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OFFICE OF RESEARCH ADMINISTRATION

RESEARCH PROJECT INITIATION

Date: October 18, 1974

Project Title: **Development and Application of Some Fast Neutron Dosimetry Techniques Utilizing Plastic Track Detectors for Therapeutic and Health Physics Related Applications**

Project No: **E-26-613**

Principal Investigator: **Dr. K. Z. Morgan/Dr. M. Sohrabi**

Sponsor: **Atomic Energy Commission, Oak Ridge Operations**

Agreement Period: From 9/1/74 Until 8/31/75

Type Agreement: **Contract No. AT-(40-1)-4814**

Amount: **\$31,371**

Reports Required: **Publication Preprints; Publication Reprints; Progress Report; Final Report**

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GEORGIA INSTITUTE OF TECHNOLOGY
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RESEARCH PROJECT TERMINATION

Cont. AT-(40-1)-4814

Date: December 16, 1975

Project Title: **Development and Application of Some Fast Neutron Dosimetry Techniques
Utilizing Plastic Track Detectors for Therapeutic and Health Physics**

Project No: **Related Applications
E-26-613**

Principal Investigator: **Dr. K. Z. Morgan/Dr. M. Sohrabi**

Sponsor: **Energy Research and Development Administration**

Effective Termination Date: 8/31/75

Clearance of Accounting Charges: 8/31/75

Grant/Contract Closeout Actions Remaining: **Certified Expenditures Report**

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Terminated Project File No. E-26-613

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PROGRESS REPORT

on

Contract No. AT-(40-1)-4814

with

Georgia Institute of Technology

for the

DEVELOPMENT AND APPLICATION OF SOME FAST NEUTRON DOSIMETRY
TECHNIQUES UTILIZING PLASTIC TRACK DETECTORS FOR THERAPEUTIC
AND HEALTH PHYSICS RELATED APPLICATIONS

Karl Z. Morgan and Mehdi Sohrabi

Co-Principal Investigators

May 19, 1975

Scope of Investigation

This research as outlined in the initial research proposal was to accomplish four principal objectives which may be summarized briefly as follows:

1. Optimization of electrochemical-etch-foil technique for application to neutron dosimetry
2. Development of a fast neutron dosimetry technique that is capable of registration of neutron dose by charged particle recoil tracks produced directly in foil materials
3. Application of this technique in the measurement of steep neutron dose gradients at bone-tissue and air-tissue interfaces
4. Comparison of these measurements with previously reported experimental and theoretical results.

Most of the experimental research on 1 and 2 above has been completed and some progress has been made on 3 and 4. We estimate that all four proposed investigations will be completed within the contract twelve month schedule. In the course of this research several additional, closely related new studies have been suggested and as a result a proposal is being submitted at this time to ERDA for the continuation of these studies and the renewal of this contract. We believe the additional proposed research is contiguous with the objectives of this contract and would greatly enhance the value of the present research effort. The following report is a brief summary of some of the progress we have made to date and describes the more significant results some of which have been reported in scientific publications as indicated by the attached reprints.

Significant Results from the First Six Months Operation of This Contract

The first step in this program was to develop an electrochemical etching chamber which would demonstrate that it is practical to process a number of foils simultaneously and that reproducible results can be obtained and easily applied in a routine personnel monitoring program or other similar applications. Several sizes and designs of etching chambers were constructed and tested. The quality and sensitivity of etching depended importantly upon the size and angle of the electrodes and these parameters were optimized. There was, however, little dependence on size of chamber, amount of electrolyte and type, and location of the electrodes. For our further studies, we used two Lucite cylindrical chambers (diameter, 11 cm, length, 11 cm), each holding approximately one liter of electrolyte. The foils to be etched were placed between the chambers each held by two rubber "O" rings to isolate them electrically. The chambers were then filled with the etchant and a high voltage at a high frequency (e.g. 2 kHz) was applied by means of two circular electrodes about 6 cm in diameter. The planes of the electrodes were kept parallel to that of the foil plane. Platinum, palladium, and stainless steel electrodes each gave satisfactory results but in several respects stainless steel is preferred.

The selection of a proper power supply is essential in obtaining satisfactory results with the electrochemical-etch-foil technique. The usual audio oscillator is not satisfactory because it does not have the capacity to maintain the required potential across the electrodes and in some cases cannot maintain constant frequency. We used two systems to obtain the desired voltages and frequencies. One was a stable audio frequency generator coupled to a push-pull power amplifier which we constructed in our laboratory. We got best results with this system so it was used in most of our experiments

reported here. Another power supply was also designed. In this case the audio oscillator was connected to a 70 watt Heathkit amplifier (push-pull) stepping up the output to the desired voltage with a transformer. With both systems, and especially the second method, we have sufficient capacity to process a large number of foils simultaneously and to adjust the voltage and current to any desired level of interest for proper etching. Also, with this arrangement we are able to choose the desired frequency within the audio range.

With the above equipment we determined the most efficacious potential and frequency to be applied across the electrodes. Figures 1, 2, and 3 are typical sets of data showing the variation with voltage across the electrodes of the sensitivity (number of tracks/cm².rad), the mean track diameter and the optical density, respectively, when the other parameters (as shown on the graphs) were maintained constant. As can be seen these three variables (each of which relates to sensitivity) tend to reach a semi-plateau above about 600 volts where the voltage of the system is much less critical and does not require unusual circuitry for regulation. In our studies we found that we obtained completely satisfactory and reproducible results when operating at any fixed potential between 600 and 1200 volts. At lower voltages, satisfactory results probably could be obtained, however, if etching time were increased appropriately.

Proper operation of the electrochemical-etch-foil technique depends very critically upon the frequency of the voltage applied to the etching chambers and across the foils that separate the chambers. Figures 4, 5, and 6 indicate a resonant frequency exists at which we obtain maximum etching efficiency. Under these conditions the resonant frequency is at about 2 KHz where we obtained a maximum number of recoil tracks, maximum track diameter

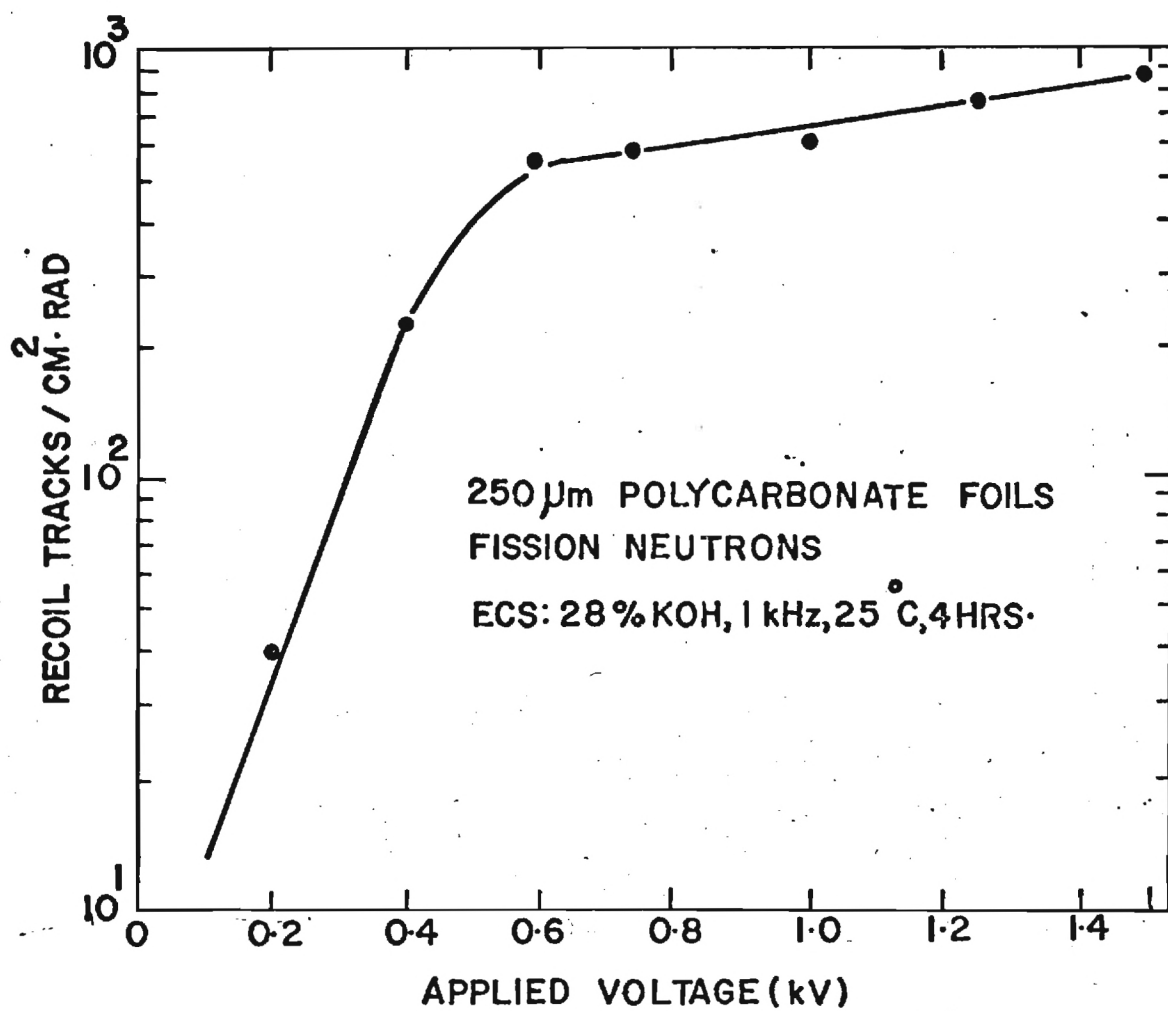


Figure 1. Track Density per Rad of Fission Neutrons as a Function of Applied Voltage at Indicated Etching Conditions

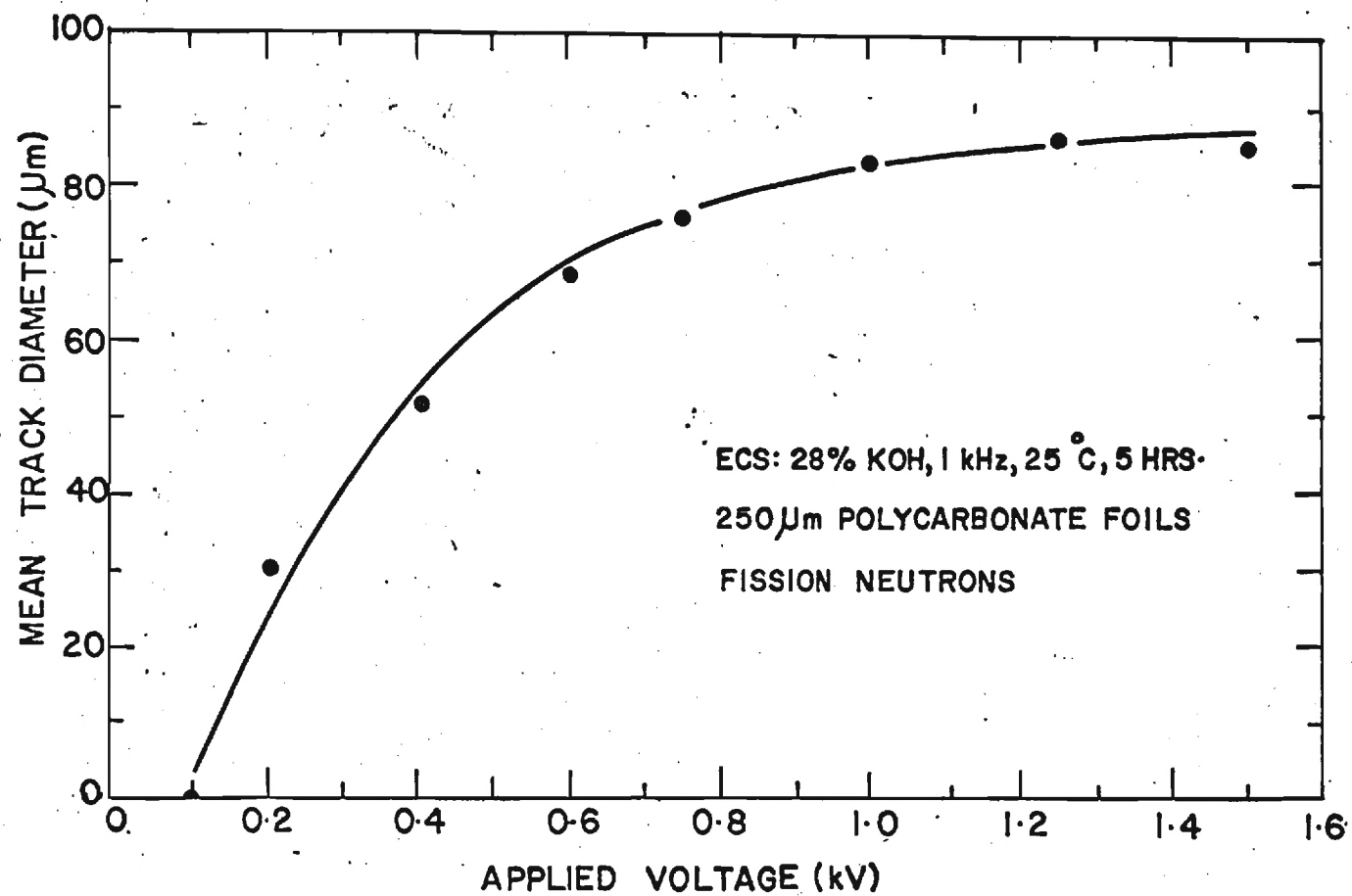


Figure 2. Mean Track Diameter as a Function of Applied Voltage at Indicated Etching Conditions

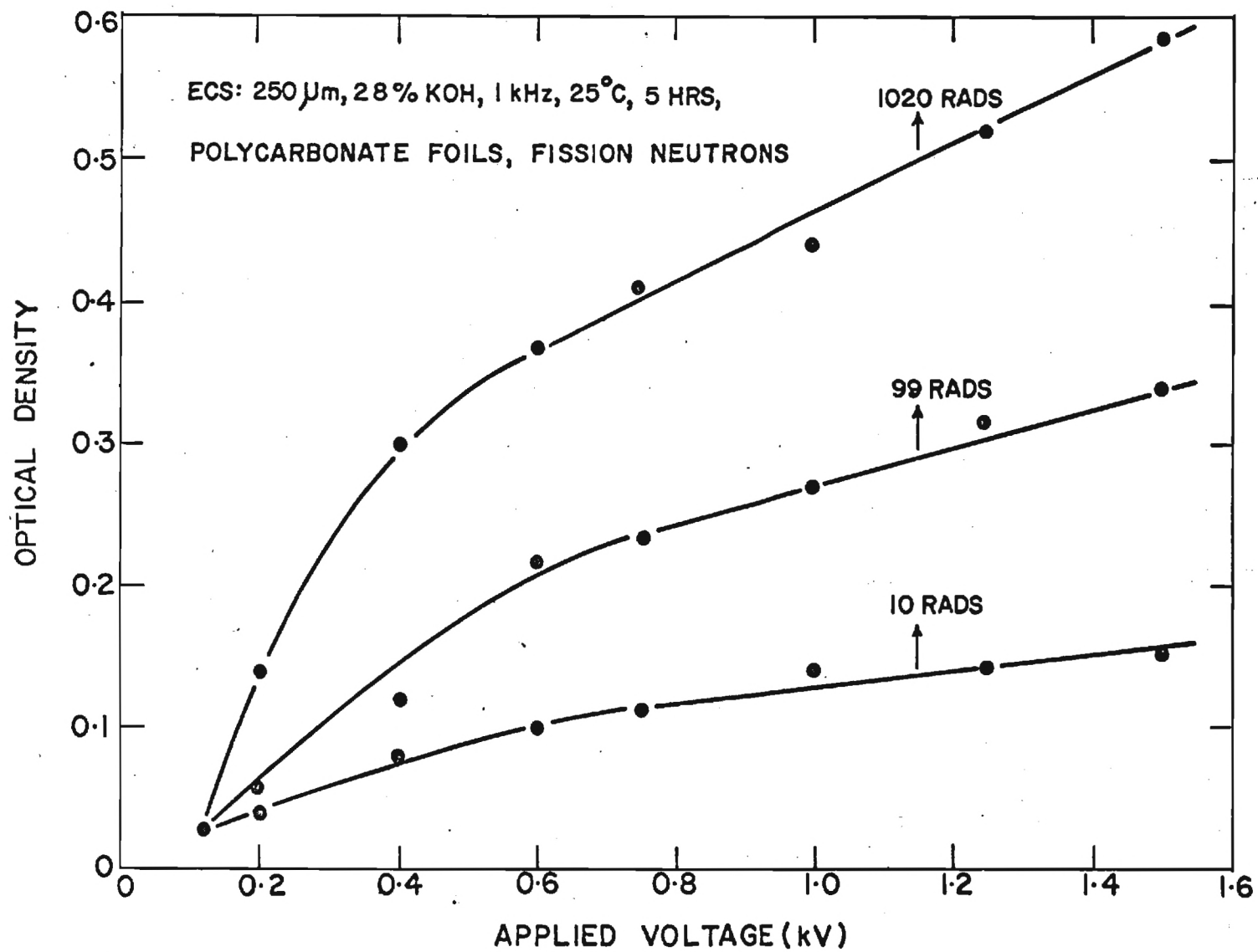


Figure 3. Optical Density as a Function of Applied Voltage
at Indicated Etching Conditions

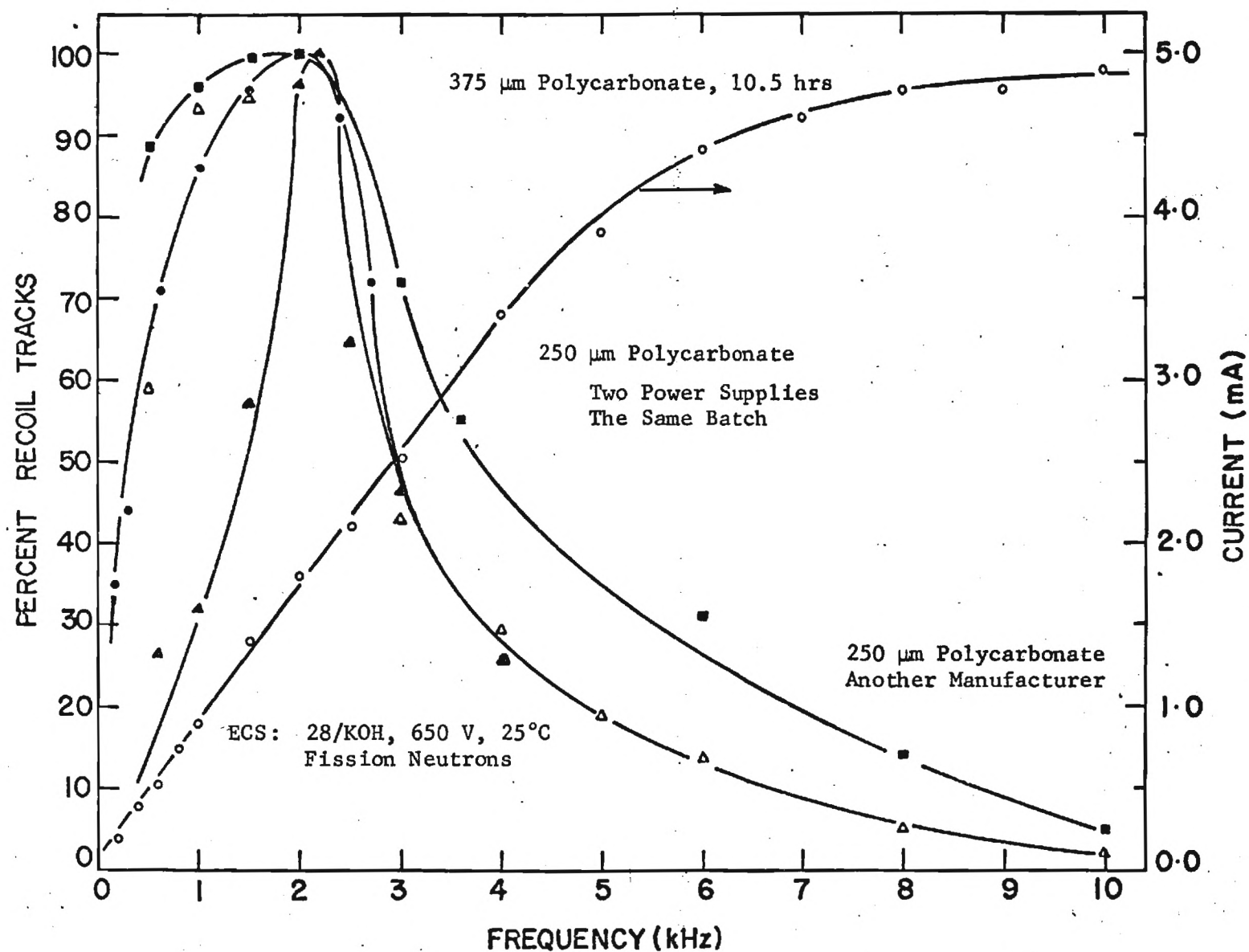


Figure 4. Percent Recoil Tracks and Current as a Function of Frequency of the Applied Voltage in Polycarbonate Foils of Different Thicknesses, Different Manufacturers Using Two Different Power Supplies

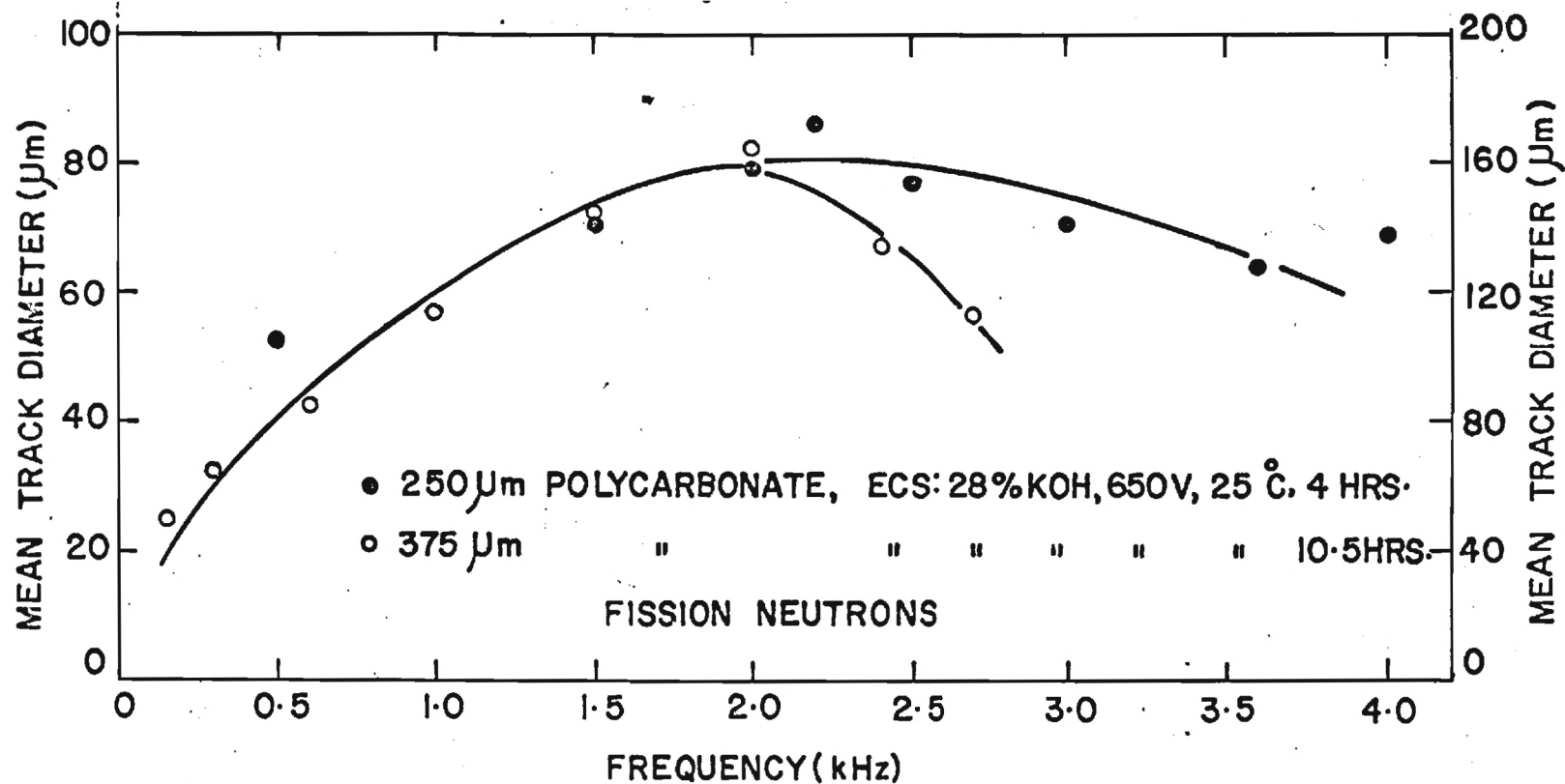


Figure 5. Mean Track Diameter as a Function of Frequency of the Applied Voltage at Indicated Etching Conditions

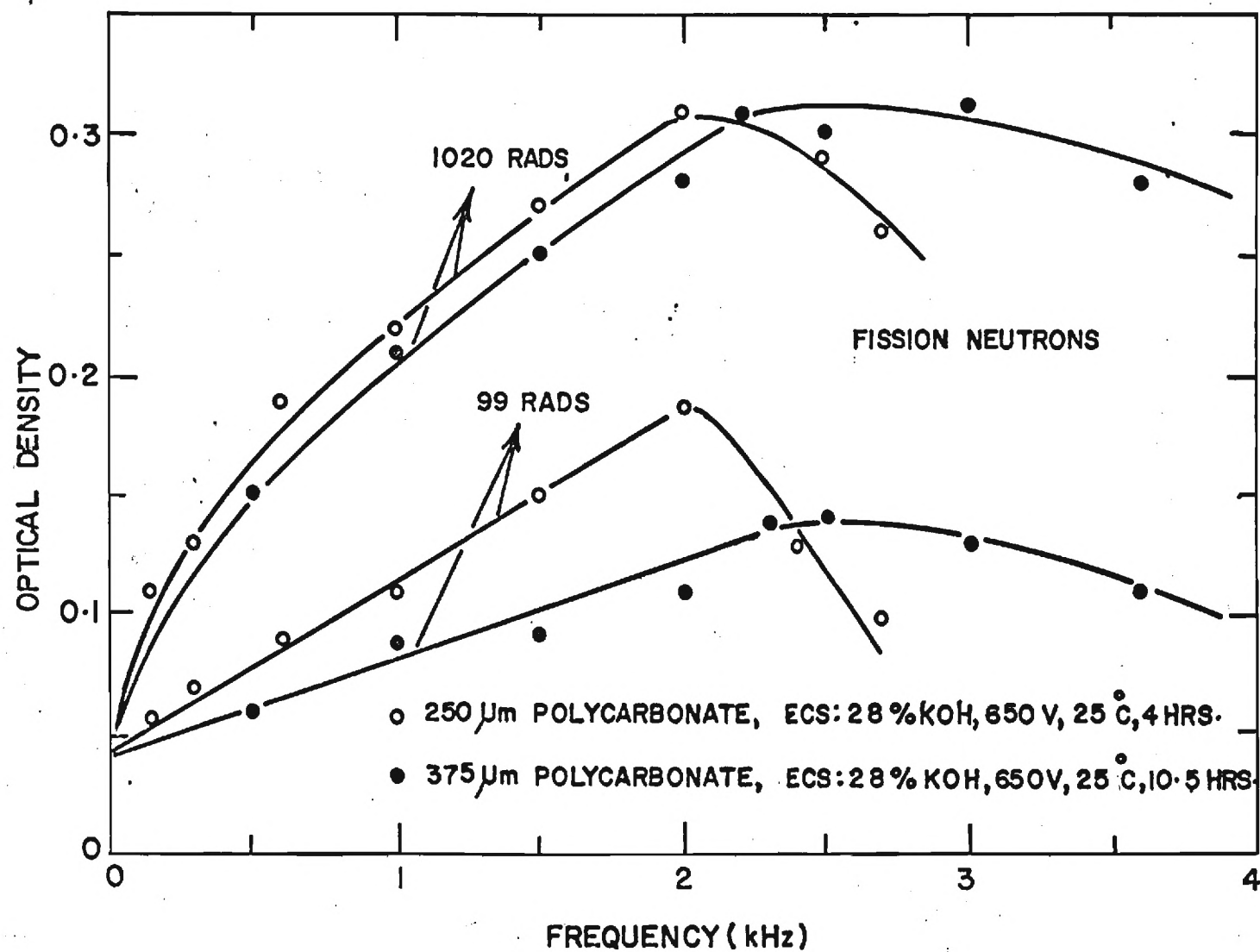


Figure 6. Optical Density as a Function of Frequency of Applied Voltage in Polycarbonate Foils of Different Thicknesses for Different Fission Neutron Doses

and maximum optical density as shown in Figs. 4, 5, and 6, respectively. Our first thought was that these resonance peaks occur at the electrical resonance frequency given by $f = 1/(2\pi\sqrt{LC})$ in which f is frequency in Hz, L is inductance in Henries, and C is capacity in farads. This would have provided a very simple explanation because at resonance the current I_o is a maximum and is given by

$$\bar{I}_o = \frac{\bar{E}_o}{\sqrt{R^2 + \left(2\pi fL - \frac{1}{2\pi fC}\right)^2}}$$

in which the potential $\bar{E}_o$ is in volts and current $\bar{I}_o$ is in amperes. However, upon checking the values of L and C and measuring the current $\bar{I}_o$ as plotted also in Fig. 4, it became evident that the resonant peak for maximum etch efficiency (~ 2 KHz) occurs far to the left of the very broad electrical resonance peak and the current $\bar{I}_o$ at maximum etching efficiency is only about 1/3 of the current at electrical resonance. Thus we had to seek some other explanation for this resonance in the etch efficiency curves. This explanation follows.

With no potential applied across the chambers the ions in solution have a random walk. With an applied d.c. potential the ions move in one direction, i.e. toward the electrode of opposite charge. With an alternating potential the ions move back and forth changing direction twice each cycle and this oscillating motion is superimposed upon the random walk motion. At low frequency (below resonance) the average distance or amplitude of motion of the ion each half cycle is greater than the unetched track length, for example, at 1 Hz it is about 300 μm or many times the length of the tracks in the foil before etching. At high frequency (above resonance); for example, at 10,000 Hz, the ions move only about 0.03 μm per half cycle or a small fraction of the

range of the tracks in the foil. At this frequency the ion drift per cycle is about 10^{-4} that at 1 Hz. Therefore, at high frequencies the ions tend to remain relatively stationary. At resonance for maximum etching efficiency, however, or at 2000 Hz the ions move about $0.2 \mu\text{m}$ each half cycle. Since the range of the unetched tracks is of the order of $1 \mu\text{m}$, one would expect the oscillating ion to deliver maximum energy to the foil track cavity at this frequency. Hence, the resonant curves in Figs. 4, 5, and 6. In future studies we propose to study the effect of bringing the electrical resonance peak into resonance with the maximum etching efficiency peak to further improve this technique.

The other variables which must be optimized to obtain best results in the electrochemical-etch-foil technique are the type of foil and its thickness. A number of types of foils from various factories and vendors were investigated. Some of these polymers were cellulose acetate, cellulose nitrate, cellulose acetate butyrate, cellulose triacetate, and Lexan polycarbonate. It would take many pages to review these findings, but suffice it to say in this brief report that the Lexan polycarbonate seems to give the best results in terms of reproducibility, size, and number of tracks per rem exposure to neutrons. Therefore, Lexan polycarbonate foils were used in most of our studies. Polycarbonate foils of a number of thicknesses were studied as indicated already in Figs. 4, 5, and 6. As demonstrated in Figure 7 the etching time should be reduced as the foil thickness is decreased when the other parameters are fixed for otherwise the foils will break through. When these co-adjustments of etching time and foil thickness are made, it is seen in Fig. 7 that the sensitivity ($\text{tracks}/\text{cm}^2 \cdot \text{neutron}$) decreases and the track diameter increases with increasing foil thickness and etching time. Figure 8 indicates how sensitivity ($\text{tracks}/\text{cm}^2 \cdot \text{rad}$) increases with etching time for four foil thicknesses. Figure 9 indicates that

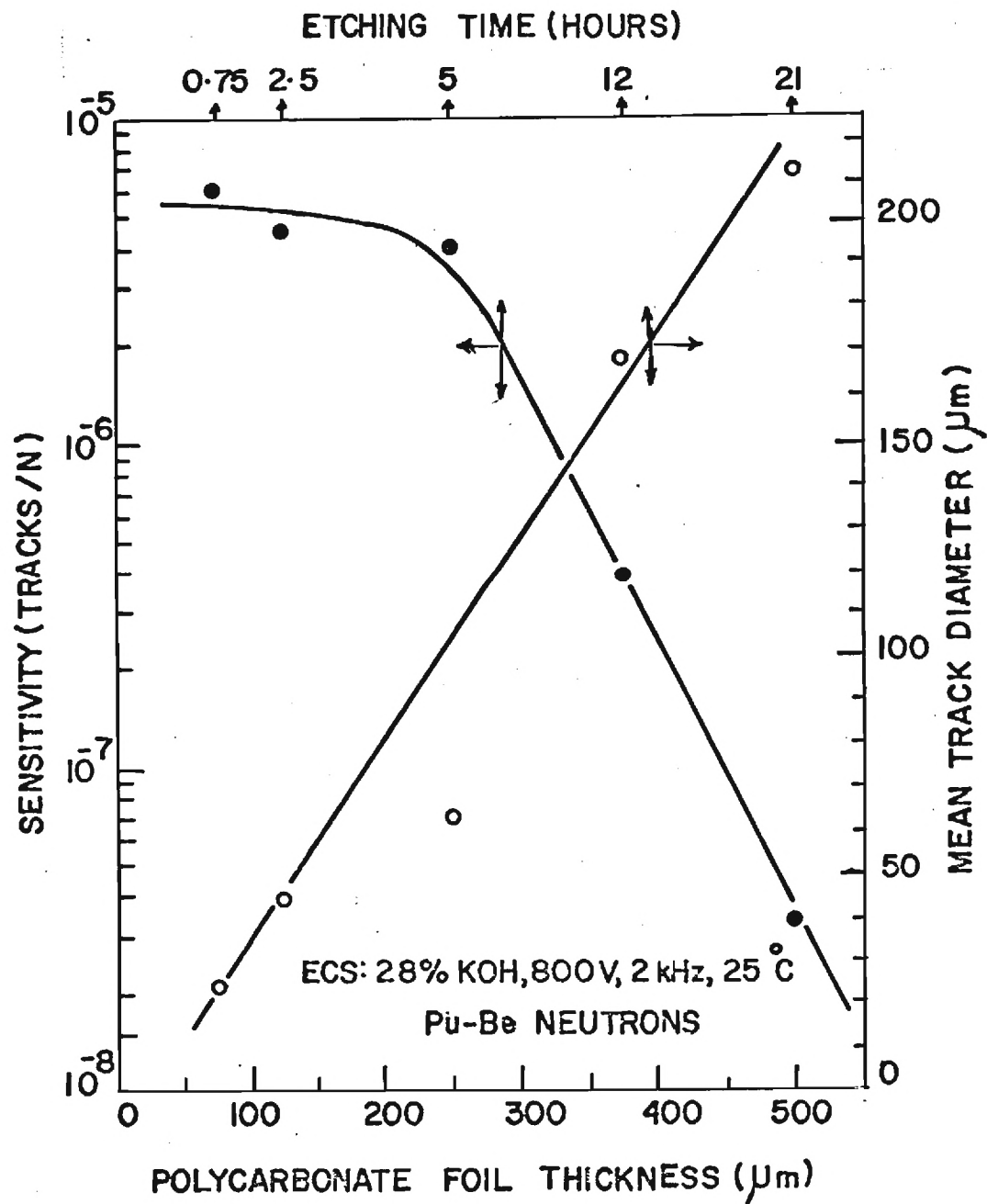


Figure 7. Sensitivity and Mean Track Diameter as a Function of Polycarbonate Foil Thickness at Optimum Etching Times

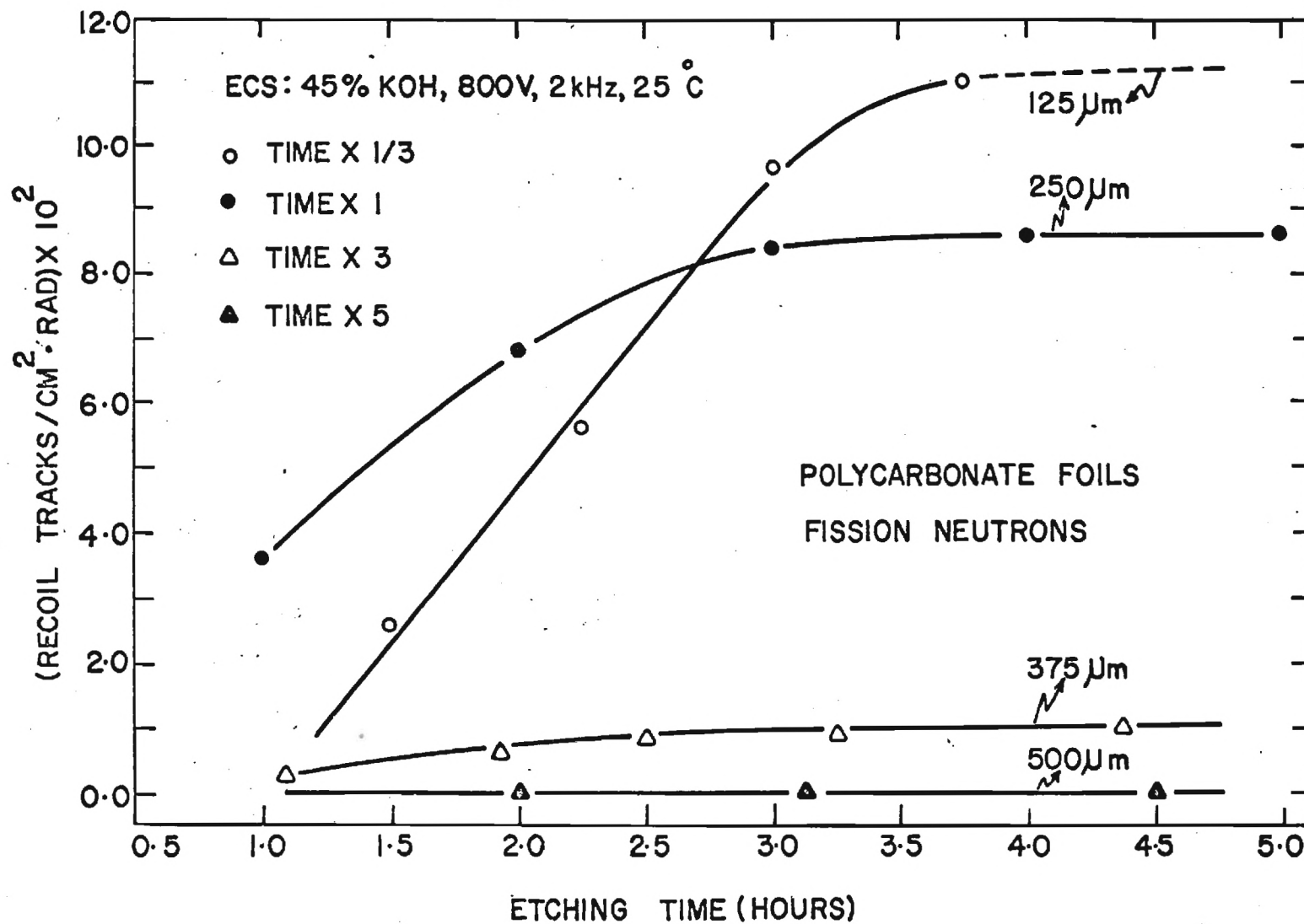


Figure 8. Track Density per Rad as a Function of Etching Time in Polycarbonate of Different Thicknesses

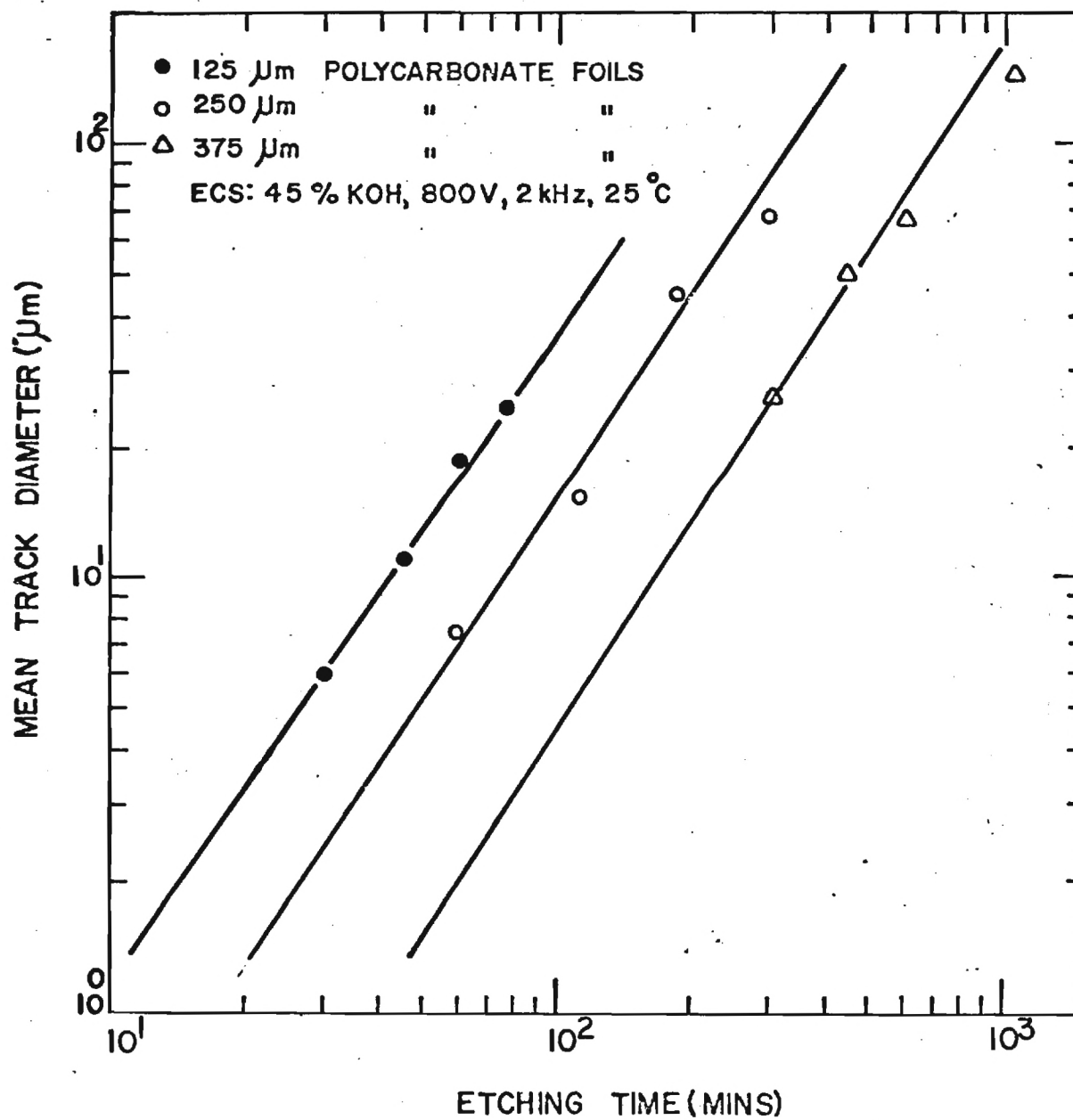


Figure 9. Mean Recoil Track Diameter as a Function of Etching Time in Polycarbonate of Three Different Thicknesses

the mean track diameter is given approximately by the equation

$$D \approx Ct^{1.5}$$

in which D = track diameter (μm), t = time (min), and C has values of 0.033, 0.014, and 0.0046 for foils of thickness 125 μm , 250 μm , and 375 μm , respectively.

Alkali hydroxide solutions seem to be the best etchants; however, the effectiveness of the different alkali ions varies due to differences in ion radii and mobility in the etchant. It was expected from these studies and those of others that the etching rate would increase with molecular weight, i.e. increase in going from LiOH to NaOH to KOH to RbOH and to CsOH. The radii of the alkali ions increase in the above order while the hydration numbers decrease with this order. Thus Li^+ with the highest hydration number has the least mobility of the above ions and appears to be the least efficient of the above etchants. Figure 10 indicates for example that we were able to obtain much larger tracks with KOH than with NaOH solutions in a shorter etching time. However, as shown in Figure 11, if the etching time is increased above about 7 hours, the sensitivities of the two systems (tracks/ $\text{cm}^2 \cdot \text{rad}$) become about the same even though the tracks using NaOH are much smaller and more difficult to count.

Also, the concentration of the alkali hydroxide solution is very important in electrochemical etching. Figure 12 indicates how the sensitivity increases with concentration of KOH when the etching time, voltage, and frequency are maintained constant. The relationship for the least squares fit in this case is given by the equation

$$S = 25.08C - 207.2$$

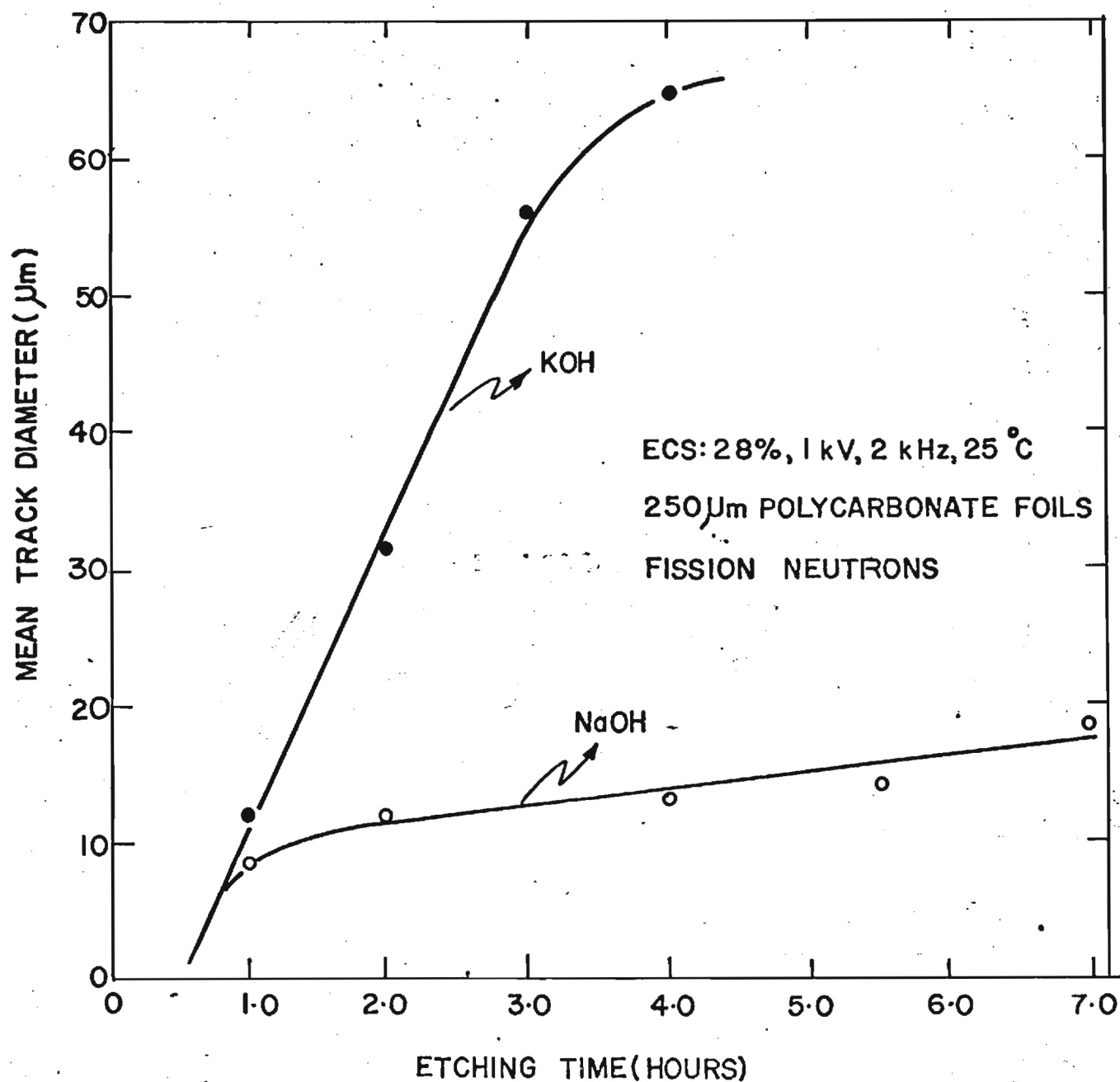


Figure 10. Mean Recoil Track Diameter as a Function of Etching Time in 250 μm Polycarbonate Using NaOH and KOH Solutions of 28% by Weight Concentrations

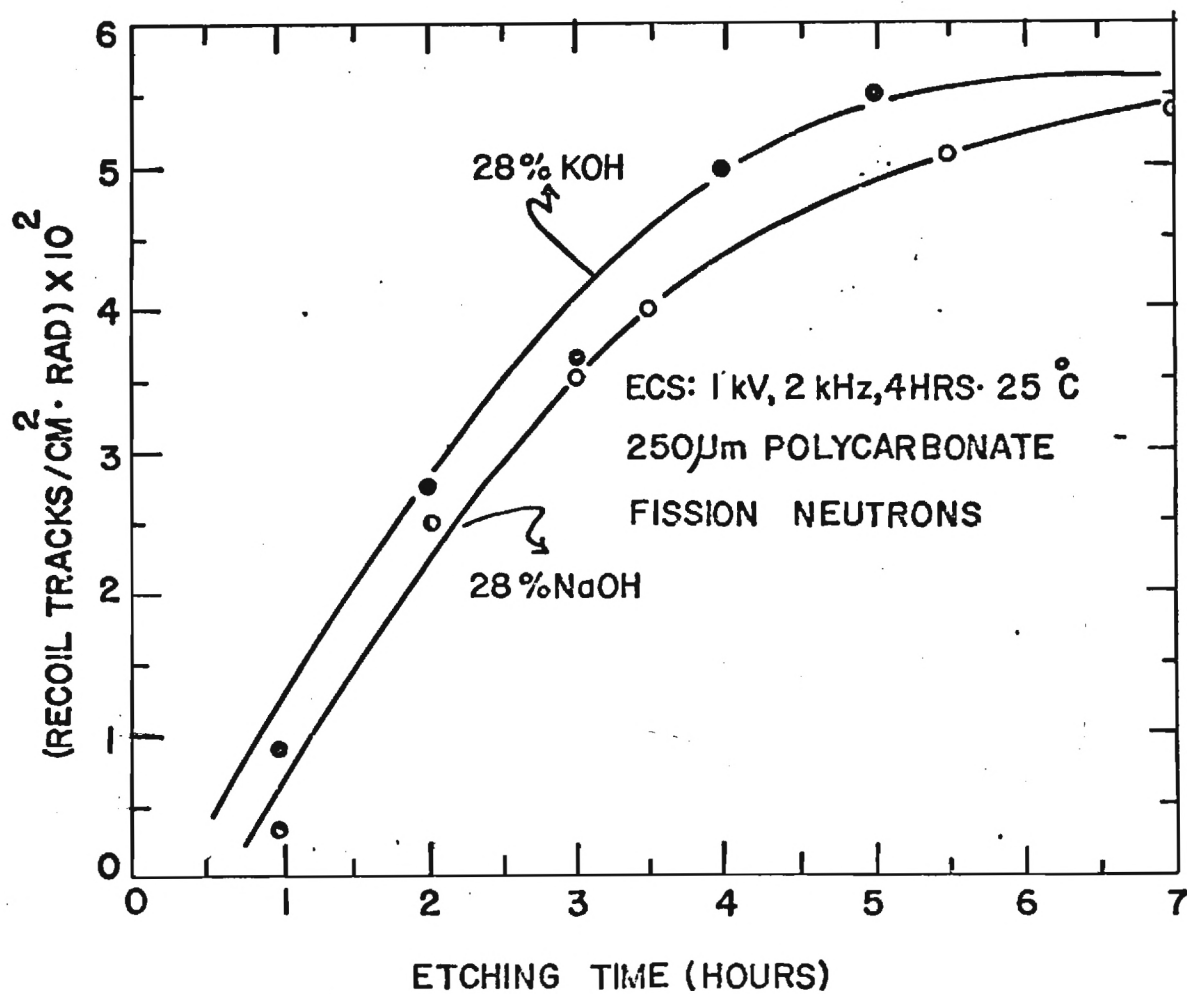


Figure 11. Fission-Neutron-Induced Recoil Track Density per Rad as a Function of Etching Time in 250 μm Polycarbonate Foils Using NaOH and KOH Solutions of 28% by Weight Concentration

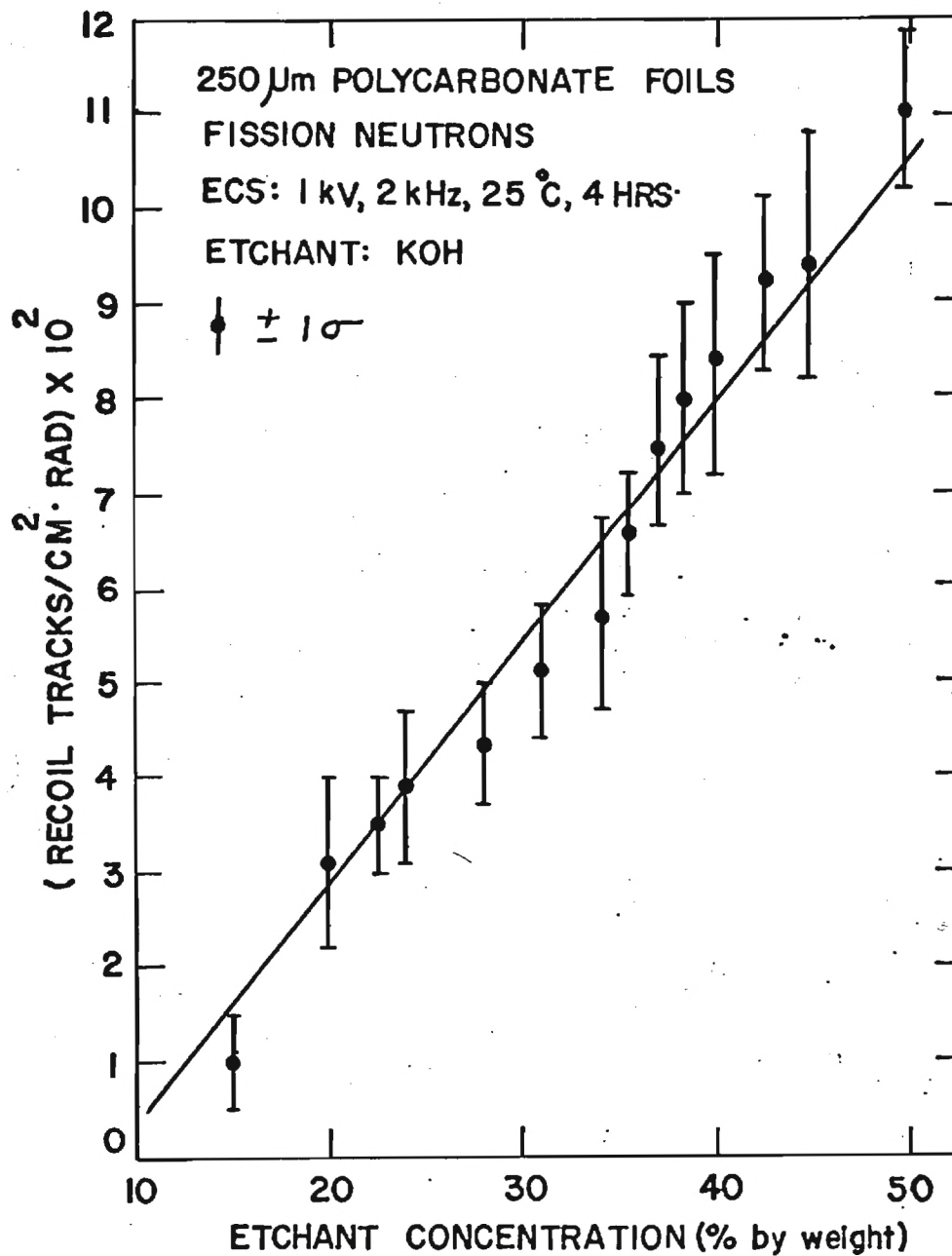


Figure 12. Fission-Neutron-Induced Recoil Tracks per Rad in 250 μ m Thick Polycarbonate as a Function of Potassium Hydroxide Concentrations

in which S = sensitivity (recoil tracks/cm²·rad) and C = etchant concentration (% by weight).

Figure 13 shows track density as a function of etching time before (lower curve) and after (upper curve) optimization of etching conditions. The sensitivity has increased by a factor of two while the etching time has decreased by a factor of two.

Both the sensitivity (tracks/cm²·rad) and mean track diameter increase linearly with the etchant temperature as indicated in Figure 14. In this case the best fit equations are

$$S = 9.5T - 228$$

$$D = 1.47T - 35$$

in which S is sensitivity (tracks/cm²·rad), T is temperature (°C), and D is track diameter (μm). Figure 15 indicates the importance of operating at a higher temperature (e.g. 60°C) when the etching time and other parameters are fixed. However, by increasing the etching time, sufficient sensitivity can be obtained at lower temperatures (e.g. 25°C) even for processing films that have received low neutron exposures.

In the course of these studies a number of neutron sources have been used to irradiate the foils in order to measure dependence on energy of the neutrons. Table 1 summarizes the sources used in these experiments.

Now that the optimum conditions for using the electrochemical-etch-foil technique for neutron monitoring have been fairly well determined the studies in the application of this dosimetry technique to the measurement of steep neutron dose gradients at interfaces in bone and tissue surfaces are getting underway.

In the course of these studies several papers have been presented at scientific meetings describing the findings of this research and some of the

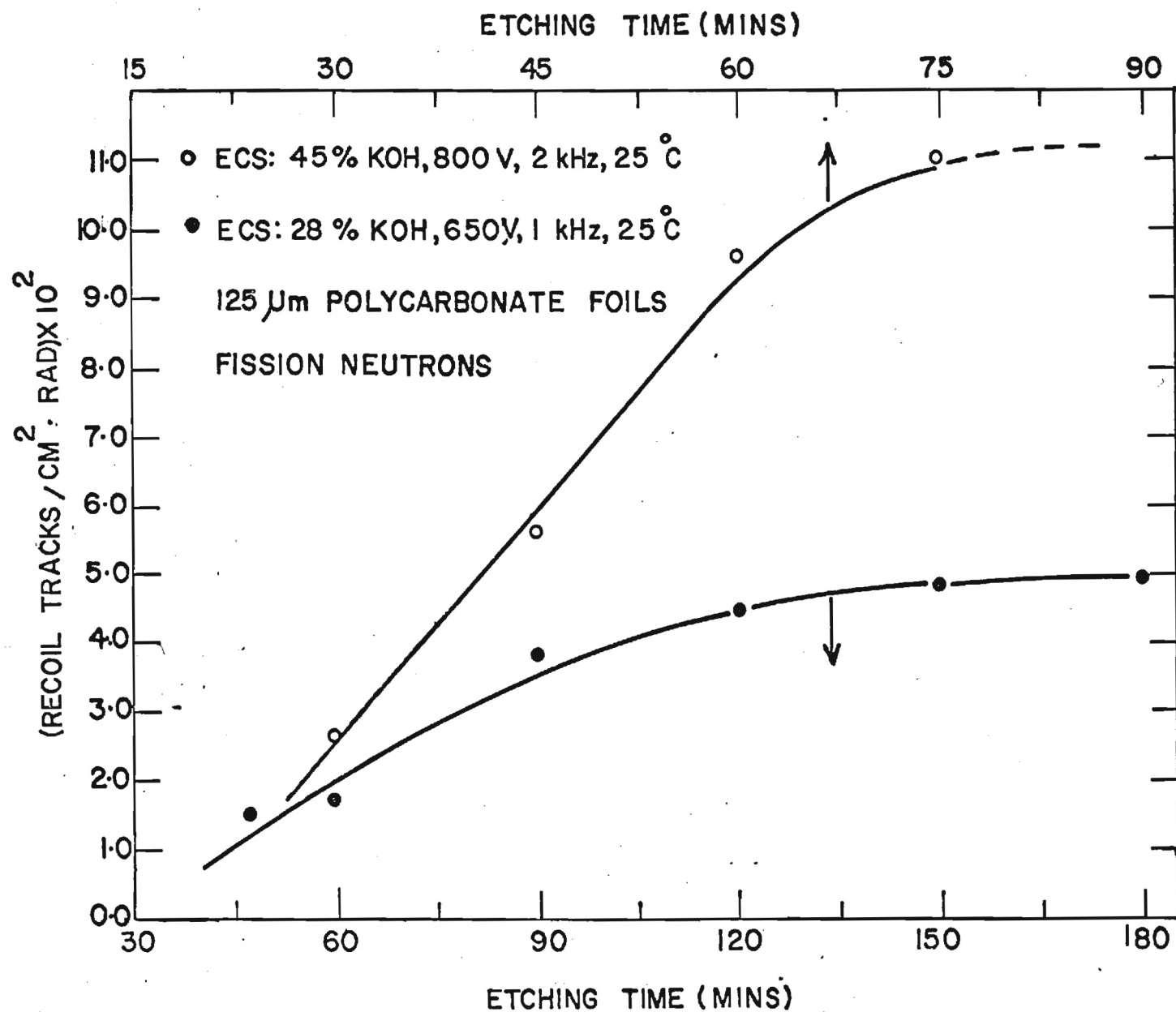


Figure 13. Fission-Neutron-Induced Recoil Track Density in 125 μm Thick Polycarbonate as a Function of Etching Time at Two Different Etching Conditions as Indicated

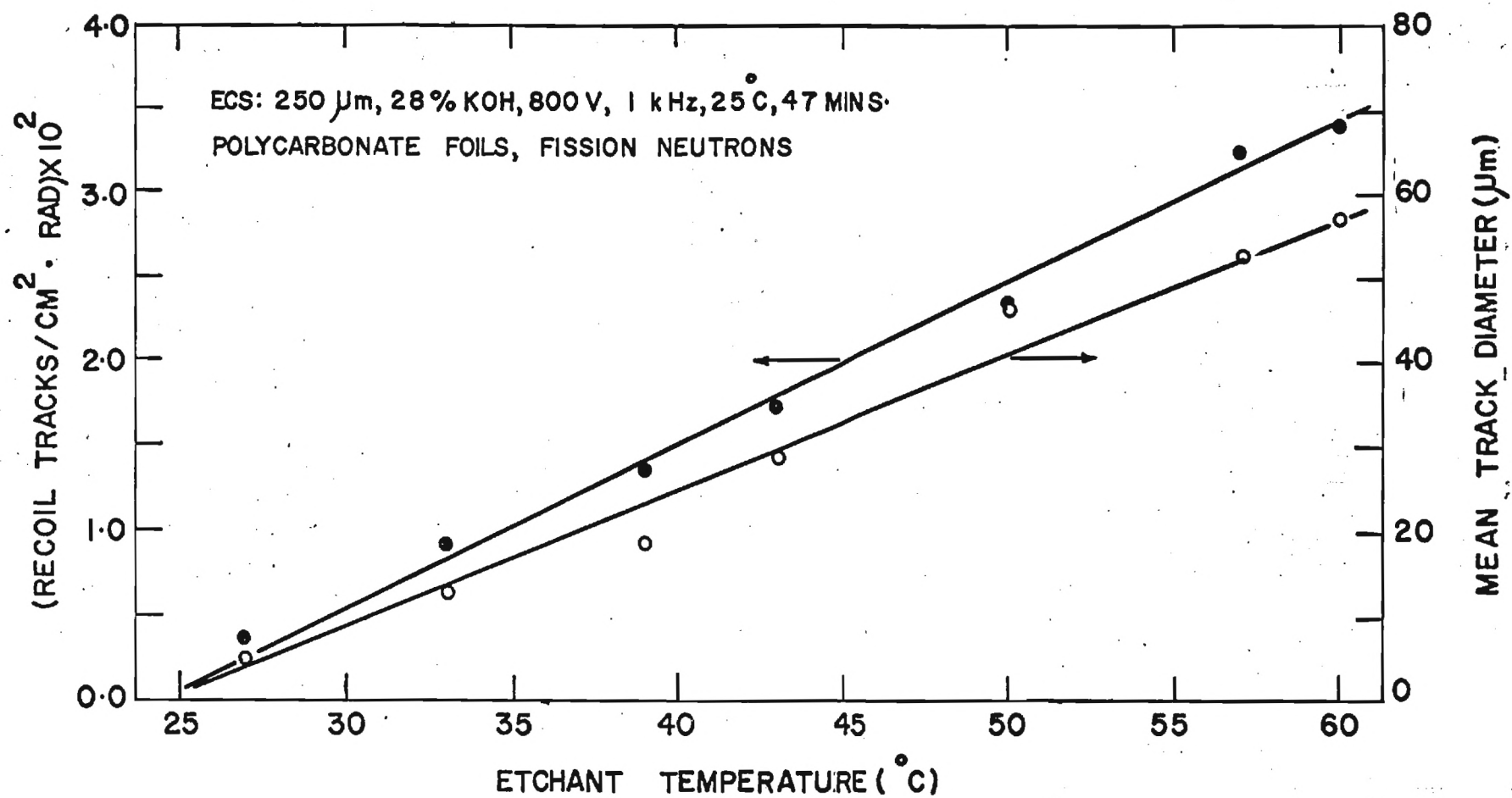


Figure 14. Fission-Neutron-Induced Recoil Track Density and Mean Track Diameter in 250 μm Polycarbonate as a Function of Etchant Temperature

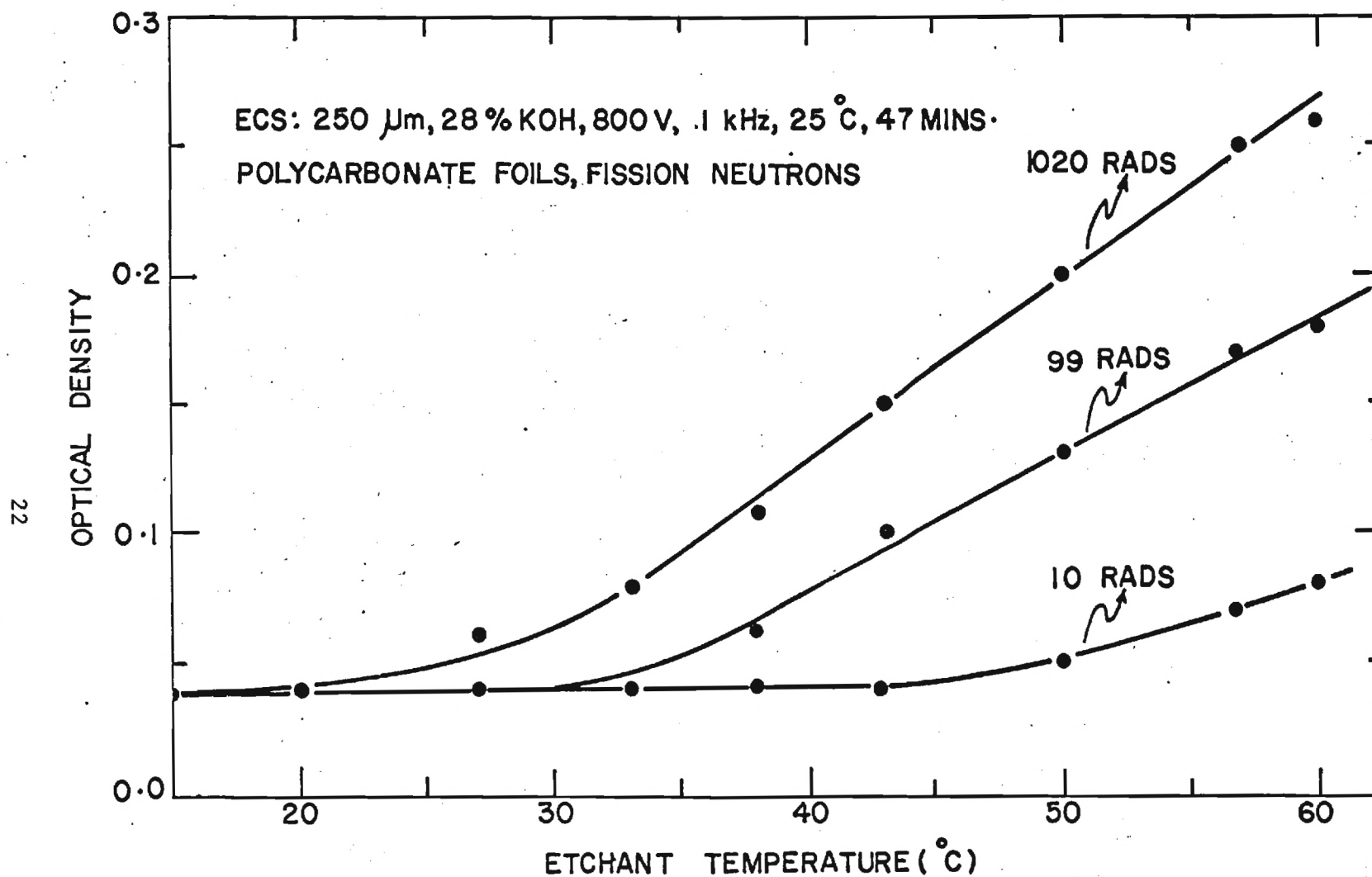


Figure 15. Optical Density as a Function of Etchant Temperature for Different Fission Neutron Doses

Table 1. Summary of Neutron Facilities Used During the Course of Our Research

Type of Facility	Kind of Source	Neutron Energy (at maximum MeV)	No. of Runs for Each Facility
1. HPRR	Fission neutrons	~ 1	~ 20
2. Isotopic source	Pu-Be	~ 4	5
3. Texas A&M	16 MeV d^+	~ 7	12
Cyclotron	50 MeV d^+ on Be target	~ 20	
4. Univ. of Wash- ington Cyclotron	16 MeV d^+ on Be target	~ 7	6
5. NRL Cyclotron	35 MeV d^+ on Be target	~ 15	6
6. Emory University Betatron	Photoneutrons pro- duced by 25 MeV x-rays	~ 2	4
7. Georgia Tech Research Reactor (Biomedical Facility)	Thermal neutrons	thermal	4

results have been published in the scientific literature; other papers are in preparation for publication in the near future. Altogether we have some 50 sets of data and graphs and so this report is only a sampling and summary of our most significant results. Our public presentations and publications include:

1. "Electrochemical Amplification of Recoil Particle Tracks in Polymers and Its Application in Fast Neutron Personnel Dosimetry," Health Physics, 27, 598 (1974).
2. "Recent Developments in Fast Neutron Personnel Dosimetry Using Track Etch Methods," Proc. 3rd European Cong. IRPA, Paper 14, Amsterdam, The Netherlands (1975). See also Transactions of the American Nuclear Society Student Conference, p. 24 (1975). NOTE: This paper was awarded the citation of being the best paper of the Health Physics Session at the American Nuclear Society Student Conference.
3. "Some Observations on the Optimization of Electrochemical Etching Parameters for the Amplification of Fast-Neutron-Induced Recoil Particle Tracks in Polymers," in preparation for publication.
4. "Fast Neutron Personnel Dosimetry Using Electrochemically Amplified Recoil Particle Tracks in Lexan Polycarbonate," in preparation for publication.
5. Ph.D. Dissertation in preparation.

Summary

The electrochemical-etch-foil technique continues to look extremely promising for neutron monitoring of personnel and applications in medicine and research. We have determined what we believe are some of the most important parameters that must be controlled and at present are in the process of determining the best combinations of these parameters to provide a simple and reliable means of fast neutron monitoring.

Attachments

Included with this progress report are reprints and copies of papers that have been presented publicly. These have attracted wide scientific interest and have resulted in many requests for reprints and letters asking for details of our findings so that similar studies can be conducted elsewhere and so these persons can apply these techniques in their neutron dosimetry applications.