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FOCUS: HONORING R. G. COOKS' ELECTION TO THE NATIONAL ACADEMY OF SCIENCES: EDITORIAL

A Focus Honoring R. Graham Cooks' Election to the National Academy of Sciences

Congratulations to Graham Cooks, a long-time member of the ASMS and a former president of the society, who was recently elected to the National Academy of Sciences of the U.S.A. He joins other eminent American mass spectrometrists, including John Fenn, Fred McLafferty, Klaus Biemann, Carl Djerassi, John Hayes, and Jack Beauchamp in this honor. This *JASMS* focus of original science articles, contributed by his many protégés, honors his accomplishments.

I have known Graham, or about him, for 48 years. It was during the winter 1968 as a postdoctoral research associate with Fred McLafferty at Purdue University that Fred was helping me locate an academic position. Since I am a Midwesterner, we were restricting considerations to the Midwest. Fred indicated that he knew of a position at Kansas State University, but then he backed away and reported that Kansas State was about to appoint a "star" who had recently finished his second Ph.D. studies at Cambridge in England. This was my first introduction to the name R. Graham Cooks, and "star quality" was and still is correct. I didn't apply at KSU, for obvious reasons, and some months later accepted my first independent position at the neighboring University of Nebraska-Lincoln.

Later I met Graham, possibly via a seminar invitation or at a Midwest meeting, before he went to Purdue. I am quite sure he reviewed some of my early papers, where he was immensely helpful and supportive, especially important at the start of one's career. One communication I submitted to *J. Am. Chem. Soc.* is entitled "The unusually slow loss of hydrogen from ionized 1,5-hexadiyne" (1973) on the esoteric (by today's standards) subject of the slow decomposition of a benzene isomeric radical cations. I think it was Graham who wrote "this is what a communication should be – an uncluttered description of a novel result." I always suspected but never knew for sure that Graham was the reviewer, but even if Graham didn't write this, he would have, had he been asked. Comments like this are of immense importance to young investigators because they are enthusiastic and supportive. One of Graham's outstanding virtues is to help and support his students, coworkers, and colleagues.

Ideas flowed from his active mind and continued from his early approaches using MS/MS, collisional and surface activation to understand ion structure. More importantly, Graham put his own touch on this area by conducting complex mixture analysis by MS/MS. Everyone from that era recalls his direct MS/MS analysis of coca leaves and strawberry jam, admiring his boldness to eschew chromatography and advocate putting such complex samples in pristine mass spectrometers. This work is a harbinger of his present efforts on direct mixture analysis, ambient ionization, and small mass spectrometers.

Figure 1. Graham Cooks

Graham has impacted nearly all aspects of our subject. His kinetic method yields physical chemical properties (e.g., proton affinities) of complex molecules, taking as subjects molecules well beyond the ken of the chemical physicists of the day who remained fascinated by detailed measurements of simple hydrocarbons. His invention of desorption electrospray ionization (DESI) has become an important complement to electrospray ionization and an opportunity to do "ambient mass spectrometry." Then there are "paper spray" and "leaf spray." Once he formed ions by various approaches, some by his own methods, he contributed considerably to our understanding of collisional activation and to developing surface-induced

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Table 1. Titles of a selection of RG Cooks papers that have received at least 100 citations

Year	Title	Journal
1969	Bond Formation upon Electron Impact	*Org. Mass Spectrom.*
1971	Structure and Fragmentation Mechanisms of Organic Ions in Mass Spectrometer	*Org. Mass Spectrom.*
1973	Design and Performance of a Mass Analyzed Ion Kinetic-Energy (MIKE) Mass Spectrometer	*Anal. Chem.*
1974	Kinetic-Energy Distributions from Shapes of Metastable Peaks	*Proc. Royal Soc. A*
1977	Intrinsic Basicity Determination Using Metastable Ions	*J. Am. Chem. Soc.*
1978	Direct Analysis of Mixtures by Mass Spectrometry	*Anal. Chem.*
1981	Proton Affinities from Dissociations of Proton-Bound Dimers	*J. Am. Chem. Soc.*
1982	Mass Spectrometry of Large, Fragile, and Involatile Molecules	*Science*
1982	Consecutive Reactions in Triple Analyzer Mass Spectrometry and Applications to Mixture Analysis	*Anal. Chem.*
1987	Mechanisms in Molecular SIMS	*Chem. Rev.*
1991	Operation of a Quadrupole Ion Trap Mass Spectrometry to Achieve High Mass Charge Ratios	*Int. J. Mass Spectrom.*
1994	Reactions of Ions with Organic Surfaces	*Accts. Chem. Res.*
1997	Soft-Landing of Polyatomic Ions at Fluorinated Self-Assembled Monolayer Surfaces	*Science*
2000	Copper(II)-Assisted Enantiomeric Analysis Of D,L-Amino Acids Using the Kinetic Method: Chiral Recognition and Quantification in the Gas Phase	*J. Am. Chem. Soc.*
2000	Miniature Mass Analyzers	*J. Mass Spectrom.*
2001	Collisions of Ions with Surfaces at Chemically Relevant Energies: Instrumentation and Phenomena	*Rev. Sci. Instrum.*
2001	Chiroselective Self-Directed Octamerization of Serine: Implications for Homochirogenesis	*Anal. Chem.*
2003	Preparing Protein Microarrays by Soft-Landing of Mass-Selected Ions	*Science*
2003	Chiral Analysis by MS	*Anal. Chem.*
2004	Mass Spectrometry Sampling Under Ambient Conditions with Desorption Electrospray Ionization	*Science*
2004	Rectilinear Ion Trap: Concepts, Calculations, and Analytical Performance of a New Mass Analyzer	*Anal. Chem.*
2005	Mass Spectrometric Profiling of Intact Biological Tissue by Using Desorption Electrospray Ionization	*Angew. Chem.*
2005	Ambient Mass Spectrometry Using Desorption Electrospray Ionization (DESI): Instrumentation, Mechanisms, and Applications in Forensics, Chemistry, and Biology	*J. Mass Spectrom.*
2006	Droplet Dynamics and Ionization Mechanisms in Desorption Electrospray Ionization Mass Spectrometry	*Anal. Chem.*
2006	Rapid Trace Detection of Triacetone Triperoxide (TATP) by Complexation Reactions During Desorption Electrospray Ionization	*Chem. Commun.*
2007	Development of Capabilities for Imaging Mass Spectrometry under Ambient Conditions with DESI	*Int. J. Mass Spectrom.*
2007	Principal Component Analysis of Urine Metabolites Detected by NMR and DESI-MS in Patients with Inborn Errors of Metabolism	*Anal. Bioanal. Chem.*
2008	Desorption Electrospray Ionization (DESI) Mass Spectrometry and Tandem Mass Spectrometry (MS/MS) of Phospholipids and Sphingolipids: Ionization, Adduct Formation, and Fragmentation	*J. Am. Soc. Mass Spectrom.*
2008	Desorption Electrospray Ionization Mass Spectrometry: Imaging Drugs and Metabolites in Tissues	*Proc Natl. Acad. Sci. U.S.A.*
2008	Latent Fingerprint Chemical Imaging by Mass Spectrometry	*Science*
2010	Paper Spray for Direct Analysis of Complex Mixtures Using Mass Spectrometry	*Anal. Chem.*

Figure 2. R. G. Cooks at ASMS conferences. Top Row: 2015, 2010, 2015; Middle Row: 2014 (with wife Maria Cooks), 2006 (with Gary Glish), 2000; Bottom Row: 2014 (with fellow ASMS Presidents); 2015 (with Bob Murphy and wife Maria). Photos, except ASMS Presidents, by Sue Weintraub

Figure 3. Top Row: Coventry/Warwick for the Jennings/Scrivens 2012 Birthday Celebration. Middle Two Top: with son Barry, End top: with Pat Williams, wife of the late Dudley Williams; Middle Row: with wife Maria in Kyoto IMSC, 2012; Middle and Right: Coventry; Bottom Row: with Nico Nibbering and Helmut Schwarz at the Berlin Wall, 1984; with Alison Ashcroft and Nico Nibbering, Siena Int'l MS School, 2013. Photos by M. L. Gross

dissociation. He promoted extensively ion traps and Orbitraps for mass analysis, recognizing more quickly than most of us the importance of these instruments compared with the large magnetic sectors and Fourier transform ion cyclotron resonance (ICR) instruments that captured our imagination in the 1980s and 1990s. His far-sighted interest has continued with the development of miniature mass spectrometers, even those that can fit in a back pack or a surgical operating theater.

To illustrate the subjects to which he has contributed, I decided to look at his citation record to assess what the community has been "saying" about the importance of his work. I reasoned that if colleagues and competitors thought the work was important, they would cite the paper. The following table is the outcome of my survey and lists a subset of his papers that have received at least 100 citations. It is organized from the beginning of his career to the present. A glance at the titles and years show the interesting

Figure 4. At the 2012 William Nichols Award Symposium and banquet, New York, honoring Alan Marshall. Top Right: with Dick Zare; Bottom Right: with Michael Gross, Alan Marshall, and John Baldeschwieler. Photos by M. L. Gross

evolution of his thinking about our subject. A look to the journal titles reveals that he has supported many journals ranging, from high impact general journals to specialist journals in our field. A long-distance view demonstrates his ongoing contributions that have continued for nearly 50 y.

His work, like that of many in his era, begins with the **fundamentals of gas-phase ions**, their fragmentation, the release of internal energy as kinetic energy, and their collisional activation. This field, of diminishing attention today, follows from the inherently interesting fragmentation of isolated gas-phase ions that underpins mass spectrometry and physical organic chemistry. The mass spectrometer is a "test tube" for ion reactions.

Accompanying these fundamental studies are studies in **instrument design and MS/MS** – a theme that runs through all of his work in direct sample introduction in the mass spectrometer and the opportunity for direct and later ambient analysis.

Emerging from his fundamental studies is the **kinetic method**, which is one of the key ways to measure proton affinities, particularly of molecules that are involatile and not suitable for gas-phase equilibrium. Another subject of interest is the analysis of chiral compounds, culminating in his work on serine oligomers. A follow-up on MS/MS is surface-induced dissociation and the fundamentals of ion–surface interactions and "soft landing" experiments.

Recently, **ambient ionization mass spectrometry** and small, even portable mass spectrometers have occupied his attention. He has developed new ionization approaches (DESI, DESI imaging, paper spray) that set the stage for rapid analysis in simple instrument settings. Accompanying these fundamental and instrumental advances are papers showing **applications**.

Graham sees opportunities to use mass spectrometers for many problems, even to do organic synthesis and teach organic synthesis with no hoods or solvent disposal. Preparing interesting materials prompted his development of soft landing ideas and related strategies of synthesis. A summary of his work is captured with some references by: http://en.wikipedia.org/wiki/R._Graham_Cooks.

To honor him, the *JASMS* editors decided on a Focus to which would be invited his former coworkers, both students and postdoctoral researchers, who are in a position to contribute a scientific article. Graham has educated over 125 Ph.D. students, and a large fraction of them are in academic or related settings wherein they can do mass spectrometry research. *JASMS* captured 20 articles from Graham's protégés to comprise this Focus Section. In this way, we draw attention not only to his research accomplishments but also to his human legacy of students and coworkers. A listing of the articles is as follows:

Ion Chemistry Developments

1. Online Monitoring of Methanol Electro-Oxidation Reactions by Ambient Mass Spectrometry by Si Cheng, Qiuhua Wu, Howard D. Dewald, Hao Chen
2. Formation of Pyrylium from Aromatic Systems with a Helium:Oxygen Flowing Atmospheric-Pressure Afterglow (FAPA) Plasma Source by Sunil P. Badal, Tyree D. Ratcliff, Yi You, Curt M. Breneman, Jacob T. Shelley
3. Charge-Tagged *N*-Heterocyclic Carbenes (NHC): Direct Transfer from Ionic Liquid Solutions and Long-Lived Nature in the Gas Phase by Thyago S. Rodrigues, Denis Lesage, Wender A. da Silva, Richard B. Cole, Günter Ebeling, Jaïrton Dupont, Heibbe C. B. de Oliveira, Marcos N. Eberlin, Brenno A. D. Neto
4. Metal Cationization Extractive Electrospray Ionization Mass Spectrometry of Compounds Containing Multiple Oxygens by Kenneth D. Swanson, Sandra E. Spencer, Gary L. Glish
5. Gas-Phase Oxidation via Ion/Ion Reactions: Pathways and Applications by Alice L. Pilo, Feifei Zhao, Scott A. McLuckey
6. Investigation and Applications of In-Source Oxidation in Liquid Sampling-Atmospheric Pressure Afterglow Microplasma Ionization (LS-APAG) Source by Xiaobo Xie, Zhenpeng Wang, Yafeng Li, Lingpeng Zhan, Zongxiu Nie

Instrumentation Developments

7. Analytical Validation of a Portable Mass Spectrometer Featuring Interchangeable, Ambient Ionization Sources for High Throughput Forensic Evidence Screening by Zachary E. Lawton, Angelica Traub, William L. Fatigante, Jose Mancias, Adam E. O'Leary, Seth E. Hall, Jamie R. Wieland, Herbert Oberacher, Michael C. Gizzi, Christopher C. Mulligan
8. A Simple Method for Improving the Spatial Resolution in Infrared Laser Ablation Mass Spectrometry Imaging by Juha-Pekka Hieta, Anu Vaikkinen, Samuli Auno, Heikki Räikkönen, Markus Haapala, Gianmario Scotti, Jaakko Kopra, Petteri Piepponen, Tiina J. Kauppila
9. High Mass Ion Detection with Charge Detector Coupled to Rectilinear Ion Trap Mass Spectrometer by Avinash A. Patil, Szu-Wei Chou, Pei-Yu Chang, Chen-Wei Lee, Chun-Yen Cheng, Ming-Lee Chu, Wen-Ping Peng
10. Isobar Separation in a Multiple-Reflection Time-of-Flight Mass Spectrometer by Mass-Selective Re-Trapping by Timo Dickel, Wolfgang R. Plaß, Wayne Lippert, Johannes Lang, Mikhail I. Yavor, Hans Geissel, Christoph Scheidenberger

Applications

11. Laser-Induced Acoustic Desorption/Electron Ionization of Amino Acids and Small Peptides by Tiffany M. Jarrell, Benjamin C. Owen, James S. Riedeman, Boone M. Prentice, Chris J. Pulliam, Hilkka I. Kenttämaa
12. Assigning Peptide Disulfide Linkage Pattern among Regio-Isomers via Methoxy Addition to Disulfide and Tandem

Mass Spectrometry by Kirt L. Durand, Lei Tan, Craig A. Stinson, Chasity B. Love-Nkansah, Xiaoxiao Ma, Yu Xia
13. Ammonium Bicarbonate Addition Improves the Detection of Proteins by Desorption Electrospray Ionization Mass Spectrometry by Elahe Honarvar, Andre R. Venter
14. Characterization of Lipid A Variants by Energy-Resolved Mass Spectrometry: Impact of Acyl Chains by Christopher M. Crittenden, Lucas D. Akin, Lindsay J. Morrison, M. Stephen Trent, Jennifer S. Brodbelt
15. Estimating the Efficiency Of Phosphopeptide Identification by Tandem Mass Spectrometry by Chuan-Chih Hsu, Liang Xue, Justine V. Arrington, Pengcheng Wang, Juan Sebastian Paez, Yuan Zhou, Jian-Kang Zhu, W. Andy Tao
16. Monitoring Toxic Ionic Liquids in Zebrafish (*Danio rerio*) with Desorption Electrospray Ionization Mass Spectrometry Imaging (DESI-MSI) by Consuelo J. Perez, Alessandra Tata, Michel L. de Campos, Chun Peng, Demian R. Ifa
17. MS/MS-Assisted Design of Sequence-Controlled Synthetic Polymers for Improved Reading of Encoded Information by Laurence Charles, Gianni Cavallo, Valérie Monnier, Laurence Oswald, Roza Szweda, Jean-François Lutz
18. Characterization of ZnO Nanoparticles Using Superconducting Tunnel Junction Cryodetection Mass Spectrometry by Logan D. Plath, Zongyu Wang, Jiajun Yan, Krzysztof Matyjaszewski, Mark E. Bier
19. Detection of Metastatic Breast and Thyroid Cancer in Lymph Nodes by Desorption Electrospray Ionization Mass Spectrometry Imaging by Jialing Zhang, Clara Feider, Chandandeep Nagi, Wendong Yu, Stacey A. Carter, James Suliburk, Hop S. Tran Cao, Livia S. Eberlin
20. Ambient Ionization Mass Spectrometry Measurement of Aminotransferase Activity by Xin Yan, Xin Li, Chengsen Zhang, Yang Xu, R. Graham Cooks

Congratulations, Graham, on your election to the U.S. National Academy of Sciences. The ASMS is proud of your accomplishments and history and pleased to regard you as a friend, coworker, and colleague.

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