CONF-790728--5

MASTER

THERMAL NEUTRON DOSIMETRY USING ELECTROCHEMICAL ETCHING

Shian-Jang Su, Micheal E. Sanders, and Karl Z. Morgan
Georgia Institute of Technology
School of Nuclear Engineering
Atlanta, Georgia 30332

Paper Presented To:

24th Annual Meeting of the Health Physics Society
Philadelphia, Pennsylvania
July 10, 1979

DE-AS05-76 EV04814

Introduction

The availability of intense neutron fluxes from high-energy accelerators, nuclear reactors, and isotropic neutron sources in recent years has provided a major tool for both physical and biomedical applications. Subsequently, these technological strides have led to an ever increasing need for improved neutron dosimetry in problems associated with area monitoring, personnel monitoring, accident dosimetry, and a variety of other health physics and biomedicine related situations.

Significant contributions in the measurement of the fast neutron dose have been advanced independantly by Sohrabi[1], Su[2], and others. Their methods utilize recoil particle track registration in polycarbonate foils exposed to fission spectrum neutrons and then amplified by using improvements in Tommasino's[3] elegant electrochemical etching technique. These researchers have shown that the polycarbonate/electrochemical etching technique for the measurement of fast neutron dose is highly reliable, simple, rugged, inexpensive, insensitive to gamma reactions, non-fading, and applicable over a wide exposure range.

One of the only disadvantages of using polycarbonate as a neutron dosimeter is its insensitivity to the thermal neutron dose--the threshold energy for recoil particle track registration being approximately 1-MeV in polycarbonate. This study shows that by utilizing the $^{6}Li(n_t,alpha)^{3}H$ reaction in specially prepared LiF radiators an accurate indication of the thermal neutron dose can be achieved by measuring the high LET alpha and triton induced track densities registered in electrochemically etched low background Lexan polycarbonate foils.

Methods and Materials

A LiF radiator in tablet form was prefered over a powder layer because tablets are more reproducible and easier to handle during experiments. The radiator thickness was desired to be greater than the range in LiF of the emitted tritons and alpha particles in order to maximize the number of particles that could reach the polycarbonate foil and produce tracks. The alpha-triton pairs liberated from the $^{6}Li(n,alpha)^{3}H$ reaction move in approximately opposite directions and the 4.78 MeV of energy produced is divided as kinetic energy between the triton and alpha in approximately the inverse ratio of their masses. The range of the 2.7 MeV tritons in LiF was calculated to be 12 microns and the range of the 2.0 MeV alphas to be 6.6 microns. The radiator tablets were prepared from three different enrichments of commercial TLD powder obtained from Harshaw Chemical Company (specifically TLD-100, TLD-600, and TLD-700). We desired the radiator tablets to be made from only LiF, however because those made without a binder or lubricant were extremely fragile, 25% by weight of KBr was used as a binder and added to the LiF. Each of the ingredients was finely ground and uniformly mixed by mortar and pestle before pressing into tablets approximately 1.2 cm in diameter.

A thin tablet was desired in order to minimize the self-shielding effect, but since a 0.3 mm tablet was too fragile to handle, tablets consisting of two layers were made. The first layer, which would be in contact with the polycarbonate foil during irradiation, was made 0.3 mm thick. The ^{6}Li abundance of this layer was varied by using different mixtures of TLD-600 (95.62% ^{6}Li) and TLD-700 (99.93% ^{7}Li). The second layer of the tablet (0.50 mm thick) was made by the mixture of 0.1125 g of TLD-700 and 0.0375 g of KBr. Thus, the whole 0.80 mm tablet was made of LiF and KBr, but the self-shielding effect of the ^{6}Li was kept low since it was added to only the thin layer. The ^{6}Li abundance in this layer of radiator was varied between 0.07% and 95.62% with increments of 0.07, 15.95, 31.90, 47.85, 63.77, 80.85, and 95.62%. The second layer was pressed with a small amount of force to form a smooth surface to which the thin layer could be added. Then a larger force was applied to press the entire powder into a tablet.

A preliminary study performed at the Georgia Tech Research Reactor confirmed that the LiF radiator and Lexan polycarbonate foil combination would exhibit a reasonable track density response under a thermal neutron flux irradiation of $5.94 \pm 0.96 \times 10^{6}$ n/cm^{2}. Therefore, a series of thermal neutron exposures were initiated on the HPRR of the Health and Safety Division of Oak Ridge National Laboratory.

In each radiator/polycarbonate foil arrangement irradiated, the LiF radiator faced the thermal neutron source and the first or thin layer containing the differing ^{6}Li abundance was in intimate contact with a 250 micron thick foil of low background, Lexan polycarbonate. The radiator/foil sandwich was enclosed and held in place with a standard, polyethelene TLD "snap-close" holder. The irradiated foils were electrochemically etched in an equal volume solution of C_2H_5OH and 45% KOH by applying 1000 V at 2 kHz for 90 minutes. The reaction particle track density was then determined for each foil by counting the number of tracks in each of five random positions on the microscope at 100X magnification. The actual counting area of each view under magnification was equal to 0.0147 cm^2.

Results

Three periods of irradiation, 10, 20, and 60 seconds, corresponding to thermal neutron integrated fluxes of $(1.02 \pm 0.21) \times 10^7$, $(2.14 \pm 0.37) \times 10^7$, and $(6.80 \pm 0.66) \times 10^7$ n/cm^2 were used. Track counts for the foils with 0.07% ^6Li abundance indicated that the radiators exhibited a background response to thermal neutron irradiation. Duplicate runs were done by irradiating two different radiators at the same time. Since the deviation in each test was not more than 10%, we assumed the runs could be averaged. The averages of each group are plotted on Fig. 1. Thermal neutron track efficiency converted from Fig. 1. as a function of ^6Li abundance in the radiators is plotted on Fig. 2. The curves shown in these two slides indicate the track efficiencies reached a semi-plateau at approximately 60% ^6Li abundance. The data for the 63.77%, 80.85%, and 95.62% enriched radiators was converted to track densities (tracks/cm^2) and thermal neutron fluence (n/cm^2) and are shown in the next slide, Fig. 3. A line can be fitted through each set of data with a slope of 7.91×10^{-4} tracks/neutron, with errors less than 10.3%. The sensitivity of the LiF radiator/polycarbonate dosimeter to thermal neutrons is 755 ± 77 tracks/cm^2 per millirem.

As discussed earlier, the 0.3 mm thick first layer of radiator was much thicker than the effective range of the tritons and alphas liberated during the $^6Li(n,alpha)^3H$ reaction. Therefore, only particles produced in the last 12 microns, which is called the sensitive layer, can reach the polycarbonate foil and produce tracks. The track density generated in the polycarbonate foils should be proportional to the product of the thermal neutron fluence present in the sensitive layer of the radiator and the ^6Li abundance in that layer. As was expected for a given irradiation time, the track density increased with increasing ^6Li abundance until self-shielding had the effect of decreasing the thermal neutron fluence in the sensitive layer as the ^6Li abundance increased.

THERMAL NEUTRON TRACK DENSITY

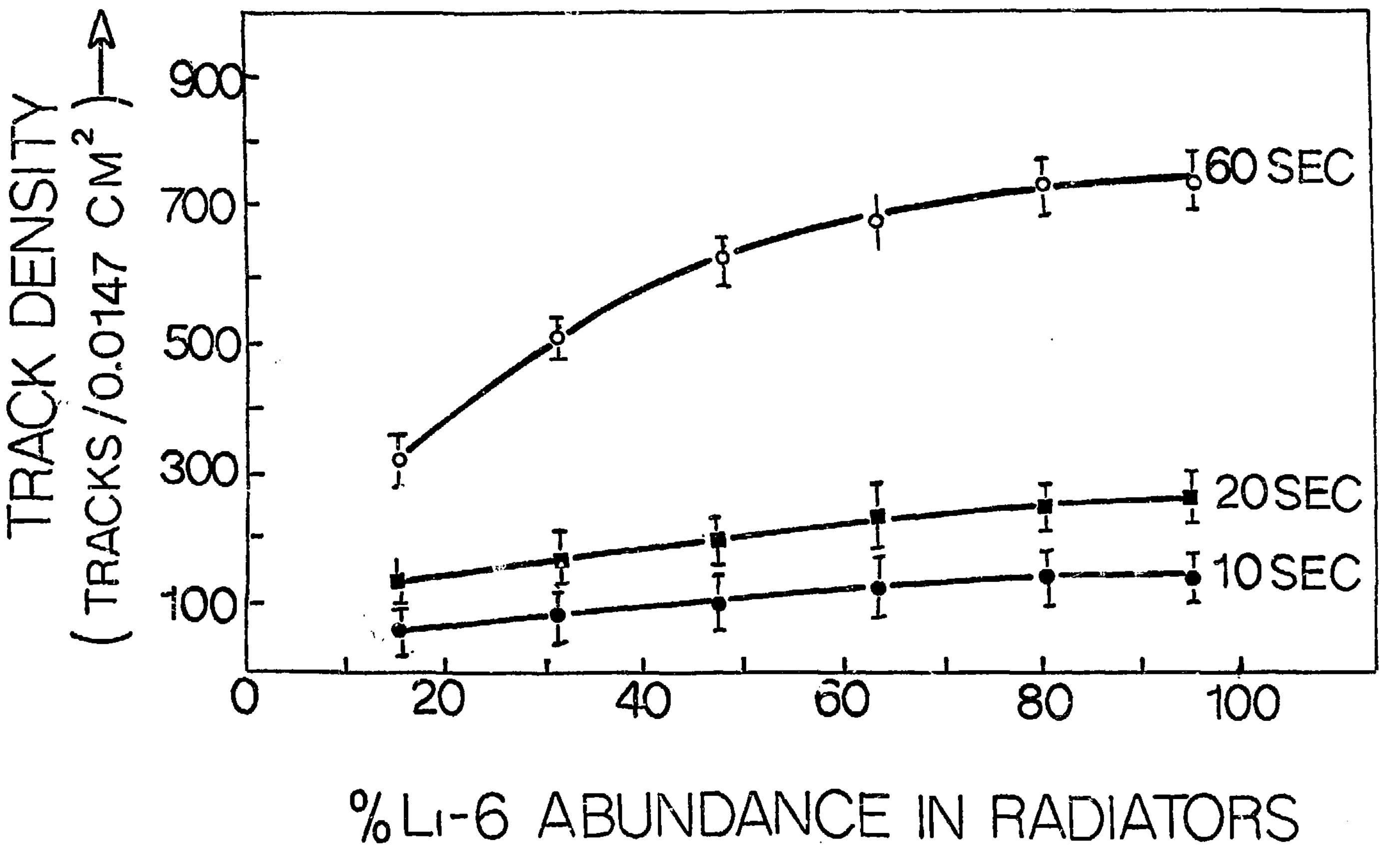

FIGURE 1

THERMAL NEUTRON INTERACTION

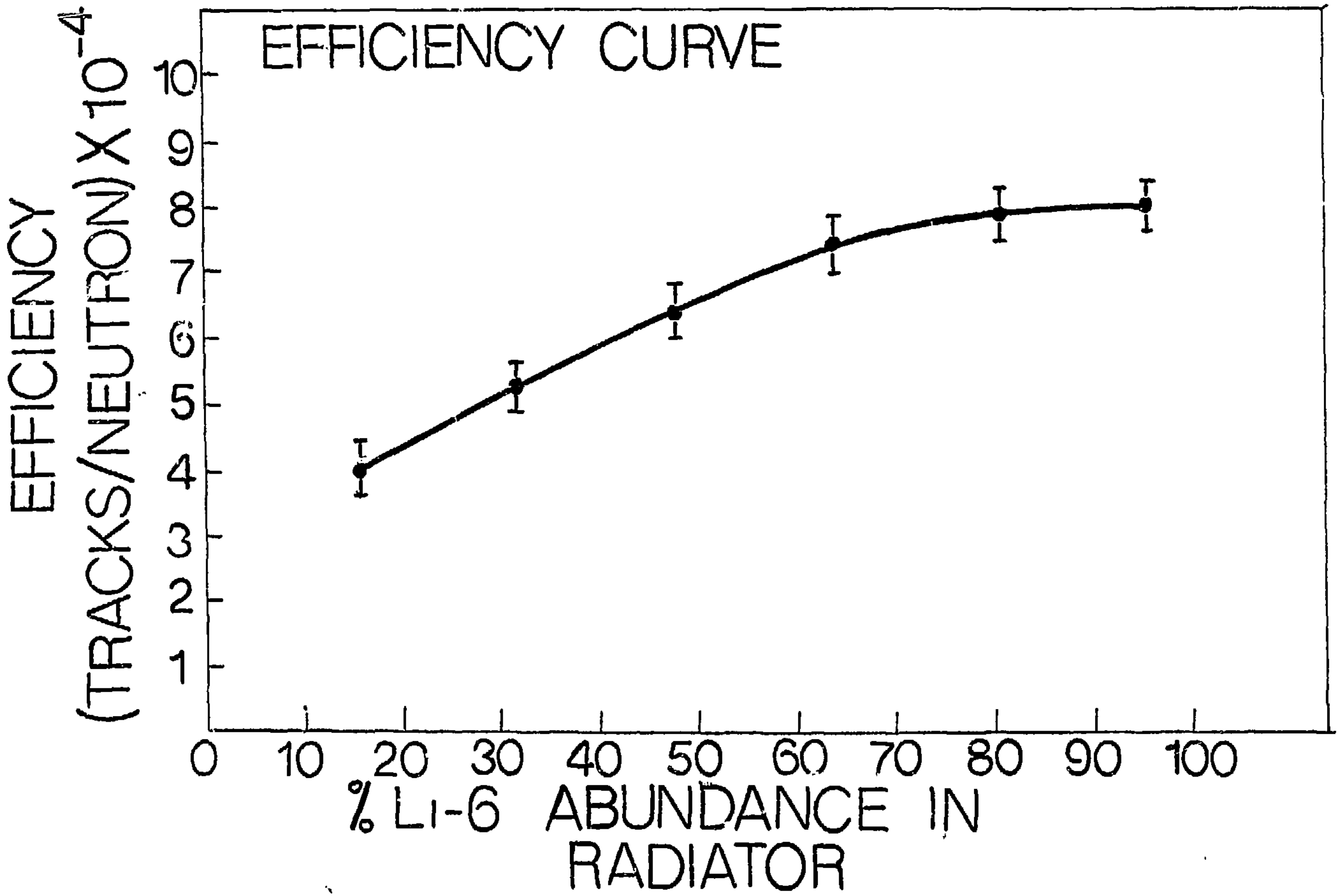

FIGURE 2

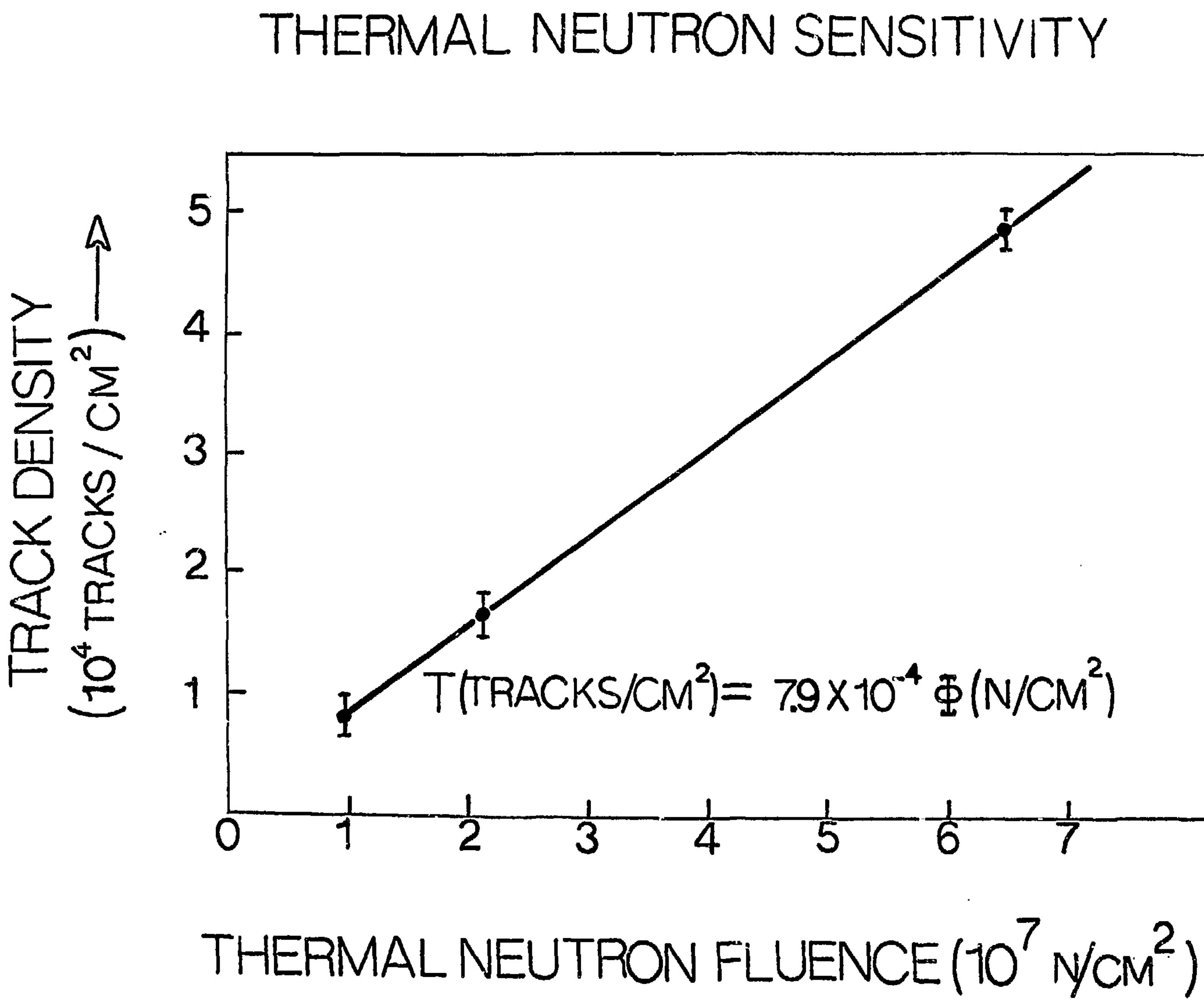
THERMAL NEUTRON SENSITIVITY
TRACK DENSITY
(10⁴ TRACKS / CM²) ⟶
5
4
3
2
1
0
1
2
3
4
5
6
7
T(TRACKS/CM²)= 7.9 X 10⁻⁴ Φ (N/CM²)
THERMAL NEUTRON FLUENCE (10⁷ N/CM²)

FIGURE 3

A supplementary experiment was performed to demonstrate the self-shielding phenomenon of the radiator (next slide). Radiators with seven different ^{6}Li abundances were placed in the polyethelene holders with a gold foil in contact with the first layer of the radiator. The second layer of the radiator (with all TLD-700 and KBr) faced the beam portal of the GTRR biomedical facility. After irradiation, the gold foils were counted and the thermal neutron flux densities were calculated.[4] The relative thermal neutron flux densites as a function of the ^{6}Li abundance are presented graphically here in Fig. 4. It is obvious from this representation that the thermal neutron flux density decreases as the ^{6}Li abundance increases, as expected.

The next slide Fig. 5. shows the LiF/polycarbonate thermal neutron dosimeter's directional dependence. The measured directional dependence of 0°/90° sensitivity is approximately 2.1. This ratio is more pronounced than those of the fission-fragment techniques of Cross and Ing[5] and recoil particle registration techniques of Becker[6] and Piesch[7].

Fig. 6. exhibits the optical density of the foils as a function of neutron dose equivalent in 250 micron low background, Lexan polycarbonate etched in C_2H_5OH and 45% KOH at 22°C by applying 1000 V at 2 kHz for 90 minutes. This slide shows that the optical density is a linear function of thermal neutron dose equivalent over a wide range.

There are three reactions which produce tracks in the polycarbonate foils used in this technique: (a) direct interaction of fast neutrons producing recoil oxygen and carbon nuclei in the foil, (b) the thermal neutron $^{6}Li(n_t,a)^{3}H$ reaction where both the alpha and triton produce tracks, and (c) the fast neutron $^{7}Li(n_f,alpha\text{-}n)^{3}H$ reaction where tracks can be produced by tritons, alphas, and the reaction neutron products. The cross-section for the fast neutron reaction in ^{7}Li is 0.4 barns with a threshold energy of 5 MeV (Sethn et al)[8], while sigma for the thermal neutron ^{6}Li reaction is 945 barns. The ratio of fast neutrons with energy greater than 5 MeV to thermal neutrons in each of three different runs on the HPRR were; 0.93 for the bare HPRR, 0.25 for the steel shielded HPRR, and 0.037 for the Lucite shielded HPRR. The relative concentration of ^{7}Li to ^{6}Li in the first layer of the 80.85% ^{6}Li LiF radiator is 0.24. So the ratio of the fast neutron reaction to the thermal neutron reaction in the LiF radiator for the three different reactor runs is 9.3×10^{-5}, 2.5×10^{-5}, and 3.6×10^{-6} respectively. Thus we deemed it reasonable to neglect reaction (c).

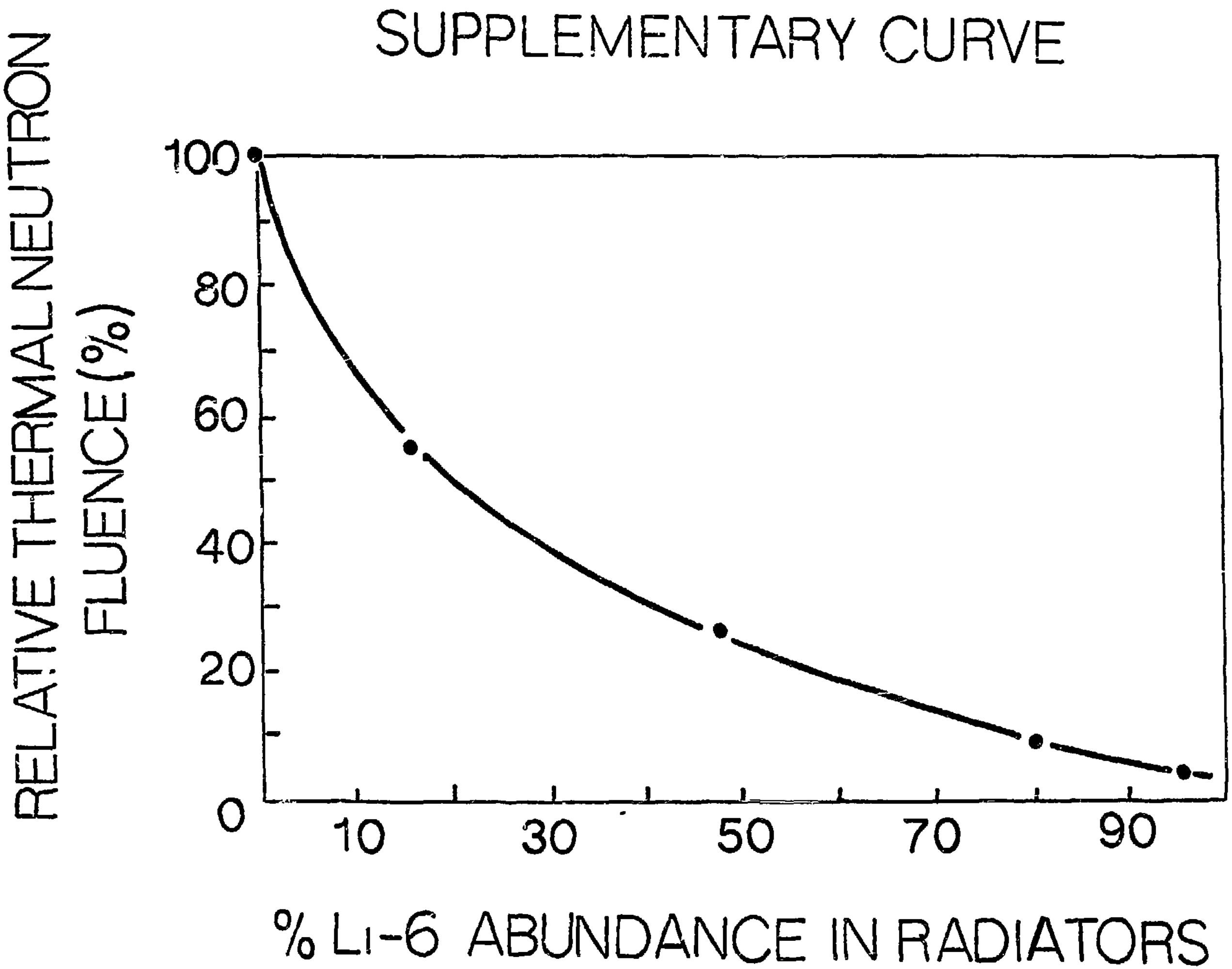
SUPPLEMENTARY CURVE
RELATIVE THERMAL NEUTRON FLUENCE (%)
100
80
60
40
20
0
10
30
50
70
90
% Li-6 ABUNDANCE IN RADIATORS

FIGURE 4

THERMAL NEUTRON DOSIMETER DIRECTIONAL RESPONSE

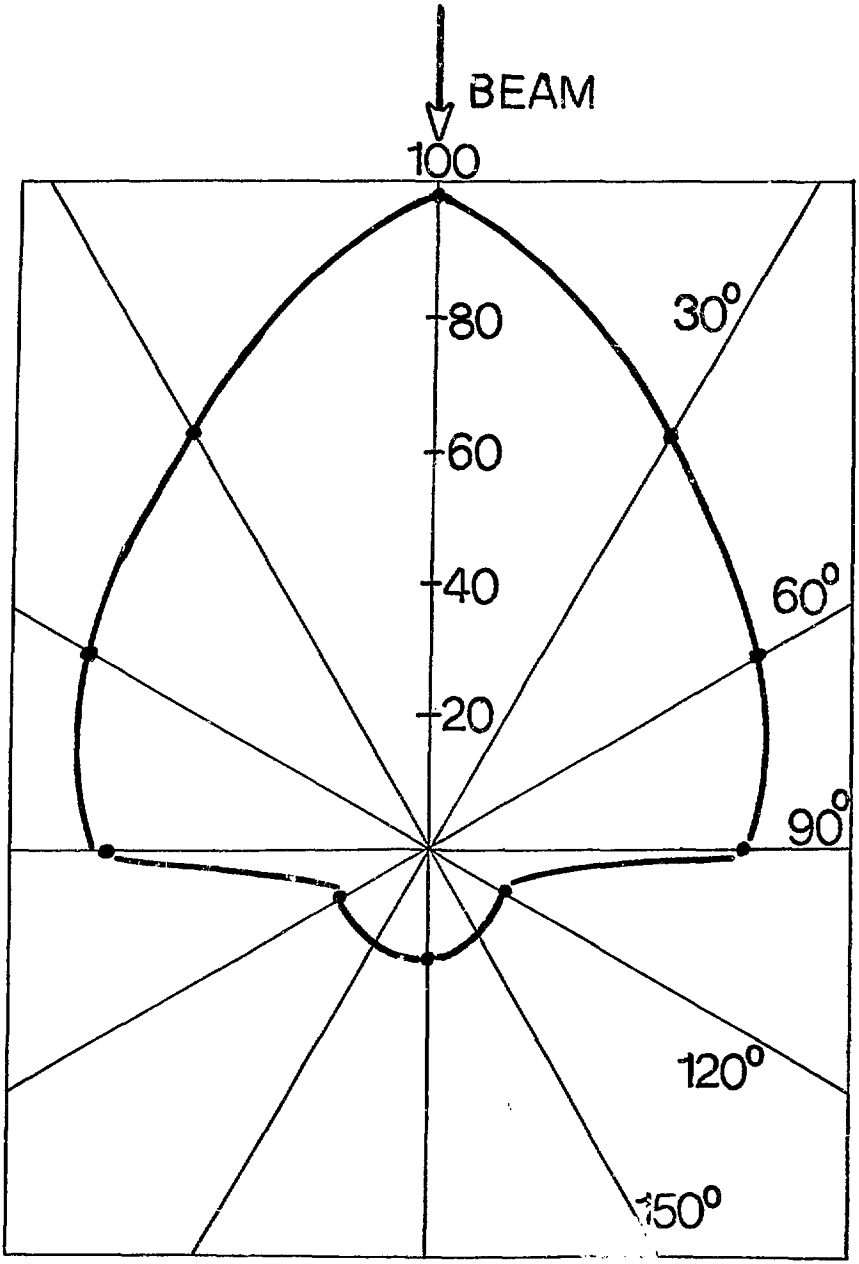

FIGURE 5

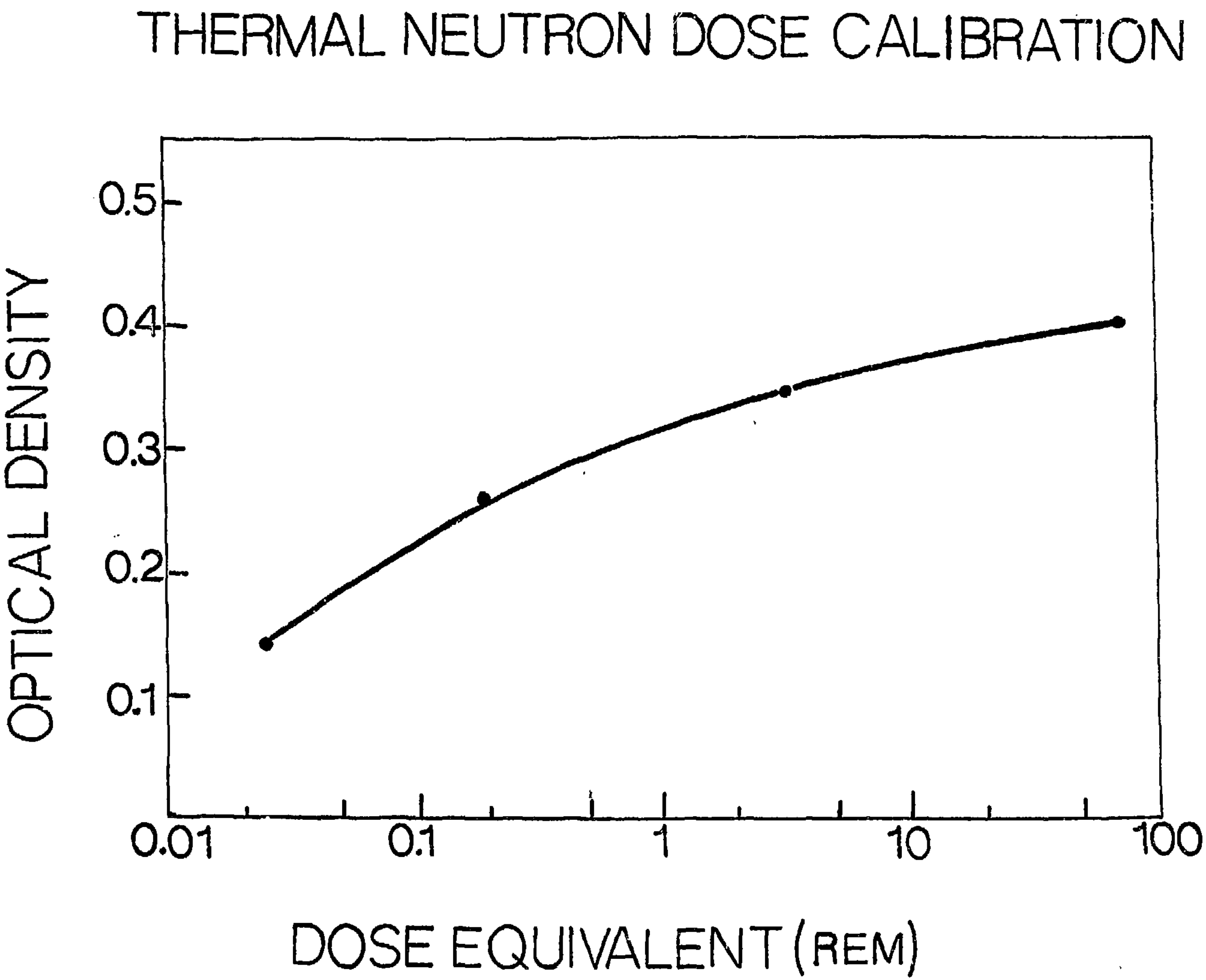
THERMAL NEUTRON DOSE CALIBRATION
0.5
0.4
0.3
0.2
0.1
OPTICAL DENSITY
0.01
0.1
1
10
100
DOSE EQUIVALENT (REM)

FIGURE 6

RUN	SHIELD	MEASURED (MREM)	CALCULATED (MREM)
1	BARE HPRR	H_{TH} 2.16 ± 0.36	1.54
2	HPRR STEEL SHIELD	H_{TH} 5.71 ± 1.04	4.94
3	HPRR LUCITE SHIELD	H_{TH} 45.7 ± 4.1	61.0

FIGURE 7

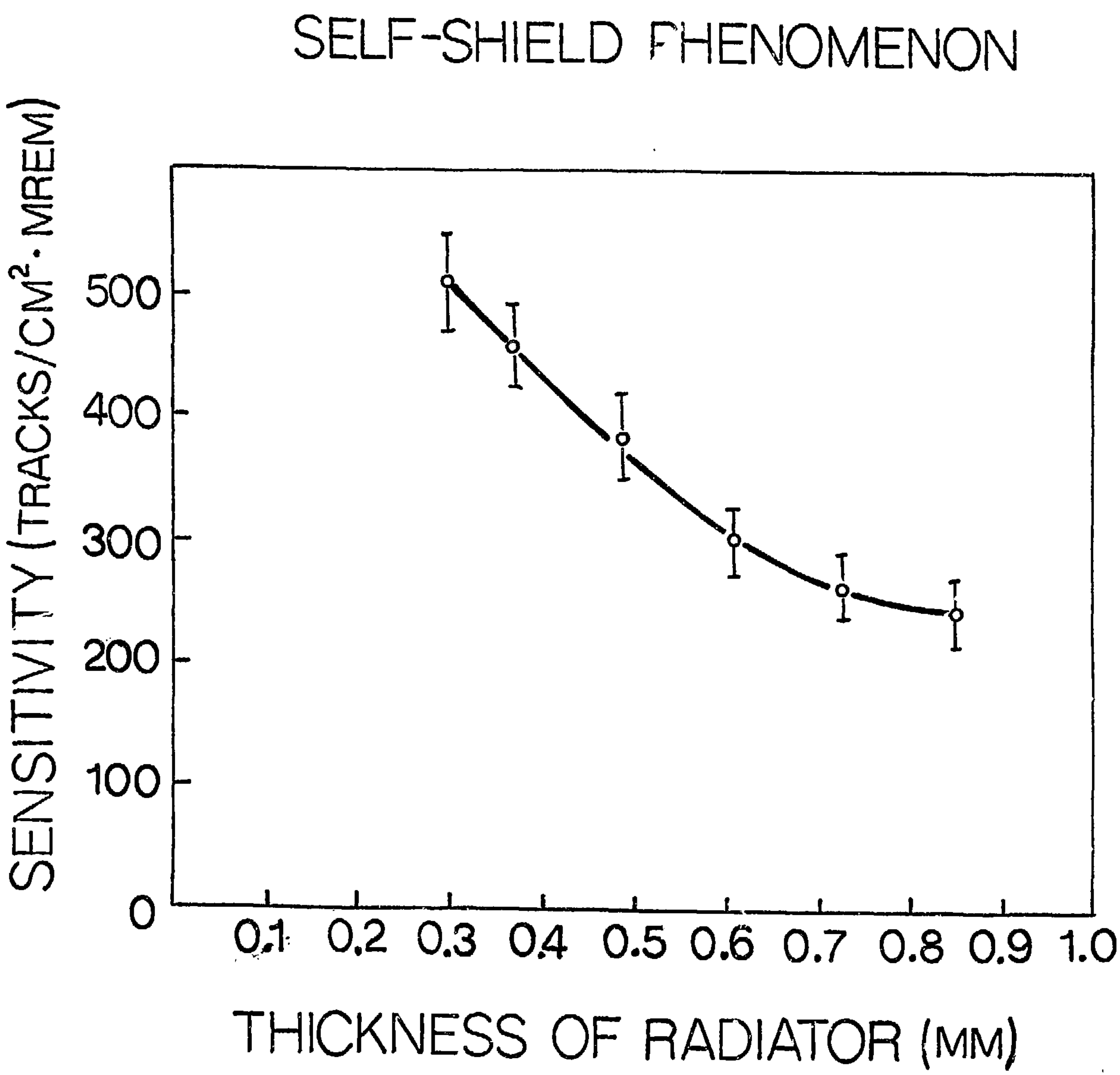
SELF-SHIELD PHENOMENON
SENSITIVITY (TRACKS/CM²· MREM)
500
400
300
200
100
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
THICKNESS OF RADIATOR (MM)

FIGURE 8

Two LiF radiators containing 80.85% ^{6}Li in the first layer were placed in the thermal neutron dosimetry position adjacent to 250 micron polycarbonate foil as previously described. A 0.08 cm cadmium disc was placed in front of one of the radiators to absorb all of the incident neutrons of energy below o.5 eV, i.e., the thermal neutrons (Brown et al)[9]. From the etched foils the thermal neutron dose was determined by subtracting the unshielded track density from the shielded and dividing this value by the thermal neutron conversion factor previously determineded to be 755 ± 77 tracks/cm^2 per mrem and the directional coefficient as described in Fig. 5. Fig. 7. shows the correlation obtained for this run between the measured neutron dose equivalent and the calculated neutron dose equivalent values.

Our future thermal neutron studies using this technique will utilize boric acid radiators and the $^{10}B(n,alpha)^{7}Be$ reaction which has a cross-section of 3837 barns. Because of this high capture cross-section, its lower cost, availability, and according to another supplementary test represented in my final slide (Fig. 8.) has a comparable thermal neutron sensitvity of approximately 500 tracks/cm^2 per mrem, boric acid is most probably a better choice of radiator.

Conclusions

This study demonstrates the feasibility of using high LET particle radiators to determine the thermal neutron dose by reaction particle registration in low background polycarbonate foils using electrochemical etching. When used in conjunction with the already proven fast neutron recoil particle track registration technique, a viable fast and thermal neutron dosimeter is realized with the advantages of being:

* non-fading
* insensitive to low LET radiation reactions
* inexpensive in both processing and materials
* usuable over a wide dose range
* a permanate record and good reproducability
* high sensitivity
* tissue equivalent and a dose equivalent response over a wide range

But most importantly, it finally provides a simple and reliable dosimeter for both the fast and thermal neutron components.

Finally I would like to extend my thanks along with those of Dr. S-J Su and Dr. K.Z. Morgan to the Division of Biomedical and Environmental Research, Department of Energy for the support and financial assistance granted under DOE Contract No. E-26-642.

BIBLIOGRAPHY

1. Sohrabi, M. "Electrochemical Etching Amplification of Low-LET Recoil Particles Tracks in Polymers for Fast Neutron Dosimetry", Ph. D. Dissertation, Georgia Institute of Technology (1976)

2. Su, S-J. "Neutron Dosimetry Using Electrochemical Etching", Ph. D. Dissertation, Georgia Institute of Technology (1979)

3. Tommasino, L. "Electrical Etching of Damaged Track Detectors by H.V. Pulse and Sinusoidal Waveforms", Proc. Int. Colloq. Corpuscular Photog. Solid Detectors, 7th Barcelona (1970)

4. Appendix A and B this paper

5. Cross, W.G. and Ing, H. "Directional Dependence of Fast Neutron Fission Track Dosimeter", P/44, 20th Annual Meeting, Health Physics Society, Buffalo, New York (1975)

6. Becker, K. "Neutron Perdonnel Dosimetry by Non-Photographic Nuclear Track Registration", Proc. ENEA Symp. Rad. Dose Measurements, Stockholm (1967)

7. Piesch, E. "Developement for the New Detectors for Accident Dosimetry", Symp. New Dev. Phy. Biol. Rad. Detectors, VM-143/131 (1970)

8. Steh, I.R., M.D. Goldberg, and B.A. Magurno, "Neutron Cross-Sections", Vol. 1, Suppl. 2, BNL-325, Brookhaven National Laboratory (1964)

9. Brown, J.B., F.M. Gayton, J.A. Hall, J.R. Harvey, and G.A.M. Webb, "Recommendations Concerning the Use of the Personnel Neutron Albedo Dosimeter", RD/BR-828, Berkeley Nuclear Laboratory (1967)