

Periodic Table and its utility for predicting, as Mendeleev did, the properties of elements yet to be discovered in the Table's empty spaces. The authors reject any simple identification of "elements" with simple substances, atoms, or nuclei, and instead propose a consistent definition of element that applies at both the macroscopic and microscopic levels.

The essays in this book examine the concept of "element" from a variety of schools of thought, and they should prove interesting and informative to philosophers and historians of science in addition to practicing scientists (especially chemists) with a philosophical bent. While the level of background knowledge assumed on the part of the reader varies from essay to essay, for the most part the authors do a commendable job of illustrating the

historical and philosophical points using chemical concepts that should be understandable to anyone who has completed a first-year undergraduate chemistry course. Similarly, most philosophical concepts used, with the exception of the notation of formal logic in Chapters 6 and 12, are explained for the benefit of readers trained in natural science but not academic philosophy. The book is thus suitable for readers with a wide range of interests and academic backgrounds, and will surely stimulate many useful further discussions and debates.

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*Robert Le Rossignol: Engineer of the Haber Process*, Deri Sheppard, Springer Biographies, Cham, Switzerland, 2020, xxiii, 547 pp, ISBN 978-3-030-29714-5, \$97.

As I write these lines, it is clear that all of us have witnessed one of the most astounding accomplishments ever in medicine. A world-wide pandemic surfaced in January 2020, and in an approximately ten-month interval multiple vaccines for Covid-19 were discovered, developed, and brought into distribution to the public, with other vaccines now following. This astonishing result came about because of strong cooperation among government, industry, and academia. Whenever the scientific accomplishments of the 21<sup>st</sup> century are listed, this development of Covid-19 vaccines has to rise to the top of the list.

What are the great accomplishments of the century just past, the 20<sup>th</sup> century? Perhaps the most important was the discovery of artificial nitrogen fixation in the early years of that century. Nitrogen based fertilizers were vital for food production for an ever growing world

population, but the supply of nitrate fertilizer from natural sources in South America was under increasing pressure. At the beginning of the 20<sup>th</sup> century, starvation of millions was not that many years away. This was highlighted by English scientist Sir William Crookes in an 1898 speech to the British Association for the Advancement of Science. Crookes also proposed the solution—converting the nitrogen in the air to nitrogen based fertilizer. However, this was far more easy to propose than to actually accomplish.

You readers know that artificial fixation of nitrogen did in fact take place in the first decade of the 20<sup>th</sup> century. HIST recently recognized the accomplishment with one of its Citation for Chemical Breakthrough Awards. The name that all associate with this accomplishment is that of German chemist Fritz Haber. Those with a little more knowledge would add the name of Carl Bosch. Haber's synthesis of ammonia by catalytically reacting hydrogen with nitrogen was vital, but the scale up of the synthesis for industrial use was accomplished by German engineer Bosch. Both individuals were Nobel Prize winners for

nitrogen fixation, although Bosch's award (1931) trailed Haber's by 13 years. However, there was yet another significant individual needed for the discovery. That person was Haber's assistant, Robert Le Rossignol! How do we know that Le Rossignol was crucial to the process? Haber arranged for 40% of his royalties from the German company BASF for the discovery to go to Le Rossignol! Haber's giving that much money away testifies more than any written paragraphs of praise to the importance of Le Rossignol's contributions.

Le Rossignol had no desire for self promotion, so he wrote little about his years with Haber. It is mainly because of author Deri Sheppard's eight years of research that we now have a reasonably complete picture of this little known yet important chemist.

While Rossignol's name indicates French origin, for many years the family had been English citizens on the Channel Island of Jersey. Le Rossignol was born into an established, professional family. His father was a physician. Robert's early education was at Victoria College on the island of Jersey. This was not a college as we would understand it in the US, but a secondary institution modeled on English public schools. When he attended Victoria College, he benefitted from outstanding training in analytical chemistry under chemistry teacher Frederick Woodland Toms, who was also Official Analyst to the state of Jersey. In 1901 Le Rossignol went on to University College London (UCL) to study chemistry. There he worked with future Nobel Laureate (1904) William Ramsay. When Le Rossignol graduated in 1905, he was the winner of the UCL gold medal, now known as the Ramsay medal. Along with his chemical training, he also had studied mechanical engineering for two years during the sequence. This knowledge was to prove invaluable in his work with Haber. Le Rossignol stayed on at UCL one more year to pass examinations for an Associateship of the Institute of Chemistry in organic chemistry. Now came the time for postgraduate studies, and many English chemists would perform those studies in Germany. Ramsay chose to place his most promising students either with Richard Abegg at Breslau or Fritz Haber at Karlsruhe. Le Rossignol decided to work with Haber. He later admitted that a big factor in his decision was that Karlsruhe was near the Black Forest, which he wanted to visit.

Fritz Haber has been the subject of many biographies because of his two contrasting accomplishments. The "good" Haber carried out nitrogen fixation to prevent millions from starving, while the "bad" Haber facilitated chemical warfare in World War I. Sheppard confines

himself mainly to Haber's studies on nitrogen fixation, in which Le Rossignol played a key role.

Haber was raised in an affluent Jewish family. He became interested in both chemistry and physics. He began university studies in Berlin and spent some time in Heidelberg before returning to Berlin, where he did his doctoral studies in organic chemistry! Sheppard notes that some German sources credit Haber with the synthesis of 3,4-methylenedioxymethamphetamine. You readers may recognize that the street name of the compound is *ecstasy*. However, his fellow student Richard Abegg introduced Haber to physical chemistry. Haber was captivated by this newer aspect of chemistry, which drew his permanent attention. He became interested in entering academic science. His efforts were fruitless until finally in his middle 20s he obtained a position with the Technische Hochschule at Karlsruhe. It was there where Haber threw himself wholeheartedly into physical chemistry, blossoming into a chemist of the first rank. Haber first did landmark work on hydrocarbon pyrolysis, followed up with significant work on the electrochemical reduction of nitrobenzene. Then came his revolutionary textbook on electrochemistry, *Outline of Technical Electrochemistry Based on Theoretical Foundations*.

Over the next five years Haber continued important work in electrochemistry, but aspects related to chemical thermodynamics increasingly drew his attention. In 1905 he published the influential book, *The Thermodynamics of Technical Gas Reactions*, which moved him deeply into chemical thermodynamics. The book was translated into English by Arthur Lamb in 1907. Interestingly, Le Rossignol was given the task of translating one of the appendices of Haber's book. Sheppard notes that in the preceding year Haber probably started dipping his toe into the nitrogen fixation problem, although Haber didn't recognize its significance at that time. The Margulies brothers of Österreichische Chemische Werke in Vienna had detected small but reproducible traces of ammonia in their chemical plant, so they wondered if they had accidentally stumbled upon a viable method of making ammonia. The brothers offered Haber financial support to investigate the situation, so he undertook experimental studies with student Gabriel van Oordt in 1904. They studied the reaction of nitrogen and hydrogen to give ammonia as well as the decomposition of ammonia to its components at 1020 °C with an iron catalyst. They found that this was a true equilibrium process, but also that the amounts of ammonia obtained were too small to be practical.

Le Rossignol arrived in Karlsruhe in September 1906, one month after Haber had been appointed as full Professor of Physical Chemistry at the institution, the highest level achievable there. Haber was at the top of his game, but trouble lurked on the horizon. Walther Nernst, of impending Third Law of Thermodynamics fame, had written Haber in the fall of 1906 that Haber's results on the ammonia equilibrium gave ammonia concentrations far too high when compared to Nernst's calculated predictions. To defend his work, Haber would have to reexamine the ammonia equilibrium. New graduate student Le Rossignol came equipped with knowledge of high pressure equipment gained in the chemistry department at UCL and expertise in chemical kinetics and equilibria gained from working with George Donnan as well as Ramsay at UCL. It was probably in February of 1907 that Haber and Le Rossignol began their ground breaking research.

The Nernst-Haber confrontation took place at a Bunsen Society meeting on May 7, 1907, in Hamburg. In the interim after first writing Haber, Nernst had done experimental studies on the ammonia equilibrium with his assistant Fritz Jost. They definitely had formed ammonia at high pressure by reaction of hydrogen and nitrogen. Nernst used his Heat Theorem to calculate ammonia values in agreement with his experimental values, but these calculations involved generous assumptions. By contrast, the work of Haber and Le Rossignol involved many more experiments carried out in meticulous fashion. They had newer, more accurate calculations for ammonia concentrations than previously done with van Oordt, and their values compared more favorably with Nernst's theoretical calculation than did Jost's. However, Haber gave a very defensive presentation, so people attending the meeting probably thought that Nernst had carried the day.

After the Hamburg fiasco, Haber and Le Rossignol went back to the laboratory to restudy the problem. They found that Nernst's experimental values for the equilibrium were just flat out wrong. Le Rossignol had designed a conical valve with which he could achieve precise control of gas flows through the apparatus. The two men conducted 56 pressurized experiments in the temperature groups of 700, 800, 900, and 974 °C. Rather significant was the conclusion that Nernst's experiments gave ammonia values 30 to 50% lower than those obtained by Haber and Le Rossignol. The deficiencies of Nernst's previous experimental work were pointed out by Haber and Le Rossignol in a hard ball, 16 page paper published in 1908 in *Zeitschrift für Elektrochemie und angewandte*

*physikalische Chemie*. The experimental strength of their results showed that the position of ammonia equilibrium at pressure had been unequivocally decided in favor of Haber and Le Rossignol. Now the prestigious German chemical company BASF entered the action.

The history of BASF goes back to 1865. The company's initials stand for Badische Anilin- und SodaFabrik (Baden Aniline and Soda Factory). Baden was the German state where the company was located. Haber had previously carried out electric arc studies on formation of nitric oxide. BASF was interested in chemical innovation and already had a program going on nitrogen fixation. After some negotiations, Haber signed two contracts with BASF which gave him far more financial support than could ever be obtained from his academic institution. However, he was not allowed to publish without BASF's permission, to divulge technical details, or to work with other firms without BASF's approval. Pretty much the way it is for today's chemist working in industry.

The first contract dealt with the synthesis of nitrogenous gases made from nitrogen and oxygen. The second contract, the one that BASF was reluctant to sign, was to support Haber's research on ammonia synthesis. Le Rossignol's salary was essentially paid by BASF, but only through grants to Haber. These contracts were signed in March 1908. Unknown for many years was the private contract between Haber and Le Rossignol of May 1, 1908, which promised Le Rossignol 40% of the royalties that Haber was to gain from the BASF patents.

Despite their thorough previous studies, Haber and Le Rossignol had to overcome still more barriers to come up with a useful technical process. They needed high pressures, 100-200 atmospheres, combined with a lower temperature plus a useful catalyst, all this combined with a useful circulation apparatus. They steadily worked away, and in the third week of March 1909, they were able to see beads of liquid ammonia in the collecting vessel. Haber ran through the laboratory shouting, "Come down, there's ammonia—you have to see how the ammonia is pouring out." Later a convincing demonstration of the process was given to BASF management. Carl Bosch of BASF told his management "I think it can work," and so the process to scale up this new technology was under way, with future Nobel Laureate Bosch as the main actor.

Thus far we have only covered Part 1 (one third) of a 547 page book. The rest of this review will go much more quickly. To gain some perspective, let me cite an apocryphal story of a speech given by the Harvard

football coach to his team on the eve of their football game with Yale. "Gentlemen, you are about to play in a Harvard-Yale football game. You will never again in your life do anything so important." We may chuckle at the lack of perspective of the Harvard coach, but it is quite true that Le Rossignol never did anything as important again. That might be true for a lot of us. Our graduate research was exciting, but much of our follow up work was mundane. Le Rossignol lived a long productive life in science, but no one succeeding discovery ever had the impact of nitrogen fixation. How could it!?

Le Rossignol left Haber and Karlsruhe in August 1909. He joined the "Auer" company in Berlin, where he carried out industrial research on electric lighting. In 1910 he married Agnes Emily Hedwig Walter. The marriage lasted 65 years until her death. The couple had two sons. The older, John Augustin, died in World War II as a fighter pilot. The younger, Peter Walter, died while in college by suicide.

When World War I broke out in August 1914, the Le Rossignols were clearly enemy aliens, because upon marriage Emily was assumed to gain the citizenship of her husband. Internment of aliens began in November 1914. The internment camp was the disused Ruhleben race course a few miles from the center of Berlin. Fortunately for the Le Rossignol family, women and children were not interned, so only Le Rossignol was subject to this treatment. Conditions at the camp were quite harsh at first, although things got better with time. Suddenly on March 15, 1915, Le Rossignol was released for the purpose of going back to his old occupation. His release came about because of Haber's direct intervention. For the remainder of the war Le Rossignol worked on the improvement of the use of electric lamps, obtaining patents in this area.

Le Rossignol's work for an enemy industry during war time might seem questionable to us. Of course, none of the products he made had war use. However, Le Rossignol's association with Haber and Haber's association with chemical warfare would cause suspicion. Eventually the British government decided to give him a passport and not hold his place of employment against him. With the signing of the armistice ending World War I, Le Rossignol and his family left Germany for good on December 6, 1918. Through methods that are still not clear, Le Rossignol was able to eventually transfer his share of the royalties from BASF to England, somehow escaping the ruinous inflation of German currency that took place after the end of the war.

The 1918 Nobel Prize for Chemistry was not given in that calendar year. There was support for Haber in the Nobel Chemistry Committee, but, with the war just ending, a Nobel Prize would have been very controversial considering Haber's association with chemical warfare. In addition, the invention of nitrogen fixation had given Germany two vital assists to carry on the war. Nitrogen was not only crucial for fertilizers for agriculture, but it was also needed for the manufacture of munitions. The British blockade had cut off German access to South American nitrates, so nitrogen fixation had been vital for Germany continuing the war.

In 1919 matters had changed. The Swedish Committee members, strongly sympathetic to German science, focused on Haber alone, no thought of Le Rossignol or Bosch. Haber was thus awarded the unshared 1918 Nobel Prize in chemistry on November 13, 1919. Considering the attitudes in the Sweden scene at that time, the sharing of the prize between Haber and an Englishman would have been unthinkable. The year 1919, therefore, was the last year Le Rossignol had much of a public presence. However, in his Nobel Prize Award talk, Haber gave full recognition to Le Rossignol's vital contributions to the nitrogen fixation process.

Prior to World War I, English industry had been strongly dependent on the science coming out of Germany. The war showed the need for a stronger English scientific base. As part of this recognition, the English General Electric Company, GEC, not connected with the US company of the same name, constructed and staffed a research laboratory at Wembley to provide this needed research. One of the initial hires in 1919 was Le Rossignol. His research there gradually moved from electric lamps to valves. Probably even more significant than the Le Rossignol valve he had invented for Haber was his work on the CAT valve. CAT valves (or tubes, as they were called in North America) consisted of an integrated glass envelope and cylindrical anode enclosing the grid and filament, the leads to which were taken to outlet points in a further glass cylinder attached to the envelope of the anode and forming a continuation of it. Its construction entailed the making of a number of glass-to-metal joints, which were extremely difficult to make. These valves provided crucial for radio transmission.

With the outbreak of World War II, GEC's Wembley laboratory turned its expertise to the war effort. They and Le Rossignol made important contributions to mobile radar. Le Rossignol's efforts during the war were mainly on the administrative side rather than the technical. His

remaining son John Augustin, an RAF pilot, was killed on September 5, 1943, in Sicily. Le Rossignol's retirement took place a few years after the end of the war at age 65. In his last two years before retirement, he obtained three final patents. During his career he had 39 patents total, of which eight of the earliest were on ammonia synthesis.

Le Rossignol retired on his 65<sup>th</sup> birthday, April 27, 1949. He and his wife moved to a new home near the small village of Penn. The Le Rossignols supported a number of benevolences. Le Rossignol's wife Emily died on October 26, 1975. In her will she gave the residue of her estate to five UK charities. Le Rossignol died nine months later on June 26, 1976 at age 92. Fortunately Dr. Ralph Chirnside interviewed Le Rossignol on March 29, 1976, recording his final memories of the work with Haber, and author Sheppard was able to make use of the transcript of that interview.

Graduate students and post docs almost never share prizes with their mentors, no matter how significant their input may have been. Probably the poster child for unjustified omission was graduate student Jocelyn Bell Burnell, who by all rights should have shared the 1974 Nobel Prize in physics for the discovery of pulsars with mentor Antony Hewish. Sheppard certainly has made a strong case for the vital part Le Rossignol played in this ground breaking discovery. If we consider the three parts of this accomplishment to be experiment, engineering, and theory, Le Rossignol was the main worker for the

first two parts of the three. Would this have justified the sharing of the Nobel Prize? Perhaps in this day and age, yes. Back then, no.

While this book might seem like just one more telling of the nitrogen fixation discovery, the scientific and engineering detail separate this book from the crowd. I strongly recommend its purchase to anyone with a suitable technical background interested in seminal chemical discoveries. The chemical thermodynamics are heavy going, but Sheppard does his best with clear explanations and detailed drawings plus four appendices that take the reader again through the thermodynamics. Footnotes for each chapter are collected at the end of the chapters, and those footnotes further clarify difficult concepts. As a bonus, the reader gets a shortened but thorough biography of Haber. Do I have any complaints about the book? One. There is NO index! However, that is more of a problem for the book reviewer than for the reader.

Overall I was also impressed with the work that author Sheppard carried out to bring to life this little known figure of nitrogen fixation. Sheppard's eight years of work resulted in showing us something of the real person behind the name of Robert Le Rossignol. Unlike many of us, he was able to touch greatness. Was he great himself? With this exhaustively researched book, you readers can decide for yourself.

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### **Rabinovich presents Chemistry on Stamps Talk for American Philatelic Society**

University of North Carolina at Charlotte professor and Past Chair of HIST, Daniel Rabinovich, made a presentation "The World of Chemistry on Postage Stamps" for the APS Stamp Chat series. Rabinovich has frequently presented philatelic talks at HIST symposia, illustrating how the topic of the symposium has been commemorated in postage stamps. His APS presentation is an example of outreach in the other direction, showing chemistry to philatelists. The presentation is available on YouTube at [www.youtube.com/watch?v=SCve5FxtoxU](http://www.youtube.com/watch?v=SCve5FxtoxU).