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GEORG WITTIG (1897–1987)

A life of chemical fantasy become reality

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In scientific research, as Albert Einstein has emphasized, imagination is more important than knowledge. Groping after the unknown, the scientist must be able to conjure up some guiding image of the elusive quarry. In this philosophical sense, any great discovery must first exist in the mind before it can be found to exist in external reality. It follows that great scientists give their fantasies both free rein and respectful attention. Such a man was Georg Friedrich Karl Wittig, who lived to see his chemical fantasies become tangible scientific facts.

As Wittig set off on his chemical odyssey in the 1920s, such chemical structures as carbenes, carbon radicals, charged carbon centers and hypervalent compounds were widely considered as chimeras of "unsound" chemists. But over the next six decades of the twentieth century Wittig would contribute to making carbenes, arynes, ylide complexes, unusual carbanions and hypervalent compounds of groups 15–17 routine reagents in organic chemistry.

Such achievements and analogous successes by other chemists in the areas of free radicals, carbonium ions, multiple bonds between two metals or two nonmetals and novel strained rings have ushered in the era of reactive intermediates in organic

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chemistry. As the twentieth century draws to a close, the attitude now reigning in chemical research is pronouncedly euphoric: "Whatever structures a chemist's imagination can properly conceive, an ingenious and persistent experimentalist can probably prepare in the laboratory."

Although many chemists throughout the world have made brilliant contributions to the rapid progress of chemistry, Georg Wittig stands out as one of the select few who have truly blazed new trails in research. In fact, from the beginning of his professional career Wittig pursued lines of research well outside the mainstream of conventional organic chemistry and gave little heed to fashions in research, technical applicability or outside recognition. Indeed, he was to spend half his career (1926–1953) quietly pursuing his curiosity on the behavior of organolithium compounds, organic free radicals, autooxidation, ylides and hypervalent compounds, before the implications of his research achievements attracted much outside attention. As if in compensation, however, honors came to Georg Wittig in swelling numbers after the early 1950s. The Nobel Prize, which he shared with Herbert Brown in 1979, was the culmination of the tardy recognition he was to receive during his entire career. Yet professional recognition was a reward that his modest nature would never seek and once it came, it found him unchanged. Throughout his professional life he retained a zest for keen experimental work and a burning zeal to impart such a love of research to his many graduate students and postdoctoral associates. He often cautioned his students not to be in a hurry to publish many scientific articles, nor to be anxious to have an experiment turn out in the intended manner. He saw in slow, careful laboratory work and in disappointing experimental failures not just a cause for frustration, but rather a rare opportunity to discover something really novel, something totally unexpected. In this romantic view of research is revealed Wittig's artistic temperament.

Born in Berlin on June 16, 1897, Wittig was reared in an environment where the artistic muse was revered [1]. His father was an artist and Professor in the Kassel Applied Art Academy and his mother's devotion to music was to be emulated by the young Georg. He enjoyed playing the piano and even tried his hand at composing an overture. Indeed, at an early stage music vied with chemistry for his professional affection. His affinity for the arts and humanities was further augmented by completing the *Abitur* at the humanistic Gymnasium at Kassel in 1916. A further influence on his developing humanity can be discerned in his love of nature, which was awakened by hiking excursions undertaken while a member of the *Wandervögel* (migratory birds), a German youth group akin to the Boy Scouts of other countries. This acquired sense of the beauty and liberating spirit of the great out-of-doors was to remain keen throughout Wittig's life and to be manifest in his love of mountaineering. Periodically during his arduous professional life, he found that an excursion into the mountains would purge his mind of technical issues and permit him to gain a fresh and wider perspective. The painting by Caspar David Friedrich (1774–1840), which shows a lonely climber on a mountain peak looking out over the fog-shrouded slopes below him, conveys that sense of serenity and detachment that Wittig always found in Nature.

The same interest in Nature ultimately proved decisive in his choice of chemistry as a career. From his earliest experiments in his home laboratory, he was fascinated by the color changes that simple household items such as vinegar and washing soda could cause in a water extract of red cabbage. This and similar experiments were described in a book, "Chemische Experimente aus dem Alltag" (Chemical Experiments from Everyday Life). It is noteworthy that many famous chemists, such as Wittig's contemporary, Karl Ziegler, were drawn to chemistry by such home laboratory experiences. This illustrates the importance of science experimentation at an early age, before a child's inborn curiosity is stifled by rote learning.

Before he could pursue the study of chemistry, however, Wittig was inducted into the German Army after leaving the Gymnasium in 1916. He saw the end of the Great War as a prisoner of war under the English and it was 1920 before he could commence his studies at the University of Marburg. His experiences with the difficulties of being a returning war veteran with limited resources was to make him unusually empathetic with the war veterans he was to receive as students after 1945. His own doctoral research was directed by Karl von Auwers (1863–1939) on the preparation and properties of aryl-substituted quinones. Upon the awarding of the doctorate in 1923, Wittig was advised by von Auwers to undertake a *Habilitationsarbeit*, an independent program of research that is the prerequisite for a young academician who aspires to teach at a German university. In Wittig's case he completed a preparative study in the area of chromones and coumarins and in 1926 became a Privatdozent. He subsequently spent six years at Marburg, where he began to develop an interest in stereochemistry, free radicals, ring strain and the reactivity of organometallic compounds. Wittig's growing awareness of the theoretical aspects of organic chemistry was fostered by friendships struck at Marburg with Karl Ziegler (1898–1973), with Walter Hückel (1895–1973) and with Hans Meerwein (1879–1965), the last of whom was von Auwers' successor at Marburg.

In 1932 Wittig was appointed Ausserplanmässiger Professor and Division Head at the Technische Hochschule in Braunschweig. Here he finally had the freedom and resources to launch his independent research and he found in Karl Fries, Head of the Institute, a most stimulating colleague for his developing ideas. Unfortunately, the next five years was to be a time of great turmoil, as the Nazi State began to intrude into the operation of the universities. When Fries came under sharp attack in 1937 by the National Socialists, who wished to introduce party doctrines into the university, Wittig stood resolutely at Fries' side. It is melancholy to note that in 1938 Karl Fries was forced to relinquish his professorship for political reasons and his research activities were brought to a halt *. But his name lives on in the world chemical literature for the structural rearrangement he discovered.

Wittig was subsequently able to escape this oppressive atmosphere when he was appointed Professor Extraordinarius in Freiburg. Here the Head of the Institute, Hermann Staudinger (1881–1965), the future Nobel laureate, opened up a new perspective in Wittig's thinking, namely macromolecular chemistry. The atmosphere that Staudinger helped create at Freiburg was most conducive to the development of Wittig's scientific ideas and political interference by the German Government was barely noticeable. Then, in 1944, Wittig received a call as Professor Ordinarius and Head of the Institute at the University of Tübingen. Finally, in 1956 he assumed the position as Head of the Chemical Institute at the University of Heidelberg, where he

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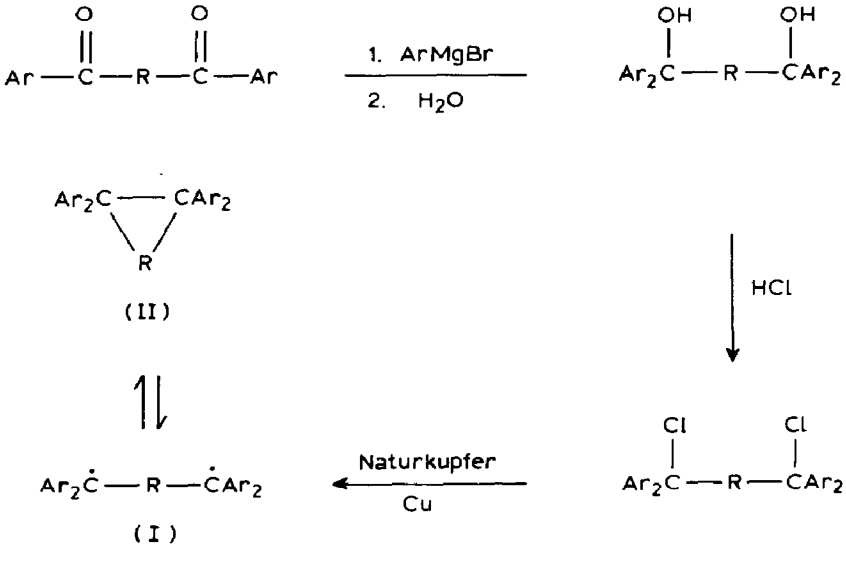
* An appreciation of the contributions of Karl Fries to chemistry and a short biography have been published by Dr. Herbert Bestian, one of Fries' last research students: Chem. Ber., 117 (1984) XXIII–XLI.

remained until his formal retirement from administration in 1967. However, he continued to be active in research and publication for more than a decade thereafter. His love of putting his mechanistic and structural ideas to the experimental test remained keen throughout his career and was the priceless legacy he bestowed on all his students and associates.

The unfolding of Georg Wittig's scientific career is instructive for young chemists about to set out on a life of research and wondering how to identify significant problems worthy of study. Topics of research, like styles in clothes, are subject to fashion and what is today le dernier cri is tomorrow's tedium. Wittig, on the other hand, generated the evolving themes of his research from his own observations in the chemistry laboratory. He brought to such observations a mind greatly influenced by the ideas of his colleagues, whose opinions and critical judgments he so zealously sought out early in his career. From Karl Ziegler, whose studies with organolithium compounds and carbon-centered radicals were to make him a chemistry prodigy in his 30s, Wittig was made aware of trivalent and anionic carbon intermediates; from Hans Meerwein and his masterly researches in carbon-skeletal rearrangements, Wittig came to appreciate the role of positively polarized carbon and other nonmetal salts; from Walter Hückel and his contributions to structural theory, Wittig recognized the importance of stereochemistry and bond strain on the relative stability of isomeric structures. Further contacts with Karl Fries and Hermann Staudinger deepened his interest in such fundamental themes as rearrangements, divalent carbon and the association of monomeric organic molecules into larger aggregates. But although one can trace the seminal sources of Wittig's ideas, it was his genius to integrate these concepts and apply them to the unusual reactions he encountered in the course of investigations carried out with an entirely different goal in prospect. It was with this in mind that Wittig often counseled his students to be disappointed if an experiment runs strictly in accord with expectations. He pointed out that such an outcome probably arose from a prosaic, unimaginative posing of an experiment; he realized that to an imaginative, well-informed chemist,



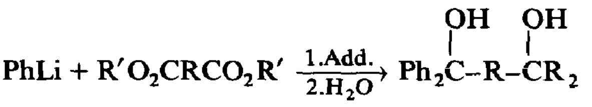
Scheme 1

an unexpected experimental result is invariably more exciting and probably more significant. Such results reveal what is truly new.

To Wittig, then, it mattered not what aspects of chemistry one began to investigate, so long as genuine curiosity and careful observation were brought to bear. These attitudes were bound to yield rich rewards of interesting and significant findings, if their implications were pursued with imagination and persistence. To illustrate this process, Wittig could have done no better than to trace his own pilgrimage through the maze of early twentieth century organic chemistry. Beginning with an extension of his Habilitationsarbeit on chromones, he was curious about the possibility of preparing 1,3-diketones from the action of metal alkoxides on such chromones. From that study emerged an interest in the stereochemistry of the dioximes of such ketones and how the structure of the possible oximes could be established [2]. Undoubtedly through contact with his close friend and colleague, Karl Ziegler, Wittig then investigated how a variety of diketones could be utilized for preparing a variety of biradicals by the well-established sequence depicted in Scheme 1 [3–6]. In a wide-ranging variation in the nature of the R-group, Wittig and co-workers studied the influence of the spatial separation of the radical centers on biradical formation (I) or covalent ring formation (II) [7–9]. He was to return to the theme of highly arylated hydrocarbons later in his career when he studied the transition metal-promoted coupling of *o*-dilithiobenzene and of 2,2′-dilithiobiphenyl for the formation of unusual hydrocarbons rings [10]. The availability of such potentially biradical hydrocarbons as II prompted Wittig to probe the role of radicals in autooxidation [11,12] and radical-chain polymerization [13].

But it was in a search for a reagent to convert esters more efficiently into glycols, which were intermediates in such radical systems (Scheme 1), that Wittig found his favorite class of reagents, organolithium compounds. Noting that in 1930 Ziegler had discovered a highly convenient route to organolithium compounds, from the organic halides and lithium metal and ether (Scheme 2) [14], Wittig prepared phenyllithium in this manner and found it greatly superior to the Grignard reagent

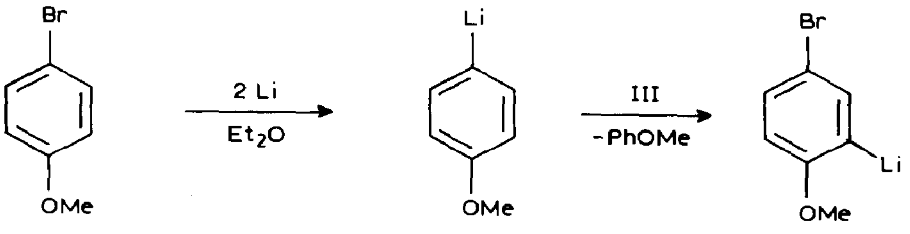
$$\mathrm{PhBr} \xrightarrow[\mathrm{Et_2O}]{\mathrm{2Li}} \mathrm{PhLi \cdot LiBr}$$



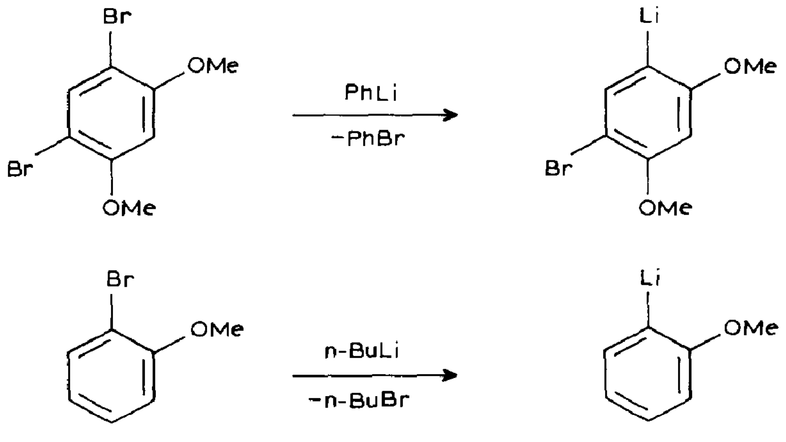
$$\mathrm{PhLi + R'O_2CRCO_2R'} \xrightarrow[\mathrm{2.H_2O}]{\mathrm{1.Add.}} \mathrm{Ph_2\overset{OH}{\overset{|}{C}}{-}R{-}\overset{OH}{\overset{|}{C}}R_2}$$

Scheme 2

for converting diesters into glycols (Scheme 2) [15]. When he attempted to use this approach for appending *p*-anisyl groups to carbonyl centers, Wittig encountered the unexpected: *p*-bromoanisole (III) was converted, in part, not only into *p*-anisyllithium (IV), but also into 2-bromo-5-anisyllithium (V) (Scheme 3) [16].



Scheme 3



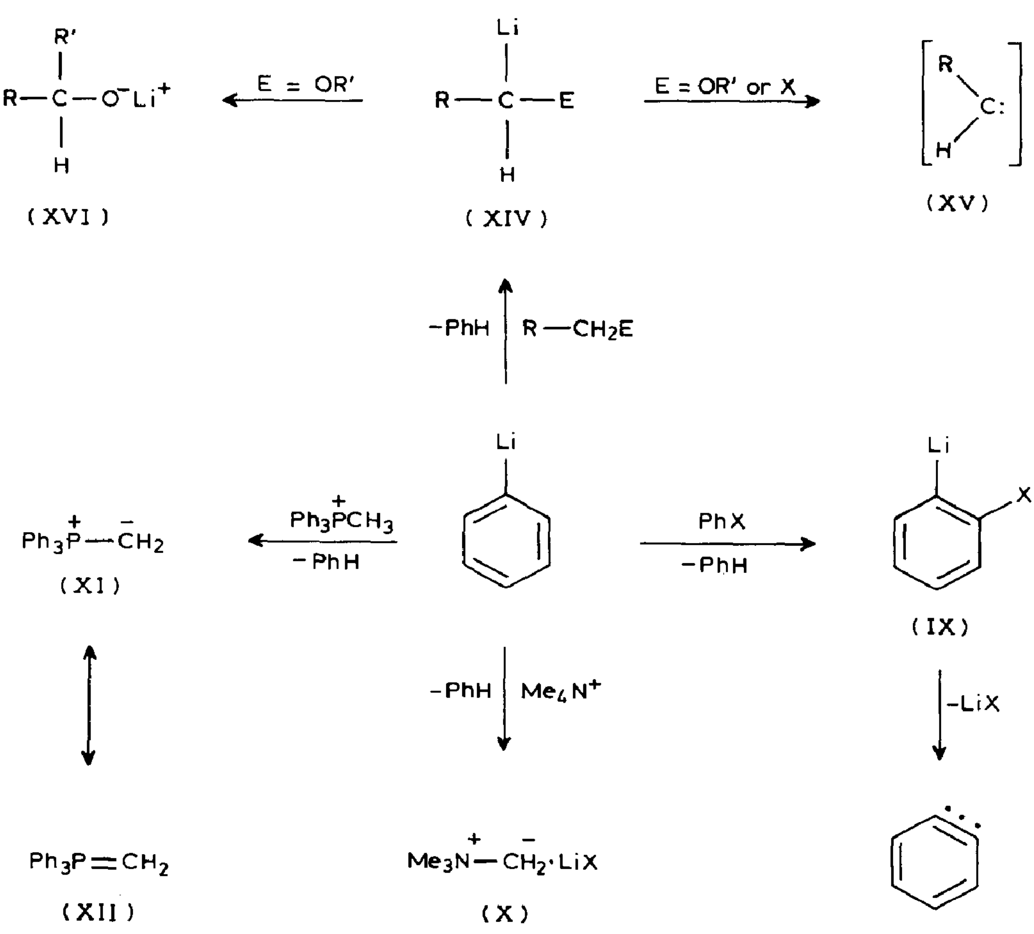
(VI) (VII)

Scheme 4

Wittig correctly deduced that a hydrogen–lithium exchange had taken place between IV and III to generate V and anisole. Such a lithiation at an unactivated C–H bond had been discovered in Henry Gilman's (1893–1986 [17]) research group in 1934 [18], but had been described in an unpublished doctoral dissertation. Gilman and Bebb finally did publish such a lithiation of dibenzothiophene in 1939 [19] and Gilman, Moore and Baine a lithiation of dibenzofuran in 1941 [20,21]. Nevertheless, Wittig's discovery, which was published in 1938 [16], must be recognized as a parallel, independent contribution. In the same article, moreover, Wittig also reported the discovery of the halogen–lithium exchange reaction, when he observed that phenyllithium reacted smoothly with 4,6-dibromoresorcinol dimethyl ether in the manner shown in Scheme 4. Here again, he was co-discoverer with Henry Gilman of an extremely important organolithium transformation, for in the same year Gilman described the conversion of VI into VII by n-butyllithium [22,23].

In Wittig's hands, phenyllithium now served as the "master key" for opening up hidden possibilities in areas of unusual, highly reactive carbanionic intermediates (Scheme 5) [24]. Through phenyllithium's proton-abstracting ability toward weak carbon acids, Wittig was able to convert haloarenes into *o*-lithiohaloarenes, which were shown to serve as aryne precursors (IX) [25]; quaternary ammonium salts were converted in trialkylammonium methylides (X) [26]; phosphonium salts were analogously transformed into phosphonium ylides (XI) or phosphoranes (XII) [27]; and alkyl ethers or halides (XIII, $E = OR$ or X) underwent α-lithiation [28]. The lithiated intermediate (XIV) was shown to undergo either α-elimination to produce carbenes (XV) or rearrangement in the case of ethers to yield alkoxides (XVI) [30]. The latter novel transformation was recognized in the chemical literature as the Wittig ether rearrangement [31].

Such anionic rearrangements, as first uncovered by Wittig and co-workers and subsequently developed by others [32–34], inspired a host of studies into the mechanistic basis of such transformations and their utility in organic synthesis (Scheme 6). From such investigations, it has become evident that although such rearrangements clearly involve carbanions as initial intermediates, free radicals are

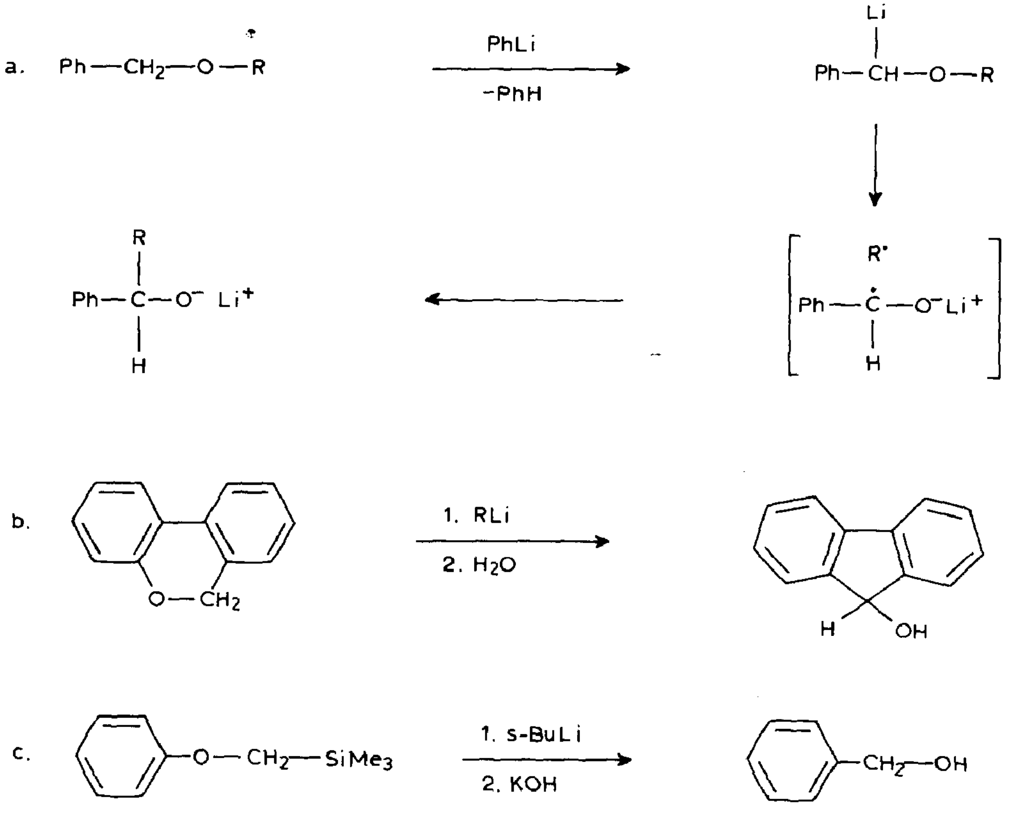


Scheme 5

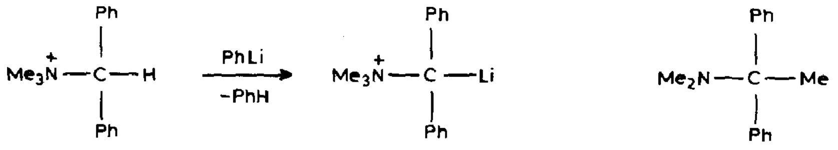
generated in the course of reaction. In subsequent extensions to nitrogen analogues of ethers, Wittig was able to demonstrate the role of carbanions both in the Stevens (eq. 1) and the Sommelet (eq. 2) rearrangements [35], although here again radicals may be the crucial intermediates [36].

By uncovering the possibilities of carbanionic rearrangements, Wittig demonstrated the rearrangements possible with negatively polarized carbon [37], thus establishing a pleasing parallel to the chemistry elucidated by his colleague, Hans Meerwein, for positively polarized carbon. The latter has likewise gone into the literature as a name reaction, the Wagner–Meerwein rearrangement [38].

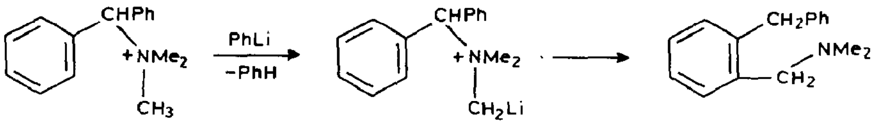
Besides carbanionic rearrangements, however, all other transformations depicted in Scheme 5 have had a profound impact on different aspects of modern organic chemistry. First, the α-elimination of an α-substituted alkyllithium to generate divalent carbon (XIV → XV) has been employed in countless studies for the synthesis of carbenes and subvalent intermediates of many other elements (RN, R_2Si, RB, etc.) [39]. Secondly, the β-elimination of an *ortho*-lithiated haloarene to form an aryne has become a standard route to produce benzynes for synthetic applications or mechanistic studies [40]. Furthermore, strained C=C or C≡C bonds in rings or in highly substituted sites can be formed by analogous β-elimination, as



Scheme 6

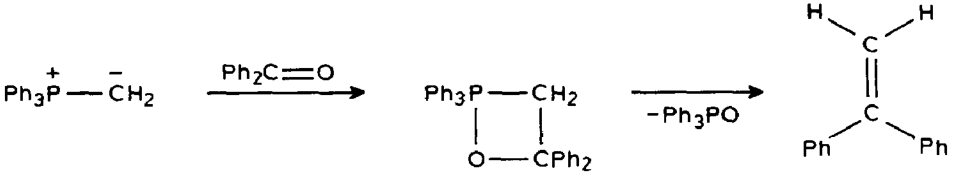


(1)



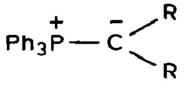
(2)

Wittig himself was able to show [41]. Thirdly, the generation of zwitterions involving carbanionic carbon (X and XI) has had far-reaching consequences, in addition to their role in rearrangements (eqs. 1 and 2). The role of such ylides, as Wittig has termed them, in organic synthesis has been the subject of monographs [42] and lengthy reviews [43], and the nature of the positive center in these zwitterions has been varied among N, P, As, Sb, S and neutral Si, Al or a transition metal. Possibly the single most astonishing reaction of such ylides is the olefination reaction. In the prototypical example discovered by Wittig and Geissler in 1953 [44], the phosphorus ylide XI smoothly converted a carbonyl compound into an olefin (eq. 3). Since its

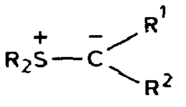


(3)

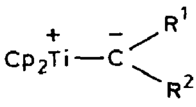
discovery, the Wittig reaction, as it was termed by appreciative colleagues, has found numerous applications in the synthesis of natural products, such as vitamin A, especially since the substituents on the anionic carbanion can be varied widely (XVI). In effect, it serves as a source of the carbene unit, CR$_2$, in synthetic transformations. As already alluded to, the phosphorus center can be exchanged for a wide variety of nonmetals (XVII) and metals (XVIII), to yield ylides of methylenes of modified reactivity [43].



(XVI)



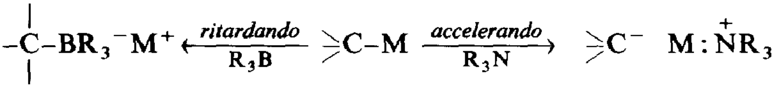
(XVII)



(XVIII)

The discovery of the ylides was made during Wittig's attempts to synthesize, not the carbene sources he found, but rather unusual, hypervalent compounds of main-group elements [45]. The nitrogen and phosphorus ylides, X and XI, were encountered in experiments designed to produce the pentavalent derivatives, R$_5$N and R$_5$P. Although he never succeeded with nitrogen, Wittig did eventually succeed, with the aid of his favorite reagent, phenyllithium, in preparing Ph$_5$P [46], as well as Ph$_5$As [47], Ph$_5$Sb [48], Ph$_5$Bi [49], Ph$_4$Te [50] and Ph$_3$I [51]. The explosive character of triphenyliodine showed that the limits of kinetic stability had been reached.

These investigations into hypervalent compounds aroused Wittig's interest in the complexes that such phenyl derivatives might form with each other. Noting that phenyllithium is dimeric in ether and that it forms 1/1 complexes both with diphenylberyllium and with triphenylborane, Wittig viewed such associations as charge-separated metal cation-metallate anion complexes (XIX–XXI), or "ate" complexes [51]: $\mathrm{Li^+LiPh_2^-}$ (XIX) < $\mathrm{Li^+BePh_3^-}$ (XX) < $\mathrm{Li^+BPh_4^-}$ (XXI). The chemical reactivity decreased from XIX to XXI because of the greater electronegativity of boron and its completed electron octet. Wittig discerned parallels and contrasts between such "ate" complexes or associations and the well-established "onium" complexes, as exemplified by tetraalkylammonium salts. He then employed the complexation leading to "ate" or "onium" complexes to elucidate the effect of Lewis acids and bases on the reactivity of the carbon–metal bond (XXII). In this view, an amine should accelerate reaction by forming an "onium" complex with the metal and thus liberating the carbanion (XXIII). A Lewis acid, such as Ph$_3$B, should, on the other hand, retard reaction by forming a stable complex with the carbanion (XXIV). It reveals Wittig's humanistic education that he chose the musical terms, *accelerando* and *ritardando*, to describe these effects:

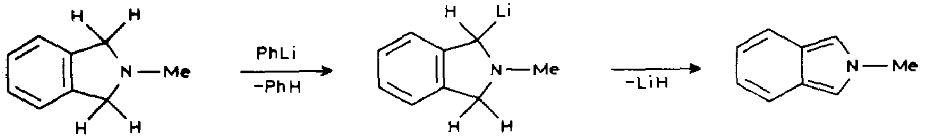


$$-\overset{|}{\underset{|}{C}}-BR_3{}^-M^+ \xleftarrow[R_3B]{\textit{ritardando}} >C-M \xrightarrow[R_3N]{\textit{accelerando}} >C^- \quad M:\overset{+}{N}R_3$$

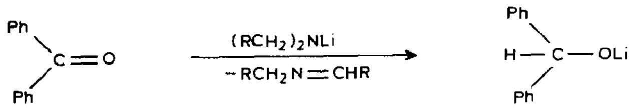
(XXIV) (XXII) (XXIII)

Wittig's analysis of the effects of electron donors and acceptors on reaction rates in organometallic chemistry has proved to be extremely valuable in understanding solvent effects on carbanionic reactions. Where carbanion formation is less likely, as with organoaluminum compounds, Wittig's predictions do not apply. Ethers and amines, for example, strongly retard the addition of R_3Al to olefins or acetylenes [52,53].

What was characteristic of Wittig's research was that interesting observations made in the course of one investigation then determined the direction of future studies. Not only his excursions into ether rearrangements, ylides and arynes, but also his work on amine rearrangements yielded abundant clues on undiscovered, promising chemistry. Through studies of the Sommelet and Stevens reaction [35] (eqs. 1 and 2), Wittig considered that anionic nitrogen intermediates might eliminate hydride ion. From this realization stemmed the development of a novel synthesis of the isoindole nucleus [54] (eq. 4) and of lithium dialkylamides as reducing agents [55] (eq. 5).

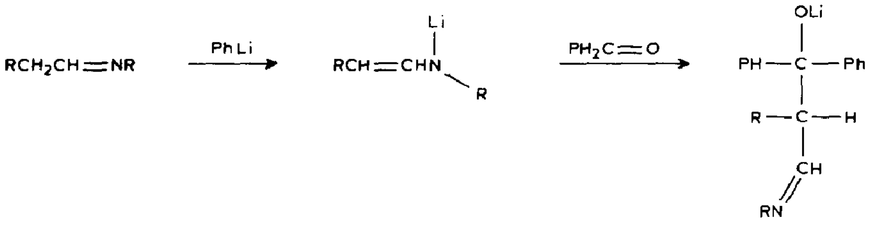


(4)



(5)

The ability of aldimines, $RCH_2CH{=}NR$, in turn, to undergo lithiation then opened up the possibility of conducting directed (that is, component-controlled) aldol-like condensations (eq. 6) [56]. As was evident in all his work, Wittig seldom failed to grasp the significant implications of one finding for future experimentation.



(6)

During his career Georg Wittig explored the implications of his tireless and enthusiastic experimentation with over 300 students who came to his laboratory from throughout the world. The results of those fascinating investigations have been published in almost 300 articles and reviews that emerged from Wittig's pen over a

56-year period. Every Wittig publication bears the hallmark of the absolute primacy given to careful experimentation and scrutinizing observation. Prevailing theory was always acknowledged but never was considered as a reason for not performing an appealing experiment. His students learned that the court of last resort in any scientific discussion was not the textbook, but the laboratory.

The scientific community was slow to recognize the importance of Wittig's research. He was 55 before the first major recognition was bestowed, the Adolf von Baeyer Memorial Medal in 1953. A large number of honorary memberships in learned societies followed over the next two decades, and then finally a series of awards, the Otto Hahn Prize in 1967, the Paul Karrer Medal in 1972 and the Karl Ziegler Prize in 1975, culminating in the Nobel Prize in 1979, awarded jointly with Herbert C. Brown. The Journal of Organometallic Chemistry was fortunate to claim him as a founding member of its Editorial Board.

It would certainly be superfluous to add to the applause of Wittig's achievements, now when all of their implications are evident. It is more to the point, I judge, to applaud the characteristics of Wittig's work that have gone into these discoveries: namely, the courage to undertake unfashionable areas of research, solely for the love of fundamental truth; the humility to be guided by experiment, rather than pressing upon Nature some preconceived notion; and the logic and perseverance of following the leads that Nature points out.

Despite his dedication to chemistry, Wittig the scientist never overshadowed Wittig the humanist. As any of his students or colleagues can recall, he maintained a lively interest in the arts and in Nature throughout his life. With his wife, Waltraut, he found in these activities the revivification his spirit and imagination sought. With her death in 1978 Wittig lost a soulmate, who shared his enthusiasms for science and the arts. For this reason, he encouraged his students to seek recreation outside the laboratory, confident that they would thereby return to their research alert and enthusiastic. As this author can personally attest, whether in correspondence or conversation, Georg Wittig was an open, receptive and courteous partner in any discussion. He radiated his fascination and keenness for the life of a chemist and he left his listeners optimistic for the future of chemical research.

Wittig's passing at the age of 90 on August 26, 1987, signals the end of an era, a century in which chemical structure determinations were largely achieved by means of an ordered series of chemical diagnostic reactions. Such techniques have now been supplanted, in great measure, by the physical methods of spectroscopy. Although there is no doubt that chemistry gains by the introduction of physical methods, there are also losses to be noted. With such an array of instrumentation available, the chemist now has less occation to exercise the imagination and genius of a Georg Wittig in devising chemical traps for reactive intermediates, like benzynes or ylides, or in proving structures by conducting reactions. The career of Georg Wittig should serve to show the modern chemist how many reactions might be overlooked if we become too closely bound to our instruments for studying chemical reactions. Let us run our instruments, but let our instruments not run us.

Acknowledgments

In preparing this appreciation of Georg Wittig's career, I have drawn upon biographical material helpfully provided by Professor Manfred Schlosser of the

Université de Lausanne, Switzerland, which included appreciations by former Wittig students, who are now themselves well-known professors: Professor Manfred Schlosser, Professor Ulrich Schöllkopf of the Universität Göttingen and Professor Dieter Hellwinkel of the Universität Heidelberg. I also had available a list of Professor Wittig's 297 publications and his photograph, which were likewise supplied by Professor Schlosser. I especially wish to acknowledge a most informative biographical note on Georg Wittig, published by the late Professor Ralph E. Oesper, in 1954 [1]. Finally, my impressions of Georg Wittig came from fortunate contacts I have had with him over the last thirty years, contacts that were anticipated with pleasure and are treasured in retrospect.

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