

GEORGIA INSTITUTE OF TECHNOLOGY
OFFICE OF CONTRACT ADMINISTRATION
SPONSORED PROJECT INITIATION

206

Date: March 11, 1977

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

Project No: *E-26-626*

Project Director: *Dr. Karl Z. Morgan*

Sponsor: *Energy Research and Development Administration; Oak Ridge Operations*

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Sponsor Contact Person (s):

Technical Matters

Contractual Matters
(thru OCA)

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Research Contracts, Procedures and Reports Branch
Contract Division
U.S. Energy Research and Development Administration
Oak Ridge Operations
P.O. Box E
Oak Ridge, TN 37830*

NOTE: *Continuation of E-26-623*

Defense Priority Rating: *none*

Assigned to: *Nuclear Engineering* (School/Laboratory)

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GEORGIA INSTITUTE OF TECHNOLOGY
OFFICE OF CONTRACT ADMINISTRATION
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Date: May 24, 1978

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

Project No: E-26-626

Project Director: Dr. Karl Z. Morgan

Sponsor: Department of Energy; Oak Ridge Operations

Effective Termination Date: 11/30/77

Clearance of Accounting Charges: 11/30/77

Grant/Contract Closeout Actions Remaining:

- Final Invoice and Closing Documents
- ~~Final Report~~ Certified Expenditure Statement
- Final Report of Inventions
- Govt. Property Inventory & Related Certificate
- Classified Material Certificate
- Other _____

NOTE: *Continued by E-26-632*

Assigned to: Nuclear Engineering (School/Laboratory)

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PROPOSAL TO CONTINUE DEVELOPMENT AND APPLICATION
OF SOME FAST NEUTRON DOSIMETRY TECHNIQUES
UTILIZING PLASTIC TRACK DETECTORS
FOR RADIOTHERAPY AND HEALTH PHYSICS

Submitted to the
Development of Energy

by the

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PROPOSAL TO CONTINUE DEVELOPMENT AND APPLICATIONS
OF SOME FAST NEUTRON DOSIMETRY TECHNIQUES
UTILIZING PLASTIC TRACK DETECTORS
FOR RADIOTHERAPY AND HEALTH PHYSICS

by

G. B. Stillwagon, S. J. Su and K. Z. Morgan

Abstract

Research in the area of electrochemical etching has been attracting increasing interest for many health physics and for radiotherapeutic applications. A few of the problems of interest which we propose to carry out in the coming year include the rem response determination, improved track reading techniques, fast neutron threshold energy determination, some radiobiological applications, background reduction, thermal neutron dosimetry and large scale etching techniques. Investigation of these problems is expected to make a major contribution to fields of alpha particle and thermal neutron dosimetry. Of special interest in these studies is the development and application of these improved techniques to microdosimetry of plutonium which is localized in the sensitive endosteal surfaces of bone.

Introduction

The complete results to date of our research in the field of electrochemical etching of polycarbonate foils for the purpose of dosimetry have been accumulated and appear in (ORO-4814-5) and our most recent progress report dated August 1977. These results show the intensive research done by the investigators in the field as well as some recommendations on new problems to be solved for the advancement of this recent and interesting avenue of approach for many dosimetry applications. Some of the recommended studies are already being investigated and preliminary results have been presented at several scientific meetings including the American Physical Society, Auburn, Alabama, 1975; Health Physics Society, Denver, Colorado, 1976 and San Francisco, 1976; International Radiation Protection Association, Amsterdam, The Netherlands, 1975; the American Industrial Hygiene Association, Atlanta, Georgia, 1976; and the Health Physics Society, Atlanta, Georgia, 1977. (Copies of these reports have been sent to the Washington office of ERDA.)

Many of the problems we have listed under this contract are being investigated actively at present, including calibration of this dosimeter for alpha particles from plutonium, proton track formation in polycarbonate, a new technique to simultaneously detect fast and thermal neutrons by means of polycarbonate foils using electrochemical etching technique and the determination of microdistribution of α -dose deposited by ^{239}Pu in the radiosensitive 10 μm layer on the endosteal surface of bone. The last two problems are in the preliminary stages of examination and will be actively pursued during the new contract year as well as some other interesting areas of research proposed below.

Proposed New Research

The following proposals for continued research into the infant field of electrochemical etching of polycarbonate foils are but a few of the promising avenues of research and application of this technique. This new development offers the promise of information hitherto unachievable in the field of health physics over prevailing methods such as the thick photographic emulsions with proton track counting (a technique which one of us [KZM] first introduced to health physics in 1944). These problems and other parameters to be evaluated in polycarbonate are as follows.

Rem Response Determination

The response of tissue to fast neutrons, called rem response, provisionally estimated by methods of approximation in the past. However, it was normally estimated only for the first collision dose without considering the results of second, and higher order collisions. These latter neutrons, sometimes referred to as albedo neutrons, could be very important in determining the total in vivo dose. Extensive measurement of the neutron dose distribution in more realistic non-homogeneous phantoms, i.e. in media simulating the complexity and heterogeneity of a human body is of considerable interest and importance to neutron therapy, human radiography and radiation protection. Attempts will be made to find the dose distribution, including the effect of albedo neutrons, in a heterogeneous phantom.

Track Reading Techniques

Many different track reading techniques have been tried to read these polycarbonate foils as accurately as possible with a minimum of eyestrain in order to make this method of dosimetry more attractive for mass

application or for commercial utilization. We have mentioned in the past our success with the microfiche reader and the overhead projector (see August 1977 Progress Report). In addition to these techniques there is an almost limitless number of other conceivable track reading techniques; among these would be the use of select portions of the optical spectra, polarized light and special microscope devices. The latter is mentioned because now we are interested in counting very small track diameters ($\sim 1 \mu\text{m}$) which require oil emersion or phase contrast microscopy. We will examine these possibilities this contract year.

Fast Neutron Threshold Energy

The actual threshold energy a neutron needs to just begin to create tracks in polycarbonate has been widely debated in the literature. The difficulty in determining this threshold is caused by the wide number of parameters which affect the final result such as etching conditions and treatment of the foil before etching. To attempt to shed more light on this threshold for fast neutron track production we will examine this threshold in greater detail.

Radiobiological Applications

Polycarbonate electrochemical dosimetry has the potential of making one of its greatest contributions in this area. In the past it has only been possible to determine the location of bone seeking radionuclides in osseous tissue using tedious photographic emulsion techniques. These suffer from many drawbacks such as fading, long exposure times and high sensitivity to variations in developing technique. Using the polycarbonate dosimeter we can determine the location of these nuclides by observing

the position of alpha particle tracks or fission fragment tracks (after thermal neutron irradiation) in these foils. These foils possess the desirable characteristics of no fading, shorter irradiation times and a simpler, more reliable, developing procedure. Not only do we believe this can be done but also we believe we can go further and determine the micro-distribution of the dose delivered in the 10 μm layer on the sensitive endosteal surface of bone. Hitherto this has been accomplished only by calculational methods involving theoretically derived equations with assumptions applied to obtain a usable equation. We believe we can determine, experimentally, the dose received by this 10 μm thick layer on the endosteal surface of bone which is so important when considering biological effects such as osteosarcoma. It is likely that these data can be useful in determining a more meaningful or relevant maximum permissible body burden for ^{239}Pu and provide an operational procedure usable to obtain similar data for many bone-seeking radionuclides. The International Commission on Radiological Protection cautioned in its publications (ICRP No. 8-1966, ICRP No. 11-1967, ICRP No. 14-1969 and ICRP No. 19-1972) that the present practice of calculating the permissible body burden of the actinide elements by averaging the dose over the entire skeleton should be discontinued in favor of averaging the dose over the 10 μm endosteal surface layer of trabecular bone. Unfortunately this has never been done because past dosimetry techniques do not provide the necessary spacial resolution.

Background Reduction

In our previous Progress Reports we mentioned the low background levels we have achieved, which appear to be among the lowest that have been reached

operationally at this time. We would like to lower this background still further, especially to try to eliminate some anomalies that are sometimes noticed in the foils. Very low backgrounds are sometimes important; for example, when neutron personnel monitoring films are processed only on an annual basis or when one must obtain the neutron contamination in a beam of accelerator produced x-rays used in medical therapy.

Thermal Neutron Dosimetry Using Polycarbonate Foils

Both theoretical and experimental results show the polycarbonate foil is only sensitive to fast neutrons when electrochemically etched. The aim of this study is to employ nuclides having a high (n,α) cross section, e.g. ${}^6\text{Li}$ and ${}^{10}\text{B}$ as radiators for thermal and epithermal neutron dosimetry. The main advantage of using ${}^6\text{Li}$ or ${}^{10}\text{B}$ as a radiator is that slow neutron and fast neutron responses could both be permanently recorded on one polycarbonate foil because of the possibility of distinguishing between them for example the fast neutron carbon recoils and the $\alpha + {}^7\text{Li}$ thermal neutron recoils from ${}^{10}\text{B}$ or $\alpha + {}^3\text{H}$ recoils from ${}^6\text{Li}$.

Large Scale Etching

We have reported previously our efforts to etch a greater number of foils at one time than possible before. This step is important in showing that the polycarbonate dosimeter can be used for routine, large scale dosimetry purposes. We will continue our efforts to improve this step toward large scale etching capabilities.

Proton Irradiations

There is still some concern surrounding the ability of protons to cause etchable tracks in polycarbonate as a result of the protons own

ionization. This problem will be examined by irradiating foils with protons possessing several different energies. We will then attempt to reach a final conclusion in answering this intiguing question.

Duration of the Proposed Research

The proposed research is intended to be completed within a period of 12 months which will last from December 1, 1977 to November 30, 1978.

Project Personnel

Karl Ziegler Morgan was born in Enochville, North Carolina on September 27, 1907. He received the A.B. and M.A. degrees from the University of North Carolina in 1929 and 1930, respectively, and the Ph.D. degree in physics from Duke University in 1934. From 1934 to 1943 he was chairman of the Physics Department of Lenoir Rhyne College, and during this period did research in cooperation with Duke University in the field of cosmic ray showers, the meson lifetime, etc. in Linville Caverns, on Mt. Mitchell, Beech Mountain, Mt. Evans, etc.

He joined the Metallurgical Laboratory staff at the University of Chicago in the spring of 1943. Here he was one of a group of six persons who developed and established the new science and profession of health physics. During the fall of 1943, he transferred to Oak Ridge National Laboratory where, until the latter part of 1972, he was Director of the Health Physics Division which was engaged in research, engineering, and applied problems. Upon leaving Oak Ridge National Laboratory, he joined the faculty of the Georgia Institute of Technology as Neely Professor in the School of Nuclear Engineering.

In 1956, he was the first president of the Health Physics Society, and from 1966 to 1970 was the first president of the International Radiation Protection Association which has about 10,000 members in some 65 countries. He now is President Emeritus of IRPA, an Emeritus member of

the Main Commission of ICRP, and a member of NCRP. For 20 years he was chairman of the committees of ICRP and NCRP dealing with maximum permissible internal dose of radioisotopes. He has published over 300 papers dealing with subjects of cosmic rays, radiation protection, instrumentation, internal dose, and general health physics. He is past editor of the journal HEALTH PHYSICS.

In 1962, K. Z. Morgan and W. Binks (England) were awarded the first gold medals for meritorious work in the field of radiation protection by the Royal Academy of Science of Sweden.

Dr. Morgan received the Distinguished Alumni Award and the honorary Doctor of Science Degree from Lenoir Rhyne College, honorary membership in Sigma Pi Sigma from Berea College, and the First Distinguished Service Award of the Western Chapter of the Health Physics Society. He is a member of the Health Physics Society, the International Radiation Protection Association, the American Public Health Association, the American Association for the Advancement of Science, the American Industrial Hygiene Association, the Research Society of America, the Radiation Research Society, the Society of Nuclear Medicine, the American Association of Physicists in Medicine, the American Association of Physics Teachers, an Associate Fellow of the American College of Radiology, a Fellow of the American Physical Society and of the American Nuclear Society, and an Affiliate of the Royal Society of Medicine.

Major Publications of the Last Five Years

1. "Common Sources of Human Exposure to Ionizing Radiation in the United States," American Engineer, July 1968.
2. "Ionizing Radiation: Benefits Versus Risks," Annual Meeting of the Health Physics Society, June 16-20, 1968, Denver, Colorado; and published in HEALTH PHYSICS, Vol. 17, No. 4.
3. "Assumptions Made by the Internal Dose Committee of the International Commission on Radiological Protection," Sixth Annual Meeting of the Gesellschaft fur Nuclearmedizin, Wiesbaden, Germany, September 26-28, 1968; and published in Proceedings, 1969.
4. "Redirecting Health Physics Studies to Areas of Greatest Interest," First European Congress of the International Radiation Protection Association, Menton, France, October 9-11, 1968; and published in Proceedings, 1968.
5. "The Need for Standardization Procedures in the Application of Ionizing Radiation to Medical and Dental Patients," Seminar sponsored by the National Center for Radiological Health, Rockville, Maryland, November 15, 1968, Seminar Paper 003.
6. "The Proper Working Level of Radon and Its Daughter Products in the Uranium Mines of the United States," Hearing on Radiation Standards for Mines, Washington, D. C., November 20, 1968; Congressional Record, 1968.
7. "Supplemental Statement on the Proper Working Level of Radon and Its Daughter Products in the Uranium Mines of the USA," Supplement to Testimony presented on November 20, 1968, Washington, D. C.; Congressional Record, 1968.
8. "Future Opportunities in Health Physics," Health Physics Society Mid-year Topical Symposium, Los Angeles, California, January 29-31, 1969.
9. "Risks from Diagnostic X-Rays," Yale Scientific, Vol. XLII, No. 5, February 1968; Reprinted from Yale Scientific in the Journal of the American Radiography Technologists, Vol. XIV, No. 4, Winter 1969.
10. "Radiation Standards for Reactor Siting," Testimony presented before the Joint Committee on Atomic Energy at its Hearings on Environmental Effects of Producing Electrical Power, Phase 2, January 1970; Congressional Record.
11. "Energy Pollution of the Environment," Midyear Symposium of the Health Physics Society, Louisville, Kentucky, January 28, 1970; and published in Proceedings in USPHS-BRH Series.
12. "A Time of Challenge to the Health Physicist," Presidential Address presented before the Second International Congress on Radiation Protection, May 8, 1970, Brighton, England; Health Physics, Vol. 20, May 1971, pp. 491-498.
13. "My Opinion — You Can Drastically Cut X-Ray Exposure Below Today's Levels," Consultant, March/April 1970.

14. "History of the Health Physics Society," published as part of the RSNA Symposium on the Critical History of American Radiology (Nov. 1970).
15. "History of the International Radiation Protection Association," published in Proceedings of the RSNA Symposium on the Critical History of American Radiology, November 1970.
16. "Criteria for the Control of Radioactive Effluents," IAEA Symposium on Environmental Aspects of Nuclear Power Stations, UN Building, New York, August 1970, Proceedings published, this paper is IAEASM-146/10; Synopsis published also in Environmental Studies, 1971.
17. "Maximum Permissible Levels of Exposure to Ionizing Radiation," International Summer School on Radiation Protection, Boris Kidric Institute of Nuclear Sciences, Cavtat, Yugoslavia, September 20-30 1970; and published in Proceedings under title of "Radiation Dosimetry," 1971.
18. "President's Report to the General Assembly of IRPA," Brighton, England, May 1970, Health Physics, Vol. 20, No. 5, 1971.
19. "History of Radiation Protection," Symposium Ccmmemorating the 75th Anniversary of the Discovery of X-Rays," Milwaukee, November 13-14, 1970; Materials Evaluation, Vol. XXIX, No. 3, March 1971.
20. "Why the 1968 Act for Radiation Control for Health and Safety Is Required," Radiology, Vol. 99, No. 3, pp. 569-588, June 1971.
21. "Excessive Medical Diagnostic Exposure," Third Annual National Conference on Radiation Control," Scottsdale, Arizona, May 3, 1971; and published in Proceedings.
22. "Health Physics and the Environment," International Symposium on Rapid Methods for Measurement of Radioactivity in the Environment, Neuherberg, Federal Republic of Germany; (Proceedings published by IAEA).
23. "Adequacy of Present Radiation Standards," presented at the Environmental and Ecological Forum, Silver Springs, Maryland, January 20, 1971; Proceedings of Forum published in 1972.
24. "Proper Use of Information on Organ and Body Burdens on the Assessment of Radioactive Organ and Body Burdens, Stockholm, Sweden, November 22-26, 1971, IAEA/SM/150-50; Proceedings of Symposium published by IAEA.
25. "Health Physics Measures to Implement New USAEC Regulations Relating to Radiation Exposure of the General Public," Budapest, May 1971; and published in Proceedings.
26. "The Need to Reduce Medical Exposure in the United States," outline of testimony presented before the Health and Welfare Subcommittee of the Senate Committee on Labor and Public Welfare on Senate Bill S.3327, May 15, 1972, Washington, D. C.; published in Congressional Record, 1972.
27. "Environmental Impact of Natural and Man-Made Ionizing and Non-Ionizing Radiations," Second Health Physics Summer School, Herceg Novi, Yugoslavia, August 1973; and published in Proceedings, 1974.

28. "The Need for Radiation Protection," Radiologic Technology, 44, No. 6, pp. 385-395, 1973.
29. "Possible Consequences of Excessive Medical Exposure in the United States," Given before the Fachverband für Strahlenschutz at the Bern, Switzerland meeting, March 21, 1973 and published in the Fachverband Proceedings, 1973.
30. "Transportation of Radioactive Material by Passenger Aircraft," Committee report to the Joint Committee on Atomic Energy of Congress, Congressional Record, September 17, 1974.
31. "Biological Effects of Ionizing Radiation," published in Proceedings of Short Course, September 9, 1974.
32. "Reducing Medical Exposure to Ionizing Radiation," Journal American Industrial Hygiene Association, 36, 5, p. 358, May 1975.
33. "Appropriateness of Regulations for Air Shipment of Radioactive Materials," Miami Beach, Florida, September 23, 1974, International Conference on Shipping of Radioactive Material; and published in Proceedings, 1975.
34. "Health Physics — Past, Present and Future," Published in Proceedings of First Asian Congress of the International Radiation Protection Association, December 1974.
35. "Suggested Reduction of Permissible Exposure to Plutonium and Other Transuranium Elements," Journal of American Industrial Hygiene Association, July 1975.
36. "Effects of Radiation on Man," Proceedings of Short Course on Energy and the Environment — Cost-Benefit Analysis published by Pergamon Press, (1975).
37. Chapters on "Ionizing Radiation Exposure," and on "Exposure to Non-Ionizing Radiation," in text, Environmental Problems in Medicine, Ed. W. D. McKee; publisher, Charles C. Thomas, 1974.
38. "Recent Developments in Fast Neutron Personnel Dosimetry Using Track Etch Methods," Proc. 3rd European Congress IRPA, Paper 14, Amsterdam, The Netherlands, May 1975, with Mehdi Sohrabi.
39. "Development and Application of Some Fast Neutron Dosimetry Techniques Utilizing Plastic Track Detectors for Radiotherapy and Health Physics," USERDA Contract No. AT-(40-1)-4814, 1975, with Mehdi Sohrabi.

Biographical Sketch

STILLWAGON, GARY BOULDIN--Ph.D. Candidate, Georgia Institute of Technology, School of Nuclear Engineering

Education

| | |
|--|--------------|
| B.S., Physics, Georgia Institute of Technology | 1974 |
| M.S., Nuclear Engineering, Georgia Institute of Technology | 1975 |
| Ph.D. Candidate, Georgia Institute of Technology | 1976-Present |

Employment History

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|---|--------------|
| Methodist Hospital, Memphis, Tennessee, Medical Physicist | 1974 |
| Georgia Institute of Technology | |
| Graduate Research Assistant | 1975-1976 |
| Graduate Research Assistant and Co-Principal Investigator | 1976-Present |

Experience Summary

In 1974, Mr. Stillwagon performed the duties of a Medical Physicist at the Methodist Hospital in Memphis, Tennessee. His duties included calibration of radiation therapy machines, calibration of nuclear medicine instruments, writing procedures for the calibration and upkeep of these instruments to be used routinely by technicians and teaching classes in X-ray and Radium Physics. From 1975 to 1976, Mr. Stillwagon worked as a research assistant on a project concerned with the transmutation of transuranic actinide elements as an alternative to waste disposal. From 1976 to the present, Mr. Stillwagon has been pursuing the Ph.D. degree in Nuclear Engineering with a speciality in Health Physics. During this time, he has also been teaching, performing research and serving as co-principal investigator on ERDA and DOE projects with the Georgia Institute of Technology.

Professional Activities

Member, Health Physics Society
Member, American Association of Physicists in Medicine
Member, American Nuclear Society
Past Member, Georgia Society of Professional Engineers and Society of Physics Students
Graduate Student Senator

Reports, Publications and Papers

1. Stillwagon, Gary B., "Void Fractions in Cruciform-like Flow Channels," Tran. Am. Nuc. Soc. Stud. Conf., Atlanta, Georgia (1975).
2. Stillwagon, Gary B., "The Importance of Void Geometry Representation for Plutonium Recycle Analysis In Boiling Water Reactors," Trans. Am. Nuc. Soc. Stud. Conf., University of Virginia, Charlottesville, Virginia (1976).

Reports, Publications and Papers (Continued)

3. Stillwagon, Gary B., "The Gas Core Actinide Transmutation Reactor," Trans. Am. Nuc. Soc. Stud. Conf., University of Virginia, Charlottesville, Virginia (1976).
4. Stillwagon, Gary B., and R. W. Carlson, "The Importance of Void Geometry Representation for Plutonium Recycle Analysis in Boiling Water Reactors," Trans. Am. Nuc. Soc. Conf., Toronto, Canada (1976).
5. Stillwagon, Gary B., "Adapting the Polycarbonate Detector and Electrochemical Etching to the Microdosimetry of ^{239}Pu In Bone," Trans. Am. Nuc. Soc. Stud. Conf., Raleigh, N. C. (1977).
6. Proposed Research for the Ph.D. degree, "The Microdosimetry of ^{239}Pu in Bone Utilizing the Polycarbonate Dosimeter and Electrochemical Etching," in partial fulfillment of the Ph.D. degree, Georgia Institute of Technology (1977).
7. Stillwagon, Gary B., S. J. Su and K. Z. Morgan, "Adapting the Polycarbonate Detector and Electrochemical Etching to the Microdosimetry of ^{239}Pu in Bone," Trans. Health Physics Society Conf., Atlanta, Georgia (1977).
8. Su, S. J., G. B. Stillwagon and K. Z. Morgan, "Neutron Dosimetry Using Electrochemical Etching," Health Physics Society Conference, Atlanta, Georgia (1977).
9. Stillwagon, Gary B. and K. Z. Morgan, "Calibration of the Polycarbonate Detector for the Microdosimetry of ^{239}Pu Alpha Particles in Bone," Trans. Health Physics Society Conf., Atlanta, Georgia (1977).

Biographical Sketch

SU, SHIAN-JANG--Ph.D. Candidate, Georgia Institute of Technology, School of Nuclear Engineering

Education

| | |
|--|--------------|
| B.S., Nuclear Engineering, National Tsing Hua University | 1968 |
| M.S., Nuclear Science, National Tsing Hua University | 1970 |
| Ph.D. Candidate, Georgia Institute of Technology | 1975-Present |

Employment History

| | |
|--|--------------|
| Atomic Energy Council, Republic of China, Research Assistant | 1968 |
| Army of Republic of China, 2nd Lieutenant | 1970-1971 |
| Institute of Nuclear Energy Research, Researcher | 1971-1975 |
| Karshrue Nuclear Center, West Germany, Practical Training | 1972 |
| Georgia Institute of Technology, Graduate Assistant | 1976-Present |

Experience Summary

While working at the Institute of Nuclear Energy Research, Republic of China, Mr. Su was responsible for radiation dosimetry in the health physics section. He attended many programs such as construction of whole body counters, hot cell shielding design, personnel monitoring and instrument calibration for a 50 MW heavy water reactor and radioisotope applications. In July 1972, under the sponsorship of the Atomic Energy Council, Mr. Su went to West Germany and attended practical training at the WAK fuel reprocessing plant. Since 1975 he has been pursuing a Ph.D. degree in Nuclear Engineering with a specialty in Health Physics. He is a member of the China Nuclear Society and the Health Physics Society.

Reports, Publications and Papers

1. B.S. Thesis, "Fast Neutron Flux Measurement of 1MW Tsing Hua Open Pool Reactor," National Tsing Hua University, 1968.
2. M.S. Thesis, "Study on Taiwan Natural Background," National Tsing Hua University, 1970.
3. Su, Shian-Jang, "A Preliminary Calculation of Argon-41 Gamma Dose From TRR," Nuclear Science Journal, 11, 58 (1974).
4. Su, Shian-Jang, "Final Report for Whole Body Counter," INER, Taiwan (1975).
5. Su, Shian-Jang, "Neutron Dosimetry Using Electrochemical Etching," Paper presented in 1977 American Nuclear Society Eastern Region Student Conference, Raleigh, N. C., 1977.
6. Literature review of the proposed research, "Neutron Dosimetry Using Electrochemical Etching," in partial fulfillment of the Ph.D. degree, Georgia Institute of Technology, 1977.
7. Su, Shian-Jang, "Neutron dosimetry using Electrochemical Etching," Paper P/21 presented in 22nd Annual Meeting of the Health Physics Society, Atlanta, Georgia, 1977.
8. Su, Shian-Jang, "Detection of Hot Fallout on Taiwan in the Period 1971-1975," Health Physics, 33, 241 (1977).

GEORGIA TECH FACILITIES

The facilities most closely related to teaching and research in the nuclear engineering programs at Georgia Tech are located in the Frank H. Neely Nuclear Research Center and the Cherry L Emerson Building.

Nuclear Research Center

The Frank H. Neely Nuclear Research Center, valued at \$10 million dollars, was completed in 1964. The Georgia Tech Research Reactor (GTRR) which serves as the key component to the Center is a heterogeneous, heavy-water moderated and cooled reactor, fueled with enriched uranium. It is similar in design to the Argonne Research Reactor, CP-5, which has demonstrated excellent research capability and dependable service. The GTRR has been designed to develop a sustained power output of five megawatts with a corresponding thermal neutron flux of about 10^{14} n/cm²-sec. The reactor has a variety of experimental facilities (such as beam tubes, through-tubes, pneumatic tubes, fast flux facility, biomedical facility, thermal column, and gamma facility) to provide a wide range of experiments. The GTRR currently operates at a power of five megawatts two or three shifts per day.

The reactor and associated systems are housed in a steel containment building eighty feet in diameter and sixty-five feet in height. The adjoining 24,000 square foot, two-story, air-conditioned laboratory and office building contains laboratories for radiochemistry, health physics, materials preparation, nuclear spectroscopy, low-level counting, radiobiology, electronics, as well as a machine shop and a large animal quarter.

Equipment at the Frank H. Neely Nuclear Research Center includes:

5 megawatt, heavy-water research reactor

Neutron diffractometers (3)

Time-of-flight neutron spectrometer

Experimental setup for pile oscillator measurements

100,000 curie Co^{60} facility

50,000 curie capacity hot cell with master-slave manipulators

20' x 6' x 18' deep storage pool joined to hot cell by transfer chute

Neutron generator--200 keV TMC Activation Model 211, (d,n) reactor with tritium producing 2×10^{14} MeV neutrons/sec

Gamma facility--spent fuel element storage in pool provides gamma rates to 10^6 R/hr depending on sample geometry

Intermediate level radiochemistry laboratory (18' x 26') with radioisotope hoods, shielded glove boxes

Facilities for treatment, storage, and handling of solid and liquid radioactive wastes, including retention tanks of 8000 gallon capacity

Model 33 teletype terminal providing remote computer access on and off campus

Data acquisition system

The data acquisition system consists of three data processors (PDP-8, PDP-8/I, and PDP-12), with appropriate peripheral devices for use in sophisticated experiments requiring on-line analysis and control, as well as data collection. One of the processors is located within the reactor containment building and is intended for use by experimenters with a minimum of setup time. A rather sophisticated interface for accepting both digital and analog signals is operational. There is a communications link to the other processors, located in the laboratory portion of the Center, which acts as

a data collection center for several experiments and a secondary processor. Cell processors function (with lower priority) as a data reduction and preprocessing facility, and as such are generally available for use 24 hours per day.

Emerson Building

The School of Nuclear Engineering also occupies about 24,000 square feet in the Cherry L. Emerson Building. This space contains ample classrooms, student laboratories and offices as well as a periodical library, staff machine shop, electronics shop, seminar room, computation laboratory and a large, two-story high bay with 10 ton crane. The latter space houses the nuclear engineering laboratory. The counting rooms and other laboratories have been well equipped with conventional radiation detection instruments (including several multichannel analyzers) in sufficient numbers to accommodate laboratory sections of 12-15 graduate students.

Specialized equipment available in the Emerson Building to nuclear engineering students includes:

An AGN-201 training reactor

A natural uranium-light water subcritical assembly

Korad K-15 pulsed ruby laser

Three EAI model TR-20 analog computers

A 12,000 curie Cs-137 gamma source

One MeV Van de Graaff positive particle accelerator with beam analysis system

50 and 250 KVP x-ray machines

Several gas chromatographs

Eleven radioisotope fume hoods and three glove boxes

Vacuum deposition system

The training reactor is located in the nuclear engineering laboratory. It is used in laboratory exercises and for special student problems and theses. The GTRR continues to be available for all pedagogical experiments and research which require its higher flux.

Rich Electronic Computer Center

The Georgia Tech Rich Electronic Computer Center was established in 1955 and its facilities are an integral part of the academic and research programs at Georgia Tech. The Office of Computing Services operates two large-scale digital computers, a Control Data Corporation CYBER 74 system and a Control Data Corporation 6400 system. Both systems operate under the NOS 1.0 operating system, thus providing back-up capabilities.

The CDC CYBER 74 has the following configuration: Two central processors (one 6400 and one 6600), fourteen peripheral processors, 131,072 words of core memory (60 bit words), 944 million characters of disk storage, eight magnetic tape drives, two card readers (1200 cpm), three line printers (1200 lpm), one card punch (250 cpm), 64 ports for time-sharing users, and an operator console. A CALCOMP Model 763 Digital Incremental Plotter provides high quality.

The following software packages are available: COMPASS, ALGOL, COBOL, FORTRAN (four different compilers are available), APL, RPG, MIMIC, MIX, plus many others.

A Radiation, Inc. Analog-to-Digital conversion system is available in an off-line mode. It multiplexes up to 16 channels of analog magnetic tape input in a sequence determined by a programmable scanner, digitizes the samples at a rate up to 50,000 samples per second and to an accuracy of 11 bits plus sign, and produces a digital magnetic tape output.

Small Computer Applications Laboratory

The Small Computer Applications Laboratory was established to provide expertise for the development of complete hardware-software systems, particularly those systems associated with nuclear instrumentation and data reduction.

The laboratory has both general-purpose and specialized facilities for the application of small computer systems to a wide variety of research, development, instrumentation, and educational areas. It is equipped with four general purpose computers (a PDP-12, PDP-8/I, GT 42, and PDP-8) with various peripheral equipment specifically suited to small computer applications and system development. The 8K PDP-12 with its high speed disk storage, line-tape storage, and a graphics facility plays an important role in software systems development and data storage. In addition, the PDP-12 is equipped with multiple analog-to-digital converters, relays, a programmable KW-12A clock, and an I/O access panel to provide convenient development, fabrication, testing, and simulation of the interfacing of special equipment to the computer.

Library

The Price Gilbert Memorial Library is a centralized scientific, technical, and management collection of 730,000 volumes, plus 690,000

microtext and other bibliographic units. Outstanding collections in the fields of science and engineering have been developed to support graduate study and research. It is housed in adjoining structures totaling 240,000 square feet of space. It can house over one million volumes and can seat two thousand users. The library's United States Patent specification collection is the only one in the Southeastern area. In 1962, the library was designated one of twelve Federal Scientific Report Centers and its collection of reports from the Atomic Energy Commission, the Department of Defense, the National Aeronautics and Space Administration, the Clearinghouse for Scientific and Technical Information now totals over 500,000 titles. Extensive files of standards issued by American associations and societies and a complete file of U.S. Military Standards and Specifications and of British Standards are maintained. The library is also a depository for United States government publications issued by the Government Printing Office, and for maps issued by the Army Map Service. Available on microfilm are more than 6,000 company catalogs providing information on industrial components.

Other Facilities

Georgia Tech has many other facilities closely allied with the nuclear engineering program including, for example, various facilities operated by Divisions of the Engineering Experiment Station. Among these are specialized laboratories utilizing x-ray equipment, optical and electron microscopes, emission and absorption spectrometers, analog computers, microwave systems, cryostats and many other research devices. In addition, close cooperation exists between the School of Nuclear Engineering and other Schools of the Engineering College and the School of Physics.

DEVELOPMENT AND APPLICATION OF THE
ELECTROCHEMICAL TRACK ETCHING TECHNIQUE

by

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Progress Report

on

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Summary

Further studies are reported on the development and application of charged particle track amplification in polycarbonate foils by electrochemical etching for fast neutron and alpha particle dosimetry. The first results are reported in the effort to use these foils to determine the microdosimetry of ^{239}Pu in bone. The initial work has been involved with calibrating this dosimeter for alpha particle dosimetry; it has been calibrated for fast neutron dosimetry already. A device called the Vacuum-sealed alpha-calibrator was designed and built for this calibration. Dog bone samples from dogs with body burdens of $^{239}\text{Pu}(\text{IV})$ have been obtained from Dr. W. S. S. Jee at the University of Utah. Human bone samples of autopsied material from plutonium workers have been obtained from Dr. B. D. Brietenstein associated with the U. S. Transuranium Registry in Hanford. In addition, rat bone samples are being processed at Oak Ridge National Laboratory for use in this research. Initial results are presented also in the examination of proton track formation to determine the existence of these tracks and the threshold for their formation (one theory suggests proton recoils do not contribute significantly to tracks observed in these foils following neutron irradiation). Comments concerning the two groupings of tracks observed after alpha particle irradiation are offered.

The results obtainable using two foil reading techniques are presented. First the microfiche reader, providing a magnification of 50X yielded track diameters 2-4 mm and the transparency projector, providing a magnification equal to 14X at a screen to projector distance of 5 meters resulting in track diameters of 0.6 mm. The results of our annealing study to reduce the background and a study leading to the conclusion that the background seen

on these foils follows a Poisson distribution are given also. Results from the operation of our large chamber capable of etching 35 foils are reported. Two studies are presented concerning the system itself. The first involves the optimization procedure followed to obtain the best etching conditions for the foils presently in use; it indicates 800 volts, 2 KHz, 4 hours etching time and 45% KOH are reasonable for this chamber. The second involves an attempt to determine the "figure of merit" for the Lexan polycarbonate dosimetry system.

Introduction

This is a short-term progress report on further development and application of charged particle track formation in polymers such as polycarbonate amplified by electrochemical etching for fast neutron and alpha particle dosimetry. Briefly, the technique involves irradiation of a sensitive polymer by either charged particles, such as protons or alpha particles, or fast neutrons followed by an electrochemical etching treatment to reveal the induced recoil, (n, C), proton or alpha particle tracks. Electrochemical etching, proposed by Tommasino,¹ when applied to recoil particle tracks overcomes many shortcomings of conventional etching methods leading to a new avenue in fast neutron dosimetry (Sohrabi).^{2,3} Investigations of procedures to calibrate this electrochemical etching system for alpha particle microdosimetry for use in the osseous microdosimetry of ²³⁹Pu led to the design and construction of the vacuum-sealed alpha calibrator (Stillwagon, et al).^{4,5} Studies were conducted to examine any desirable effects obtained by annealing the foils as well as results expected from two foil reading techniques which we developed, Su, et al.⁶ Progress realized in bone microdosimetry studies and proton track formation studies are presented here. Past results concerning the fast neutron sensitivity and physiochemical characteristics of this approach are now available in the literature by Sohrabi and Morgan.⁷⁻¹¹ The results are all in favor of supporting and using this technique for a number of applications in health physics, radiotherapy and even radiography. Some of the results were reported in previous progress reports and other publications, some in this report and some will be studied during the remaining contract period.

Results of current studies as reported here are divided into ten categories:

1. To study the calibration of this dosimeter for alpha particles.
2. To indicate progress completed to date in the determination of microdosimetry of ^{239}Pu in bone.
3. To study proton track formation in polycarbonate.
4. To examine two foil reading techniques which we developed.
5. To investigate the effects of annealing these foils and foil selection with respect to the observed background.
6. To determine the optimal etching conditions for the current polycarbonate foil in use with a large chamber (35 foils).
7. To comment on results observed after alpha particle irradiation of these foils.
8. To verify the existence of a Poisson distribution of background tracks.
9. To define a figure of merit for the electrochemical system by examining the etching parameters.
10. To attempt to operate a large chamber capable of etching 35 foils.

Osseous Microdosimetry

The effects of alpha particle irradiation realized in tissue can be quite severe indeed, even at a fairly low, macroscopically calculated dose. Carcinoma involving epithelial cells (cells of the sinus cavity and surrounding bone), pulmonary neoplasia, fibrosis, necrosis, spontaneous bone fractures, tooth and tooth socket effects, osteomyelitis, osteoporosis, osteosarcoma, hemorrhage, edema and even leukemia are all possible results of alpha irradiation in vivo. Death can be caused by an average skeletal dose of only 60 rads (Mays and Lloyd)¹² in beagles when injected with ^{239}Pu citrate intravenously, of 90 rads in humans injected with a ^{224}Ra solution (Spiess and Mays);¹³ or by an average skeletal dose of 1200 rads in humans ingesting ^{226}Ra (Evans, et al).¹⁴ The primary concern here is the nuclide ^{239}Pu .

The public today is becoming increasingly disturbed over the nuclide ^{239}Pu which is utilized in the nuclear industry. ^{239}Pu is produced in great quantities in $^{235}\text{U} - ^{238}\text{U}$ fueled reactors so potential exposure to man will be present during the handling of spent fuel during reprocessing or prior to disposal. Also ^{239}Pu is produced for use in weapons and the production of these weapons presents a possible source of human exposure. Lastly, due to weapons tests in the atmosphere, approximately 300 KC of ^{239}Pu were distributed globally from megaton range tests in the early sixties, according to Eisenbud.¹⁵ Given its availability, the great toxicity exhibited by ^{239}Pu is probably one of the major reasons for interest in this nuclide. It is known from work by Jee, Thompson, Bair, Mays, and others^{12,16-40} that microgram quantities of ^{239}Pu administered to experimental animals by various methods are capable of producing lethality in these animals.

Plutonium, like all other actinide elements, radium and strontium, is basically a bone seeker. Although the distribution of ^{239}Pu found in the body is dynamic, at any one point in time it will be determined by such factors as route of entry into the body, chemical form and chemical state of the plutonium, species of the animal, its age and general state of health (Jee).¹⁷ When dealing with particulate ^{239}Pu , the particle size distribution and method of creating the particles is important (Bair).³⁰ Studies by Hamilton and Durbin⁴¹⁻⁴⁸ at the University of California and Jee, Mays and others^{17,18} at the University of Utah using the intravenous mode of administration have shown that a hydrolyzable, polymeric plutonium compound will be taken up preferentially by the liver but a monomeric plutonium compound will be preferentially concentrated by the bone. Unlike others, bone seekers such as radium and strontium, plutonium distributes in the bone in a highly uneven manner. Plutonium tends to concentrate on the surfaces of osseous tissue, Figure 1,⁴⁹ and therefore is called a surface seeker. The distribution is not homogeneous with respect to this surface distribution. Plutonium is retained preferentially on the endosteal with an endosteal to periosteal concentration equal to about 4 to 1 (Jee).¹⁷ Once deposited, the plutonium essentially remains in place until removed either by resorption or apposition. This surface deposition has been verified by detailed autoradiographs which show clearly this distribution, Figure 2.⁵⁰ The cells concerned with bone resorption, osteoclasts, progressively concentrate ^{239}Pu . This plutonium is later relocated to macrophages (as osteoclasts are removed) which travel throughout the marrow irradiating the marrow and the endosteal surface or the ^{239}Pu can be recycled into the blood (Durbin),⁴⁵ (Jee).¹⁷ The most sensitive gauge of ^{239}Pu hazard to mammals is the osteosarcoma or bone tumor. Only 3.6 rads delivered to the whole skeleton of a rat was sufficient

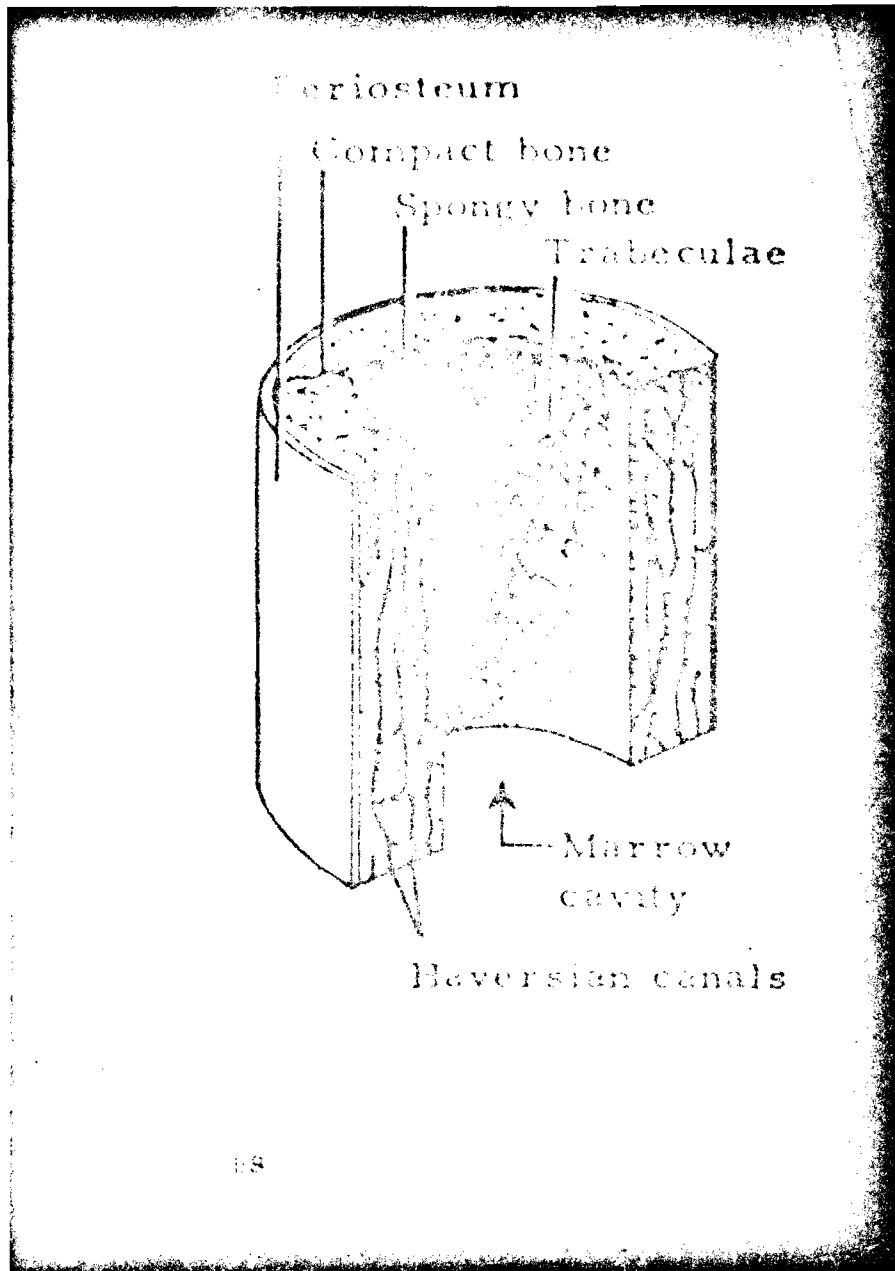


Figure 1. Cutaway of the shaft portion of bone showing the location of the periosteal and endosteal surfaces.

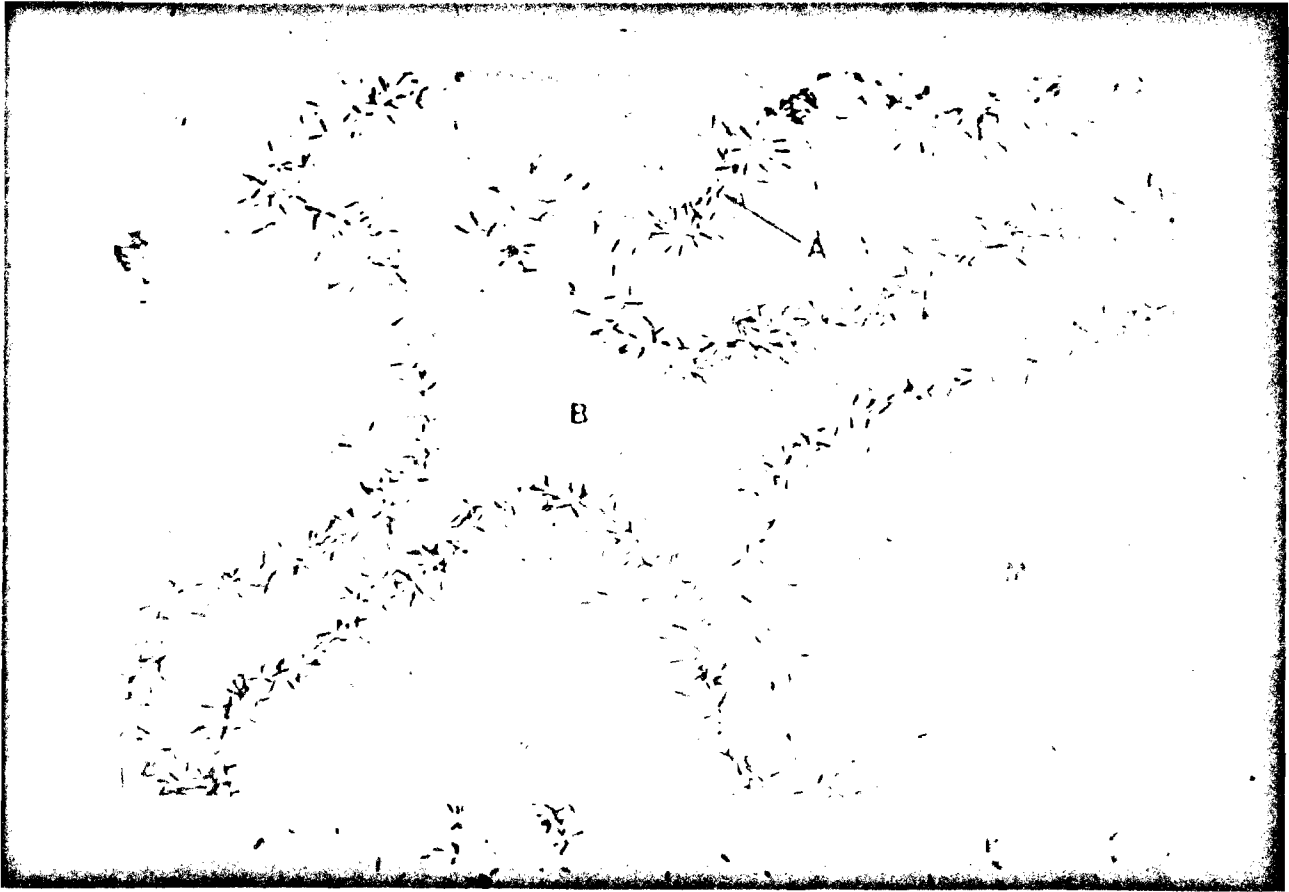


Figure 2. Detailed neutron-induced autoradiograph of portions of four trabeculae, showing fission fragment tracks distributed mostly upon surfaces in the lumbar vertebral body. Bones are from a dog injected with $0.0158\mu\text{C}$ of $^{239}\text{Pu}/\text{kg}$ Pu(IV) citrate and sacrificed five days later. B denotes bone, M denotes marrow and A denotes buried ^{239}Pu (x210).

to cause a tumor (Jee).¹⁷ The risk of tumor formation appears to be related to three factors: (1) the volume of cells at risk from the emitted alpha particle, which is determined by its range, (2) the amount of alpha irradiation impinging upon the bone surface, and (3) the proliferative potential of the cells under irradiation (ICRP).⁵¹

The ICRP recommends the limiting dose rate to the bone surfaces be set at 15 rem/yr out to a distance 10 μ m removed from these surfaces (ICRP).⁵¹ Unfortunately, however, the direct experimental data allowing this recommended limit to be implemented are not available. As a consequence, the old value of the maximum permissible organ burden (MPOB) for ²³⁹Pu is (based on an assumed uniform distribution of Pu in bone) still in use both nationally and internationally.

We are presently applying our dosimetry system developed by Sohrabi and Morgan⁷ to this microdosimetric method and the details of this usage for the Lexan polycarbonate system will be given in the next section. Here, let us mention, we have obtained from Dr. W. S. S. Jee samples of bones from dogs injected intravenously with ²³⁹Pu (IV) citrate at the University of Utah, human bone samples from plutonium workers from Dr. B. D. Brietenstein associated with the U. S. Transuranium Registry at Hanford and rat bone samples are being processed at Oak Ridge National Laboratory for use in this research. It is hoped the results of these studies will provide data which will make it possible to calculate the MPOB in accordance with the recommendation of ICRP and these data may be useful in leading to the establishment of a new and more reliable MPOB in bone for ²³⁹Pu.

Alpha Particle Calibration Studies

The alpha particle is emitted in natural radioactive decay by numerous nuclides including the one of interest here, ^{239}Pu . Physically, the alpha particle is a helium nucleus, ${}^4_2\text{He}^{++}$, and as such is considered to be densely ionizing as it passes through an absorbing medium. The ionization produced by an alpha particle is confined to a region very close to the track of the particle (Vaughan, et al).⁴⁰ In fact, ninety percent of the ionization is contained in a cylinder of radius $0.01\mu\text{m}$ whose axis lies along the track. From the previous section we know that the critical organ for ^{239}Pu is bone. Therefore, when concerned with the dosimetry of ^{239}Pu , one would be concerned with bone dosimetry. Due to the inhomogeneous distribution of ^{239}Pu in bone, we are actually concerned with its microdosimetry. The microdosimetry of ^{239}Pu as it resides in osseous tissue has proven to be a difficult problem to solve for a variety of reasons. Some of the difficulty is caused by the concentration of ^{239}Pu upon the bone surfaces, especially the endosteal surface, where the most proliferative cells are found, as indicated in the previous section. It is known that the endosteal surface to periosteal surface concentration is about 4 to 1 (Jee).¹⁷ Since ^{239}Pu is an alpha particle emitter, maximum damage can be inflicted upon bone as a result of this endosteal surface concentration from the short ranged and densely ionizing alpha particle. To date, the maximum permissible organ burden for ^{239}Pu in bone is still based upon its effect in animals relative to that of ^{226}Ra and relative to a uniform distribution in bone instead of being based upon a separate set of experimental data. In fact, there has been little experimental data which would allow the MPOB for ^{239}Pu in bone to be refined in some fashion. This situation has caused the present MPOB in

bone, currently $0.04\mu\text{C}$, to come under fire in recent years by scientists such as Edsall⁽⁵²⁾ and Morgan⁽⁵³⁾ who would like to see the MPOB for ^{239}Pu in bone have its own base in experimental data.

When looking at the various microdosimetric techniques applied to alpha emitters encased in bone, one finds there are basically three methods which have been used over the years to obtain an estimate of the spatial distribution of the dose delivered to the endosteal face of bone. These methods are similar in that they employ various photographic emulsions to determine the spatial distribution of the deposited activity in bone. After the exposure of the emulsion to ^{239}Pu contained in situ in bone has been completed, different techniques are used to find the activity or fluence at various locations. These measurements are then related to dose by several of the available mathematical formulations derived by Spiers,⁵⁴ Kononenko⁵⁵ and others. Rowland and Marshall⁵⁶ used direct counting of the tracks at various locations on the emulsion to find activity. Twente and Jee¹⁶ used densitometric techniques to determine activity, thus avoiding the difficulties associated with direct counting but requiring a calibration procedure using a standard source. A final method, used by James and Kember,⁵⁷ involved the use of a microscope equipped with an eyepiece marked with small five micron diameter circles. Knowing the depth of the emulsion and the circle diameter, fluence could be found if this cylindrical volume was related to an equivalent spherical volume.

There are several characteristics we would consider desirable for any new osteogenic cell microdosimeter; we would like it to:

1. allow the desired spatial distribution of the dose to be determined.
2. be sensitive down to the low dose expected to be encountered at the MPOB.

3. be tissue equivalent so rems can be read directly from the dosimeter.
4. resist fading so that rechecking of results at various intervals would be possible.
5. operate without the use of residual or induced activity to prevent radiation hazards and avoid restrictions to use of systems containing ^{237}Np or ^{239}Pu .
6. be economical and simple to use.
7. operate without an excessively complex procedure involved to obtain data, thereby expediting reproducibility.
8. have a small background correction.

The photographic emulsion techniques fail on several of these points. They have a low sensitivity (rad range), are not tissue equivalent, suffer serious fading and can require extremely long exposure periods - ranging into the months. It is felt new possibilities are offered by the dosimeter developed here at Georgia Tech by Sohrabi and Morgan⁷ and described in detail by Sohrabi.⁸ In the present system, 250 μm Lexan polycarbonate foils serve as dosimeters receiving damage by the ^{239}Pu alpha particles. These foils will then be etched using an electrochemical etching technique first proposed by Tomissino¹ and developed to its present form by Sohrabi and Morgan.⁷ Figure 3 shows a typical etching chamber and Figure 4 shows the chamber connected to our electrochemical etching apparatus. These foils offer several advantages over the previous methods of osseous microdosimetry. They are tissue equivalent and have been shown to follow the ICRP rem response curve for fast neutrons. Also, these polycarbonate foils resist fading, are sensitive down to the few mrad range, operate at low background, they are easy and economical to use and have been shown capable of resolving information at the distances desired by Beach and Becker.⁵⁸ Although calibrated for fast neutron dosimetry, the foils have not been calibrated for alpha



Figure 3. A Plexiglass etching chamber.

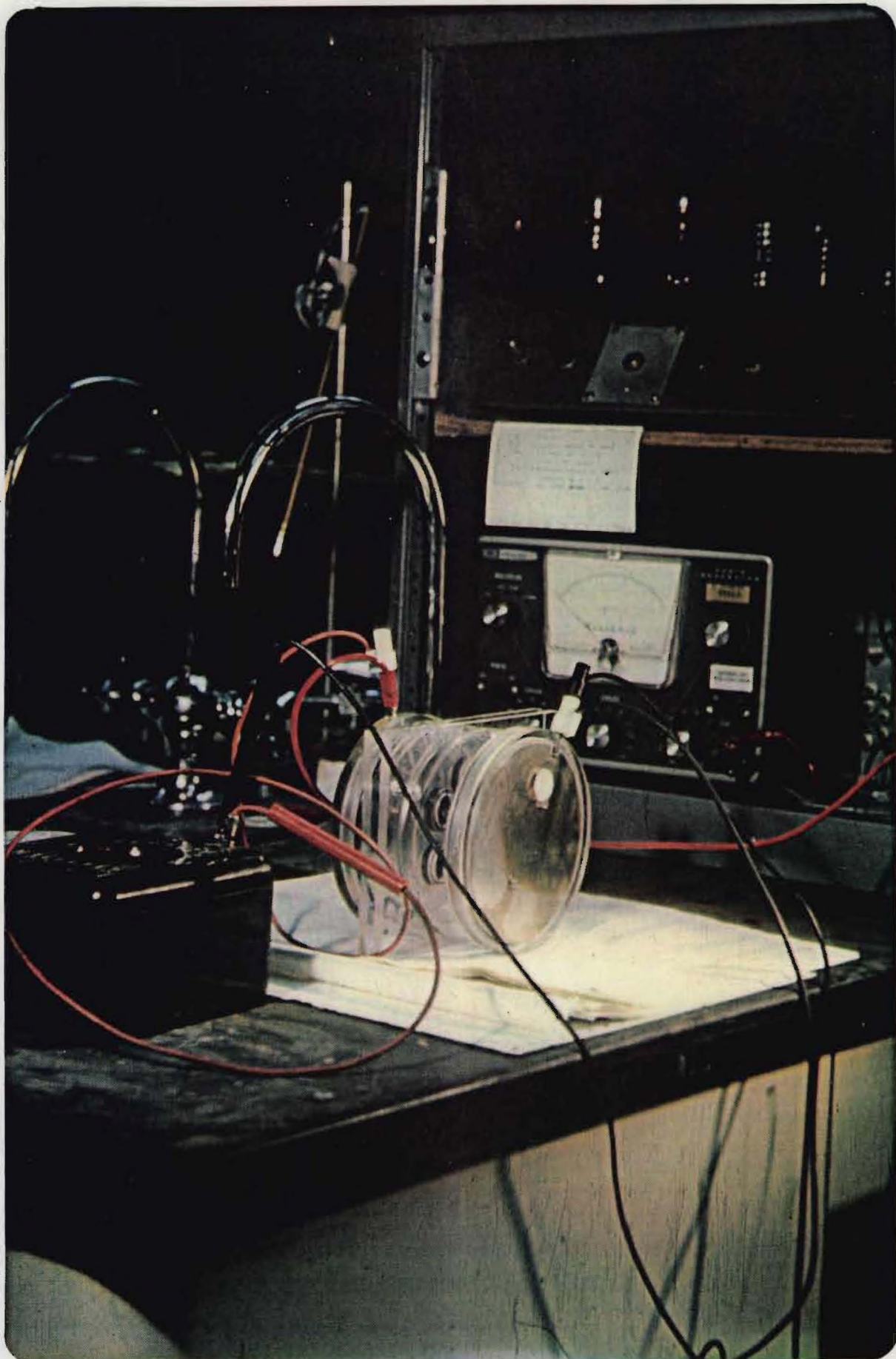


Figure 4. Electrochemical etching apparatus showing etching chamber, audio generator, amplifier and volt meter.

particle dosimetry. This calibration is one subject of concern in this research; that is, to find a factor to relate numbers of tracks appearing on the foils to dose equivalent and absorbed dose.

Determination of the Alpha Calibration Factor

Basically, the problem is being approached in the following manner. An NBS ^{239}Pu standard solution was obtained and evaporated onto a planchet. The dose at various heights above the source was found by observing the number of counts received by several surface barrier detectors providing input into a multichannel analyzer and then converting to dose using a small computer program written for this purpose. The centerpiece of the experiment is a device designed by this research called the Vacuum-sealed alpha calibrator which will handle a rather loosely mounted source requiring a vacuum in order to facilitate counting, Figure 5. Here a surface barrier detector is shown as the detector but a separate top is also used which holds the polycarbonate foils. The entire experimental setup is shown on Figure 6. The vacuum gauge was used to maintain a continuous record of the vacuum. The vacuum varied between 701 and 711 mm of mercury.

Initially, it was desired to know how the alpha calibrator would compare with other, more widely used, surface barrier detector systems. Here a Whitey system was selected for this comparison. When equal source to detector distances and counting times were used to count an ^{241}Am commercially available check source (0.1 μC), the Whitey system gave a full width at half maximum (FWHM) of 1.33% and the alpha calibrator 1.23%. Later this latter figure was lowered to 1.13% with additional practice. Ortec surface barrier detectors were used throughout this part of the work. After counting the ^{239}Pu source at various heights above the source with the surface barrier

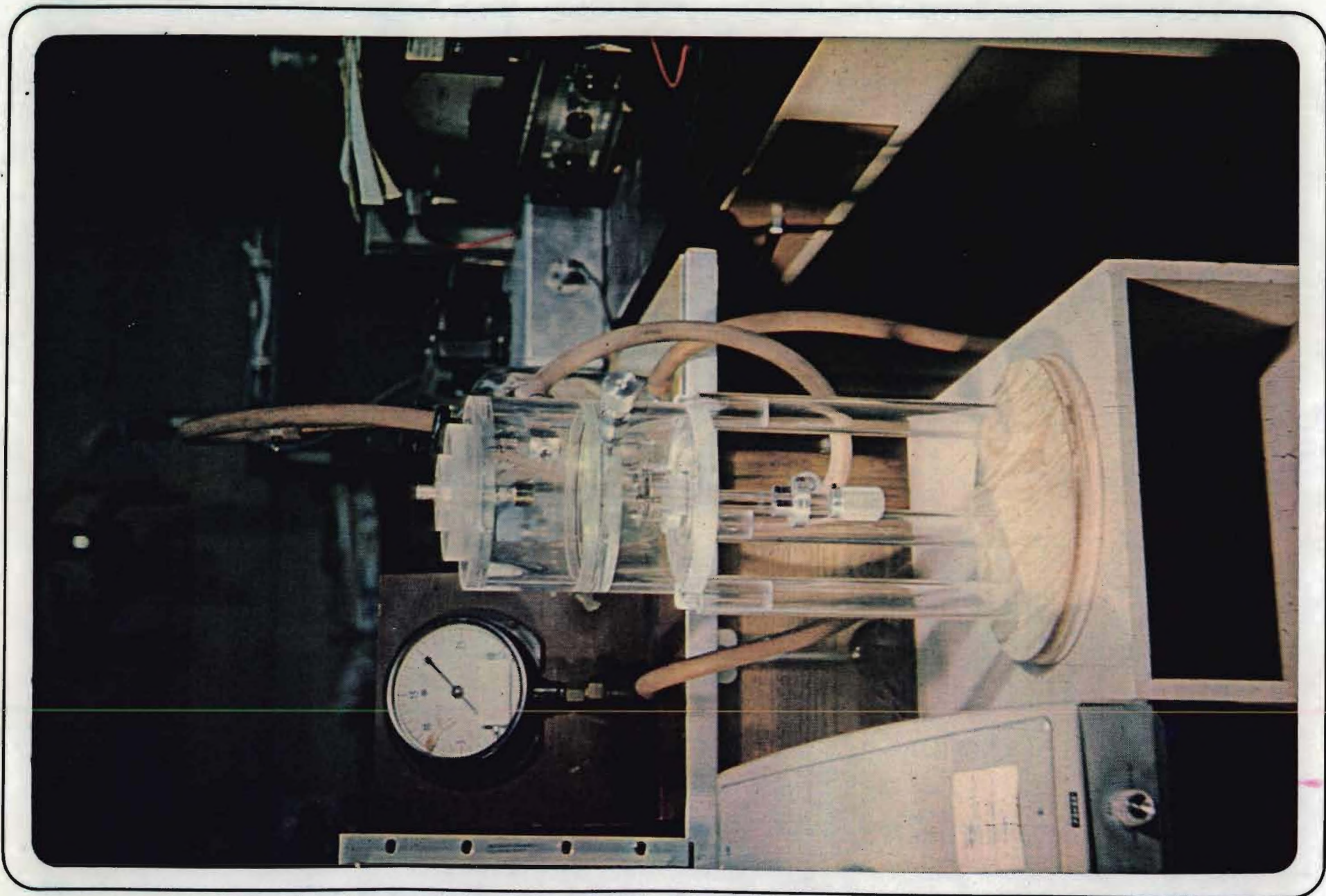


Figure 5. Closeup of the vacuum-sealed alpha calibrator showing the vacuum hookup, two compartment design and the calibrated source stand.

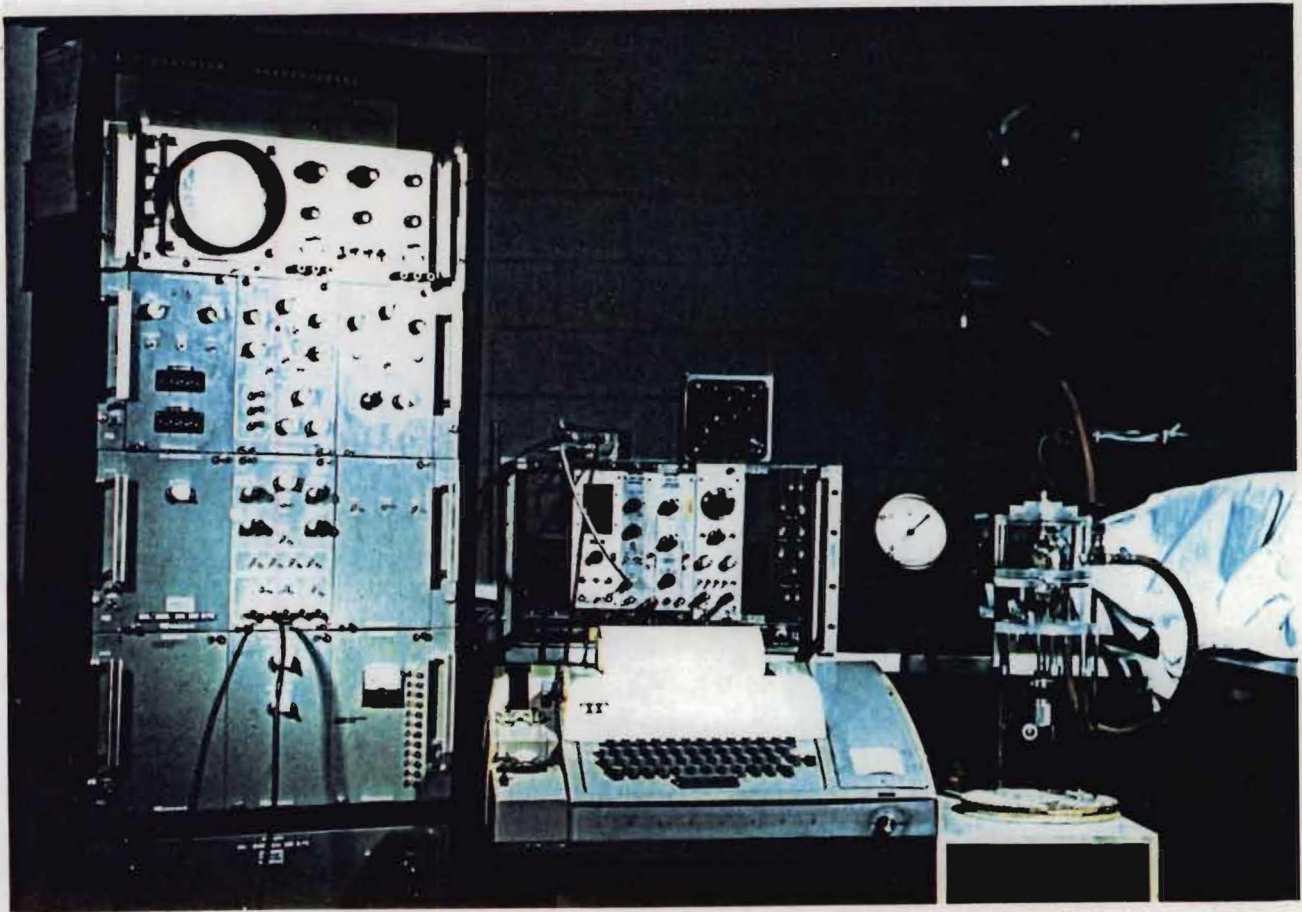


Figure 6. The experimental setup is shown above. Visible is the ND2200-MCA counting electronics, vacuum gauge, particulate trap, teletype and the Vacuum-sealed alpha calibrator.

detector, it was required to duplicate these conditions using the polycarbonate foils. A donut-shaped ring was constructed to match the dimensions of the portion of the surface barrier detector in front of the sensitive area. Now the foils could be irradiated at the same distances from the source and a reasonable comparison between the foils and surface barrier detector was made. A large Princeton Gamma-Tech detector was also used. Here the diameter was large enough for the foils to fit directly over the exposed sensitive area allowing very similar irradiation conditions to be effected between this detector and the foils.

The foils along with some background controls were etched utilizing the chamber and setup previously described. The initial results appear to indicate an alpha calibration factor of about 10 mrad/track using data from the small detector (10 mm diameter) and some data from the large detector (22 mm diameter).

To refine this number, ceramic rings have been obtained from the Ortec Company which are the ones actually used during construction of the surface barrier detector used by the work here. These will enable the geometries and physical conditions existing between the surface barrier detector countings and the foil irradiations to more closely approximate each other. Also, we are examining the possibility of using a grid system to simulate an inhomogeneous distribution of the alpha emitter during calibration.

Proton Irradiations

In order to study the possibility of proton responses in Lexan polycarbonate, 250 μ m polycarbonate foils were irradiated with different energies and doses of protons by the accelerator at the Department of Physics and

Astronomy, University of Florida, Gainesville, Florida and the Georgia Tech Ion Implanter at the School of Physics. The energies and fluences range are given in Table I.

The Georgia Tech Ion Implanter is a type 200 MPR constructed by Accelerators, Inc. of Austin, Texas. It can be operated within a usable range from about 50 to 200 KV; the beam current was set to be 2×10^{-9} amperes. The irradiated times were 0.516, 2.58, and 5.16 seconds for 10^9 , 5×10^9 and 10^{10} protons/cm² respectively. The irradiated foils were one inch squares.

The proton accelerator at the University of Florida is a 3 MeV Van de Graff accelerator. The protons were given energies equal to 1 MeV, 2 MeV and 3 MeV. The beam current was around 10^{-9} amperes. The irradiation times were 10 seconds for each irradiation. The irradiated foils were about 0.35 inches in diameter.

In our preliminary results, no proton tracks were observed in foils irradiated at 200 KeV and below. So there exists a threshold energy greater than 200 KeV for proton track registration in polycarbonate foils. Varnagy et al. found that cellulose acetate (Cellit) was suitable for detecting protons with a threshold of about 300 KeV.⁵⁹

For highest proton energy irradiated foils, we did not peel off the mask covering the foils. The range of 1 MeV protons in a polycarbonate foil is only 33 μm ; it is smaller, therefore, than the thickness of the mask (38 μm). So no tracks were found in foils for 1 MeV proton irradiations. There were clusters of tiny tracks observed in the 2 MeV and 3 MeV proton irradiated foils after they were electrochemically etched under 45 percent KOH solution at 25°C applying 800 V at 2 KHz for four hours (see figure 7).

Table I. The Energies and Fluences of Proton Used During the Polycarbonate Foil Irradiations

| <u>Proton Energy</u> | <u>Fluence (protons/cm²)</u> | <u>Irradiated place</u> |
|----------------------|---|-------------------------|
| 50 KeV | $10^9, 5 \times 10^9, 10^{10}$ | Georgia Tech |
| 100 KeV | $10^9, 5 \times 10^9, 10^{10}$ | Georgia Tech |
| 200 KeV | $10^9, 5 \times 10^9, 10^{10}$ | Georgia Tech |
| 1 MeV | 1.16×10^{10} | U. of Florida |
| 2 MeV | 8.56×10^{10} | U. of Florida |
| 3 MeV | 9.06×10^{10} | U. of Florida |

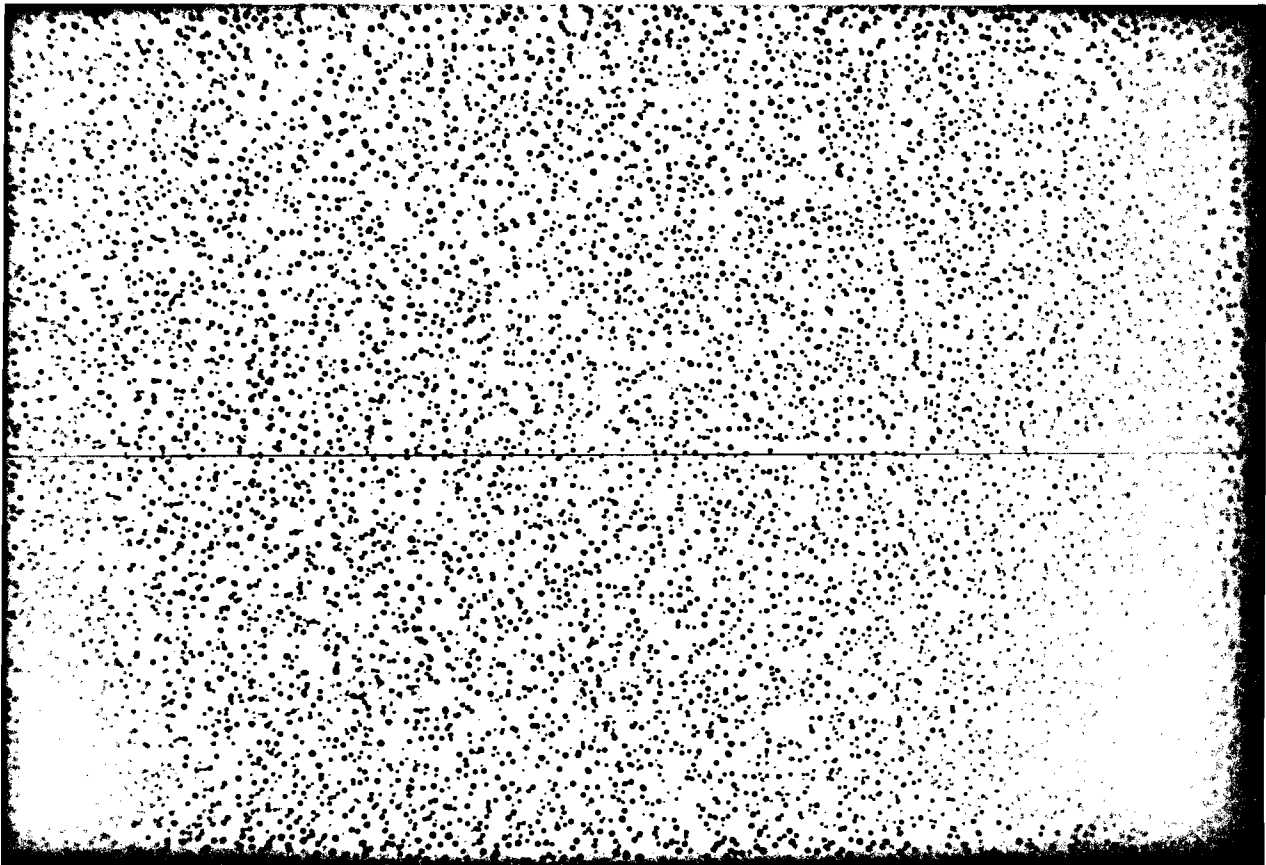


Figure 7. Tracks created by 2 & 3 MeV protons and etched in 45% KOH for 4 hours using 800 volts applied at 2 KHz.

The tracks were found only on the side of the foil facing the proton beam. Additional quantitative proton studies need to be performed.

Counting Techniques

Two new techniques were developed to facilitate counting of the polycarbonate foils. Both techniques employ optical devices almost every laboratory and university possess in readily obtainable supply --- the microfiche reader and the overhead (transparency) projector. These two techniques offer several advantages over the microscope for low count density and thick foils (Tripier, et al).⁶⁰ (Spark counting requires very thin foils (Johnson and Becker)⁶¹ and techniques such as densitometry measurements require a large number of counts - high dose). Advantages effected during utilization of these two techniques over our earlier conventional microscope counting are:

1. A larger foil area is actually counted; the overhead projector providing greatest viewing area, microfiche reader second, and microscope third.
2. Image is reviewed on a screen and not through an eyepiece causing less eye strain.
3. Less time is required per foil for counting.
4. No elaborate equipment is required other than these inexpensive pieces of equipment most institutions possess anyway.

To facilitate counting with the transparency projector a grid was constructed on the screen upon which the image was projected; Figure 8. The grid consisted of two concentric circles of radius 4.3 and 7.0 cm on the screen which were cut into sectors with radii drawn every 20° (18 total). These yielded 36 small areas which could be counted easily, their sum

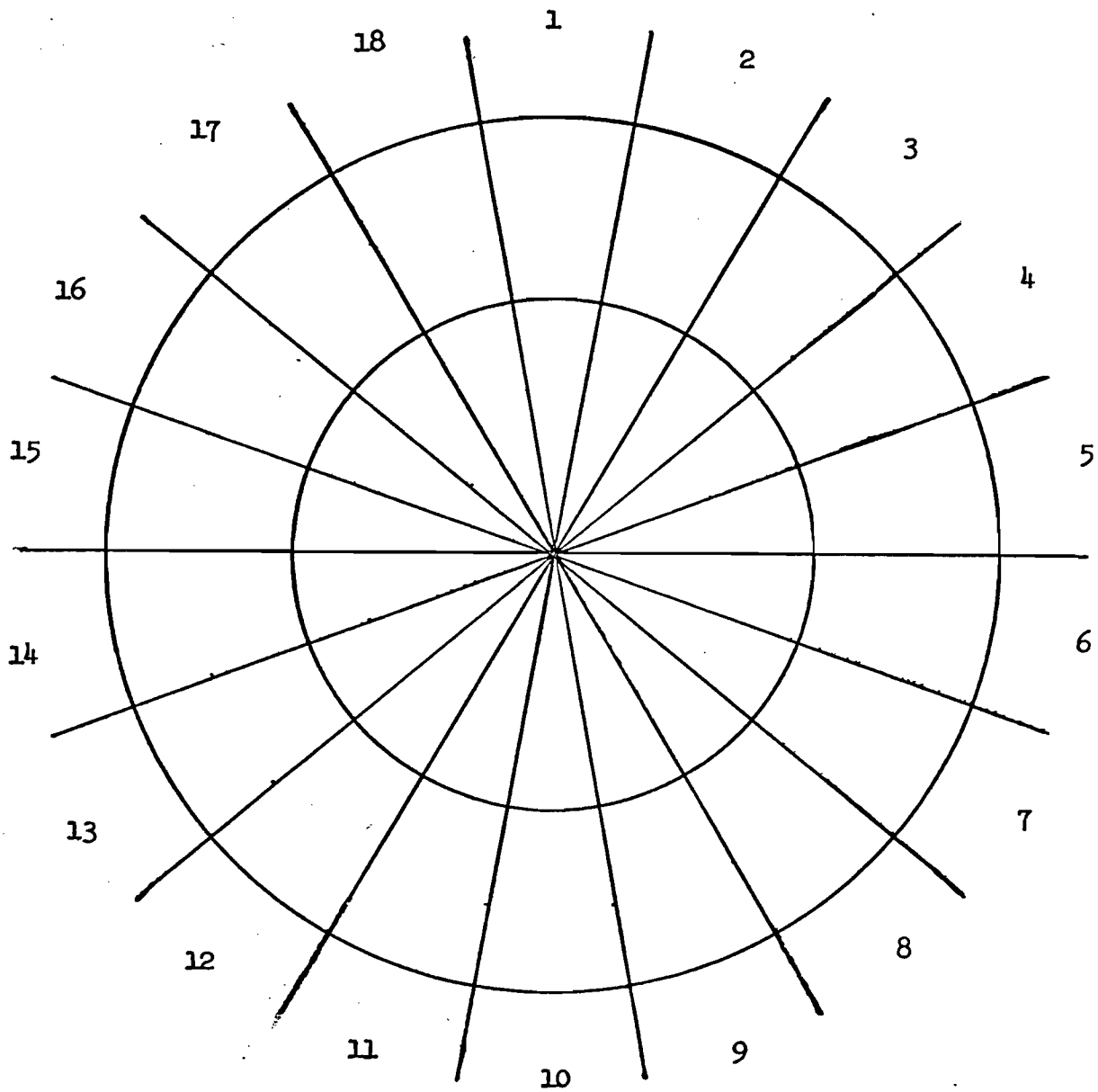


Figure 8. GRID USED IN FOIL COUNTING

equalling the total number of counts contained within the enclosed area, 154 cm^2 . The counts appear on the screen as black dots on a clear background so the viewer encounters little difficulty in recognizing them.

Figure 9 is a photograph of the screen of the overhead projector with the image of a foil containing high background projected upon it. The number of counts obtained for this foil within the encircled area was 685 counts. This number was obtained by counting the tracks within the 154 cm^2 circular area; then making a random movement of the foil (so a different area of the foil was superimposed over the grid), recounting the number of tracks and finally, calculating a numerical average of these two trials. Foil magnification was 14X with a screen to projector distance of 16 ft. 5 in. and track diameters were about 0.6 mm. Of course, almost any magnification could be obtained depending on the distance of separation of the projector and screen.

The second technique developed - the microfiche reader - offers other possibilities. Figure 10 is a photograph of a small rectangular portion of the microfiche reader screen. The actual area of the screen photographed was 17 by 13 cm. The horizontal line traversing the upper section of the photograph is a component of the "cross-hair" placed upon the microfiche reader screen to partition the screen into quarters and thereby facilitate counting. Foil magnification here was 50X and track diameters were in the range of 2-4 mm. The foil shown yielded 142 counts. This value was obtained by counting one quarter of the screen, then moving the foil in a random fashion and recounting the same screen quarter (but now a different

Figure 9. Portion of Transparency Projector Screen Showing High Background Foil

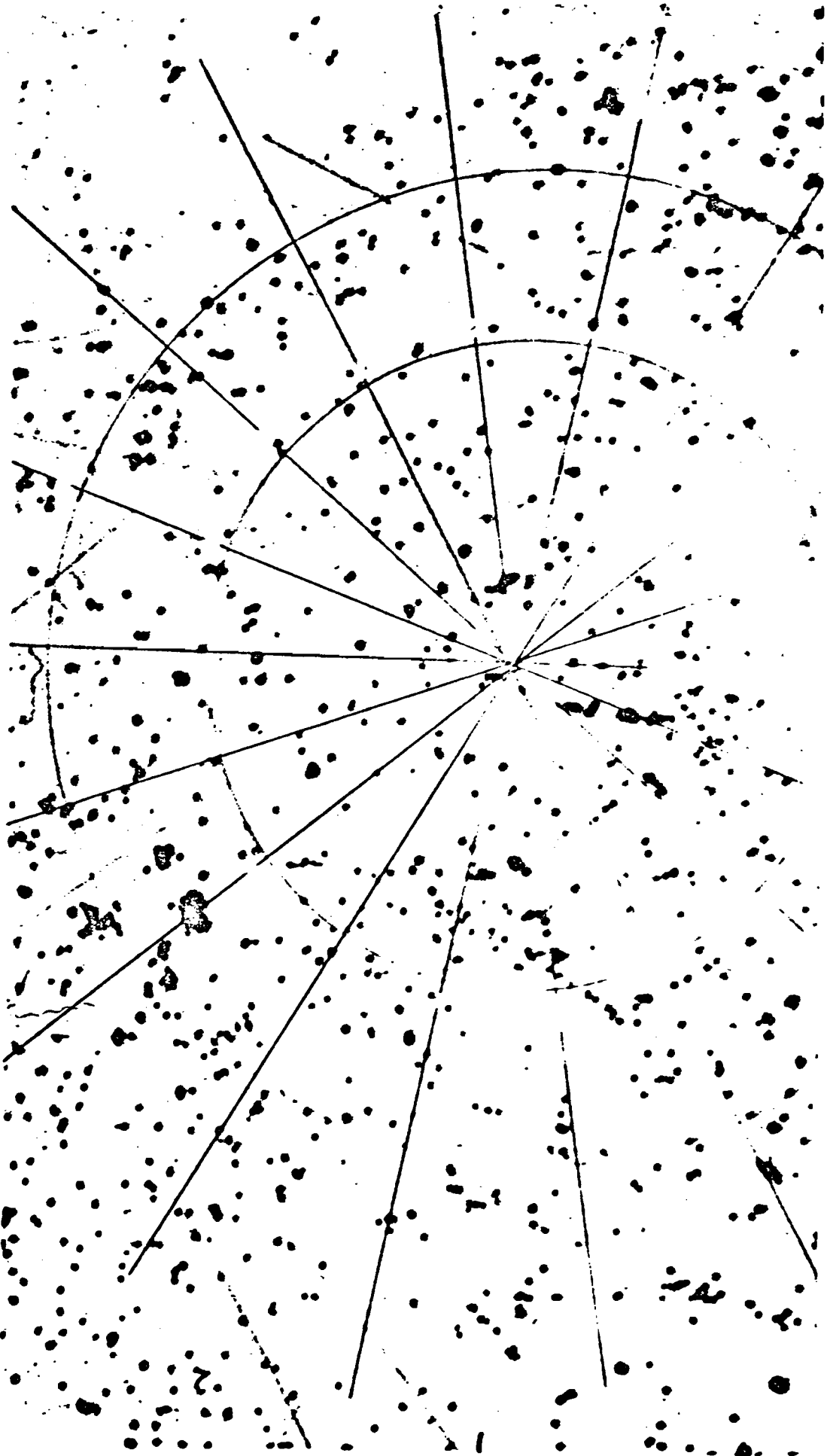
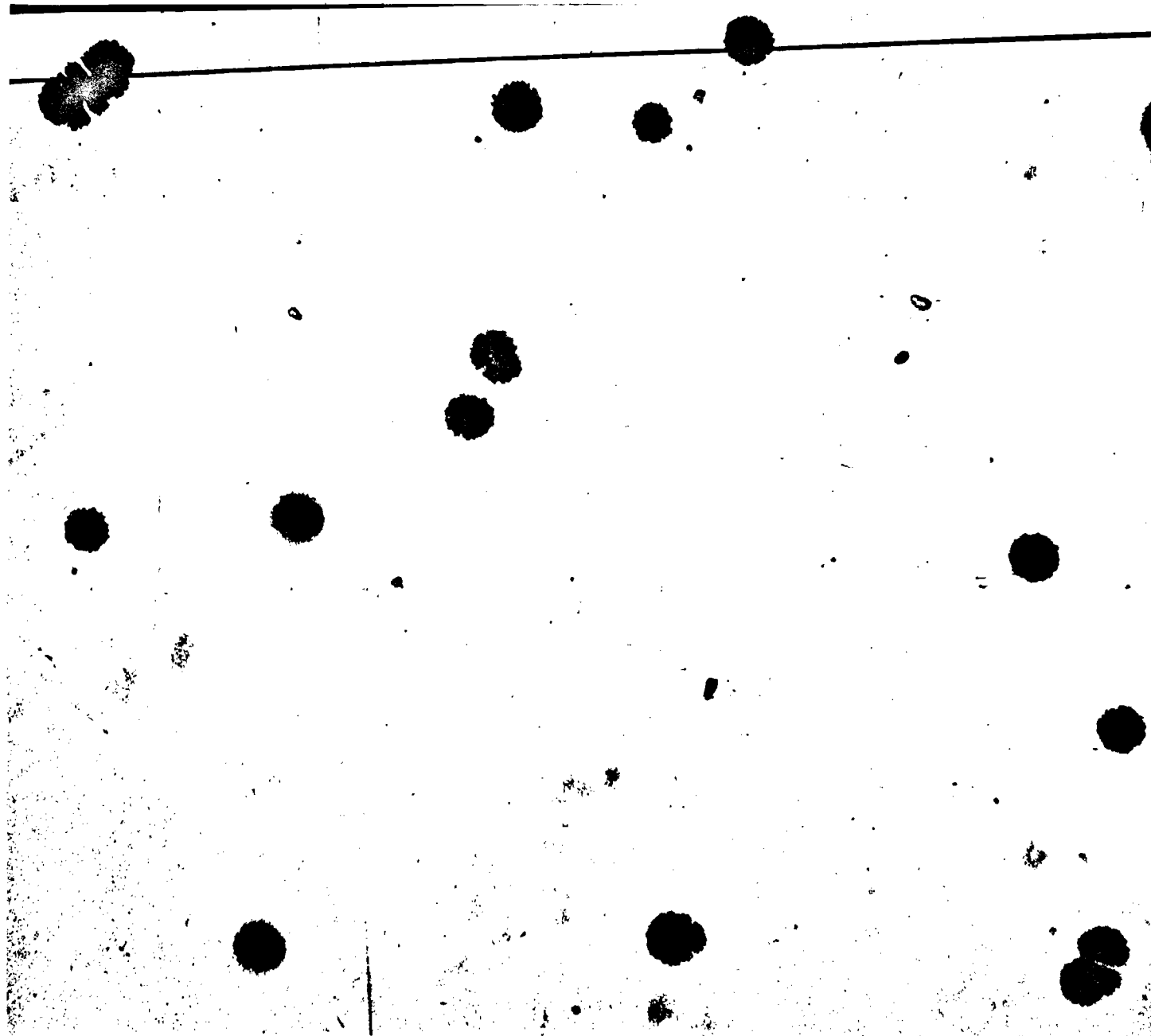


Figure 10. Photograph of Transparency Projector Screen Showing High Background Foil



portion of the foil would be projected beneath the cross-hairs). This procedure was repeated until four quarters were counted at which time a sum was calculated.

Since the magnification was 50X with the microfiche reader, the tracks were viewed in greater detail than when viewed with the transparency projector. The star-like outcropping from individual tracks became visible, similar to the appearance of the tracks seen by the microscope. Also, not unlike the case with a microscope, the focus on the microfiche reader can be adjusted so the plane of focus includes only tracks in one plane or on one side of the foil or the other. The tracks on the side opposite the side being visualized appeared as unfocused blurs. This would explain why some tracks were viewed with distinction and others were out of focus in Figure 10. A smaller area of the total foil was visible on the microfiche reader than on the transparency projector screen but the tracks were more readily distinguishable from other stray marks on the foil.

It is felt these two techniques will speed up counting and greatly enhance the efficiency of eliciting data from the Lexan foils.

Background Track Density Reduction

Presence of a high background can sometimes be a problem with polycarbonate foils. This problem is thought to occur as a result of two factors. Although the duration of a particular irradiation measurement may be fairly short, the foil has been accumulating etchable background from both background radiation (cosmic rays, trace uranium, etc.) and surface abrasions since its production in a factory. Therefore, interest

exists to maintain this background as low as possible. We have tried several methods, including annealing, washing with isopropyl alcohol and selection of a foil with an inherently low background.

In our annealing experiments, six groups of foils, seven to a group, were heated to various temperatures for twenty minutes. Then one foil from each group was placed in the seven foil chamber along with a control which had not been annealed. Etching conditions used throughout this study were

1. Etching solution - 30% KOH
2. Voltage - 700 Volts
3. Frequency - 2 KHz
4. Etching time - 4 hours.

It was found a significant reduction in background and increase in overall track homogeneity resulted if the foils were washed with isopropyl alcohol and rinsed with distilled water prior to annealing. The six temperatures used to anneal the foils were 60⁰, 105⁰, 130⁰, 140⁰, 155⁰ and 170⁰C. The foils were counted with the microfiche reader and transparency projector. Finally, the data were arranged so that the counts for each temperature and control could be examined for the seven runs and an average value calculated and plotted, Figure 11. The background in reference to controls dropped to 69 percent at 155⁰ and decreased rapidly until it was reduced to 54 percent at 170⁰. Other foil types were then used to examine the effect observed on the operational background. The masked foil, 250 μ , marketed by Transilwrap, Atlanta was utilized in an experiment involving 143 background foils plus controls. These foils were etched at 800 volts for 5 hours in a 45 percent KOH solution. This resulted in a background of 2.2 tracks/cm². Therefore it was decided to use this foil exclusively

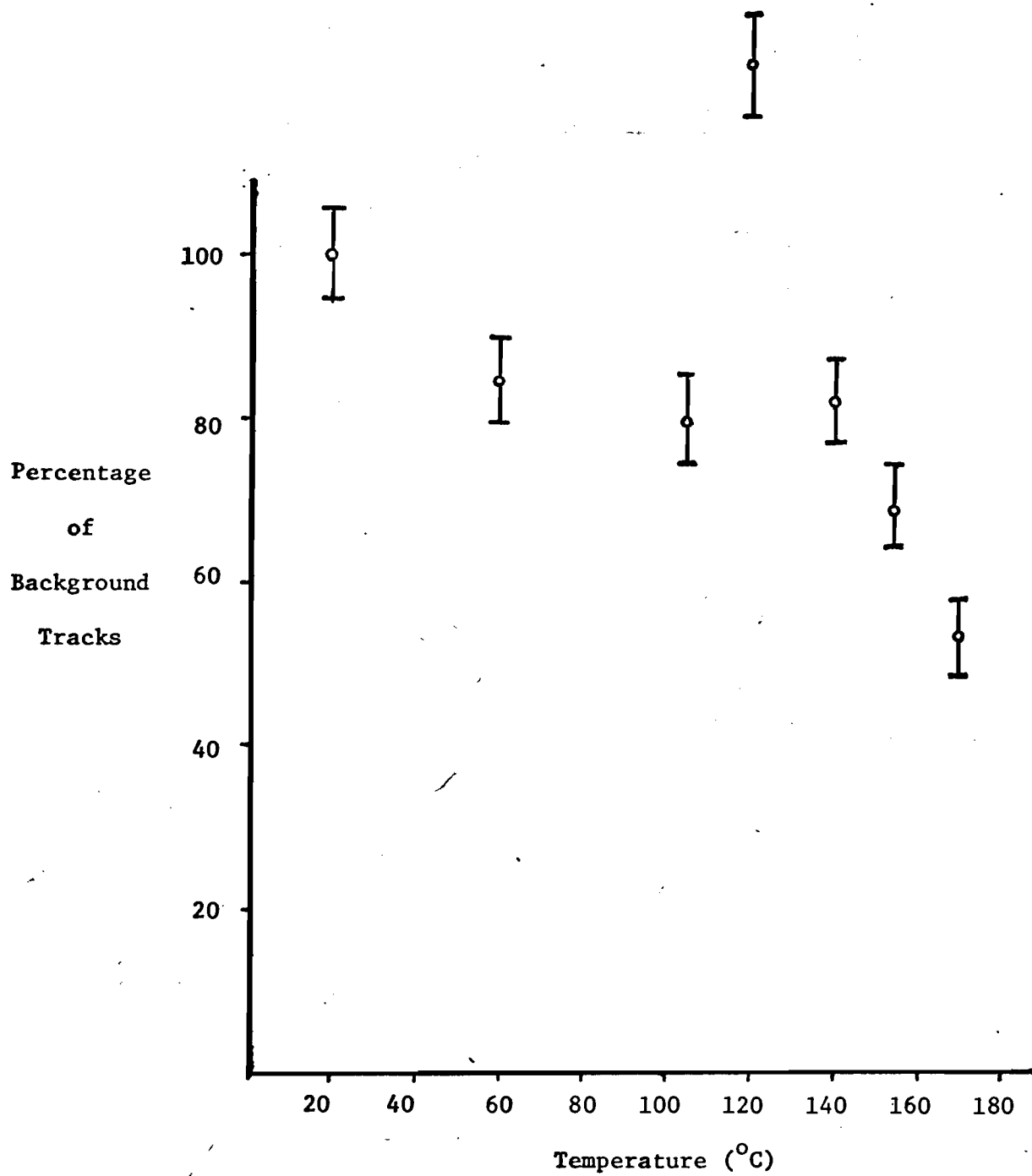


Figure 11. Percentage of Background Tracks After Annealing for 20 Minutes at Different Temperatures before Etching

avoiding any pretreatment of the foils, in agreement with earlier work by Sohrabi.⁸

Poisson Distribution

Visual counting techniques in general, including the aforementioned two, suffer due to the presence of the human element which can introduce a significant systematic error affecting the accuracy of the final result. For precision work, a great deal of effort must be expended during track counting to insure the elimination of any personal bias which might exist. One assumption applied in this type of counting is the existence of a Poisson distribution describing the counts (Price).⁶² This assumption was experimentally checked with data obtained from a series of measurements of one of the foils. For this measurement, the tracks were randomly scanned and counted over an area equal to one quarter of the microfiche reader screen. One hundred such random scans and countings were made producing a total of 3514 tracks, yielding a mean value of 35.14 tracks per scan. The frequency histogram, representative of these data is shown on Figure 12. The equivalent Poisson distribution is also shown on this figure and is

$$P_n = 500 \frac{m^n e^{-m}}{n!}$$

The normalization constant, 500, is obtained by multiplying five times the number of trials (100) and m is the mean value of the data. The standard deviation can be obtained directly from the histogram and is about 7, and the value obtained for the standard deviation directly from the Poisson is $(m)^{\frac{1}{2}} = (35.14)^{\frac{1}{2}} \approx 6$. Since these two values are so close we can conclude the Poisson distribution assumption employed in the present counting experiments is valid.

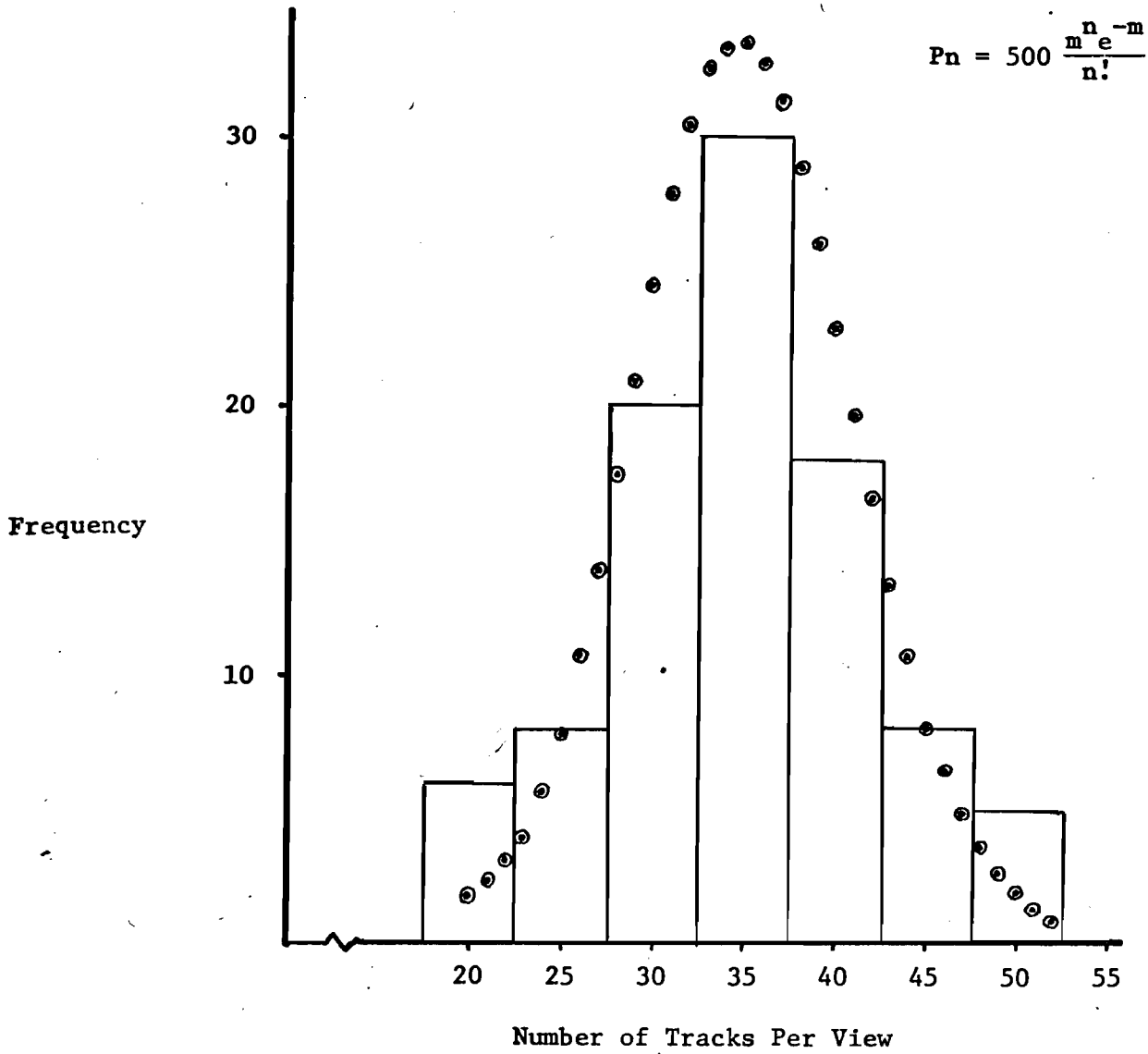
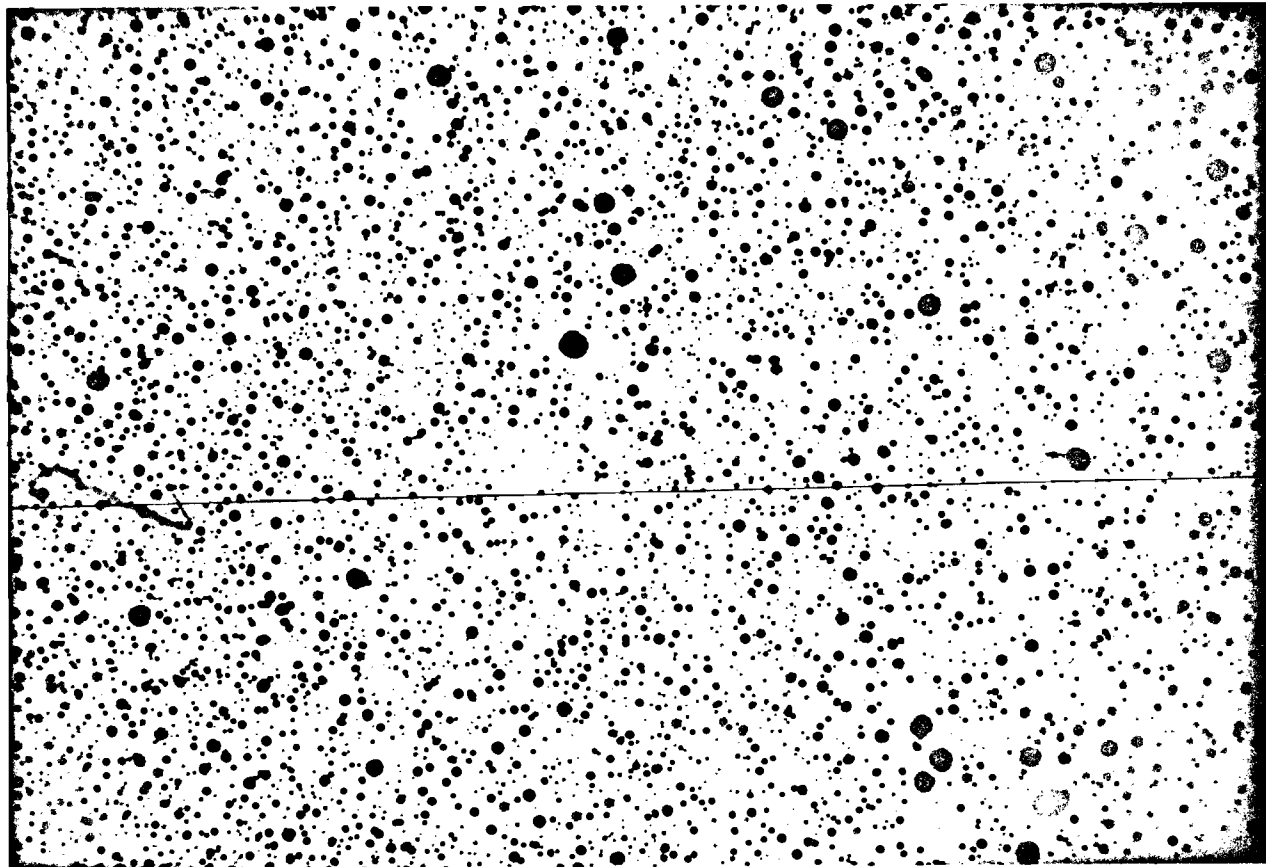


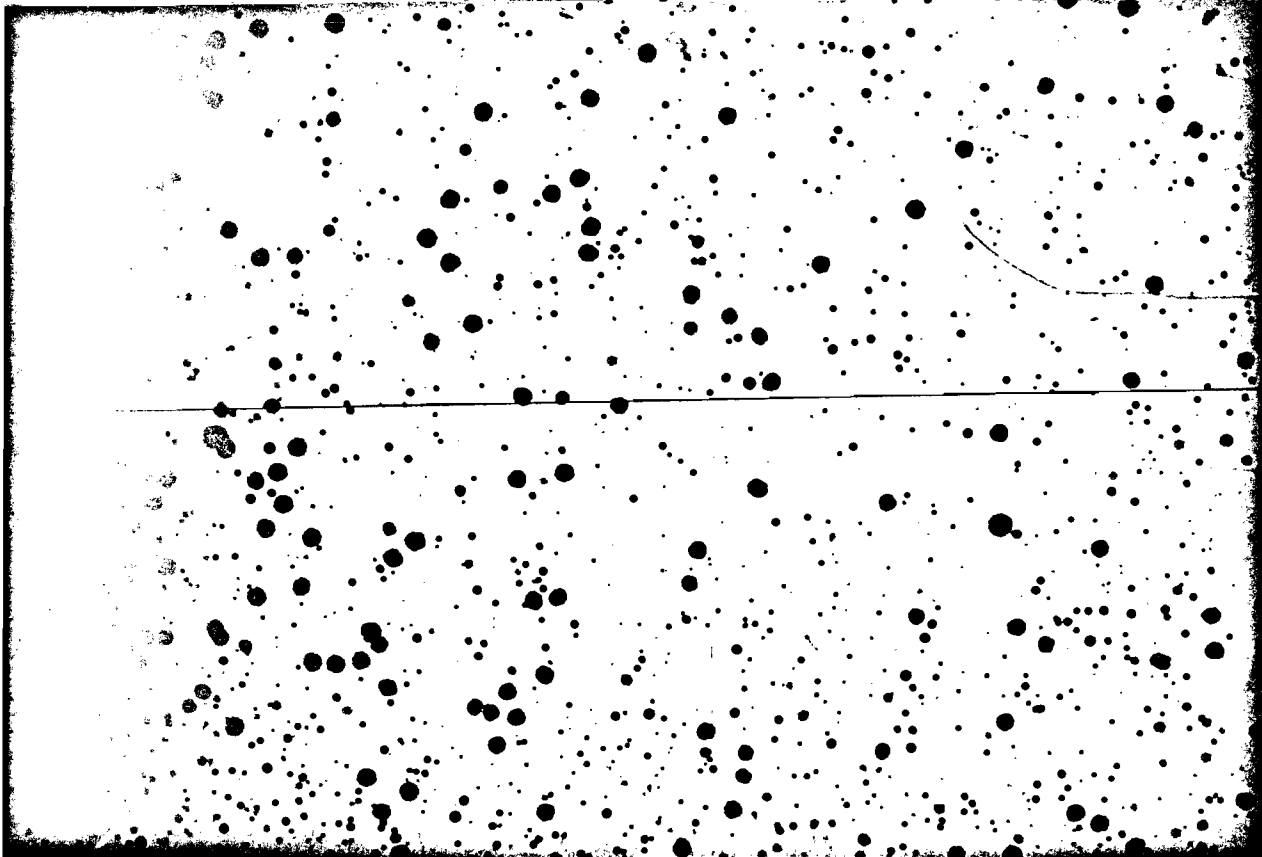
Figure 12. Comparison of the Frequency Histogram of Observed Number of Tracks Per View with the Poisson Distribution P_n Corresponding to the Observed Mean Value ($m = 35.14$)

Alpha Particle Irradiations

Before attempting the calibration of the polycarbonate dosimeter for alpha particles, it was desired to observe the results of direct alpha particle bombardment followed by the electrochemical etching process on these foils. Several foils were irradiated for fourteen (14) hours with a $0.1\mu\text{C}$ ^{241}Am source ($E_{\alpha} = 5.49 \text{ MeV} - 85\%$ and $5.44 \text{ MeV} - 13\%$) at a source to dosimeter distance of 2 mm (in air). Foils were irradiated both with and without the masking in place. After irradiation the foils were etched for 5 hours at 800 volts and 2 KHz in 45 percent KOH as the etchant. These foils were then read on the microfiche reader and an interesting result was noted. The track diameters fell into two rather distinct groupings, Figure 13. The difference in the diameter was more apparent in the foils irradiated with the masking in place - 5.8 mm average track diameter for the wider tracks and 1.5 mm average track diameter for the narrow track group - but the number of tracks within each group was greater for the narrow track group. The average track diameters for the alpha irradiations without foil masking were 5.7 mm and 1.9 mm for the wide and narrow track groups, respectively. These track diameters were measured directly from the microfiche reader screen and would need to be divided by a factor of 50 (the reader magnification) to obtain the actual foil track diameter. Also the actual track density appeared to be greater for the foil irradiated with the masking in place. These effects were explained in the following manner. We know from earlier work (Fleischer, et al)⁶³ alpha particles are capable of producing tracks in polycarbonate as a result of their own ionization. We know also, from the work of Sohrabi,⁸ collisions between incident particle and the nuclei composing the foils are possible which result in tracks being formed by



B



A

Figure 13. Tracks created by the 5.44 MeV alpha particles of ^{241}Am at a source to dosimeter distance of 2 mm in air. Irradiation time = 14 hours. A. Without foil masking. B. With foil masking. Etching parameter- 45% KOH, 800 volts, 2 KHz and etching time 5 hours.

the recoil nuclei (carbon and oxygen). Therefore, it is conceivable the smaller tracks were produced by alpha particles themselves and the large tracks by carbon and oxygen recoil nuclei produced after collisions with the incident alpha particles. A similar effect was noted by Sohrabi in his search for proton tracks.⁸ This explanation was discussed and received support (Sohrabi).⁶⁴ We are also aware of the greater sensitivity of polycarbonate to alpha particles as the energy of an initially high energy alpha particle is degraded from the work of Harley and Pasternack.⁶⁵ Since the foil masking is about 38 μ thick, the alpha particle energy loss would be approximately on the maximum of the dE/dx versus absorber thickness curve according to the data of Harley and Pasternack.⁶⁵ Therefore, more alpha particles would be capable of producing etchable tracks at this degraded energy than at the peak energy where energy loss is on the flat portion of the curve.

Optimization Study

Once the type of foil to be used in these studies had been determined, the etching conditions yielding the best results were required. Based upon the data of Sohrabi,⁸ a series of foils were etched with controls to systematically examine the effects observed when the various parameters were changed. Different combinations of the following selections were used: etchant, 30 and 45 percent KOH; etching time, 4, 5 or 6 hours; voltage, 700 or 800 volts. The etchings were performed as indicated on Table II. The most optimal set of conditions to be used was a trade-off between several desired effects. It was desired to have the largest track diameter possible, of course, to expedite counting but at the longest etching times two facts became evident. First, at the longest etching time reproducibility suffered because the

Table II. The Set of Conditions Used During Each Run to Determine the Most Optimal Etching Conditions

| Run | Percent KOH | Etching Time Hours | Voltage |
|-----|-------------|--------------------|---------|
| 1 | 45 | 4 | 700 |
| 2 | 45 | 5 | 700 |
| 3 | 45 | 6 | 700 |
| 4 | 45 | 4 | 800 |
| 5 | 45 | 5 | 800 |
| 6 | 45 | 6 | 800 |
| 7 | 30 | 4 | 700 |
| 8 | 30 | 5 | 700 |
| 10 | 30 | 4 | 800 |
| 11 | 30 | 5 | 800 |
| 12 | 30 | 6 | 800 |

frequency of premature shutdowns increased due to etching through of large tracks and leakage. Second, the background increased because deeper layers of the foils were exposed and available for etching at the longest etching time. Therefore, we selected the most optimal etching conditions to be 45 percent KOH, 800 volts and 5 hours etching time except when background is crucial relative to the large track diameter and consequent ease of track counting. In this latter case, 4 hours was decided to be the best etching time.

Large Scale Etching

One of the problems to be solved in shaping this dosimetry system into a practical personnel dosimetry system is increasing the number of foils that can be etched in one batch. Toward this end Dr. M. Sohrabi designed a large etching chamber capable of etching several times the number of foils now possible with our laboratory scale chambers. The laboratory scale chambers will etch seven (7) and the large chamber will etch thirty-five (35) foils at one time. This latter chamber (Sohrabi and Morgan)⁶⁶ has fixed stainless steel electrodes (24.5 cm in diameter and 75 μ in thickness) and 10 cm total length. The diameter of the chamber is 25.5 cm. There are two holes in the bottom of this chamber connected to two hoses for draining. As pointed out by Sohrabi and Morgan in an earlier report,⁶⁶ this chamber did not yield a large degree of reproducibility due to its tendency to allow etchant to leak around the O rings, stopping the etching process.

At the beginning of this contract period, almost half the etching positions leaked severely. During this contract period, attempts were made to correct this problem. The first attempt culminated in the construction of a vice-

like instrument to apply pressure directly to the O rings at each etching position and thereby reinforce the watertight seal. This succeeded in reducing the leakage from eighteen down to three or four positions, but the upper limit had been reached as to the amount of pressure that could safely be applied to the chamber. The problem must be arising from another source. It was determined the recesses into which the O rings fit into the chamber had small but detectable differences in their machined depth. The O rings used were constructed from a reasonably hard material and they were thin (3 mm) so it was considered an uneven seal might be created when these rings were present together. New O rings were ordered to custom specification (not a standard size, as before) which were constructed 4.5 mm thick and with a softer material to facilitate formulation of the watertight seal when pressure was applied. This method has been more successful than previous ones but leakage still occurs with a greater frequency than is observed in the small chambers. It is believed with more work this chamber can be made stable enough to use routinely and will significantly increase the number of foils etched per cycle.

Some Observations on Electrochemical Parameters

In electrochemical etching, a high voltage at a certain frequency is applied across an etching chamber filled with an etchant. Each diffusing ion in the etchant experiences a force which is equal to

$$\vec{F} = Z_i e \vec{E}$$

where \vec{F} = the induced force, Z_i = the number of elementary charges, e = the unit electric charge, and \vec{E} = the electric field at the location of the ions. This force is superimposed on the random walk of the diffused ions. Thus

a positive ion is accelerated toward and away from an electrode as its potential oscillates from minus to plus, respectively. Likewise, the negative ion is accelerated to and from an electrode as it alternately becomes an anode or cathode respectively.

Applied high voltage is a parameter of importance in affecting the electrochemical etching amplification results in terms of sensitivity (tracks/neutron) and track diameter. Larger track diameter and good contrast are specially necessary for the microfiche reader due to its single magnification (50X).

Figure 14 shows the track diameter distribution appearing on the microfiche reader screen. These tracks were created by ^{252}Cf fast neutrons impinging upon 250 μm polycarbonate foils and then etched in 45% KOH solution for 4 hours applying various voltages at 2 KHz. As can be seen, the most probable frequency of track diameter on the microfiche reader screen shifts from 3 mm to 5 mm when the applied voltage increased from 800 V to 1500 V. It is particularly recommended that higher voltage should be applied to get larger track diameter and well defined tracks for counting by a microfiche reader.

Table III are sets of data showing the variation of the background, the sensitivity and the mean track diameter with applied voltage across the electrodes. The other parameters (250 μm polycarbonate foils etched in 45% KOH solution applying voltage at 2 KHz for 4 hours) were maintained constant. All of these three variables showed increased magnitude with voltage. The error of each value was assumed to follow the Poisson distribution. The figure of merit is defined for the comparison of different applied voltages.

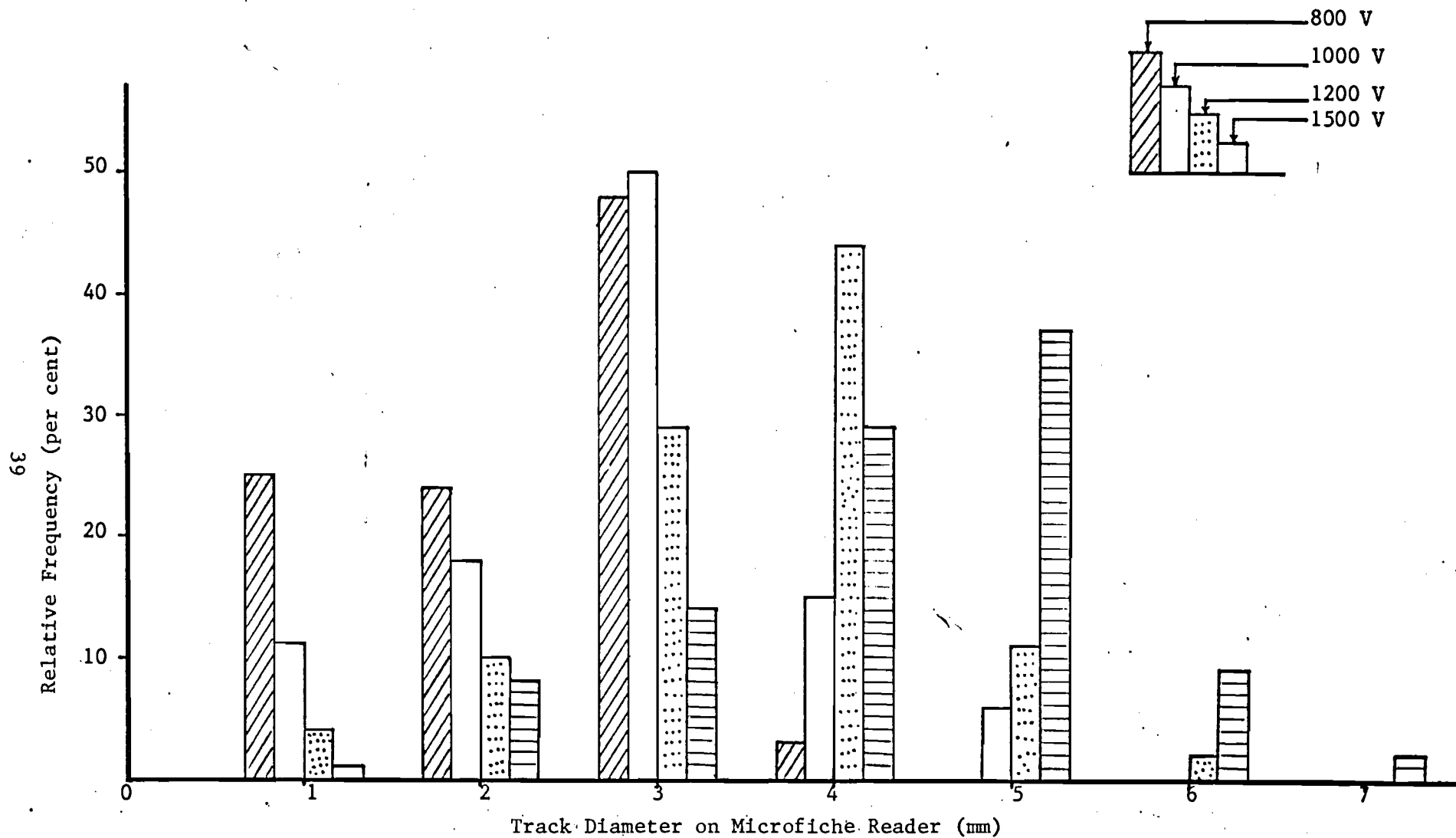


Figure 14. Track diameter distribution of tracks (measured in mm from microfiche reader screen) created by ^{252}Cf fast neutrons. Foil thickness = 250μ . Etching parameters - 45%, 2 KHZ, various voltages and etching time was 4 hours.

Table III. Background, Sensitivity, Figure of Merit, and Mean Track Diameter Variation with Applied Voltage

| Background ₂ (tracks/cm ²) | Sensitivity (tracks/neutron) | Relative figure of merit (sensitivity ² /Background) | Mean Track diameter (μm) |
|--|---------------------------------|---|--------------------------------|
| 600V 1.2±1.1 | (2.3±1.1)×10 ⁻⁷ | 0.044 | 26 |
| 800V 3.0±1.7 | (1.6±0.24)×10 ⁻⁶ | 0.85 | 43 |
| 1000V 11±2.3 | (3.6±0.34)×10 ⁻⁶ | 1.2 | 54 |
| 1200V 14±3.7 | (4.4±0.38)×10 ⁻⁶ | 1.4 | 63 |
| 1500V 27±5.2 | (6.7±0.46)×10 ⁻⁶ | 1.7 | 81 |

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