Preface

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George Andrew Olah (1927–2017)

This issue is dedicated to George A. Olah, who passed away on March 8, 2017 at his home in Beverly Hills, CA, USA. He was the recipient of the 1994 Nobel Prize in Chemistry for his groundbreaking work on superacids that led to isolation and observation of long-lived carbocations. He has made fundamental and far-reaching contributions to the field of acid catalysis.

Olah was one of the preeminent scientists in the second half of the twentieth century and the early part of the twenty-first century. His pioneering contributions to Chemistry spans through one of the most magnificent era of science. Olah was born on May 22, 1927 in Budapest, Hungary to Jewish parents, Julius Olah, a lawyer and Magda Krasznai a homemaker. He finished his school years in the Gymnasium of the Piarist Fathers (one of the best schools in the Roman Catholic religious teaching order in Budapest at that time) while the World War II was raging. In his autobiography “A Life of Magic Chemistry” he describes his horrifying experiences of the time. He and his parents survived the war

with the help of friends, but he lost his older brother, Peter. After the war, he studied chemistry at the Institute of the Technical University of Budapest and completed his PhD in 1949 under the supervision of Géza Zemplén, a carbohydrate chemist trained under the legendary Emil Fischer. In 1949, he married Judith Lengyel, a technical secretary at the University, who later studied chemistry and became his partner in research and life. He began his academic career as an Assistant Professor in the same department at the Technical University and was promoted in 1954 as the Head of Department of Organic Chemistry and Associate Scientific Director of Central Research Institute of the Hungarian Academy of Sciences. However, these years in Hungary were really tumultuous in his life due to postwar Soviet rule. Amid the turmoil and aftermath of a failed uprising against the regime in 1956, the future seemed bleak and he decided to join the torrent of about 200,000 Hungarian refugees and fled his native Hungary with his wife and 2-year old first son. After a brief stay in Vienna and England, they moved to Canada.

Olah, was a true legend, a giant of a chemist and a great visionary, who had a prophetic approach for solving challenging problems. In 1957, he joined the Dow Chemicals at Sarnia in Canada and continued his research work there. One of the major areas at Dow Chemicals at that time was Friedel-Crafts-type chemistry for the production of ethylbenzene from benzene and ethylene (for the conversion to styrene for polystyrene manufacture), which involve arenium (Wheland) cation intermediates. It was during this period (1957–1964), that his breakthrough work on long-lived carbocations, such as *tert*-butyl and isopropyl cations in superacids was carried out.

In 1965, Olah was recruited to academia as a professor in Western Reserve University, which later became Case Western Reserve University by merging with Case Institute of Technology. Based on the work of Ron Gillespie in Canada, he had realized that fluorinated Bronsted superacids such as HF and FSO₃H in combination with Lewis superacids lead to conjugate Bronsted superacids, which are billions and even trillion times stronger than 100% sulfuric acid. In the late 1950s he had demonstrated that in such superacid

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systems at low temperatures, long-lived carbocations can be generated. He coined the name “Magic Acids” for the ones made from the combination of anhydrous FSO_3H and SbF_5 . The name was coined after a postdoctoral fellow in his group discovered that these superacids cleaved Christmas candle (paraffin wax) into long lived *tert*-butyl cation (like magic!). Prior to his pioneering work, carbocations were inferred as only fleeting intermediates (with microseconds to nanoseconds of life-time) in acid catalyzed hydrocarbon transformations. It was Olah’s genius that led to the stabilization of carbocations in superacids and their structural elucidation by low-temperature NMR and later on by X-ray structural analysis. His exhaustive structural studies on long-lived carbocations gave rigor and new dimension to hydrocarbon reaction mechanisms under acid catalysis. Olah’s studies on carbocations, created clear demarcation between the trivalent *carbenium ions*, planar ions of the CH_3^+ type, and the penta- or higher coordinated *carbonium ions* of the CH_5^+ type, analogous to other *onium ions*. While trivalent carbenium ions are the key intermediates in electrophilic reactions of π -donor unsaturated hydrocarbons, pentacoordinated carbonium ions are the key to electrophilic reactions of σ -donor saturated hydrocarbons through the ability of C–H or C–C single bonds to participate in 2e–3c bond formations. The non-classical bonding in the famous 2-norbornyl cation $[\text{C}_7\text{H}_{11}]^+$, suggested by Saul Winstein in the 1940s necessitated a higher-coordinate carbon. This was the topic of the so-called classical-nonclassical ion controversy, wherein the late H. C. Brown was the main antagonist, supporting a trivalent carbenium-ion structure for the 2-norbornyl cation. It is ironic that Brown, who received the 1977 Nobel Prize for borane chemistry that is mainly centered on nonclassical structures, did not subscribe to multicenter bonding in carbon chemistry. The nonclassical structure of the 2-norbornyl cation was finally confirmed by Olah’s ^{13}C labeling work using low-temperature NMR spectroscopy and subsequent solid-state NMR at 5 K. The structure of the nonclassical 2-norbornyl cation was later unequivocally established by low-temperature X-ray structural analysis in 2013 by a German group. Olah’s studies on higher-coordinate carbon compounds led to a new area of “Hypercarbon Chemistry”. He also showed that in superacids, electrophiles can be further activated to superelectrophiles by protosolvation (superelectrophilic activation) that can initiate electrophilic reactions on weakly basic substrates.

His early work in the 1950s in fluorine chemistry in Hungary helped many researchers in the study and development of many fluorinated drugs in the pharmaceutical arena. He also developed pyridinium polyhydrogen fluoride (known as the Olah’s reagent), which is widely used as an ambient temperature stable ionic-liquid substituting for volatile HF. Many novel and practical fluorination protocols have been developed using Olah’s reagent. The same

type of reagent was later (in the 1990s) commercialized to produce high-octane gasoline in an environmentally safe process termed “ALKAD.”

In 1977, Olah moved to the University of Southern California, and founded the Loker Hydrocarbon Research Institute with the generous support of Donald and Katherine Loker, where he stayed the rest of his extraordinary life. He continued his research activities in close collaboration with the authors that included synthetic methods, catalysis, oxidations, organosilicon and organofluorine chemistry, and computations. He also became impressed with the predictive power of the computational chemistry with the prodding of his theoretician friends and used it to make important discoveries. In 1994, he was bestowed with the Nobel Prize in Chemistry for his seminal work on carbocations. (Olah was the sole recipient of the 1994 Chemistry Prize.)

Post Nobel Prize, Olah focused his efforts on solving mankind’s fossil fuel based greenhouse problem. His solution to the carbon conundrum was a new approach developed with one of us, now known as “The Methanol Economy.” The concept is based on the use of liquid methanol as an energy carrier and a convenient substitute fuel for gasoline and diesel. Moreover, it can be used as a feedstock for producing ethylene and propylene leading to all petroleum derived products. The joint work with NASA-JPL also paved the way for the development of a direct liquid methanol fuel cell (DMFC), a highly efficient portable source of electricity. He has also developed new methods for converting existing natural/shale gas (methane) and CO_2 to syngas for making methanol. Olah proposed that renewable methanol can be produced by the reaction of hydrogen (generated from water electrolysis using renewable energy) with carbon dioxide from anthropogenic and other natural CO_2 sources, thereby liberating mankind from dependence on fossil fuels. Conversion of carbon dioxide to methanol is now commercially practiced in the George Olah Renewable Methanol Plant in Iceland. More recently, based on reports of astrophysical observations of carbocations and molecules including methanol in space, Olah suggested that methanol could have played a key role in the evolution of complex building blocks that eventually led to life.

Olah’s work had a lasting impact on chemistry. His vast work on carbocations is cited in all college Organic Chemistry textbooks. His hero in chemistry was the late Hans Meerwein. He was a friend/collaborator of many notable chemists: M. Saunders, N. Arnett, R. Huisgen, R. Grubbs, K. C. Nicolau, B. Sharpless, K. Houk, H. Schwarz, J. Rabo, G. Somorjai, P. J. Stang, K. Christe, A. de Meijre, R. Gillespie, G. Schrobilgen, and the late chemists: H. Prinzbach, R. E. Williams, K. Wade, P. Schleyer, I. Ugi, J. Pople, D. J. Cram, J. D. Roberts, J. Kochi, C. Nenitzescu, A. Cotton, and many others.

Olah was prolific with close to 1500 publications, 160 patents and 25 edited books and monographs. He was a kind and generous mentor, who trained more than ~300 graduate students, postdocs and colleagues from many corners of the world, who have done exceptionally well in their chosen careers. He was a highly cited chemist with an h-index of 104! In the course of his career, besides the Nobel Prize, he won innumerable awards, including the ACS Priestley Medal, Cope Award, Roger Adam's Prize, and Eric and Israel's Sheila Samson Prime Minister's Prize for Alternatives to Transportation Fuels. He received more than 15 honorary doctorates. He was a member of many international academies, including the US National Academy of Science and Engineering.

Olah is survived by his wife Judy, two sons, George Jr. and Ronald and three grandchildren. He was a true hero of

chemistry, a great role model and a caring human being. He will be deeply missed by the chemistry community.

This special commemorative issue celebrating Olah's extraordinary achievements (he would have been 91 years old on May 22, 2018) encompasses 21 original papers and reviews focused on the field of catalysis authored by many of his friends, colleagues and students. The first article is the last paper that Dr. Olah was working on with the one of the editors. We were students and long-time collaborators of Professor Olah. We are grateful to Editor-in-Chief, Professor Gabor Somorjai for inviting us to edit this special issue. We also thank Dr. Charlotte Hollingworth (Senior Editor) and Ms. Anitha Kamaraj of Springer for making the entire editorial process efficient and seamless.