REFERENCES

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2. M. Sohrabi, Health Physics, 27, 598 (1974).
3. G. Somogyi and D.S. Srivastava, Proc. 7th Int. Colloq. Nucl. Photogr. and SSTD, Barcelona, 1970, p. 711.
4. G.M. Hassib and L. Medveczky, ATOMKI, 16, 315 (1974) and Proc. 2nd Symp. Neutron Dosimetry, Neuherberg/Munich, 1975, Vol. I, p. 535.

Table 1

Directional dependence of the registration efficiency of Makrofol E at different neutron energies

Direction	PuBe spectrum		14 MeV	
	$t/cm^2/rem$	%	$t/cm^2/rem$	%
90°	50	100	135	100
45°	30	60	100	74
0°	15	30	60	44

Etching condition : 2 kV, 3 kHz, 25% KOH + C_2H_5OH, ~ 3 hours.

FIGURE CAPTIONS

Fig. 1. Micrograph of recoil particle tracks in 500 μ thick polycarbonate etched 2 h in alcoholic solution at room temperature, 3 kV, 3 kHz (x 50).

Fig. 2. Effect of the concentration of KOH on the electrochemical etching of 500 μ thick Makrofol E.

Fig. 3. Effect of mixing 25% KOH with ethyl alcohol on the electrochemical etching of 500 μ thick Makrofol E.

Fig. 4. Effect of etching time on the electrochemical etching of 500 μ thick Makrofol E.

Fig. 5. Effect of frequency on the electrochemical etching of 500 μ thick Makrofol E.

Fig. 6. Effect of applied voltage on the neutron sensitivity of 500 μ thick Makrofol E irradiated with fast neutrons from a PuBe source.

Fig. 7. Micrograph showing the difference between the background tracks (pale) and the neutron induced tracks (black) by applying the process described here (x 50).

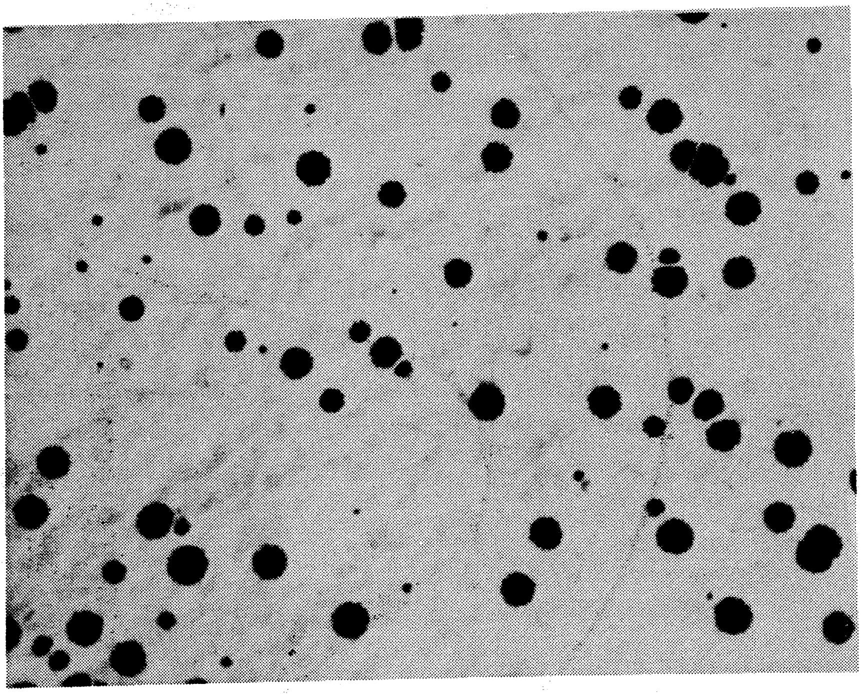


Fig:1

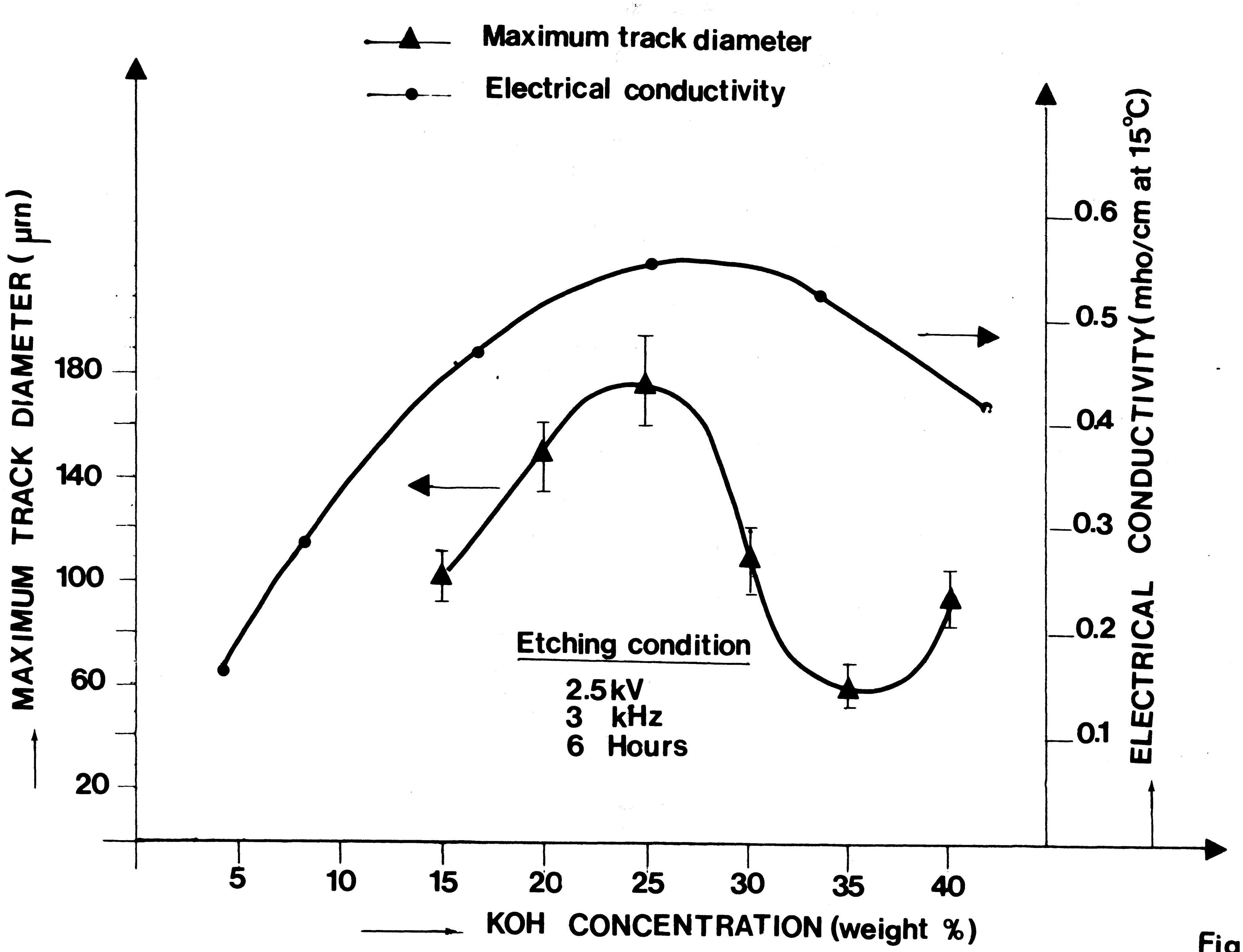
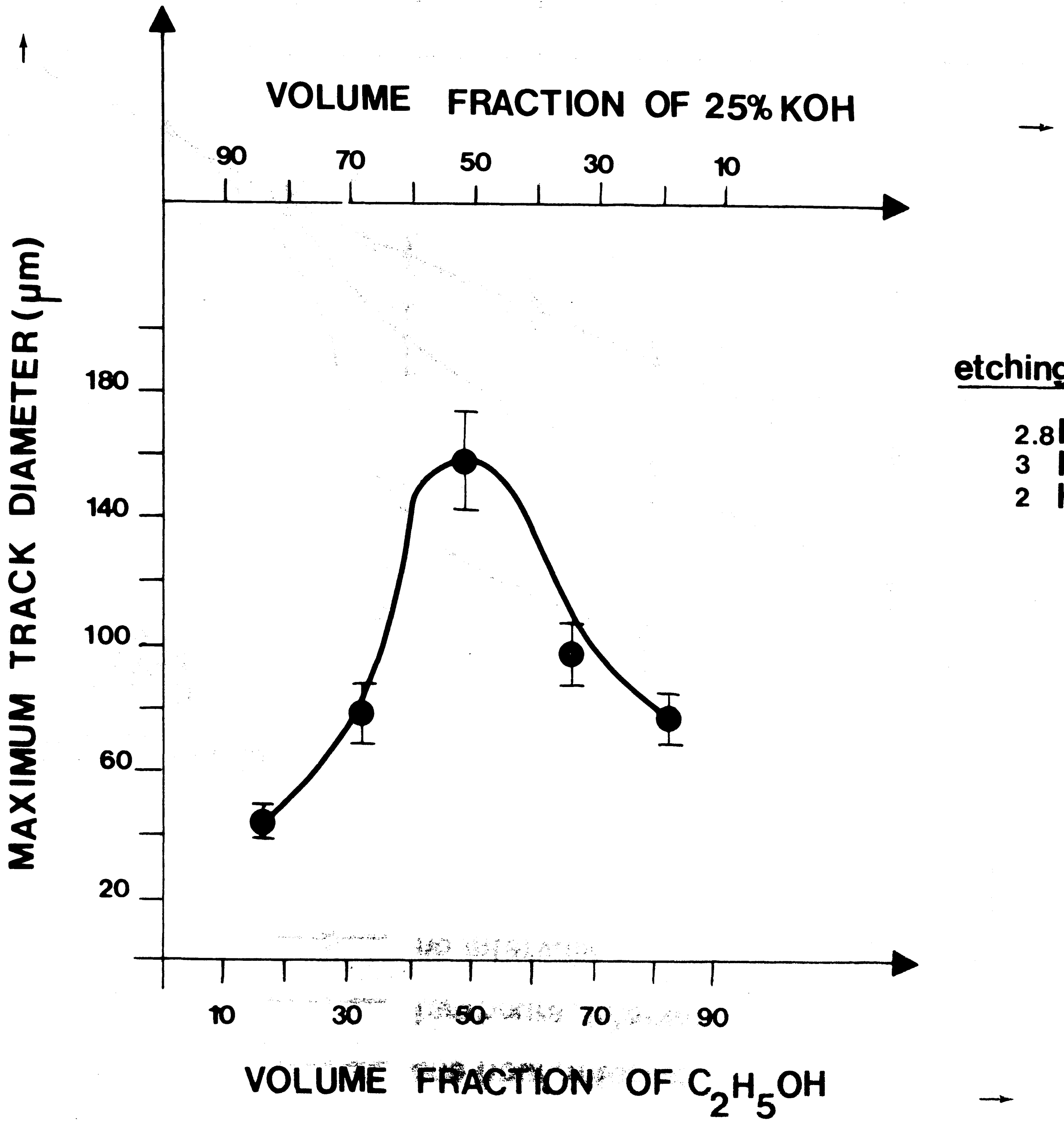

Maximum track diameter
Electrical conductivity
MAXIMUM TRACK DIAMETER (μm)
180
140
100
60
20
ELECTRICAL CONDUCTIVITY (mho/cm at 15°C)
0.6
0.5
0.4
0.3
0.2
0.1
5
10
15
20
25
30
35
40
KOH CONCENTRATION (weight %)
Etching condition
2.5 kV
3 kHz
6 Hours

Fig : 2



etching condition

2.8 kV
3 kHz
2 Hours

Fig:3

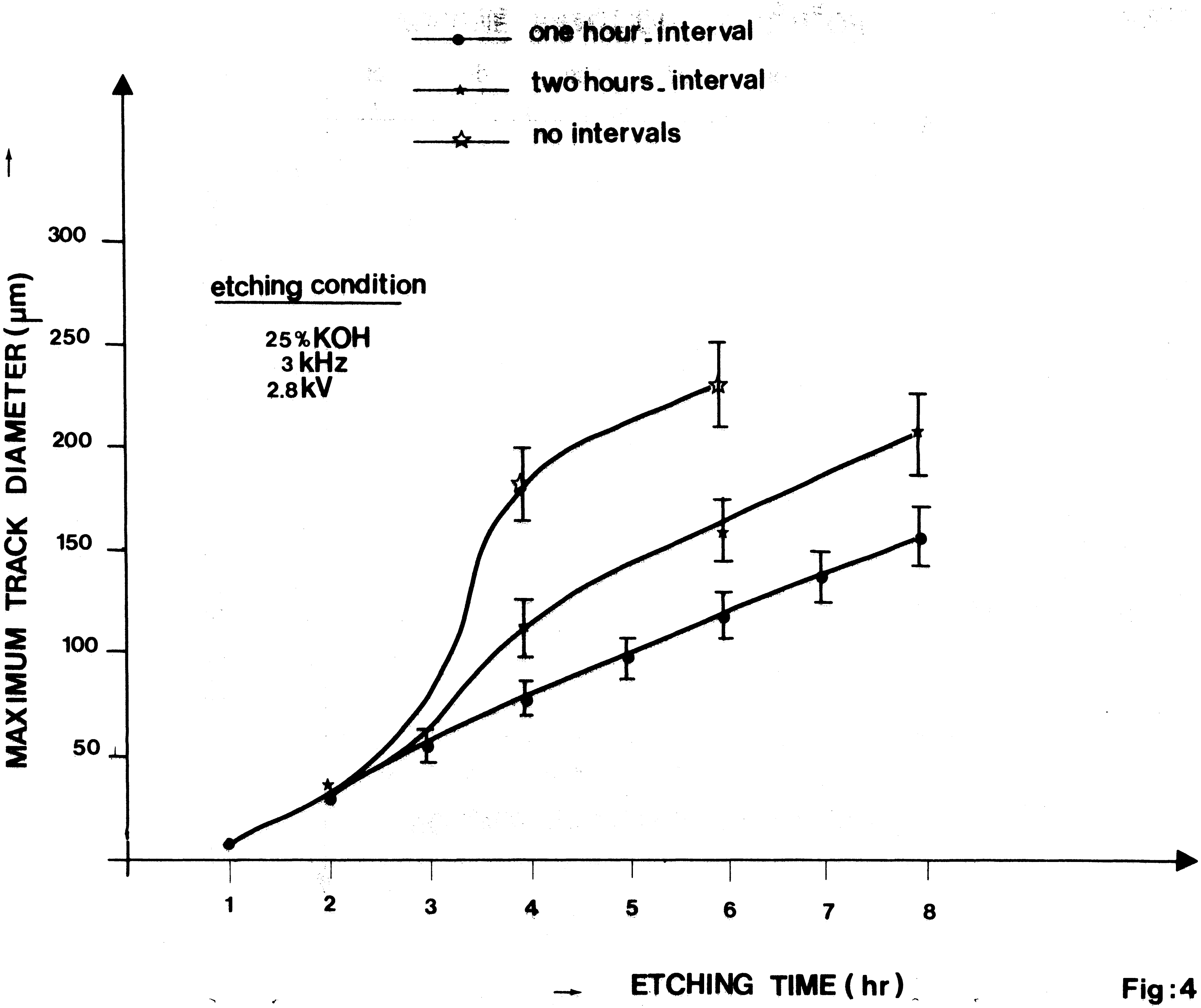

one hour_interval
two hours_interval
no intervals
etching condition
25%KOH
3 kHz
2.8kV
MAXIMUM TRACK DIAMETER (μm)
300
250
200
150
100
50
1
2
3
4
5
6
7
8
ETCHING TIME (hr)

Fig:4

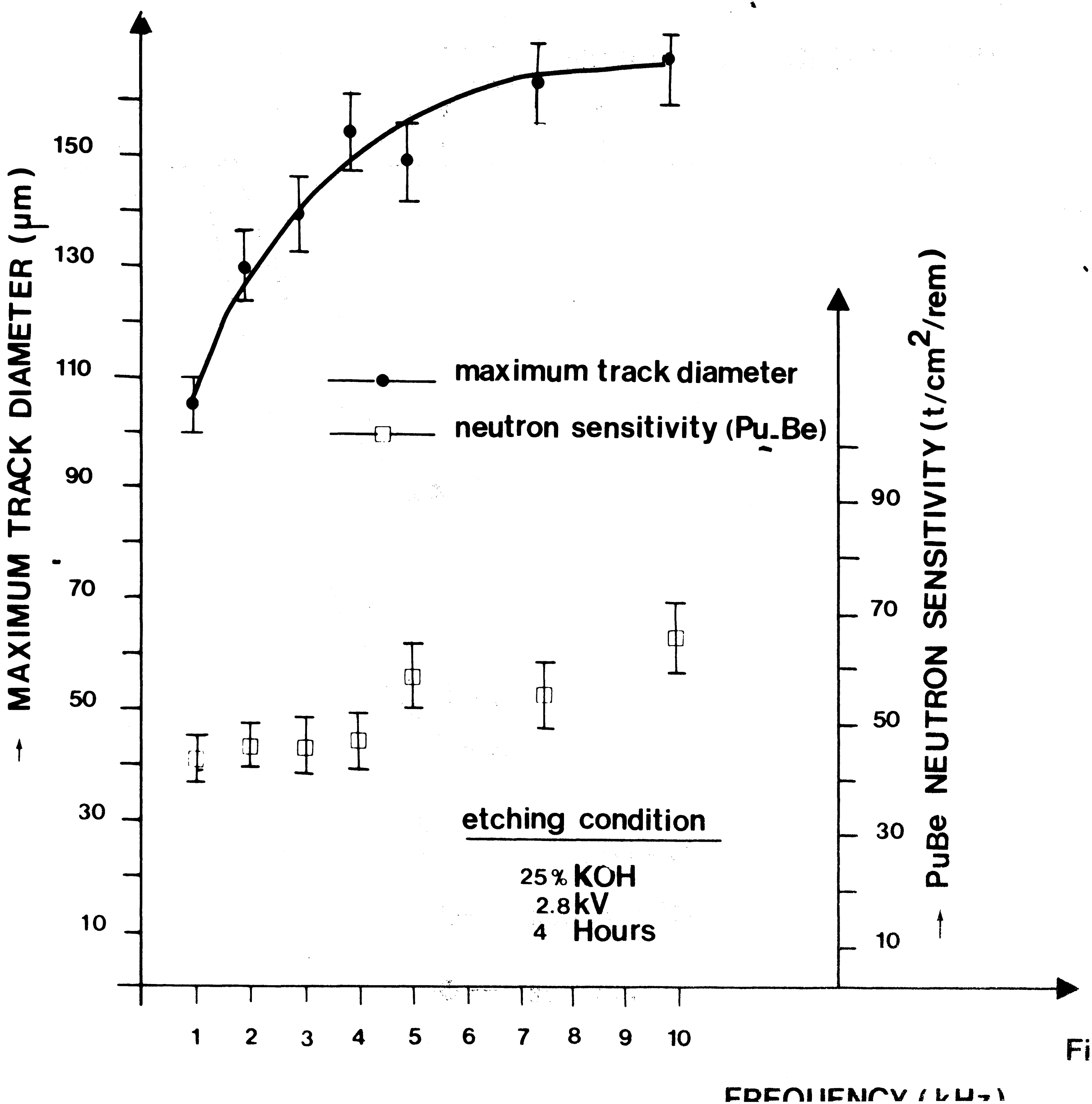
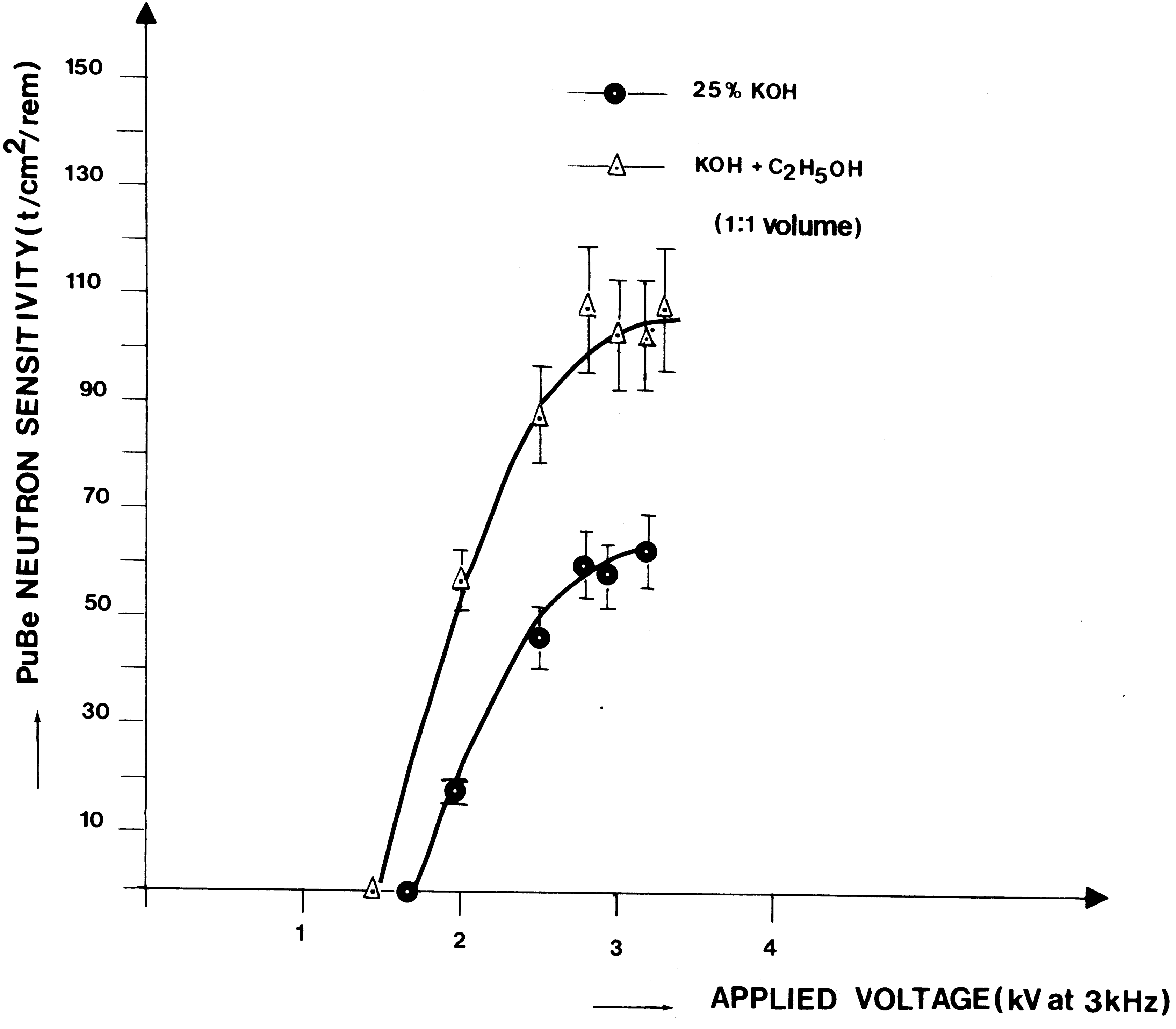

MAXIMUM TRACK DIAMETER (µm)
150
130
110
90
70
50
30
10
maximum track diameter
neutron sensitivity (Pu-Be)
PuBe NEUTRON SENSITIVITY (t/cm^2/rem)
90
70
50
30
10
etching condition
25% KOH
2.8 kV
4 Hours
1
2
3
4
5
6
7
8
9
10
FREQUENCY (kHz)

Fig:5



PuBe NEUTRON SENSITIVITY(t/cm²/rem)
150
130
110
90
70
50
30
10
1
2
3
4
APPLIED VOLTAGE(kV at 3kHz)
25% KOH
KOH + C₂H₅OH
(1:1 volume)

Fig:6

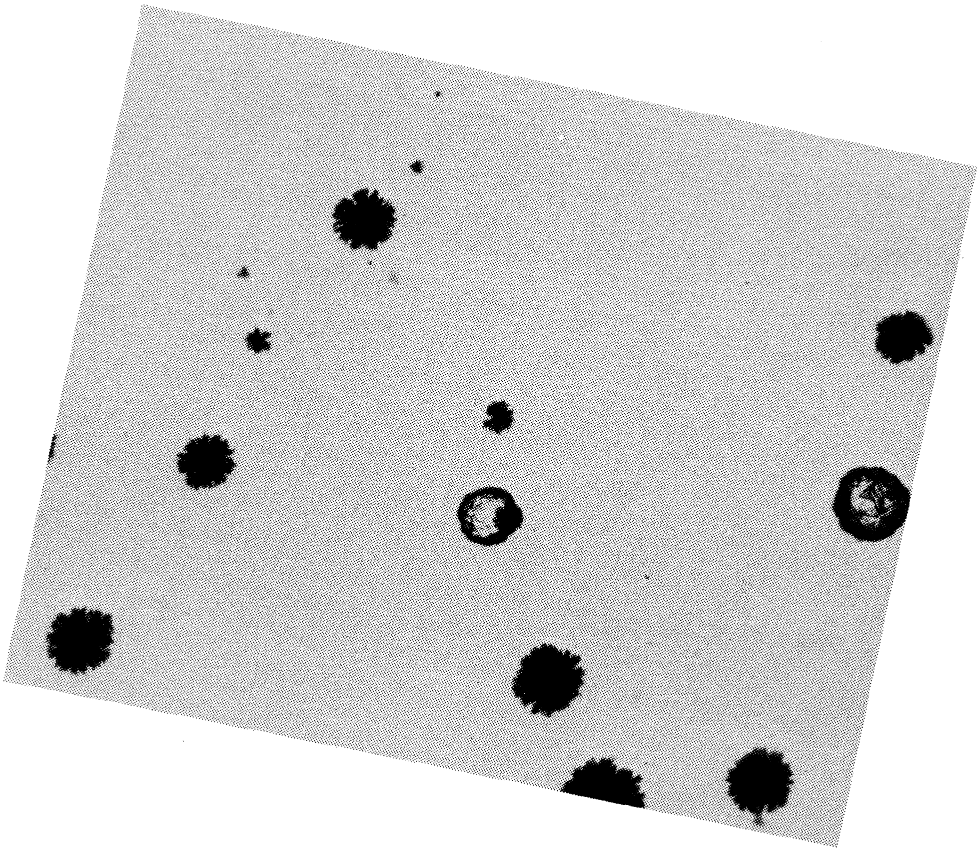


Fig: 7