

THE NUCLEUS

January 1988

Of the Northeastern Section of the American Chemical Society

Vol. LXVI, No. 4

Monthly Meeting:

*Symposium on
Mechanistic
Enzymology*

Board Minutes

October 1987

Councilors Report

New Orleans, Sept. 1987

Calendar

January Meeting

*The 705th Meeting of the
Northeastern Section of the
American Chemical Society*

*Jointly with the
Medicinal Chemistry Group*

Symposium

"Mechanistic Enzymology and Its Application to Drug Design"

Thursday, January 21, 1988

Boston College
304 Higgins Hall
Chestnut Hill, Massachusetts

- 2:30 p.m.** Refreshments
- 3:00 p.m.** Opening Remarks: Dr. James B. Mangold
- 3:10 p.m.** Dr. RICHARD B. SILVERMAN, Department of Chemistry, Northwestern University
"Mechanism of Monoamine Oxidase and Its Application to the Design of Inactivators"
- 4:10 p.m.** Dr. JOANNE STUBBE, Department of Chemistry, Massachusetts Institute of Technology
"Mechanism of Bleomycin Mediated DNA Degradation: Toward the Design of Better DNA Cleavers"
- 5:30 p.m.** Social Hour—Faculty Dining Room, McElroy Commons
- 6:30 p.m.** Dinner
- 7:45 p.m.** Keynote Address: DR. ROBERT ABELES, Graduate Department of Biochemistry, Brandeis University, "Inhibition of Serine-Proteases by Trifluoromethyl Ketones"
(Please see Abstracts and Biographies on page 4)

Dinner Reservations: No later than January 15, 1988. Please call Mrs. Karen Piper at (617) 456-8227 or (800) 872-2054. Reservations not cancelled at least 24 hours in advance will be billed for dinner price. Members, \$13.00; Non-Members, \$15.00; Students, \$5.00; Retired Members, \$8.00. THE PUBLIC IS INVITED.

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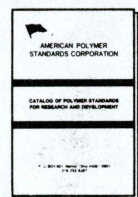
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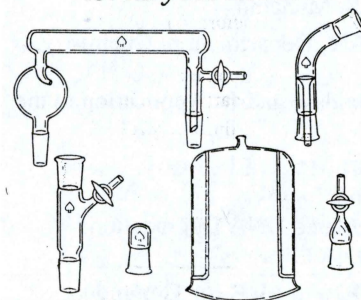
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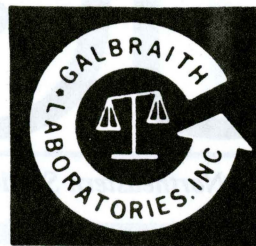


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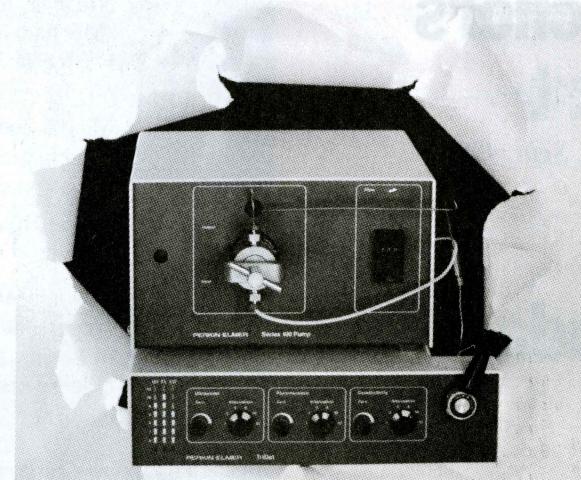
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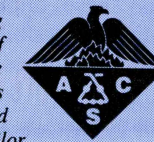
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New Orleans, September 1987

February Issue Deadline: *December 20, 1987*

THE NUCLEUS

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January Meeting

Dr. Richard B. Silverman

Biography. Professor Silverman received his B.S. in Chemistry from Pennsylvania State University in 1968 and his Ph.D. from Harvard University in 1974 (D.H. Dolphin, Advisor). Dr. Silverman did postdoctoral research at Brandeis University with Professor Abeles before accepting a position in the Chemistry Department at Northwestern University. Dr. Silverman is currently a Professor of Chemistry as well as Professor of Biochemistry, Molecular Biology and Cell Biology. Dr. Silverman has received numerous teaching and research awards, including an NIH Career Development Award (1982-1987), Alfred P. Sloan Fellow (1981-1985), DuPont Young Faculty Fellow (1976) and Fellow of the American Institute of Chemists (1985). Dr. Silverman is currently an NIH Advisory Panel Member in the Bio-Organic and Natural Products Chemistry Study Section (1987-1991).

Abstract. The flavoenzyme, monoamine oxidase (MAO), is one of the enzymes responsible for the degradation of biogenic amines; inactivators of this enzyme can exhibit antidepressant activity. This talk will focus on our studies directed at the elucidation of the mechanism of monoamine oxidase, principally via the design of mechanism-based inactivators of the enzyme. The first electron spin resonance (ESR) spectroscopic evidence for a radical intermediate will be presented. Studies with known MAO inactivators and antidepressant drugs will be described that support a radical mechanism of inactivation. On the basis of a radical mechanism new MAO inactivators have been designed. Reaction specificity for the two isozymes, MAO A and MAO B, by several inactivators, will be described. ◇

Dr. JoAnne Stubbe

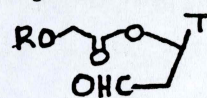
Biography. Professor JoAnne Stubbe received her B.A. in Chemistry from the University of Pennsylvania in 1968 and her Ph.D. at the University of

California at Berkeley in 1971 (G.L. Kenyon, advisor). After a brief postdoctoral stint at UCLA with Jules Rebek, she joined the faculty of Williams College, Chemistry Department from 1972 to 1977, worked with R.H. Abeles at Brandeis University, and moved to Yale University as an Assistant Professor in Pharmacology, 1977-80. She joined the faculty of the Department of Biochemistry at the University of Wisconsin-Madison in 1980, where she became a full Professor in 1985. In September, 1987 she joined the chemistry faculty at the Massachusetts Institute of Technology as Ellen Swallow Richards Professor.

Abstract. Use of specific isotopically labeled DNA models has provided extensive information about the detailed mechanism by which bleomycin (BLM) in the presence of Fe²⁺ and O₂ degrades DNA to monomeric products base and base-propenal [Giloni et al (1981), J. Biol. Chem. 256, 8608; Burger et al, (1980), J. Biol. Chem. 255, 11832]. Studies using (4'-³H)poly[dAdU] indicate that production of both monomeric products are the result of 4' carbon hydrogen bond cleavage producing a common 4'-carbohydrate radical intermediate. The product distribution then results from the O₂ dependent partitioning of this common intermediate. These results have allowed us to predict the structure of the oxidized sugar produced concomitant with base release. Using a DNA model d(CGCGCG) and stabilizing the oxidized sugar by NaBH₄ reduction, the products subsequent to enzymatic degradation have been isolated and identified as the epimeric mixture of 2-deoxy-pentitols expected from a 2-deoxy-4-pentulose precursor. The same products have recently been identified using calf thymus DNA, poly[dAdU] and poly[dGdC]. The amount of 2-deoxy-pentitols is 0.80 equivalents of the amount of base produced.

The proposed mechanisms for production of both monomeric products makes predictions about the source of "O", i.e. O₂ or H₂O, in the products produced. ¹⁸O₂ and H₂¹⁸O labeling studies have shown that 1-atom of ¹⁸O from ¹⁸O₂ is found in the carboxylate of 3'-phosphoglycolate accompanying base-propenal production. Similar ¹⁸O-labeling studies have thus far failed to show the incorporation of ¹⁸O from ¹⁸O₂ into base-propenal and the 2-deoxy-4-pentulose. Reasonable explanations for these observations are presented in terms of the proposed mechanisms.

Recent efforts have focused on investigating the detailed mechanism for the production of base-propenals and 3'-phosphoglycolate ends. Models for two of the proposed intermediates along the reaction pathway have been synthesized, 3'-O-benzoyl-5'-deoxy-4'-hydroperoxythymidine (Saito et al, 1987, J. Org. Chem. 52, 1008) and



and their chemistry studied in detail. The results of these model studies will be discussed in terms of the proposed chemistry mediated by BLM. The isotopic labeling studies and model chemistry provide a unified picture of the mechanism by which BLM mediates degradation on B from DNA. Intriguingly, recent preliminary data suggests that BLM's interaction with A-form DNA-RNA hybrids may result in chemistry at C-1' of the sugar on the deoxyribose oligomer in addition to C-4'. A consequence of this observation would be alternative product production. These findings may reflect the relative accessibilities of hydrogens in the minor grooves of B form and A form DNAs, and will be discussed in terms of the potential use of BLM as a probe of DNA conformation. ◇

Continued on page 5

Dr. Robert Abeles

Biography. Professor Abeles received his undergraduate training at Roosevelt College. He received his M.S. degree from the University of Chicago and Ph.D. from the University of Colorado. He did postdoctoral research at Harvard University before joining the faculty at Ohio State University as an Assistant Professor of Chemistry. He moved from Ohio State to the University of Michigan where he advanced to the rank of Associate Professor of Biological Chemistry. In 1964, he accepted a position at Brandeis University, where he is currently Professor and Chairman of Biochemistry. Dr. Abeles has served on numerous editorial boards, including the *Journal of Biological Chemistry*, *Archives of Biochemistry and Biophysics*, and *Biochemistry*, as well as, the NIH Biochemistry Study Section. He has been

the recipient of many honors, including Aron and Imre Tauber Chair in Biochemistry and Molecular Pharmacology (1977), Council of the American Cancer Society (1977), Advisory Board, Council of Chemical Research (1978), MIT Shen Visiting Professor (1983), Burroughs Wellcome Professorship (1983), Steenbock Lecture (1984) and most recently the Smismann Award (1987).

Abstract. We have established that peptidyl trifluoro-ketones are very effective inhibitors (transition state analogs?) for serine proteases. For instance CBz-Ala-Ala-Pro-ambo-Val-CF₃ inhibits human leucocyte elastase with K_i=1 nM. to establish the nature of the interaction between these serine proteases and inhibitors, the structure of the complex formed with chymotrypsin and N-acetyl-L-leucyl-L-phenylalanyl trifluoromethane (K_i=1.2 μM) was in-

vestigated by NMR spectroscopy and x-ray crystallography. Crystals of enzyme-inhibitor complex were prepared by allowing inhibitors to diffuse into the protein crystals at pH 5.6 (5.6-crytals), or by co-crystallization. The exact location of the P₂ residue in the crystal depends upon the methods of crystal preparation. NMR as well as x-ray crystallography showed that the carbonyl group of the inactivator forms a hemiketal with the active site serine. NMR spectroscopy shows that the OH of the hemiketal is ionized and has a pK of <4.9 as compared to 9.2 in solution. The pK of the active site His is >10. The crystallized enzyme inhibitor complex was rapidly dissolved in buffer and the rate of dissociation of the inhibitor from the complex was determined. This rate was found to be more rapid than that measured when the complex was prepared in solution. This result sug-

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Board of Directors Meeting

Northeastern Section, ACS
October 1, 1987

The October meeting of the Board of Directors of the Northeastern Section ACS was called to order at 4:45 pm by Chairman, Lloyd Taylor in the new Norris Room, MIT. The following members of the Board were present: Phyllis Brauner, Mary Burgess, Michaeline Chen, Catherine Costello, Adrienne Dey, William Foye, Esther Garber, Thomas Gilbert, Michael Hearn, Arno Heyn, Esther Hopkins, David Howell, Truman Light, John Neumeyer, Janet Perkins, James Piper, Myron Simon, MaryAnn Solstad, Lloyd Taylor, and Al Viola.

Dr. Taylor announced the receipt of a letter from Mary Good expressing appreciation for the help of the Northeastern Section with the recent IUPAC meeting in Boston. The Chairman introduced Katie Stygall who will be in charge of the Hospitality Committee in January. Dr. Taylor explained to the Board some confusion which has developed in connection with the grading of the High School Exam. He asked M. Hearn, Chairman of the Chem. Education Committee, to see if he could straighten this out. A. Heyn suggested that the section had the alternate process available of bestowing an additional Honorable Mention, along with its prerequisites.

Membership: Dr. Taylor praised the work of Dr. Duffy who has returned to Alaska. The Board was asked to look for candidates for the Membership chair.

Chemical Education (see Chairman's comments)

College and University: M. Hearn
Undergraduate Research

Symposium: Emily Dudek
Norris Summer Scholars: Sr.
Dorothy Higgins

Student affiliates: Doris Lewis
High School

High School Examination
Contest: Bette Bridges

Finance for HS Examination
Contest: vacant

Aula Laudis (Bette Bridges is covering)
Secondary School Teaching Award: Irwin Taub

A brief discussion was held about voting members of the Board. A. Heyn reported that John Neumeyer, Robert Fields, and Michael Strem were *ex officio* members of the Board because of their Divisional positions. Dr. Neumeyer already is a member of the Board, and Dr. Strem will be a member as of January, 1988.

Secretary: David M. Howell. The minutes were reviewed for the March, April, and May Meetings. Some corrections were made.

Treasurer: J. Piper presented his monthly report. It was accepted by the Board.

Board of Publications: A. Heyn
The Business manager of the NUCLEUS, W. Adams, has resigned from that position. He will continue as Advertising manager (the paid position). Russell F. McCann will be the new Business manager. Mr. McCann is a retired high school teacher from Topsfield High School. Under a new set-up, he will not have a separate checking account, but will direct the treasurer to pay monies out of the regular Northeastern Section account. Auditing of the accounts of the NUCLEUS should be greatly simplified since the amounts will be under the Treasurer's control.

A discussion was held about the financial status of the NUCLEUS, and whether there would be sufficient monies to publish the December issue. The Board approved a motion by A. Dey to publish the December NUCLEUS in the usual manner. Desktop publishing will be investigated further.

Budget: J. Piper announced that he has split the presentation of the budget so that it might be more easily understood. We still have a deficit budget.

Constitution and By-Laws: T.S. Light brought up the by-law revision proposed by the Board of Publications during the summer. After considerable

discussion, the Board approved the proposal which follows:

PROPOSED BYLAWS AMENDMENT

Proposed by the Board of Publications
May 7, 1987

(Deletions in [], additions underlined)

Bylaw III, Sec. 2(b):

"It shall be the duty of the Board of Publications to appoint annually, subject to the approval of the Board of Directors and not later than the [December] May meeting of the Board of Directors, the Editor and the Business Manager of the Official Publication of the Northeastern Section and any committee required for publication services. Appointees shall take office on [January] July first in each year."

Note: The Board of Directors, at its May meeting, voted to table the motion to amend in accordance with the above text.

Trustees: Janet S. Perkins, Myron S. Simon, Arthur Obermayer. J. Perkins reported that a new process in handling the Trustees Funds was being developed.

Archivist: E. Garber reported that the Archives were now stored in the sub-basement of the Mallinckrodt Laboratories at Harvard.

At this point, Dr. Taylor had to leave for another meeting. The gavel was passed to Dr. Gilbert, who presided over the remainder of the meeting.

Continuing Education: vacant position.

Hospitality: M. Chen. The dinner price for retired members was again considered by the Board. It was voted that the discounted price of \$8.00 per meal be continued through June 1988.

Public Relations: C. Filer, P. Samuel. No report.

Public Service: P. Brauner reported on the progress of the Chemistry Day programs. The Framingham State responses have been excellent.

Safety: M-A Solstad reported on
Continued on page 7

Councilors Report

Sept. 2, 1987 in New Orleans

Voting Issues

Councilors	Costello	Garber	Heyn	Kaufman	Light	Rock	Solstad
1. C & B Amendment on "Other" Committees of Council	Y	Y	Y	Y	Y	Y	Y
2. Resolution of Election Disputes: Recommit	N	Y	N	Y	Y	Y	N
Main Motion	Y	Y	Y	Y	N	Y	Y

Y = yes; N = no; A = abstain

Details of the issues were published in the 09/21/87 issue of C & En News.

NOBCCHE Meeting

The 15th Annual National Conference of the NATIONAL ORGANIZATION FOR THE PROFESSIONAL ADVANCEMENT OF BLACK CHEMISTS AND CHEMICAL ENGINEERS (NOBCCHE) will be held in Philadelphia April 4-8, 1988. Technical sessions will include major areas of chemistry and chemical engineering. Symposia will be organized for special topics: Materials Science, Heterogeneous Catalysis, Chemical Marketing, Organic Synthesis, and Emerging Chemical Engineering Topics. Contributed papers are invited. Send 3 copies of abstract to NOBCCHE '88, P.O. Box 588, Lafayette Hill, PA 19444. Abstract deadline is January 8, 1988. For more information contact Dr. D.M. Barnes, AT&T Bell Labs, Allentown, PA 18103, (215) 439-5680. ◇

January Meeting

Continued from page 5

gests significant structural dissimilarity between the crystalline complex and the complex formed in solution. The 5.6-crystals were exposed to pH 7.0 for two days (7.0-crystals). The rate of dissociation of the 7.0-crystals was again determined and now found to be identical with that determined in solution. X-ray crystallography shows that the conformation of the inhibitor in the 7.0-crystals is different from that in the 5.6-crystals, but no significant change in protein conformation has occurred. ◇

Nominations Sought for National Technology Medal

All ACS members are invited by the Committee on Patents and Related Matters (CP&RM) to suggest possible candidates for nomination for the National Technology Medal. Recently funded by the United States Department of Commerce and established by the Stevenson-Wylder Innovation Act of 1980, the medal is awarded annually by the President. It may be awarded to individuals, groups, companies or to institutions within the United States for outstanding contributions to technology or for the promotion of the technological workforce.

Nomination documents may be obtained by contacting the staff liaison to CP&RM, Ms. Nancy Mullens (202/872-4479); they will need to be submitted to Ms. Mullens by February 1, 1988. ◇

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Board Minutes

Continued from page 6

the Safety discussions in New Orleans. The ACS is to work with Insurance Trustees. The material put out by National was reviewed. Some of the experiments are unsafe! Interaction on this was requested.

Topical Groups:

Analytical: new chair to be announced

Medicinal Chemistry: Debra Trantolo

Beer and Wine-Makers: vacant

Retired Chemists: new chair to be announced

Adjourned at 6:45 pm. Next meeting is November 5, 1987

Respectfully submitted,

David M. Howell

Secretary

The Northeastern Section, ACS

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Calendar

Friday, January 15

Professor Angela Stacy (University of California, Berkeley)
"Connections in Solid State Chemistry: From Superconductors to Chaos"
Joint Seminar: Atoms, Molecular and Condensed Matter Series and Harvard/MIT Inorganic Series
Harvard University
Pierce Hall Room 209 at 4:00 P.M.

Thursday, January 21

Professor Richard Eisenberg (University of Rochester)
"The Twists and Turns of Hydrogenation and Hydrogen Transfer Reactions"
Harvard/MIT Inorganic Seminar Series
MIT Room 4-370 at 5:00 P.M.

Monday, January 25

Professor Barry Honig (Columbia)
"Electrostatic and Molecular Recognition in Proteins and Nucleic Acids"
Brandeis University
Gerstenzang 122 at 4:00 P.M.
Call (617) 736-2500 for more information

Tuesday, January 26

Dr. Lawrence K. Duffy (Brigham and Women's Hospital)
"Protein Biochemistry of Amyloid and its Precursors in Alzheimer's Disease"
Southeastern Massachusetts University
Science & Engineering Building Room 305 at 11:00 A.M.
Call (617) 999-8246/8232 for more information

Annual Meeting of Board of Directors

The Annual Meeting of the Board of Directors of the Northeastern Section will convene at 4:30 pm on Thursday, January 7, 1988 in the Norris Room, MIT 18-490. Members of the Section are welcome. ◇

Notices for the NUCLEUS Calendar should be sent to:
Marilyn J. Schneider
Department of Chemistry
Wellesley College
Wellesley, MA 02181
Phone: 235-0320, ext 3031
(Note: Material should be sent so that it arrives by the first of the month *prior* to the month for which the event is scheduled.)

1988 NESACS Election

The Nominating Committee needs your suggestions! If you wish to be a candidate for an office or wish to nominate someone else, please let us know:

Dr. Lloyd D. Taylor, 577-3160
Dr. Adrienne S. Dey, 444-5933
Donald Smith, 999-8232
Thomas R. Gilbert, 437-4505
Michael J. Hearn, 235-0320 x3127 ◇

It's Here Again

The Holiday Lecture, Monday Dec. 28, 1987

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