



Foreword

This special issue of the *International Journal of Mass Spectrometry* honors Professor Charles L. Wilkins for his lifetime contributions to fields of mass spectrometry and analytical chemistry on the occasion of his 70th birthday. This issue focuses only on mass spectrometry, which represents a small fraction of the breadth of Professor Wilkins' portfolio of scientific investigations. It is the field in which he has had the most recent and longest impact. The contributions to this issue reflect both new and maturing trends in "Leading-Edge Mass Spectrometry and Applications". They are organized with Fourier transform mass spectrometry (FTMS) first, because Professor Wilkins contributed so much to this field, continues with the instrumentation theme, showcasing the emerging field of ion mobility, and closes with applications and contributions from previous members of Professor Wilkins' research groups.

It seems appropriate that we, as co-editors of this special issue, write this Foreword together. Each of us knows Charlie very well and can offer unique perspectives on different aspects of his career. We provide little-known stories of how he got to his current stature in life, examples of his important contributions to analytical chemistry and to science in general, evidence of his enthusiasm for training students in research, and acknowledgement for his sometimes thankless efforts to build infrastructure at universities by establishing instrument facilities accessible to the scientific community.

1. Origins, education, and training

Charlie was born on August 14, 1938 in Los Angeles, CA, the first child to Richard and Lenore Wilkins. He was later joined by a sister, Janet, and three brothers, Kit, Creighton, and Dane. He grew up in Westchester, CA, after moving from Inglewood, CA. Later, after Charlie left for college, his house was demolished as a consequence of installation of the north runway at LAX airport. Perhaps that was an omen of the major changes that would also occur to science from Charlie's scientific career. As early as age 5, Fig. 1, he seemed to be showing signs of the direction his future would take.

Charlie received a B.S. degree from Chapman College, in Orange, CA, in 1961. He was allowed to graduate despite his unbridled enthusiasm for chemistry when flames erupted during a *highly exothermic* chemical synthesis (phosphorous pentabromide, PBr_5) that he performed on the lawn of the campus.

Charlie's own students as well as faculty accepting graduate students into their research groups often find encouragement in a story about a bump in the road towards his own Ph.D., one that does not appear on his curriculum vitae. Charlie spent a year as a graduate student at Arizona State University from 1961 to 1962.

Unfortunately, inadequate grades led to loss of financial support. Undaunted, he applied to the graduate program in chemistry at the University of Oregon in Eugene, where he was accepted, but without support. His move to Oregon in 1963 required that he get a job (it was at a laundry extracting towels) allowing him to restart his graduate studies. Lloyd Dolby, who was on the faculty in the Chemistry Department, saw a "diamond in the rough", accepted him into his research group in physical organic chemistry, and put him on a research assistantship. It was a wise investment, because only 3 years later, in 1966, Charlie received a Ph.D. after he finished not just one, but two independent research projects. One involved solvolysis studies of substituted cyclobutyl tosylates and investigation of the mechanism of the Prins reaction. A third project, aimed toward the synthesis of tetrahydrocannabinol (the active agent in marijuana), was abandoned after another group announced the successful completion of a similar synthesis.

Charlie joined Andrew Streitwieser's research group as a post-doctoral associate in 1966 at the University of California, Berkeley. He and co-workers coined the term, "triflate" [1], which in retrospect has become an important name and the synthesis of triflate esters an important strategy in the science of physical organic chemistry.

2. The University of Nebraska-Lincoln

In 1967, Charlie joined the Department of Chemistry at the University of Nebraska-Lincoln, as Assistant Professor in organic chemistry. It is interesting at this point to mention another story that should comfort new faculty members when they consider their own non-ideal starting conditions. Not only did Charlie receive only \$5000 in startup funds, he had to share laboratory space with other organic chemists and without fume hoods. His first office actually belonged to Desmond Wheeler, a colleague in Chemistry, who was on sabbatical at the time. Charlie was allocated only two drawers of filing cabinet space in that office. Once Wheeler returned, Charlie finally acquired his own office, but it was across the street from the Chemistry building in Oldfather Hall, a Social Sciences building, where he was the only chemist.

When Michael Gross arrived at Nebraska, a year after Charlie's start, Mike's first laboratory was located in a renovated men's restroom of the Chemistry building. Charlie's and Mike's early experiences at Nebraska were the beginning of a "beautiful [scientific] friendship".

Charlie was promoted to Associate Professor in 1972 and Full Professor in 1976. He spent a sabbatical leave in 1974–1975 at the Department of Chemistry, University of North Carolina where



Fig. 1. At age 5 (1944), Charlie practiced for future trips to conferences where he would give innumerable presentations on his scientific research.

he taught a beginning course in Analytical Chemistry. He and his research group made significant advances in science. Nine graduate students finished their degrees and seven postdoctoral associates worked with Charlie during that first era of his independent scientific career. His time at the University of Nebraska was marked by a number of “firsts”, including the birth of his first son, Mark.

Although his initial roots were in physical organic chemistry, the need to make accurate and precise measurements to carry out his work drew Charlie more and more deeply toward instrument development while he was at the University of Nebraska. It was then, about 5 years after he was hired, that he made the transition to analytical chemistry. His first use of mass spectrometry was as a means of measuring the isotope effect in the solution Prins reaction of styrene. He and Mike saw an application of using the isotopically labeled styrenes that Charlie had prepared for his solution chemistry as reagents in gas-phase ion chemistry. Using a drift-cell with ion cyclotron resonance (ICR), they discovered the styrene ion molecule reaction and published a few papers on the subject, one of which was of general interest and appeared as a full article in *J. Am. Chem. Soc.* [2–4].

This collaboration was the foundation of a longer lived interaction during which he and Mike developed the first analytical FTMS and demonstrated important applications in analytical and physical organic chemistry. This work followed on the heels of the pioneering efforts of Mel Comisarow and Alan Marshall to demonstrate FTMS at the University of British Columbia in Canada. The Wilkins/Gross early results were regarded with some suspicion by the MS experts of the day, especially when their narrow-band spectra did not show isotope peaks. Their mass resolving power was so much higher than that achievable on the *de rigueur* sector instruments of the day that the $A + 1$ peaks were off the slide. The work finally found good reviews at NSF, but early support from Nicolet Instruments and the Gulf Oil Foundation gave them the needed financial support to continue their research efforts (Fig. 2). They were the first to interface FTMS to chromatography (GC) and to observe self-chemical ionization (self-CI) and coined the term [5]. The collaboration continued for a few years in the 1980s, supported by both NIH and NSF, but as is usually the case, a long-distance collaboration became difficult to sustain after Charlie accepted a position at the University of California, Riverside in 1981.



Fig. 2. Early funding (\$6000 check) for development of an analytical FTMS instrument at the University of Nebraska to Charlie (left) and Michael L. Gross (right) from Gulf Oil Foundation. (Representative from Gulf Oil in center.).

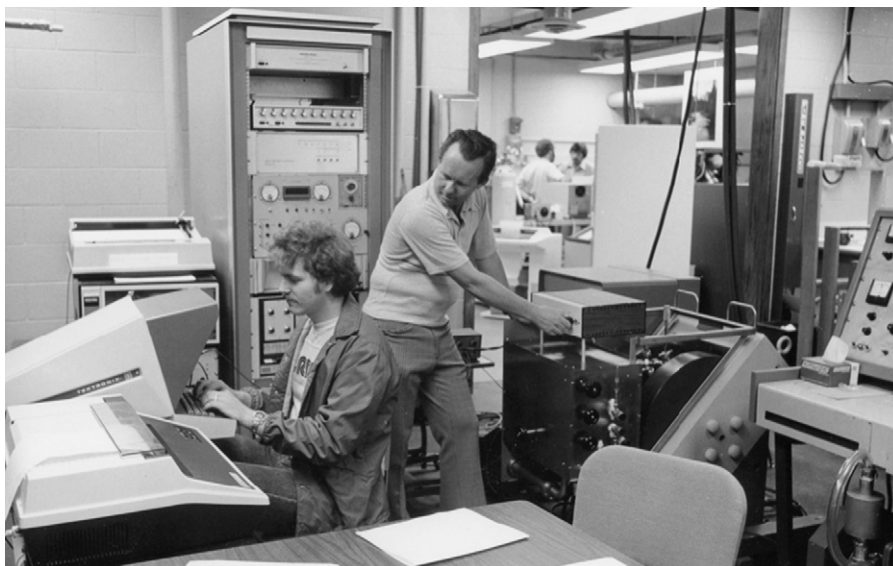


Fig. 3. Graduate student Robert L. White and Charlie are shown integrating “mini” computers with a “high field” (permanent) magnet FTMS and gas chromatography at the University of Nebraska (ca. 1980).

For almost 18 years, Charlie and his group pursued research into the development and elaboration of multidimensional gas chromatography and hyphenated multi-instrument techniques, one that was eventually the forerunner to a commercial GC-IR-MS instrument system. This work began at the University of Nebraska and continued into his tenure at the University of California, Riverside. Fig. 3 shows Charlie and one of his then graduate students, Robert L. White, working on the integration of FTMS with gas chromatography (GC) at the University of Nebraska. Computers to which they interfaced this instrumentation give a whole new meaning to the word “mini”. A pair of permanent magnets generated the “high field” for FTMS (around 1.2 T) at that time. Charlie is also noted for his pioneering work in GC-matrix isolation FTIR. He made significant contributions toward applying chemometrics to a myriad of analytical chemistry problems and developed a number of new chemometric approaches.

It was at the University of Nebraska where Charlie demonstrated his interest and skills in building facilities to benefit his scientific colleagues. There, he, together with Mike Gross and Gerry Meisels, then chair of the University of Nebraska Chemistry Department, established one of the first National Science Foundation-funded mass spectrometry facilities that was named the Midwest Center for Mass Spectrometry, recognizing its potentially broad scope. It was the beginning of a process (developing core facilities) that Charlie has repeated every time he has joined the faculty of a different university.

3. The University of California, Riverside

In 1981, he joined the Department of Chemistry at the University of California, Riverside (UCR) as Professor in Chemistry. That transition involved the transfer of a high field FTMS (7 T at the time) that was shipped across the country by moving van and arrived in working condition at Riverside. Simultaneously, a team of enthusiastic graduate students drove a rented truck across country with the rest of the laboratory equipment. Charlie was promoted to Distinguished Professor in 1997 after having served as Chairman of the Department of Chemistry (1982–1989) and having been appointed Associate Dean of the College of Physical and Mathematical Sciences (1994–1997).

Charlie’s enthusiasm for sharing science with his ever-growing research group knew no temporal boundaries. Members of his

group at UCR have vivid memories of Saturday morning group meetings. A photo of “delighted” members in attendance of one of those meetings is shown in Fig. 4. Nevertheless, 18 graduate students completed their degrees and 21 postdoctoral associates contributed to the research in his laboratory at UCR.

Charlie’s early work there involved interfacing high-performance liquid chromatography (HPLC) to nuclear magnetic resonance (NMR) detection, well before its recent popularity. Reviewers of unsuccessful proposals to the NIH were skeptical of the feasibility, practicality and usefulness of HPLC-NMR. Of course, today, 30 years later such systems are commercially available. About this time, his research group also developed some widely used mass spectrometric approaches to hydrocarbon polymer (and other materials) analysis. These include the use of silver cationization and the first applications using some unique matrices for desorption ionization. In addition, Charlie along with Peter Griffiths (also at UCR at the time) established databases and criteria for compilation of Fourier transform infrared spectra [6,7], and later Charlie headed an IUPAC Committee with a similar effort for C-13 NMR spectra [8].

Once again, Charlie built up the technical infrastructure of the university, establishing the UCR Analytical Chemistry Instrumentation Facility, which he directed from its inception until his departure from UCR (1988–1998). That facility still serves scientists in Southern California and has expanded from its original optical spectroscopy, NMR, and mass spectrometry capabilities to include X-ray crystallography.

4. The University of Arkansas

In 1998, Wilkins moved to the University of Arkansas where he is currently a Distinguished Professor. Although at a new location, Charlie’s intense passion for science continues. So far, five graduate students have completed their degrees under him at Arkansas and three postdoctoral and visiting scientists have spent time performing research in his laboratory. True to form, he has played a key role by building the Arkansas Statewide Mass Spectrometry Facility, which has been able to attract talented and well-published technical staff (see Fig. 5). That facility now includes two 9 T FTMS instruments in addition to an array of other instruments that includes a MALDI-TOF mass spectrometer, a MALDI TOF/TOF, a Q-TOF, an ion trap and two GC-MS systems along with associated



Fig. 4. Students were “fond” of Charlie’s infamous Saturday morning group meetings at the University of California, Riverside: (standing) Cathy Liub Chii Yang (Ph.D.), David A. Laude (Ph.D., now Associate Dean for Undergraduate Education, University of Texas at Austin), Steven Steiner (Ph.D., now Associate Professor, University of Wisconsin, Platteville), Lydia Nuwaysir (Ph.D., now Senior Product Manager, Applied Biosystems), Carl Ijames (Ph.D., now at Appropriate Tech Resources, Inc.), Dr. Liming Zhang (Postdoc), John R. Cooper (Ph.D., now at NOVEON/Lubrizol), Dr. Robert Brown (Postdoc, now Associate Professor, Utah State University); (sitting) Midge Coates (Ph.D., now Program Manager, This Goodly Land: Alabama’s Literary Landscape), Charlie, Dr. Kasem Chowdhury (Postdoc, now at Dow Chemical Company), and Steve Ha (Ph.D., now Research Scientist, Valent USA Corporation).

robotic sample preparation equipment. Yet a new “phase” began for Charlie at Arkansas with the birth of two more sons, Connor (in 2000) and Eric (in 2005).

Charlie’s research at Arkansas has involved direct FTMS characterization of bacteria, a subject on which he edited a book. His recent research has continued to focus on characterization of synthetic polymers, including copolymers in which Charlie has had a long-standing interest. The research at the University of Arkansas also has been devoted to analysis of lipids and phospholipids in both yeast and tissue samples. For the yeast studies a collaborative project involves investigation of ubiquitination metabolism, as well as research on the statistical treatment of the resulting data.

Phospholipid profiling of both types of samples also occupies the group’s current attention. Charlie has also become interested in the relatively new applications of ion mobility-mass spectrometry to a variety of polymer and protein analysis problems and is currently co-editing a book on the subject. A photograph of his current research group is shown in Fig. 6.

5. Awards, honors, and service

Charlie has been recognized for the seminal contributions that he has made to mass spectrometry and other fields in analytical chemistry. That work is described in over 300 publications

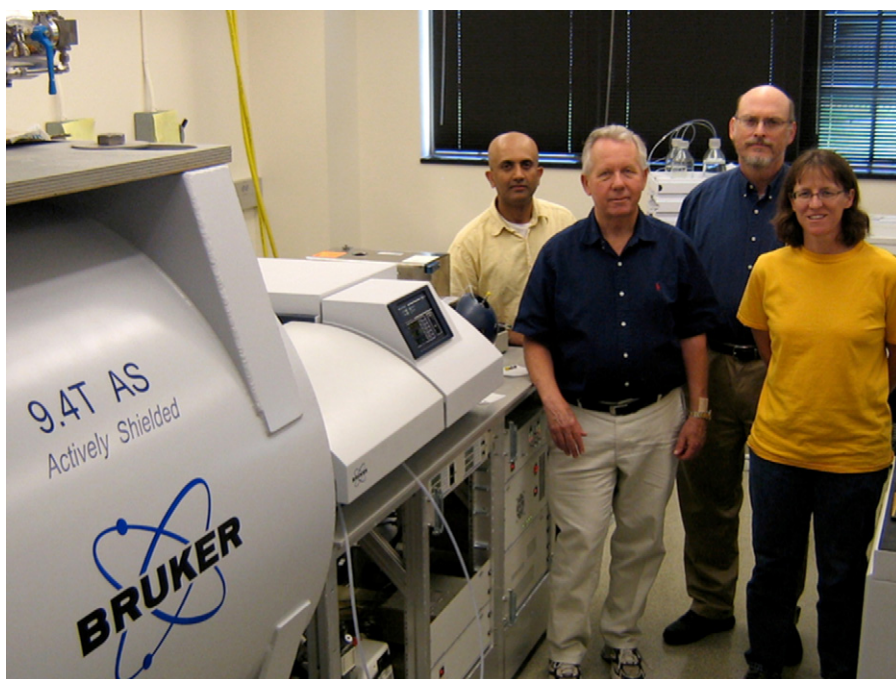


Fig. 5. Charlie’s never-ending desire to improve instrumental infrastructure at Ph.D. granting institutions where he is on the faculty continues, and has led to a state-of-the-art facility at the University of Arkansas. Charlie is pictured here at the Statewide Arkansas Mass Spectrometry Facility with technical staff: (back row) Dr. Rohana Liyanage (Ph.D. with Robert Gordon) and Dr. Jackson O. Lay (Ph.D. with Michael L. Gross), (front row) Charlie and Dr. Jennifer Gidden (Ph.D. with Michael Bowers).



Fig. 6. Charlie's enthusiasm for research and passion to share that with young people in their training as the next generation of scientists continues. Here, he is pictured with his current research group at the University of Arkansas: (back row) Sasa M. Malidinovic, Matthias Knust, Charlie, and Cynthia Kaeser; (front row) Joseph R. Smeal, S. Mariccor Banluta Batoy, and Evgenia Akhmetova.

that he has co-authored. Among his publications are eight books, ranging in topics from computer applications in chemistry to the above-mentioned direct mass spectral characterization of bacteria.

The awards that Charlie has received are numerous, including the Tolman Medal of the Southern California American Chemical Society, the Pittsburgh Analytical Chemistry Award, the UC River-

side Faculty Research Lecturer, the 1997 ACS Franklin & Field Award for Outstanding Achievement in Mass Spectrometry, the Eastern Analytical Symposium Award for Outstanding Achievements in the Fields of Analytical Chemistry, and the University of Oregon, Department of Chemistry Alumni Achievement Award, and Distinguished Awardee in Pure Science, 2004. He is a lifetime Honorary Member of the Society for Applied Spectroscopy and a Fellow of the



Fig. 7. Speakers at the Symposium to Honor Charles L. Wilkins on September 13, 2008 at the University of Arkansas: (front row) Richard M. Caprioli (Vanderbilt University School of Medicine), Catherine Fenselau (University of Maryland), Charles L. Wilkins, Catherine E. Costello (Boston University School of Medicine), Lydia Nuwaysir (Applied Biosystems); (back row) Jackson O. Lay (University of Arkansas), Michael L. Gross (Washington University in St. Louis), J. L. "Jack" Beauchamp (Caltech), Robert L. White (University of Oklahoma), and Robert S. Brown (Utah State University).

Society for Applied Spectroscopy and the American Society for the Advancement of Science.

Charlie has served his profession in many ways, in addition to establishing analytical instrumentation facilities. He has been Chairman of the Analytical Chemistry Division (1991–1992) and the Computers in Chemistry Division (1980) of the American Chemical Society. He is currently a contributing editor for *Trends in Analytical Chemistry* and a member of the editorial boards of *Mass Spectrometry Reviews*, *Applied Spectroscopy Reviews*, and the *International Journal of Analytical Chemistry*. He is also a member of the American Chemical Society, the American Society of Mass Spectrometry, the American Association for the Advancement of Science, and Sigma Xi.

Friends, colleagues, and former Wilkins-group members attended a special symposium to honor Charlie and his contributions to scientific research, infrastructure, and service held on September 13, 2008 in the Department of Chemistry and Biochemistry at the University of Arkansas, soon after his 70th birthday. Distinguished speakers (shown in Fig. 7) gave talks highlighting their research in mass spectrometry during the day and a reception and dinner took place with the appropriate “roasting” in the evening. Some of the speakers and others in attendance on this occasion are among the contributors to this special issue and we thank them for submitting their recent results.

We owe thanks to Charlie for the many reasons to put this issue together and wish him a Happy Birthday!

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