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Victor Grignard and Paul Sabatier: Two Showcase Laureates of the Nobel Prize for Chemistry**

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Grignard, Victor · history of science · hydrogenation · organic chemistry · Sabatier, Paul

In 1912 two French chemists were awarded the Nobel Prize in Chemistry: Victor Grignard and Paul Sabatier. It was the third time that a Nobel Prize in Chemistry was awarded to French scientists, after Henri Moissan (isolation of fluorine and introduction of the electric furnace to the service of science) in 1906 and Marie Curie in 1911 (discovery of the elements radium and polonium). The 1912 Nobel Prize was devoted to the field of organic chemistry, but the laureates were involved in quite different areas. Victor Grignard (Figure 1) was honored "for the discovery of the so-called Grignard reagent, which in recent years has greatly advanced the progress of organic chemistry", while Paul Sabatier (Figure 2) was recognized "for his method of hydrogenating organic compounds in the presence of finely disintegrated metals whereby the progress of organic chemistry has been greatly advanced in recent years". In 1912 Victor Grignard was 41 (the youngest laureate in chemistry up that point except for Ernest Rutherford) and Paul Sabatier was age 58. They were both working outside of the capital, Victor Grignard in Nancy and Paul Sabatier in Toulouse. It was surprising for the general public that the laureates were not from Paris, which was traditionally considered the main scientific center in France.

Figure 1. Victor Grignard.

Victor Grignard

Victor Grignard was born in 1871 in Cherbourg, a port on the west coast whose economic activity was strongly dependent on the navy. He died in 1935 in Lyon. The family of Grignard was of modest origin, his father was a worker at an arsenal. Victor Grignard lost his mother when he was very young, and he was a brilliant student at the Cherbourg High School (Lycée). Every year he received the annual first-place prize in almost all the disciplines. He was especially interested in mathematics. The family envisaged a career for him as a high school teacher. He left Cherbourg as he obtained a fellowship to work at the École Normale de Cluny. The school was closed by the government because of a lack of money in 1891, before the end of his studies and he had to finish his last year at University of Lyon where he studied mathematics. In 1894 he also completed the "licence de sciences-physiques" with the faculty of sciences of Lyon. There his friend Louis Rousset, a teacher in chemistry, suggested that he not continue in mathematics, but accept a position as a "preparateur-adjoint" in the section of general chemistry chaired by Prof. Philippe Barbier. Victor Grignard

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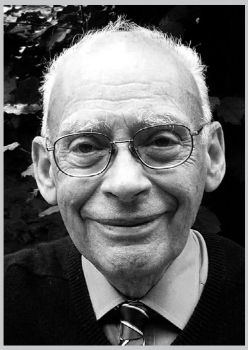
[**] I thank the Académie des sciences/Institut de France for permission to use the photos appearing in Figures 1 and 2. I acknowledge Prof. A. Lattes and Prof. F. Couderc for providing photos appearing in Figures 9–11 and Figure 12, respectively. Photos used in Figures 3–7 were taken from the book quoted in Ref. [9], and I thank Jacques Grignard, the grandson of V. Grignard for his authorization to reproduce these photos. I thank Mrs. F. Greff, head of Archives of Académie des sciences, for her help.

Figure 2. Paul Sabatier.

had never envisioned studying chemistry, which to him seemed empirical and required an excellent memory. In the research-active laboratory of Philippe Barbier, Grignard soon changed his mind and enjoyed chemistry. After the death of Louis Rousset in 1898 he succeeded him as “Chef de Travaux”.

Philippe Barbier was a well-known scientist with a strong personality. He assigned Victor Grignard a research topic on the synthesis of molecules containing both a double bond and an acetylenic moiety. In addition, in 1900 Philippe Barbier proposed that Victor Grignard reinvestigate some of his own studies done two years before as they produced erratic results. Philippe Barbier had been involved for several years in the study of metals in synthetic organic chemistry, especially in trying to extend the Saytzeff method by replacing zinc by magnesium.[1] For example he wanted to add an alkyl group to the carbonyl group of a methyl ketone $CH_3(C{=}O)$- to generate tertiary alcohols. Philippe Barbier used simultaneously methyl iodide, methylheptenone, and magnesium and obtained some of the desired product with no difference compared to the reaction with zinc.[2] Victor Grignard found the same difficulties as did his supervisor in terms of obtaining reproducible results. He decided to prepare an organomagnesium combination first and then to add the organic substrate. Symmetrical organomagnesium compounds MgR_2 were prepared a few years before in Germany but were not convenient to handle. Victor Grignard was reminded of a Frankland experiment wherein organozinc compounds were prepared in anhydrous diethyl ether. He expected that with the more electropositive magnesium the reaction would be easier. He found that the reaction $RX + Mg \rightarrow RMgX$ (R: organic radical, X: halide) was easy to realize below the boiling temperature of the solvent. Victor Grignard considered the reagent as a mixed organomagnesium compound and was able to use it as a nucleophile with many functional groups in organic molecules. Various classes of alcohols became easily available from aldehydes or ketones. The methodology was quite simple without requiring complicated equipment and experimental conditions (Figure 3).



Henri B. Kagan was born in Boulogne-Billancourt in 1930 and obtained his PhD with Dr. Jean Jacques. He worked with Prof. A. Horeau at the Collège de France, and joined the Université Paris-Sud, Orsay in 1968. Since 1999 he has been an emeritus Professor of the Université Paris-Sud. His awards include the Prelog Medal, August-Wilhelm-von-Hofmann Medal, Nagoya Medal of Organic Chemistry, Tetrahedron Prize, Wolf Prize, Ryoji Noyori Prize, Bower Award of the Franklin Institute, Horst-Pracejus Award, and Burckhardt Helferich Prize 2011.

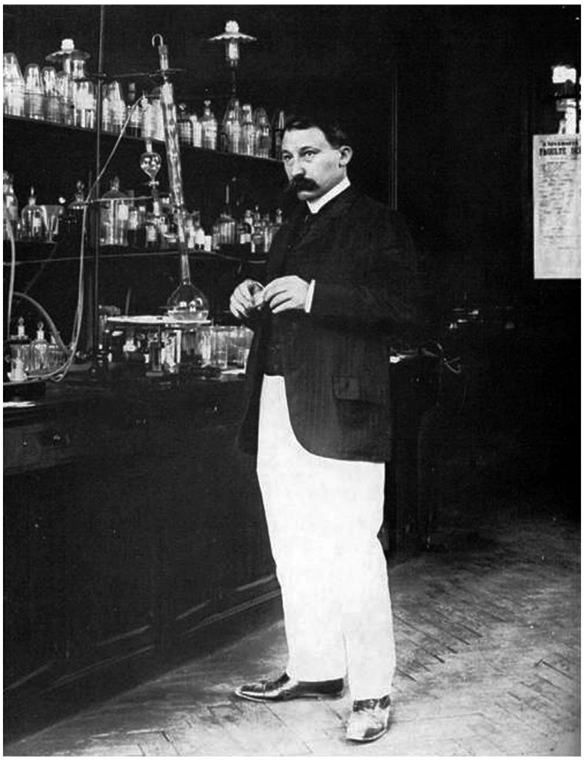
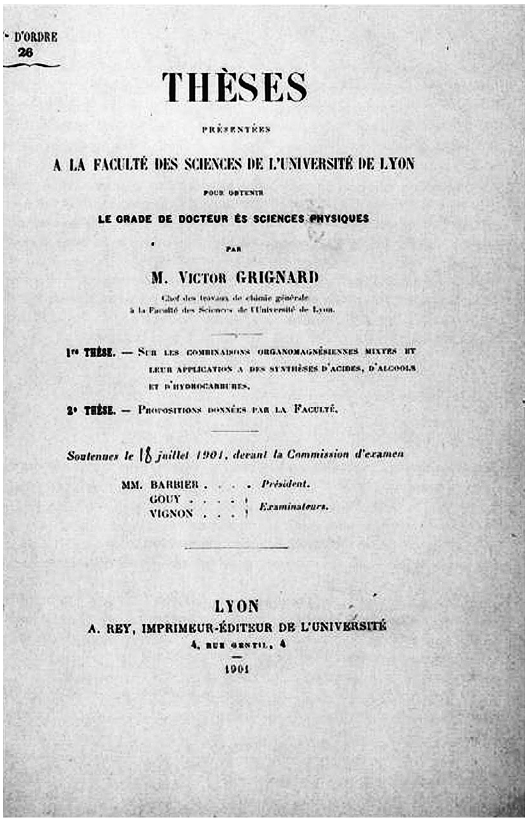


Figure 3. Victor Grignard in his laboratory.

In France and abroad many competitors began to use the “Grignard reagent”, as it soon became known as. The first paper was published in 1900 in the *Comptes Rendus de l’Académie des sciences* with Victor Grignard as the sole author.[3] Why was the name of Philippe Barbier absent? This point will be discussed in the next section. In 1900 Grignard was in the middle of his PhD work, and wanted to protect this new area of research, so he asked the advice of two important scientists at the Académie des sciences: Henri Moissan and Marcelin Berthelot. Both of them wrote to him advising that there was no way to protect a scientific discovery (apart from a patent) once it is published. They suggested that young Victor Grignard should publish as much as possible to extensively cover the area and to establish himself as a leader in the field. He did this by working nonstop—he was able to publish 14 papers between 1900 and 1904, some of which are listed in References [4,5]. In 1901 at the age of 30 he obtained a PhD (doctorat ès sciences; Figure 4).



D'ORDRE
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THÈSES

PRÉSENTÉES

A LA FACULTÉ DES SCIENCES DE L'UNIVERSITÉ DE LYON

POUR OBTENIR

LE GRADE DE DOCTEUR ÈS SCIENCES PHYSIQUES

PAR

M. VICTOR GRIGNARD

Chef des travaux de chimie générale
à la Faculté des Sciences de l'Université de Lyon.

1re THÈSE. — SUR LES COMBINAISONS ORGANOMAGNÉSIENNES MIXTES ET LEUR APPLICATION A DES SYNTHÈSES D'ACIDES, D'ALCOOLS ET D'HYDROCARBURES.

2e THÈSE. — PROPOSITIONS DONNÉES PAR LA FACULTÉ.

Soutenues le 18 *juillet 1901, devant la Commission d'examen*

MM. BARBIER . . . . *Président.*
GOUY . . . . } *Examinateurs.*
VIGNON . . . }

LYON
A. REY, IMPRIMEUR-ÉDITEUR DE L'UNIVERSITÉ
4, RUE GENTIL, 4
1901

Figure 4. Front page of Victor Grignard's PhD document (1901).

The impact of his work was enormous, and it was estimated by Victor Grignard that 700 papers on the Grignard reagents had been published by 1912 (the year of his Nobel Prize).[6] Victor Grignard developed many facets of the use of the Grignard reagents in organic chemistry. He obtained tertiary alcohols from esters, or monoacids from carbon dioxide. He described the first example of a 1,4-addition on a conjugated system, as well as the formation of the by-products R−R from RX through a Wurtz-type coupling. He prepared a variety of compounds of interest in the fragrance and pharmaceutical industry.

The prix Jecker was the most prestigious award of the Académie des sciences, and it was awarded in 1906 to Victor Grignard with strong support from Henri Moissan. Victor Grignard moved several times in his academic career from one university to another so as to get a promotion (from Lyon to Besançon, then from Besançon to Lyon, from Lyon to Nancy). In 1909 he arrived in Nancy as "Chargé de Cours". He became Full Professor in 1910 and occupied the chair of Edmond Blaise, who had recently moved to the University of Paris. In 1912 he was co-laureate of the Nobel Prize. In 1914 he was mobilized as a "caporal" (a low rank in the French army; Figure 5) and he was assigned to keep a watch along the tracks of railways in the Cherbourg area, his native land. It was a strange and bureaucratic decision of the army. In the middle of 1915 he was assigned in Paris to direct a research laboratory for the analytical analysis of gas recovered on the battle grounds, a better use of his competencies. By using trace analysis he was able to discover the method used by the German army for the preparation of a chemical weapon (yperite, $S(CH_2CH_2Cl)_2$). He also devised preparations for volatile chlorinated compounds such as phosgene, which was eventually used in the war.[7,8]

Figure 5. Victor Grignard as a "caporal" with the French army in 1914.

In 1917 he traveled as a scientific member of the "mission Tardieu" to the United States of America, which had recently joined the allied forces (Figure 6). He spent seven months there. In 1919 he returned to his laboratory. Shortly thereafter he applied for the chair of general chemistry at the University of Lyon to succeed Philippe Barbier who had just retired. Finally he accepted the position of Director of the institut de chimie physique industrielle in Lyon (today part of the CPE Lyon). It was not completely unexpected because he was always interested in the applications of chemistry to industry.

Figure 6. Victor Grignard (second row, second from left) in USA in 1917; middle of first row: T. Edison.

He died in Lyon at age 64. The life of Victor Grignard has been excellently described by his son Roger Grignard, a chemist who wrote a book for the one-hundred-year anniversary of the birth of his father.[9]

The Barbier–Grignard Dispute

Obviously Philippe Barbier (1848–1922; Figure 7) was responsible for the PhD work of Victor Grignard: he provided the research topic and the investigations were performed in his lab. There is only one paper in 1898 where Barbier and Grignard were co-authors, and it was for work not related to magnesium chemistry.[10]

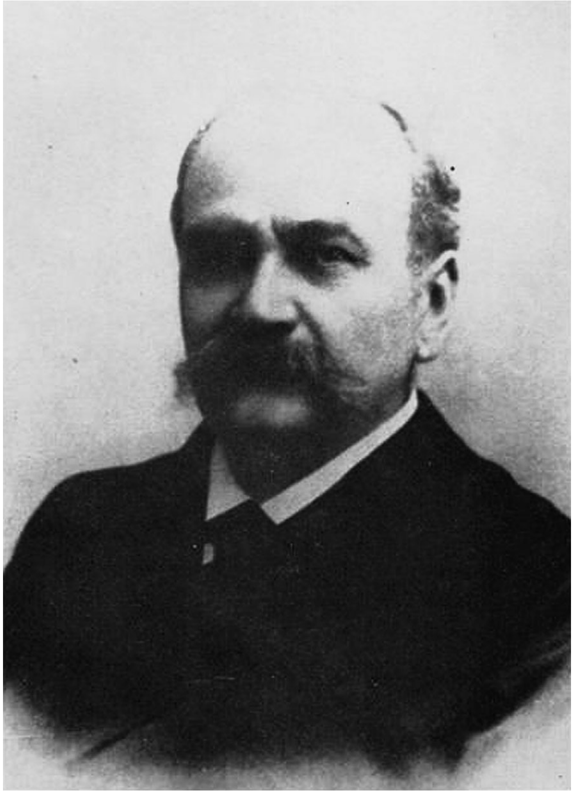


Figure 7. Philippe Barbier.

Why the absence of the name of Philippe Barbier on the various papers regarding Grignard reagents? It is difficult to answer this question but it seems to be related to the personality of Philippe Barbier.[9] He was a brilliant chemist with many new ideas. And when the reaction he envisaged was not going in the anticipated direction he would then tackle another project. Victor Grignard had difficulties developing the initial subject of his PhD work, so Philippe Barbier gave him an additional and minor topic (to his mind) to help Victor Grignard finish his doctoral work.

In 1910 Philippe Barbier complained that his name was not associated with that of Grignard in organic syntheses using magnesium.[11] He recalled that he was the first to replace zinc by magnesium in the Saytzeff methodology. Victor Grignard replied it was true, but he revendicated for himself the preparation of mixed organomagnesium compounds and their subsequent reactivity towards organic substrates.[12,13]

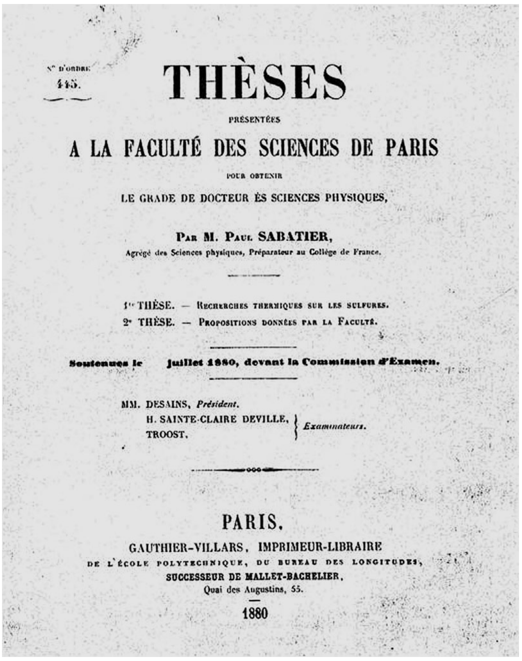
Curiously the reagents discovered by Victor Grignard are often called organomagnésiens in France, while abroad they are known as the Grignard reagents.[14] Maybe this is a consequence of the debates in France about the Barbier–Grignard issue. Victor Grignard and Philippe Barbier had a good personal relationship despite the bitterness of Philippe Barbier about the Nobel Prize of his former student. Grignard wrote in a letter to a friend that he would have preferred the Nobel Committee to award Paul Sabatier and Jean-Baptiste Senderens first, and then later award himself jointly with Philippe Barbier. He also indicated that he did nothing to influence the decision of the Nobel Committee.[9]

Paul Sabatier

Paul Sabatier was born in 1854 in Carcassonne in south of France and died in Toulouse in 1941. Just as Victor Grignard, Sabatier was raised in a modest family. He studied at a high school in Toulouse, and prepared for the entrance examination to two prestigious institutions located in Paris, the École Polytechnique and the École Normale Supérieure. He was accepted at both and chose to enter the École Normale, and graduated in first rank in 1877. He was obviously a brilliant young scientist. After one year as a high-school teacher in Nîmes he was contacted by Louis Pasteur and Marcelin Berthelot, and he decided to begin his PhD work at the Collège de France with Marcelin Berthelot, a prominent and influential chemist in France. He obtained his PhD (doctorat ès sciences) in 1880 at age 26, and at 30 (the minimum age; Figure 8) he occupied the Chair of General Chemistry at the University of Toulouse where he stayed for 45 years until his retirement.

Paul Sabatier trained with Marcelin Berthelot as an inorganic chemist. His PhD work (Figure 9) was focused on the synthesis and study of metallic sulfides. Upon becoming an independent researcher he prepared, in 1892, "nitrosylmetals", a new class of complexes considered to be analogous

Figure 8. Paul Sabatier at age 30 in 1884.



N° d'ordre
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THÈSES

PRÉSENTÉES

A LA FACULTÉ DES SCIENCES DE PARIS

POUR OBTENIR

LE GRADE DE DOCTEUR ÈS SCIENCES PHYSIQUES,

PAR M. PAUL SABATIER,
Agrégé des Sciences physiques, Préparateur au Collège de France.

1re THÈSE. — RECHERCHES THERMIQUES SUR LES SULFURES.
2e THÈSE. — PROPOSITIONS DONNÉES PAR LA FACULTÉ.

Soutenues le juillet 1880, devant la Commission d'Examen.

MM. DESAINS, *Président.*
H. SAINTE-CLAIRE DEVILLE, } *Examinateurs.*
TROOST,

PARIS,
GAUTHIER-VILLARS, IMPRIMEUR-LIBRAIRE
DE L'ÉCOLE POLYTECHNIQUE, DU BUREAU DES LONGITUDES,
SUCCESSEUR DE MALLET-BACHELIER,
Quai des Augustins, 55.
1880

Figure 9. Front page of Paul Sabatier's PhD document (1880).

to "carbonylmetals", which had been recently obtained by Ludwig Mond and co-workers.[15–18]

Paul Sabatier and Jean-Baptiste Senderens initiated a study for extending the action of metals on unsaturated molecules such as acetylene or ethylene. Indeed Henri Moissan already looked at the problem of the fixation of acetylene on metals. With Charles Moureu, Moissan found that freshly prepared nickel, cobalt, or iron did not fix acetylene but destroyed it with the evolution of gas, which was believed to be hydrogen, and the formation of a liquid suspected to be a mixture of aromatic hydrocarbons.[19]

Paul Sabatier asked Moissan for permission in 1897 to reinvestigate these experiments. He and Jean-Baptiste Senderens used a tube, heated at 300 °C, containing freshly reduced nickel, iron or cobalt (from the corresponding oxides), which were reacted with ethylene. With nickel it was found that below 300 °C nothing happens. At 300 °C the system became white-hot and gas was formed. The authors were very careful and rigorous experimentalists and managed to recover the gas formed. Analysis showed high amounts of ethane and not hydrogen as believed by Moissan. The obvious conclusion was that hydrogen generated in the partial decomposition of ethylene was transferred to the double bond of ethylene thanks to the presence of the metal. This hypothesis was immediately confirmed by using nickel, ethylene, and an excess of hydrogen. An almost quantitative formation of ethane was observed. The reaction was found to be possible at temperatures as low as 30–35 °C.[20,21] The method was soon applied to the hydrogenation of many classes of unsaturated compounds. This story was told by Paul Sabatier in a lecture given in 1939.[22]

Consequently the interests of Paul Sabatier moved from inorganic chemistry to organic chemistry. He performed the catalytic hydrogenation of many classes of organic compounds (aldehydes, ketones, nitriles, etc.). He also carried out dehydrogenations using metallic catalysts at high temperatures. He investigated reactions that could be of industrial interest and could be managed on a large scale (Figure 10). For example he described methods of catalytic cracking of hydrocarbons, methanol synthesis, methane synthesis from carbon monoxide, and hydrogenation of liquid oleic acid to solid stearic acid.

Paul Sabatier proposed a chemical interpretation of heterogeneous catalysis (in opposition to the physical interpretation made popular by Michael Faraday) with formation of unstable intermediates on the catalyst. In hydrogenation he envisaged the cleavage of the dihydrogen molecule by the metal and the formation of superficial hydrides. The importance and originality of the contributions of Paul Sabatier were analyzed in depth by Mary Jo Nye.[23]

Paul Sabatier was impressed by the German system wherein academic research was closely linked to industry or industrial problems. He supported the creation of engineering schools and he became director of the Institut de Chimie de Toulouse, today part of ENSIACET (Figure 11). He adopted the atomic theory at a time when the theory of equivalents was still strongly defended by Marcelin Berthelot in France.

The French Academy of Science recognized the value of Paul Sabatier's work and awarded him the prix Lacaze (1897) and the prix Jecker (1905). He became a correspondent member in 1901, and was invited to become a full member after being awarded the Nobel Prize. At that time full members of the Academy had to live in Paris, but this old regulation was modified because Paul Sabatier, just as Victor Grignard, did not want to move to Paris, and consequently a section of nonresident members was created by the Academy. Paul Sabatier was awarded the Davy Medal

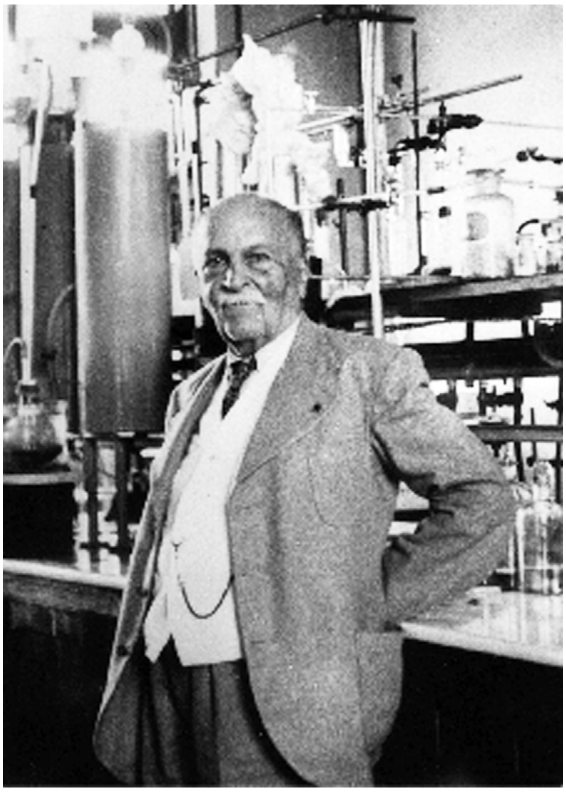

Figure 10. Paul Sabatier in his laboratory around 1935.

Figure 11. Paul Sabatier with students around 1920 at the Institut de Chimie de Toulouse, which was founded by him in 1906.

(1915) and the Royal Medal (1918) of the Royal Society, and he received the Franklin Medal in 1933 from the Franklin Institute.

The Sabatier–Senderens Dispute

Jean-Baptiste Senderens (1856–1937; Figure 12) was very gifted at scientific studies. He became a priest at the age of 24.

Figure 12. Jean-Baptiste Senderens around 1891.

He obtained in 1892 a PhD degree for his work done at the Institut Catholique Toulouse (ICT) under the guidance of Prof. Edouard Filhol, who died shortly before the end of the PhD work. He continued to work in the laboratory of ICT and collaborated, starting in 1892, with Paul Sabatier at the faculté des sciences de Toulouse. The scientific collaboration between both of them has been recently analyzed.[24] Most of the early work of Paul Sabatier on catalytic hydrogenation was done with Jean-Baptiste Senderens, and the Jecker Prize was awarded to them jointly in 1905. Jean-Baptiste Senderens left Paul Sabatier's laboratory in 1907 and was replaced by other co-workers such as Alphonse Mailhe who continued to develop catalytic hydrogenation. Senderens was unhappy that his contribution was not recognized in 1911 by Paul Sabatier in his lectures. He complained when Paul Sabatier alone obtained the Nobel Prize for catalytic hydrogenation. In his Nobel Lecture Paul Sabatier quoted his co-worker four times for his contributions.[25] He also tried to maintain a good relationship with his former coworker. In the literature the names of Sabatier and Senderens often remain associated.

Conclusions

The laureates of the 1912 Nobel Prize in Chemistry were faced with problems that are sometimes encountered concerning scientific ownership. In one case the PhD adviser was not recognized, because he unexpectedly did not publish with his student. In the other case the junior co-worker asked for more credit for the key experiments. Organic chemistry benefited a lot from the discoveries made by Victor Grignard and Paul Sabatier. These discoveries resulted from the careful observation and planning of experiments, and also took advantage of data in recent literature. Grignard gave a simple and fundamental tool to organic synthesis, especially for C–C bond formation. Sabatier introduced hydrogenation as a method which is useful in academic labs and in industry. He also proposed a mechanism, thereby recognizing the importance of unstable reaction intermediates in heterogeneous catalysis. They were strong supporters of provincial scientific leadership in France, and became nonresident Academy of Science members. They wanted to see a better cooperation between academic research and industry, and were influential not only through their publications and lectures, but also through the publication of textbooks.[26] The contributions of Victor Grignard and Paul Sabatier to organic chemistry fit perfectly with the legacy of Alfred Nobel who wanted to reward recent discoveries that are useful for mankind.

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[1] For a recent biography of A. M. Saytzeff see: D. E. Lewis, *Angew. Chem.* **2011**, *123*, 6580–6586; *Angew. Chem. Int. Ed.* **2011**, *50*, 6452–6458.

[2] P. Barbier, *C. R. Hebd. Seances Acad. Sci.* **1899**, *128*, 110–111.

[3] V. Grignard, *C. R. Hebd. Seances Acad. Sci.* **1900**, *130*, 1322–1324.

[4] V. Grignard, *C. R. Hebd. Seances Acad. Sci.* **1901**, *132*, 558–561; V. Grignard, *C. R. Hebd. Seances Acad. Sci.* **1901**, *132*, 835–837.

[5] V. Grignard, E. Urbain, *C. R. Hebd. Seances Acad. Sci.* **1902**, *134*, 849–851.

[6] V. Grignard, Nobel lecture, December 11, 1912.

[7] V. Grignard, E. Urbain, *C. R. Hebd. Seances Acad. Sci.* **1919**, *169*, 17–20.

[8] M. Dunikowska, L. Turko, *Angew. Chem.* **2011**, *123*, 10226–10240; *Angew. Chem. Int. Ed.* **2011**, *50*, 10050–10062.

[9] R. Grignard: *Centenaire de La naissance de Victor Grignard 1871–1971*, Imprimerie Audin, Lyon, **1972**.

[10] P. Barbier, V. Grignard, *C. R. Hebd. Seances Acad. Sci.* **1898**, *126*, 251–253.

[11] P. Barbier, *Bull. Soc. Chim. Fr.* **1910**, *7*, 206–207.

[12] V. Grignard, *Bull. Soc. Chim. Fr.* **1910**, *7*, 453–454.

[13] When I was a student in Collège de France between 1955 and 1960 I heard Prof. A. Horeau telling a story about the dispute between V. Grignard and P. Barbier. It was said that V. Grignard's reply to P. Barbier was: "Sir, you gave me a bone to chew, I managed to extract the marrow from the bone". I do not know if this anecdote is authentic.

[14] "Genèse et évolution de la découverte des composés organomagnésiens mixtes": R. Locquin, *Bull. Soc. Chim. Fr.* **1950**, 897–908.

[15] L. Mond, C. Langer, F. Quincke, *J. Chem. Soc. Trans.* **1890**, *57*, 749–753.

[16] Almost 100 years after Ludwig Mond's synthesis of nickel carbonyl Günther Wilke published a review on organonickel chemistry.[17] He reported on the discovery of nickel carbonyl by Ludwig Mond and co-workers. They treated nickel in a combustion tube with carbon monoxide and burned the gases to render them harmless. Ludwig Mond noticed in the flame a greenish yellow color and believed that there was some arsenic present in the carbon monoxide. Then he heated the glass tube to get an arsenic mirror. A mirror was immediately formed but it was a nickel mirror! This discovery gave rise very soon to an industrial process of preparation of pure nickel which was generated from pyrolysis of the newly discovered and volatile $[Ni(CO)_4]$. It attracted the attention of Paul Sabatier who decided to transfer on nickel unsaturated molecules other than carbon monoxide. The desired nickel/ethylene complex targeted by Paul Sabatier was finally synthesized many years later by Günther Wilke and co-workers. They prepared the unstable $[Ni(ethylene)_3]$ which was isolated as a crystalline compound.[18]

[17] G. Wilke, *Angew. Chem.* **1988**, *100*, 189–211; *Angew. Chem. Int. Ed. Engl.* **1988**, *27*, 185–206.

[18] K. Fischer, K. Jonas, G. Wilke, *Angew. Chem.* **1973**, *85*, 620–621; *Angew. Chem. Int. Ed. Engl.* **1973**, *12*, 565–566.

[19] H. Moissan, C. Moureu, *C. R. Hebd. Seances Acad. Sci.* **1896**, *122*, 1241–1243.

[20] P. Sabatier, J.-B. Senderens, *C. R. Hebd. Seances Acad. Sci.* **1897**, *124*, 1358–1361.

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[22] P. Sabatier, *Bull. Soc. Chim. Fr.* **1939**, *6*, 1261–1268.

[23] "Non Conformity and Creativity: a Study of Paul Sabatier. Chemical Theory and the French Scientific Community": M.-J. Nye, *ISIS* **1977**, *68*, 375–391.

[24] F. Couderc, V. Ong-Meang, *C. R. Chim.* **2011**, *14*, 516–523.

[25] P. Sabatier, Nobel lecture, December 11, 1912.

[26] P. Sabatier wrote a famous texbook titled *La Catalyse en Chimie Organique* and published in 1913. There were several editions including an English translation in 1923. V. Grignard was the editor of *Traité de Chimie Organique*, a 23-volume multiauthor series published between 1935 and 1954. For a historical presentation of the series, see: M. Blondel-Mégrelis, *Actual. Chim.* **2004**, 35–45.