Abstract
The life of George Olah exemplifies the fate of Hungarian scientific excellence in the 20th century. He was talented and a hard worker, but he could not remain in his country and had to seek a new life in the West. His lifelong scientific interests developed in the Budapest Technical University helped him to overcome obstacles and he became a leading chemist of worldwide fame. His qualities and flexibility in both scientific research and practical applications serve as a brilliant example for generations to come.

Keywords
early achievements, accommodating to foreign norms, scientific breakthrough, the super-acids, in the service of mankind

1 Introduction
The New York Times wrote on March 14, 2017: „George A. Olah, a Hungarian-born scientist who won the Nobel Prize in Chemistry in 1994 for his study of the chemical reactions of carbon compounds, died on Wednesday at his home in Beverly Hills, Calif. He was 89. “

Once again, a man of outstanding excellence with worldwide fame died abroad. Olah’s studies and career started in Budapest, as vividly described in his autobiography [1]. He obtained a diploma in chemical engineering at the Budapest Technical University (BTU) in 1949 and joined the Organic Chemistry Institute founded by Professor Géza Zemplén in 1913, as the first University Department for Organic Chemistry in Hungary. Zemplén, a carbohydrate chemist was the student of Emil Fisher, so Olah followed traditions and started working on natural products and carbohydrates. His first achievement was an efficient isolation of lanataglycoside C from fresh leaves of Digitalis lanata – favorably cultivated in Tihany – and its subsequent conversion to digitoxin (an effective heart medication).

2 Finding personal fields of interest
Teaching in BTU in the 1940s gave a solid foundation of laboratory skills, but in chemical theory it was incomplete. No mention was made of reaction mechanisms, or reaction intermediates. Challenged with unanswered questions, Olah prepared himself for self-study spending long hours in the library, a habit retained for life. Around 1950 he came across the name of Christopher Ingold – the pioneer of reaction mechanisms and the electronic structure of organic compounds – who defined two types of nucleophilic substitution reactions, $S_N 1$ and $S_N 2$ distinguished by the kinetic order they followed. The $S_N 1$ mechanism followed first order as a consequence of the slow, rate-determining formation of the postulated alkyl-cation intermediate (see Fig. 1). At the time, the isolation of the cation was out of the question.

1 Institute of Organic Chemistry, Research Center for Natural Sciences, Hungarian Academy of Sciences, H-1519 Budapest, P.O.B. 286, Hungary
* Corresponding author, e-mail: simonyi.miklos@ttk.mta.hu
In the lab, Olah was interested in Friedel-Crafts chemistry for alkylaton and arylation of aromatics (Fig. 2). The AlCl$_3$ catalyst – an electron-deficient Lewis acid – probably created a cation in the course of reaction. Olah was eager to find reaction intermediates.

When he read about organic fluorine compounds the idea grabbed him to apply fluorides for alkylation reactions. His first attempt to persuade his professor of the importance of fluorine chemistry was met with refusal, not without reason, however. Hydrogen fluoride is highly reactive, has a low boiling point and extremely toxic. In post-war years, fume hoods were scarce and not effective, so doing fluorine chemistry might well contaminate the laboratory atmosphere. However Zemplén allowed him to use an open balcony badly damaged during the war for his fluorine research. Olah and his associates covered and transformed it to become a "balcony laboratory" where they could start making fluorine compounds.

In 1953 he had his first own course entitled: "Theoretical Organic Chemistry" – physical organic chemistry in today’s terms and the first such course in Hungary – and it had the purpose of filling the gap between the earlier empirical approach of teaching and the new mechanistic-structural aspects and application of physical methods in organic chemistry.

While an enthusiastic teacher of chemistry, it was only secondary to Olah’s research interest in Friedel-Crafts reactions using acyl and alkyl fluorides with boron trifluoride serving to catalyze the reactions (Fig. 3).

In order to study intermediate complexes formed during these reactions Olah considered not only the reactivity of reagents and catalysts, but also the solvent: a transient ionic species would easily disappear in a reactive medium. Therefore, he applied the neat reagent, BF$_3$, as a solvent and thus succeeded in isolating arenium tetrafluoroborates of ionic nature for the first time at low temperatures (Fig. 4).

It was the real start of his long fascination with carbocationic complexes. In 1954 the Central Research Institute for Chemistry (CRICh) of the Hungarian Academy of Sciences was newly established and its head, Professor Géza Schay, invited Olah to join the Institute as deputy director, a telling sign of his reputation at the age of 27. He formed there a research group of 8 members and led a very productive research program for the next two years. It also helped him to emerge somewhat from the depressing isolation imposed upon science by the communist rule. In 1955 he attended an IUPAC meeting in Zurich, his first and only visit to a Western country from Hungary. He also lectured in East Germany (Humboldt University) and was able to see Emil Fisher’s old laboratories. He also visited the Soviet Union on one occasion, and he met there some famous scientists, including Nesmeyanov, a leading organic chemist (director of the Institute of Organoelement Compounds) and a great power in Soviet Science (president of the Academy of Sciences). Besides discussing scientific subjects, their meeting contained gloomy elements, too. “We spoke French and German... but... when somebody came in to join us, he changed to Russian and asked for an interpreter. Even he seemed to be concerned at the time not to be reported for such ‘anti-Soviet’ behavior. In private conversation, Nesmeyanov told me about the recent condemnation of Pauling’s resonance theory. He mentioned that he had not signed the official report orchestrated by the political powers, claiming illness so as not to participate in the process” [2]. Hence Olah obtained insight into a most discouraging political influence on science.

In October 1956, the Hungarian revolution broke out against Soviet rule. During those short-lived days a small revolutionary committee was formed in CRICh, to which Olah was elected, but very soon oppression and terror returned with the Soviet invasion and about 200,000 Hungarians fled the country. Olah with his family (wife and a 2-year-old son) and part of his research group joined the stream of refugees fleeing toward a new life in the West.
3 Starting a new life

The Olah family arrived first in London where a relative lived. Back in Hungary Olah had been an established researcher with recognized achievements, and at first he tried to get an academic position. He met excellent British scientists he knew from the literature (Christopher Ingold, Alexander Todd) and found them helpful, as was the British institution for refugee scientists, the Academic Assistance Counsel, which sent out letters in search of a job and resettlement for Olah. While no academic position came along an industrial research possibility opened up at Dow Chemical.

The Dow Chemical Company – having its base in Michigan US – was just establishing an exploratory research laboratory across the Canadian border in Sarnia, Ontario, where Olah was offered to join the new laboratory. It was located within a chemical plant complex and primarily served industrial interest. Olah accepted the position and kept his own practice of working extra-long hours after his company commitments. He spent the evenings and weekends pursuing personal scientific interests, reading, writing and thinking about new problems. As a fortunate coincidence, Dow used Friedel-Crafts-type chemistry in the manufacture of ethylbenzene for styrene production by the reaction of benzene with ethylene. This alkylation process assumingly involved cationic intermediates (neither observed, nor studied) and therefore Olah’s personal scientific interests first formulated in Budapest could be pursued further with industrial significance. He obtained dozens of patents for the company’s interest and was soon promoted to Research Scientist, a research position without administrative responsibility.

While working in an industrial laboratory, Olah started a seminar program and succeeded in inviting outstanding scientists despite of the location of his laboratory in a small industrial city outside the normal path of academic visitors. Olah was able to publish his scientific results, lecture at international symposia and universities, thus multiplying his personal contacts in the scientific community.

4 The first recognition and breakthrough

While at Dow Chemical, Olah edited a four-volume comprehensive monograph: Friedel-Crafts and Related Reactions published by Wiley-Interscience. In 1963 he won the American Chemical Society Award in Petroleum Chemistry for his work on Friedel-Crafts reactions, a real recognition for one who fled his native country just a few years earlier.

As for reaction intermediates, carbon was long considered to be unable to form ionic compounds, except in highly stabilized cases (triphenylmethyl dyes). In order to create a positive charge on carbon, a strong Lewis acid effectively withdrawing electrons was needed. Carbocations in the 1950s were still generally considered to be unstable and short lived owing to deprotonation to olefins (Fig. 5).

\[ (CH_3)_3C^+ \longrightarrow (CH_3)_2C=CH_2 + H^+ \]  
Fig. 5 The tert-butyl cation deprotonation

The existence of alkyl cations was indirectly deduced from kinetic studies without any reliable spectroscopic or other physical evidence for their presence in solution or in the solid state. It was realized only after Olah found a highly active agent and an inactive environment that suppressed deprotonation that long-lived stable carbocations could be obtained. The agent was SbF_5, a very strong Lewis acid that also served as the medium, and under these conditions Olah prepared tert-butyl hexafluoroantimionate, the first stable, long-lived alkyl cation salt (Fig. 6).

\[ (CH_3)_3CCOF + SbF_5 \overset{\text{CO}}{\longrightarrow} (CH_3)_3C^+SbF_6^- \]  
Fig. 6 The first stable carbocation salt

NMR spectroscopy at the time was still in its infancy, but inventive colleagues applied \(^{13}C\) labeling and the double resonance method for the spectral identification of the stable t-Bu\(^+\) cation in SbF_5, diluted by solvents, such as liquid SO_3, SO_2ClF or SO_2F_2, conditions that allowed the solution to be cooled down to –78 °C or even lower temperatures. The NMR spectra unequivocally proved the existence of the t-Bu^+ cation.

It was followed by the preparation of tert-amyl cation and isopropyl cation. Analysis of their NMR spectra also revealed that the carbon atoms of originally sp\(^3\) hybridization, in cationic form were changed to sp\(^2\), i.e. the neutral tetrahedral carbon transformed to a planar cation. These results were presented in public for the first time at the Brookhaven Organic Reaction Mechanism Conference held in 1962, followed by publications in scientific journals. Thus, the existence of alkyl cations first envisaged in Budapest and pursued further during subsequent years in America, was finally proven.

5 The superacids

After seven successful years, Olah left Dow Chemical and became professor and chairman of the chemistry department at Western Reserve University in Cleveland, Ohio. He could devote his full capacity to scientific research. As strong Lewis acids proved indispensable to produce stable carbocations, the search for superacids (i.e. acids much stronger than concentrated sulfuric acid) became a central problem for his department. Besides the originally applied SbF_5 system, they found protic superacids (FSO_3H, CF_3SO_3H) and conjugate acids (HF-SbF_5, FSO_3H-SbF_5, CF_3SO_3H-SbF_5, and CF_3SO_3H-B(O(SCF_3)_3) to be extremely useful in low-nucleophilicity solvents (SO_3, SO_2ClF, SO_2F_2). In a few years they could prepare a large variety of stable carbocations (a few samples are illustrated in Fig. 7) [3].

The Legacy of George Olah
Starting from 1962, the chemistry community witnessed a scientific debate about the structure of the norbornyl cation between Herbert Brown and Saul Winstein – both prominent chemists – and Olah became inadvertently involved in it. The facts were clear: 2-exo-norbornyl brosylate ($p$-bromobenzensulfonate) solvolyzed in acetic acid much faster than the 2-endo-stereoisomer (Fig. 8).

The adversaries in the debate gave different explanations for the same facts. According to Brown, the solvolysis rate of the endo-stereoisomer is slow due to the sterically hindered approach of acetic acid and the intermediate should be a tri-valent classical ion. On the other hand, Winstein argued that the fast reaction of the exo-isomer can be explained by assistance provided by neighboring σ-bonds resulting in a bridged carbonium ion intermediate. Neither of the two chemists could experimentally prove his claim. Since Olah’s group had developed experimental techniques to prepare long-lived carbocations, they were able to apply them to the norbornyl cation by three separate routes [4] (Fig. 9).

It was only in 1969 that Olah could perform the necessary NMR studies, and was thus able to determine the structure of norbornyl cation, which gave full credit to Weinstein’s supposition. The controversy with Brown became highly inconvenient but finally turned out to be a blessing in disguise: the norbornyl cation opened up a new domain of carbocations, in which the carbon had more than four neighbors (Fig. 10).

The hypercoordinated carbon (numbered 6 in Fig. 10) characterized by two-electron three-center (2e-3c) bonds demonstrates electrophilic reactions at single bonds in saturated hydrocarbons. Realizing this in 1972, Olah classified two distinct classes of carbocations:

1) Trivalent carbenium ions containing an $sp^2$-hybridized electron-deficient carbon atom, which is usually planar in the absence of steric interference. The carbenium carbons contain six valence electrons, and are therefore highly electron deficient. Their structure contains only two-electron two-center bonds. Their representative basic structure is $CH_3^+$.

2) Penta- (or higher) coordinate carbonium ions contain five- or higher-coordinate carbon atoms. The carbonium ion cannot be described solely by two-electron two-center single bonds. It also necessitates the use of two-electron three- (or multi-) center bonding. The carboxation center always has eight valence electrons, but overall they are electron deficient because of sharing two electrons among three (or more) atoms. Their basic representative structure is $CH_5^+$ [5]. These ions can be formed in electrophilic reactions of σ-donor saturated hydrocarbons through the ability of C-H or C-C single bonds to participate in carbonium ion formation [6]. Some characteristic two-electron three-center bonding types are represented in Fig. 11.

6 The Hydrocarbon Institute

After 12 years in Cleveland, Olah moved with his research group to the University of Southern California (USC) in Los Angeles. USC offered a limited fund to build an Institute that needed additional donations, which were provided by philanthropists (Donald and Katherine Loker), and thus the institute was later renamed as the „Loker Hydrocarbon Research Institute“.
The major mission of the new institute was the study of the fundamental chemistry of hydrocarbons in the broadest sense including hydrocarbon transformations to develop new fuels, synthetic reagents and functional polymeric materials. The work also aimed at the education and development of future generations of researchers by providing varied courses, guiding graduate students, and organizing international symposia.

The hydrocarbon research program utilized results of earlier fundamental work on superacid-catalyzed reactions in order to develop practical processes, e.g., an environmentally friendly practical alkylation of isobutylene with isobutane (Fig. 12) without the use of toxic additives.

In this alkylation, isobutane only acts as a hydride transfer agent, and the source of the tert-butyl cation formed via intermolecular hydride transfer. Superacidic catalysis proved also useful for finding new ways of hydrocracking coal, shale oil, tar sands and heavy petroleum residues.

Fig. 11 Two-electron three-center bonding types

Fig. 12 Industrial production of isoctane [7]

7 The Nobel Prize

In 1994, the Nobel Prize in chemistry was awarded to Olah as the sole recipient. The Press Release of the Royal Swedish Academy of Sciences detailed his scientific achievements:

In the early 1960s Olah and co-workers discovered that stable carbocations could be prepared through the use of a new type of extremely acidic compounds - far stronger than ‘classical’ acids like sulphuric acid. These new acids became generally known as ‘superacids’.

Olah’s discovery completely transformed the scientific study of the elusive carbocations. Since the original discovery a large number of carbocations have been prepared and their properties studied in great detail. Olah has also shown how basic knowledge on superacids and carbocations can be applied to the facile synthesis of new and important organic compounds. His work has resulted in new methods for the conversion of straight chain hydrocarbons (when used in combustion engines these have very low octane number and they are also difficult to degrade biologically) into branched hydrocarbons that have high octane numbers and are more easily biodegradable.

Olah’s extremely important contribution lies in the methods he evolved for developing carbocations in high concentrations and under conditions which give them long life. To achieve this, he used solvents which were so extremely little nucleophilic that they did not attack carbocations. Such solvents are SO₂, SO₂ClF and SO₂F₂ in which, at least at temperatures around minus 100°C, carbocations do have long life.

Investigations of the substitution reactions and rearrangements of 2-norbornyl derivatives led S.Winstein to suggest in the early 1950s that the intermediate carbocation was non-classical and contained a pentacoordinated carbon. This interpretation was questioned by H.C. Brown, who received the 1979 Nobel Prize in Chemistry for his development of boron compounds into important reagents in organic synthesis. Brown claimed that the 2-norbornyl cation did not have carbonium ion structure but was a rapidly equilibrating carbenium ion. The ensuing scientific controversy lasted until about 1980. As the structures of carbonium ions were of great theoretical interest, the problem fascinated many leading physical organic chemists, yet despite great efforts and many ingenious experiments, no definitive solution was found until the 2-norbornyl cation could be directly studied with NMR-spectroscopy. Olah and his co-workers finally observed the 2-norbornyl carbocation in a solution of SbF₅·SO₂ClF·SO₂F₂ at -158°C. Both 1,2-hydride shifts and more complicated rearrangements at this low temperature are slow enough not to disturb the interpretation of the NMR-spectra. The spectra accorded completely with Winstein’s symmetrical bridged structure, with a pentacoordinated carbon and not with a rapidly equilibrating carbenium ion. Molecules containing pentacoordinated carbon atoms are no longer an exotic curiosity in organic chemistry” [8].

The Nobel Prizes are awarded each year on December 10, the anniversary of Alfred Nobel death. Olah received his medal and diploma from the Swedish King, with feelings described in his autobiography: “The significance of the moment all at once struck me. The long journey started in Budapest flashed through my mind and also some doubt about whether all this
was real and was in fact happening.” [9]. Considering the timing of the event, Olah might have also felt something else: exactly 50 years earlier in Budapest, at the height of fascist terror and the impending siege by the Soviet Army, bare life had been at stake.

8 In the service of Mankind

The Nobel Prize puts new responsibilities and preoccupations on the winners, but Olah strongly resisted changing his earlier life and continued his research activities, instead of resting on his laurels. One new achievement was finding solid superacid systems in resins [13] and zeolites showing remarkable catalytic activity in hydrocarbon transformations. Olah’s main aim in post-Nobel years was to address problems global population would face in the 21st century: the ever increasing use of finite hydrocarbon resources, and the warming of the global climate caused by greenhouse gases, methane and CO₂.

The solution Olah and his coworkers proposed was the Methanol Economy [10]. With superacid catalysis, the conversion of CO₂ to methanol, from which ethylene and propylene could be obtained, provided raw materials for the preparation of a whole array of hydrocarbons from gasoline to polymers. Their most successful process, oxidative bi-reforming proved to be useful for industrial methanol synthesis in high yield and selectivity (Fig. 13) [11, 12]. The World’s largest CO₂-to-methanol plant, named George Olah Renewable Methanol Plant in Svartsengi, Iceland began production in late 2011 and was completed in 2012.

The industrial success was recognized in 2013 by giving the Eric and Sheila Samson’s Prime Minister’s Prize for Innovation in Alternative Fuels for Transportation to George Olah and his coworker, G. K. Surya Prakash (once a graduate student of Olah, later a successful professor in the Loker Hydrocarbon Institute) (Fig. 14). The Samson Prize was established by the State of Israel in order to reduce the global dependence on oil.

9 On a Hungarian Myth

There is a popular belief – a kind of boasting with a touch of self-pity – saying: So many Nobel Prize winners have been given by us to the world. The weakest point in that is the verb given. But there are two real questions here to be answered.

1) What has been the reason for highly talented young people growing up in a number higher than proportional to the population of Hungary? The answer is: there have always been excellent, unselfish and devoted teachers stimulating interest in their pupils, teaching them the critical essentials and letting them think and work on their own.

2) What have been the reasons why highly talented adults left the country? Let us try to find the answer by comparing Olah’s career with the fate of the Institute (CRIC) he left. Had he remained at home,

a) political reprisal would have inevitably forced him to quit his position of deputy director.

b) CRIC moved into its final premises and professor Schay strengthened by a new deputy with strong political background and moderate scientific capabilities.

c) the new deputy in CRIC grew impatient and forced Schay into early retirement. The Institute then lost a man who had been a partner of foreign visitors working in any of the various areas of chemical science.

d) the new director of CRIC had to quit for incompetence and received a university chair.

In 1969, when the structure of the 2-norbornyl cation was solved by Olah,

In 1972, when Olah defined two distinct classes of carboxations – the trivalent planar cation and the penta- or higher coordinate non-classical cation,

In the mid-1970s, when Olah moved to California,
e) CRIC was compelled by politically fattened academic bureaucracy to prepare medium-term (5 year) and long term (10-20 year) scientific plans which interfered with research activity and produced mountains of paperwork that – after some years – ended in the dust bin.

In the 1980s, when the Loker Hydrocarbon Institute started its successful history under the leadership of Olah

f) new generations having grown up in CRIC recovered from the unscientific interference of earlier years and achieved international recognition.

In 1994, when Olah received the Nobel Prize,

g) CRIC – having lost support from Hungarian pharmaceutical companies owing to privatization – found itself in miserable financial conditions. In addition, CRIC became the target of political and even professional attacks labeling it a remnant from the Stalinist era.

In the first decade of the 21st century CRIC changed its clumsy name to Chemical Research Center (CRC) and its position improved,

h) Olah visited Hungary and gave lectures both in the Academy of Sciences and in CRC about his Methanol Economy.

In 2013, when the Iceland-based „George Olah Carbon Dioxide to Renewable Methanol Plant” operated at full capacity, i) CRC became amalgamated with other Institutes into a newly established Research Center for Natural Sciences (RCNC). The reorganization was accompanied by an unforeseen event: the chemical library of CRC – enriched through more than fifty years and having become second only to the Central Library of BTU in the country – was demolished and transferred to oblivion. The reaction to that event by Olah (who used part of his Nobel money to establish a library in the Loker Hydrocarbon Institute, and also a chemistry prize in Hungary) might have been: ‘Novelties seldom happen in social life; just recall the fate of the ancient library in Alexandria’.

j) Finally a word on the newly established RCNS that proved to be a hard nut to crack for its first general director, who had to quit after three years. One is tempted to cite Alexander Todd, late professor in Cambridge (highly revered by Olah), who told Alan Katritzky, the chemistry chair of the new University of East Anglia: „if your plans work out properly, in a few centuries it indeed may become a good university” [14]. So, let RCNS develop peacefully, like the British lawn.

10 Conclusions

In a small and poor country where society has been the subject of severe political afflictions by different ideologies, most people have suffered injustice. The majority became defensive, while some found ways to compensate themselves at the expense of the community. Some even enjoyed gloating at the misfortune of others. But – fortunately – there have always been those who kept their humanity and showed solidarity with vulnerability, suffering and helped survival. All in all, the social environment in Hungary for the last one hundred years has often been inadequate for people of cultural excellence in music, fine arts and science, including Nobel Prize winners.

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