A Perspective on the Contributions to Heterocyclic Chemistry by Professor Edward C. Taylor of Princeton University

Peter A. Jacobi

Department of Chemistry, Wesleyan University
Middletown, Connecticut 06457

and

Stephen F. Martin

Department of Chemistry and Biochemistry
University of Texas, Austin, Texas 78712

For many years Professor Taylor has been one of the foremost heterocyclic chemists in the world, and there can hardly be a synthetic or medicinal chemist practicing today who has not utilized his contributions to heterocyclic synthetic methodology. In a broad sense, his vivid dramatization of the importance and effectiveness of how imagination can be exploited in heterocyclic synthesis stands as one his most important contributions. In his work, he has repeatedly demonstrated that rational syntheses of complex systems can be devised by sequential condensation, ring cleavage and rearrangement reactions. His development of novel and broadly useful synthetic methods in heterocyclic chemistry have made possible the preparation of many important natural products and biologically active compounds. His contributions are detailed in well over 400 papers, 35 patents and 74 edited or authored books.

Effective exploitation of rearrangements in synthesis has been one of his principal themes. Utilizing *o*-aminonitriles as intermediates, he developed simple routes to a broad variety of heterocyclic systems, and these extensive contributions to heterocyclic synthesis via *o*-aminonitriles were summarized in his book with Alexander McKillop, *The Chemistry of Cyclic Enaminonitriles and o-Aminonitriles* (1970). Extensions of his discovery of the base-catalyzed dimerization of *o*-aminonitriles (48, 72, 82) led to a general synthetic method for 4-aminopyrimidines and a variety of other functionalized pyrimidines (71, 75, 76, 108, 126, 127, 131, 132, 133, 134, 154). His preparation of malononitrile dimer (57) and its utilization as an intermediate for the synthesis of condensed heterocycles (58) has been the basis for much further work by other research groups. An early example of his application of rearrangement reactions to heterocyclic synthesis was his preparation of the aglycone of the antibiotic toyocamycin from tetracyanoethylene (104, 112). He elucidated the structure of and then synthesized the pyrimidotriazine antibiotic 2-methylfervenuone (MSD-92) (164), and then devised an elegant one-step synthesis of the related antibiotic fervenuin (148, 239). Extensive work on purine chemistry (28, 39, 41, 42, 49, 53, 54, 55, 64, 70, 71, 81, 87, 106, 107, 111, 160, 199, 215, 328, 372, 383, 387, 390) included an unequivocal synthesis of 9-substituted adenines

via furazanopyrimidines (199) which has become a method of choice for the preparation of these critical nucleoside bases.

Taylor was one of the first to recognize and exploit the synthetic potential of the *N*-oxide grouping in aromatic nitrogen heterocyclic chemistry (35, 40, 43, 46, 51, 53, 54, 65, 66, 69, 78, 80, 84, 94, 107, 141, 150, 165, 168, 203, 220, 249). His early synthesis of the alkaloid ricinine from 3-picoline (35), and his synthesis of xanthopterin from pterin-8-oxide (220), are outstanding examples of applications of *N*-oxide rearrangements to synthesis. He developed a general and unequivocal route to aromatic *N*-oxides based upon intramolecular capture of hydroxylamines by suitably positioned electrophiles, and applied this method to the synthesis of, *inter alia*, quinoline, purine, quinazoline and quinoxaline *N*-oxides (165). His unequivocal synthesis of pteridines (see below) is based upon pyrazine *N*-oxide intermediates. His extensive work on the photochemistry of heterocyclic *N*-oxides (88, 89, 98, 117, 129, 157, 163, 170, 172, 183, 187, 190, 191, 203) includes a review of the field (168).

Over the past forty years, Professor Taylor has contributed to the chemistry and synthesis of most of the common, and not-so-common, four-, five-, six- and seven-membered heterocyclic systems containing nitrogen, sulfur and/or oxygen, and he has introduced a host of new reagents for use in heterocyclic synthesis. He developed general procedures for the introduction of alkyl, alkenyl and epoxy groups into heterocyclic nuclei which were exploited in elegant total syntheses of the *Cinchona* alkaloids quinine, quinidine, cinchonine, and cinchonidine (210, 214, 224, 234); his strategies for the conversion of substituent primary amino groups into nitroso and nitro groups allow the ready conversion of heterocyclic amino groups into a broad variety of other types of substituents (304, 305). Certainly his early one-step synthesis of tetrahydrocannabinol (THC) was one of his more dramatic illustrations of the power of imaginative synthetic planning (121). His recent applications of intramolecular Diels-Alder reactions of monocyclic and fused 1,2,4-triazines have led to a startling array of fused pyridine and pyrimidine systems (325, 330, 331, 339, 340, 341, 348, 349, 351, 352, 357, 358, 360, 362, 363, 365, 366, 370, 374, 375, 376, 380, 391). Taylor's seminal papers on fused diazetidinones (β -azalactams) as highly strained bridgehead aza analogs of the β -lactam antibiotics (155, 156, 294, 298, 299, 301, 312, 313, 315, 317, 337, 338, 353, 399, 401) have stimulated intense interest.

Among Taylor's most significant contributions to heterocyclic chemistry have been his series of 78 papers on pteridines, and 49 papers on aza- and deaza-pteridines, extending over a period of more than 45 years. His early work dealt extensively with the development of synthetic methods to and the fundamental chemistry of these ring systems, and provided the background for much of the intensive present-day efforts in this important area of natural product and medicinal chemistry. He was one of the first to prepare 2,4-diaminopteridines (4) and to demonstrate their activity as folic acid antimetabolites, and he elucidated the nature of a number of ring opening and rearrangement reactions which have since become standard for the introduction and manipulation of substituents in this system (10-13, 25-27, 47, 56, 76). He was the first to

demonstrate the synthetic versatility of pyrazines for pteridine synthesis (16, 29, 34, 38, 41, 47, 60). This work culminated in a new synthetic approach to pteridines which makes possible the unequivocal preparation of 6-substituted derivatives uncontaminated by isomeric impurities; this "Taylor synthesis" is now considered a classic (150, 177, 218, 219). By utilization of this methodology, Taylor has described elegant syntheses of, *inter alia*, xanthopterin (220, 237, 282), isoxanthopterin (237), folic acid (293), methotrexate, aminopterin, Cyprino-Pourpre B (221), L-erythro-biopterin (233, 250), neopterin, Asperopterin B (236), euglenapterin (300), 6-formylpterin (267, 285, 356), deoxyurothione (303), urothione (373) and Form A of the molybdenum cofactor (including determination of its absolute configuration; 381, 385, 402).

Perhaps his most significant recent work in heterocyclic chemistry has come from extensions of these fundamental studies on synthesis and reactivity to the preparation of aza- and deaza-analogs of the various folate cofactors involved in metabolic one-carbon transfer reactions. A culmination of these efforts has been his design, synthesis and biological investigation of 5,10-dideaza-5,6,7,8-tetrahydrofolic acid (Lometrexol, DDATHF) (327, 328, 343, 344, 345, 350, 371, 372, 379, 382, 383, 384, 386, 387, 388, 389, 390, 393). This remarkable compound serves as a superb substrate for folylpolyglutamate synthetase (FPGS), and the resulting polyglutamate inhibits the enzyme glycylamide ribonucleotide formyltransferase (GARFT), which requires 10-formyl-5,6,7,8-tetrahydrofolic acid as its natural cofactor, and mediates the first of two formyl transfer reactions in the *de novo* purine biosynthetic pathway. Inhibition of this process leads to depletion of cellular pools of both ATP and GTP. Lometrexol, which exhibits extraordinary activity against a broad range of solid tumors, and possesses complete activity against tumors resistant to Methotrexate, is now in worldwide Phase II clinical trials. Many further analogues of Lometrexol, which have been prepared by a diversity of ingenious synthetic strategies, include 5-deaza-5,6,7,8-tetrahydrofolic acid (378), 5,10-dideaza-5,6,7,8-tetrahydrohomofolic acid (384, 408), 5,10-dideaza-10-hydroxymethyl-5,6,7,8-tetrahydrofolic acid (343), and "open-chain" analogues of Lometrexol in which the C-6 chiral center is eliminated by deletion of one of the flanking methylene groups (404, 406, 409). In a very recent and extremely promising development, Taylor designed and synthesized a pyrrolo[2,3-*d*]pyrimidine derivative which is an extremely effective inhibitor of thymidylate synthase; this new antifolate entered clinical trial in the fall of 1992 for the treatment of solid tumors (410).

Taylor has also made invaluable contributions to the general literature and teaching of heterocyclic chemistry. In 1968 he joined Dr. Arnold Weissberger as co-editor of the Wiley-Interscience series, *The Chemistry of Heterocyclic Compounds*, and *General Heterocyclic Chemistry* (Taylor has been the sole editor since Dr. Weissberger's death a number of years ago). Thus far 61 volumes have been published under his editorship. His 24-hour Film Course on *Principles of Heterocyclic Chemistry*, and his companion Audio Course, both distributed by the American Chemical Society, are educational milestones; these have been followed recently by an extensive lecture course aimed at industrial research laboratories on *Utilization of*

Heterocycles in Organic Synthesis. He is also the editor of *Advances in Organic Chemistry* (9 volumes to date) and was co-editor with Pfeleiderer of *Pteridine Chemistry* (1964).

No listing of Professor Taylor's contributions to synthetic organic chemistry would be complete without mention of his contributions to the use of thallium reagents in organic synthesis. This extraordinarily productive research program, which he carried out over a period of some 17 years together with Dr. Alexander McKillop (a former postdoctoral associate, who is now Professor of Organic Chemistry at the University of East Anglia), led to more than 130 new chemical transformations which are applicable to aliphatic, alicyclic, aromatic and heterocyclic chemistry, and have astonishing versatility, scope, specificity and manipulative simplicity. For example, Taylor and his collaborators developed new methods for the synthesis of aromatic nitriles, phenols, thiophenols, thiocyanates, amines, iodides, bromides, chlorides, fluorides, nitroso and nitro compounds; for specific labelling of aromatic hydrocarbons with deuterium and tritium; for the synthesis of symmetrical and unsymmetrical biphenyls; and for orientation control in electrophilic aromatic substitution reactions. They developed new methods for the synthesis of carbonyl compounds, for ring contractions, ring expansions, and cyclizations, for functionalization of acetylenes, imines, and other unsaturated systems, for the synthesis of allenes and acetylenes from β -keto esters, for glycol synthesis and cleavage, for the synthesis of arylacetic esters from aralkyl ketones by oxidative rearrangement, for the conversion of cinnamaldehydes to arylmalondialdehydes, and for the synthesis of both benzoxazoles and indoles from anilides. Novel, effective procedures were developed for the replacement of phenolic -OH groups by a variety of other functional groups, including carbon substituents. Taylor was able to immobilize thallium reagents on inert inorganic supports, and many of the above reactions can now be carried out simply by passing a solution of the substrate through a column packed with the supported thallium reagent. This monumental work on thallium chemistry is described in a series of almost seventy papers on "Thallium in Organic Synthesis" (see bibliography for citations) and five extensive reviews, and was recognized, together with his work in heterocyclic chemistry, in the 1974 ACS Award for Creative Work in Synthetic Organic Chemistry.

EDWARD C. TAYLOR

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Edward C. Taylor

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