

# Advances in Carbohydrate Chemistry and Biochemistry

*Editors*

R. STUART TIPSON

DEREK HORTON

Volume 31

# Advances in Carbohydrate Chemistry and Biochemistry

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## PREFACE

In this thirty-first volume of *Advances*, Williams (Swansea) surveys the deamination of carbohydrate amines and related compounds, updating earlier discussions by Peat (Vol. 2), Shafizadeh (Vol. 3), and Defaye (Vol. 25). Gelpi and Cadenas (Buenos Aires) provide a comprehensive treatment of the reaction of ammonia with acyl esters of carbohydrates; their article greatly extends that by Deulofeu (Vol. 4). A chapter by Watson (Jackson, Miss.) and Orenstein (Boston, Mass.) brings the article by Hudson (Vol. 4) on the chemistry and biochemistry of apiose up to date. Lindberg, Lönnngren, and Svensson (Stockholm) discuss the specific, chemical degradation of polysaccharides in an article that updates that by Bouveng and Lindberg (Vol. 15) and complements that by Marshall on their enzymic degradation (Vol. 30). The extensive literature on the chemistry and interactions of seed galactomannans is surveyed by Dea and Morrison (Sharnbrook, England), thus adding to our previous articles on the chemistry of a variety of polysaccharides. Claudemans (Bethesda, Md.) provides an interesting discussion on the interaction of homogeneous, murine myeloma immunoglobulins with polysaccharide antigens, and also describes the career of the late H. G. Fletcher, Jr. In a continuation of our series of bibliographic articles on carbohydrate structures that have been ascertained by crystallographic methods, Jeffrey (Pittsburgh) and Sundaralingam (Madison, Wis.) treat those structures definitively established in 1973, and list all of those determined satisfactorily before 1970.

The Subject Index was compiled by L. T. Capell.

*Kensington, Maryland*  
*Columbus, Ohio*  
*June, 1975*

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DEREK HORTON

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**碳水化合物化学与生物化学进展 第31卷**

这部进展丛书于1945年创刊,原名为《碳水化合物化学》,自1969年第24卷起改为现名,其编目情况请参见 F50/143,第25,26卷编目号为 F50/144, 145。第28卷编目号为 F74/123。

第31卷收载论文7篇,目次如下: ①糖胺类及有关化合物的测定, ②氨与糖类的酰酯的反应, ③芹菜糖的化学和生物化学, ④多糖的降解率, ⑤结晶半乳糖聚糖的化学和相互关系, ⑥同质的鼠骨髓瘤免疫球蛋白的多糖抗原的相互关系, ⑦糖、核甙和核甙酸的结晶结构的1973年书目。

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## HEWITT GRENVILLE FLETCHER, JR.

1917-1973

Hewitt Fletcher had a blackboard in the basement of his home. Written upon it were the formulas of compounds related to his current interests in chemical research. There, during a break from labors at his work bench, or from sighting a handgun ("please aim carefully, the line of fire runs only one foot past the hot-water heater"), he and his friends would discuss chemistry. He did this, not out of any forced sense of duty, but simply because chemistry was fun, and, like everything else in his life, was done energetically and with much enjoyment. Dr. Fletcher radiated thoroughness and dependability, and the qualities we traditionally expect from the people from "Down-East." His grandfather was Headmaster of the Normal School in Castine, Maine. Dr. Fletcher's father, Hewitt Grenville Fletcher, Senior, went to Amherst College and Harvard University, and later settled in Watertown, Massachusetts, with his wife, Frances Mitchell, and practised law in nearby Boston.

Hewitt, Jr., was born on May 28th, 1917, and grew up in Watertown. The only child in the family, he spent much time with his mother's parents. Despite the harmful possibilities of receiving too much attention in such a situation, he grew up tough and self-reliant, with a distrust of frills and slickness. Helen Thayer, his high-school chemistry teacher at Watertown, was impressed with Hewitt from the start. The summer before he was to take chemistry, Hewitt asked to have the textbook to be used. When the term started, there was nothing to teach him; he had devoured the contents of his book.

Before taking chemistry in high school he had, while in junior high school, set up a laboratory for himself in the basement of his parents' home. Here he experimented, keeping a notebook in which he meticulously recorded his observations and deductions. One day, he prepared sulfur dioxide, and the gas drove his mother's bridge-party out of the house. After a stern lecture by his parents concerning his chemical experimentation, he built a sort of fume-hood, to lead the noxious gases out of the basement into the open.

Unfortunately, the fumes killed his mother's best roses, and again he was made aware of his mother's displeasure. One day, he went to Helen Thayer and told her that "chemistry is at a low ebb at home today." He had asked his father to buy him a small lecture-bottle of a corrosive gas from a chemical-supply house. His father complied and took the material to his Boston law-office, where, somehow, the cylinder began to leak; one whole wing of the building had to be evacuated in the dead of winter. It is typical of the family that none of these mishaps dampened his parents' encouragement of Hewitt's scientific curiosity.

When he was a senior in high school he wanted to study organic chemistry. This was not taught, so Helen Thayer and he studied it together. The reader may wonder if he were not more bookish and industrious than is healthy for a young boy: far from it! When he became a senior in high school, his father bought him a sailboat, an original Morse Friendship Sloop called the "Lulu Belle." This was no dinghy, but a 27-foot boat of beautiful traditional design, with bunks for two. His father had done this so that his son might "learn to cope." Hewitt's first cruise was from Boston, Massachusetts, to Bangor, Maine, an ocean cruise of nearly 200 miles. As the Lulu Belle had no auxiliary engine, he was always subject to the vagaries of the wind, and spent many a night at sea, either because there was too little wind or too much for him to sail safely to shore. He also became quite well acquainted with thick New England fogs. Needless to say that Hewitt, Senior, often paced the seawall, worried about his son's being out and overdue.

Hewitt disliked dances or social affairs, but, apparently, graciously attended those he could not avoid. He did not particularly like athletics while in high school, although he was physically strong, and extremely fond of hiking and mountaineering. At 13, Hewitt met his wife-to-be, Ann Winter. Helen Thayer recalls that, on one particularly calm day, Hewitt took Ann and her for a canoe trip, from Hull, around Boston Light and outer Brewster Island, a round trip of some 20 miles over open water. Another time, during the Christmas vacation, he launched his canoe one evening in Boston Harbor, and paddled among the festively lit boats.

Following high school, he proceeded to the Massachusetts Institute of Technology in Boston for undergraduate study. During this time, Hewitt explored England by bicycle. His uncanny memory and gift for observation would allow him, some thirty years later, to recall details and lead his family to out-of-the-way places of interest when they all visited England. In 1939, he obtained his Bachelor of Sci-

ence degree, followed, in 1942, by his Doctor of Philosophy degree under Dr. R. C. Hockett. While in graduate school, he had married Ann in 1940, and the Fletchers' first child, Bradford, was born in 1942. At that time, Dr. Fletcher was an instructor at M.I.T., teaching organic chemistry and doing research as well. By this time, he had already published a few papers. It is interesting that one of his first papers reviewed the life of Augustin-Pierre Dubrunfaut, an early sugar-chemist. This typifies another of his interests: history. Later in his life, one of the most prized attractions of his home would become his magnificent collection of rare and old books; these ranged from books printed in the liberal Low Countries for religiously persecuted groups of the 17th and 18th centuries to books dealing with the history of science. During much of 1943 and 1944, Dr. Fletcher worked with Robert C. Hockett on the oxidation of polyalcohols with lead tetraacetate, and, in 1945, he collaborated with Dr. R. Max Goepf, Jr., on a number of structural problems in the field of hexitol anhydrides.

In 1945, their second son, R. Theodore, was born. Later that year, the family moved to Bethesda, Maryland, where Dr. Fletcher began work under the direction of Dr. C. S. Hudson at the then National Institute of Health. Hudson had already been at the N.I.H. (formerly called the Hygienic Laboratory) for sixteen years, and having been a student of Van't Hoff's, he had brought his knowledge of stereochemistry to bear on the problems of optical rotation and the chemistry of anomers. Dr. Fletcher entered into this collaboration with characteristic energy. His interest centered originally around synthetic methods involving the cyclitols and anhydrohexitols. During this period, Drs. Robert K. Ness, Fletcher, and Hudson were the first to use lithium aluminum hydride to prepare 1,5-anhydrohexitols from the corresponding fully acetylated hexosyl halides. Then, together with Drs. Roger W. Jeanloz and Hudson, Dr. Fletcher began his well known work on the chemistry of ribose, and developed, for synthetic use, intermediates, such as tri-*O*-benzoyl-*D*-ribopyranosyl bromide, more stable than those hitherto available. Hudson's work on the relationship between structure and optical rotation also continued in collaboration with Dr. Fletcher. In all, they published some twenty papers together. During this time, Hewitt and Ann's third son, Peter Grenville, was born in 1949.

It was obvious that Dr. Fletcher had become Hudson's right-hand man, and, when Hudson retired in 1951, Dr. Fletcher became Chief of the Section on Carbohydrates. With his excellent technician Mr. Harry W. Diehl, and in collaboration with Dr. Ness, he embarked on

a program of synthetic carbohydrate chemistry involving the development of protecting groups, as well as on the elucidation of structure of natural products, such as stevioside, the very sweet glycoside from the "Herb of Paraguay." It was at this time that Dr. Fletcher and his collaborators developed the preparation of tri-*O*-benzoyl-D-ribofuranosyl bromide, a stable intermediate of great utility in the synthesis of important glycosides and nucleosides containing the D-ribofuranosyl group. While investigating the chemistry of these derivatives, they found that, when hydrolyzed, this bromide yields 1,3,5-tri-*O*-benzoyl- $\alpha$ -D-ribofuranose which, on treatment with hydrogen halide, affords 3,5-di-*O*-benzoyl-D-ribofuranosyl halide. This compound lacks a participating group at O-2, thus opening a pathway to the synthesis of the difficultly accessible glycosides and N-glycosyl derivatives of  $\alpha$ -D-ribofuranose.

In the late fifties, the N.I.H. started the Visiting Program; under this, young scientists were invited to work for one or two years at the Institutes under the direction of staff members. Dr. Fletcher actively participated in this program. It was as a Visitor in Dr. Fletcher's laboratory that Dr. Donald L. MacDonald became interested in the preparation of phosphoric esters of carbohydrates, and his work eventually led to the MacDonald method of synthesizing glycosyl phosphates.

Also, in the late fifties, Dr. Fletcher turned to the synthesis of nucleosides, and he developed the first direct synthesis of the two anomers of 9-(2-deoxy-D-*erythro*-pentofuranosyl)adenine. In 1961, he (with Dr. Robert Barker) started his long and thorough research into the use of benzyl ethers as nonparticipating protecting-groups in carbohydrate synthesis. This work, which culminated in the subsequent publication of some twenty papers on this use of the benzyl group, was nearest to Dr. Fletcher's heart. It led to a facile synthesis for spongoadenosine, a nucleoside of great interest in cancer therapy, to a study of the mechanism of solvolysis of aldose halides, and to the preparation of a large number of extremely useful intermediates, as well as to a new method for *O*-debenzylation employing boron trifluoride.

Other avenues of research were not ignored, however, as is indicated by the preparation of the first furanose-related glycol and the study of inversions in the cyclitol series by treatment of their esters with liquid hydrogen fluoride. Then, in the mid-sixties, Dr. Fletcher began to turn to the chemistry of acylamidodeoxyhexoses. Together with Drs. Nevenka Pravdic and T.D. Inch, he prepared 1,2-oxazoline derivatives of such carbohydrates. This led to a method for

the selective transglycosylation of 2-acetamido-2-deoxy- $\beta$ -D-glucopyranosides in which the acetylated glycoside is treated with a solution of zinc chloride in butyl acetate-butyl alcohol. The butyl glycoside is formed by way of the 1,2-oxazoline intermediate. This method has the exciting potential for selective cleavage at positions in polysaccharides where 2-acetamido-2-deoxy sugars having a 1,2-*trans* configuration are located. Dr. Fletcher's death prevented him from further developing this possibility, in which he had a real interest.

Well over thirty postdoctoral Fellows and Visiting Scientists passed through Dr. Fletcher's Section over the years. The association he had with them and the sheer enjoyment he derived from working with them were perhaps his greatest pleasures. He considered this aspect of the proper continuation of science to be one of his greatest achievements. He was an excellent teacher, with a superior research philosophy, and was of unshakable integrity. These associates, who are now scattered all over the world, have expressed the fact that, to them, their years at the National Institutes of Health were their most formative ones.

To go back just a little, to 1958: when Dr. Fletcher went alone to Missoula, in Montana, to work for six weeks at the regional laboratories of the N.I.H., he caught Rocky Mountain spotted fever, but, fortunately, his friends on the scene, the Perrines, nursed him back to health. His family joined him, and then continued with him on a lecture tour out west, during which he availed himself of the opportunity of visiting Professor Gobind Khorana's laboratory. Typically, the family camped out most of the time. There was, presumably, a fair amount of trout fishing, for outdoor activities were another of Hewitt's passions. When his children were young, he was a Scout master in the local Boy Scout troop. He loved canoeing (the family had several canoes in the barn near their house) or an enjoyable weekend of hunting in Virginia. "Due to urgent business of the highest calibre, I shall be out of the laboratory," he would inform his associates. The deer he shot would be dressed by the local butcher, and, for the fortunate few, a dish of venison at the Fletchers's was a real treat.

The association between Drs. Fletcher and Pravdić, which started when she was a Visiting Scientist at the Institute in Bethesda, continued after she returned to Yugoslavia. Together they investigated the chemistry of unsaturated 2-acetamido-2-deoxy-D-glucose derivatives, and, with Emmanuel Zissis, the preparation of lactones from these sugars, work stemming from Dr. Fletcher's interest in Tay-

Sachs disease. This is a disorder in fat metabolism wherein a "hexosaminidase" (better called an acetamidodeoxyhexosidase) is missing. The lack of this enzyme causes failure of the metabolism of ganglioside  $G_{M2}$ , leading to an uncontrolled increase of this material in the body. It was known that hexosaminidases are inhibited by 2-acetamido-2-deoxy-D-glucono-1,4-lactone, but there was a virtual hiatus in the literature on the synthesis and chemistry of 2-acylaminoaldonic acids, so Dr. Fletcher began by thoroughly investigating the chemistry of the oxidation of aldoses at C-1. He unraveled the many products obtained by these oxidations, and the interconversions of these products. Glycols also exert an inhibitory effect on glycosidases, and so Dr. Fletcher began a study of the 2-acetamidoglycols as well, and he was still actively engaged in these researches to the end.

In the early sixties, the Fletchers turned again to one of Hewitt's oldest loves: sailing. Dr. Fletcher had known Dr. Louis Long, Jr., since his graduate-school days. They shared an enthusiasm for the sea, and consequently, the two families spent many a summer sailing together off the coast of Maine. Hewitt had many interests, but sailing was probably his most fervent one. Thus, it is not surprising that, in the late sixties, he decided to have a boat built for his family. It was a happy day in 1969 when they commissioned their 35-ft auxiliary ketch "Aspara." With it, the family, but more often Hewitt and Ann alone, explored the myriad tributaries and inlets of the Chesapeake Bay, or made long cruises, such as the one to Maine and back. It was from his contact with Nature, and from the great pleasure Hewitt derived from sailing, that he drew a large part of the strength needed to bear his illness so gallantly in the last year and a half of his life.

By the early seventies, his children had grown up. Bradford married, obtained his Ph.D. in literature, and then chose a life of teaching; Hewitt delighted in the resulting grandchildren. Ted, too, married; he is a chemist, and now works at the N.I.H. Their youngest son, Peter, chose a career in teaching in secondary school after he finished college, and is a member of the staff of the Friends School in Sandy Spring, Maryland.

This obituary would be incomplete without some indication of the many contributions made by Dr. Fletcher to the administration of Science, but it would be mechanical and difficult to mention *all* the honors he received. He was a member of the American Chemical Society and The Chemical Society (London). He was on many National committees, including the Science Advisory, Post Office, Na-

tional Research Council's Pioneering Research, the Chemical Literature, and the NIH Library Committee. He received the C. S. Hudson Award of the Division of Carbohydrate Chemistry of the American Chemical Society in 1968. He was on the Board of Editors of the *Journal of Organic Chemistry*, as well as the Editorial Advisory Board of *Carbohydrate Research*. He was Chairman of the first Gordon Research Conference on the Chemistry of the Carbohydrates in 1964, and was an invited lecturer at many international meetings. Dr. Fletcher was author or coauthor of more than 170 scientific papers.

Hewitt died, on October 19, 1973, of pulmonary metastases from a sarcoma in the leg. He was a man of character, one of the most distinguished carbohydrate chemists in the United States, and a wonderful and generous companion. He was truly an inspiration to all who knew him, and we shall miss him sorely.

CORNELIS P. J. GLAUDEMANS

#### APPENDIX

The following are the names of scientists who published with Dr. H. G. Fletcher, Jr.: R. Allerton, L. Anderson, J. B. Ames, R. Barker, A. K. Bhattacharya, B. Coxon, H. W. Diehl, M. T. Dienes, G. R. Findlay, I. Franjic, K. W. Freer, C. P. J. Glaudemans, R. M. Goepf, Jr., M. Haga, A. Hasegawa, R. Harrison, E. J. Hedgley, R. C. Hockett, A. G. Holstein, C. S. Hudson, T. D. Inch, T. Ishikawa, R. W. Jeanloz, R. L. Kaufman, D. Kiely, H. R. Kirshen, L. H. Koehler, H. Kuzuhara, D. L. MacDonald, R. Montgomery, R. K. Ness, C. Pedersen, T. D. Perrine, J. R. Plimmer, N. Pravdić, Y. Rabinsohn, H. E. Ramsden, N. K. Richtmyer, W. L. Salo, E. L. Sheffield, Jr., C. F. Snyder, S. Soltzberg, C. M. Sponable, J. D. Stevens, S. Tejima, E. Vis, H. B. Wood, Jr., R. C. Young, B. Zidovec, M. Zief, and E. Zissis.



