Henry Taube

1915–2005

BIOGRAPHICAL *Memoirs*

A Biographical Memoir by
Peter C. Ford
and Thomas J. Meyer

NATIONAL ACADEMY OF SCIENCES

HENRY **TAUBE**

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Elected to the NAS, 1959

By Peter C. Ford
and Thomas J. Meyer

Through a lifetime of increasingly broad and significant experiments and discoveries, Henry Taube became the world's premier mechanistic inorganic chemist (one who focuses on reaction pathways and reaction mechanisms produced during inorganic chemical changes). In his first faculty appointment, he looked at the oxidation-reduction reaction and identified questions that could only be answered through precise experimentation. He then focused on the chemistry of transition metal complexes, laying out the details of what he called "inner sphere electron transfer." He went on to compile a broad range of findings, leading to the elevation of inorganic chemistry to a major field of study.

Taube earned bachelor's and master's degrees at the University of Saskatchewan and a Ph.D. from UC Berkeley. After receiving his doctorate in 1941, he took a position as an instructor at Cornell University, where he remained until 1946. He then joined the faculty of the University of Chicago. In 1962, he moved to Stanford, where he twice served as department chair (1972-74; 1978-79) and was appointed the Marguerite Blake Wilbur Professor in 1976. He formally retired from the faculty in 1986, but continued as an active scientist long afterward.

Henry Taube was born in Neudorf, Saskatchewan, on November 30, 1915. His parents were ethnic German farmers who escaped from Russian-controlled Ukraine in 1911 for Canada. They settled initially in Winnipeg, Manitoba, where his father worked as an unskilled laborer. After four years they moved to Neudorf, where his father worked as a farm hand and his mother cleaned houses. Henry remembered that the family lived in a rented sod hut when he was born. Eventually, the family was able to rent a farm near Grenfell, Saskatchewan, which is where Henry spent his childhood. Given his background, it was not surprising that Henry occasionally referred to himself as "just a farm boy from Saskatchewan."[1] He became a U.S. citizen in 1942.

In an interview, conducted in 2003, but published in 2005,[2] Henry was asked about the people who had influenced his career and its future development. He said,

> *My father was self-taught and was known to nearly every one as 'Honest Sam Taube.' He always kept his word and I felt that he had a large influence on my life and science. Although I've often been disappointed by ideas I had that didn't work out, I learned not to fool myself, and not see things that weren't there at the disregard of the science.*

Henry's first language was Low German, and his early education was in a one-room schoolhouse. His father wanted his children to have a better life. At age 13, Henry was sent to Luther College in Regina for his secondary education. His initial thought about a career was to become a minister. But after coming across Darwin's teachings when he was a 15, he found that he had to make a change. He had a real interest in English Literature, which was promulgated by his high school physics teacher. Unfortunately, he lost the financial support from his family in his second year at Luther College, after the stock market crash of 1929. He was still able to stay on at Luther, because his chemistry teacher persuaded the school to appoint him as a helper in the laboratory. In retrospect, this event was an important step forward for scientific research.

After Luther, Henry attended the University of Saskatchewan, in Saskatoon, where he registered in chemistry and earned both the B.S. and M.S. degrees, with John Spinks, who would later become president of the university, as his advisor. Henry noted that his university stay included interactions with future Nobel Chemistry Laureate Gerhard Herzberg and remembered with relish the quality and clarity of Herzberg's lectures on atomic spectroscopy and nuclear physics. After finishing his degrees at Saskatchewan, he still was not fully committed to chemistry but decided to attend graduate school at the College of Chemistry at the University of California in Berkeley.

In the interview noted above,[2] Henry recounted the story of how he wound up at Berkeley.

> *That was arranged by the Department Chair at Saskatchewan,* Professor Thorvaldsen, *who took an interest in me. It turns out that he knew G. N. Lewis [dean of the College of Chemistry,] from their being students together at MIT. When they parted, Lewis had told him that any student he recommended would be accepted for graduate study at Berkeley. So, this is what happened in my case; he took it upon himself to recommend*

> *me to Berkeley….When he called me into his office, I was frightened, since I knew that he really had a fear of mercury poisoning and I was using mercury in the laboratory….I was quaking in my boots. It didn't help when he looked at me rather sternly and said 'Mr. Taube, please sit down.' Then he said 'I've heard from Professor Lewis that you are going to Berkeley.' I myself had been thinking about going to Columbia to do my Ph.D. with Louis Hammett.*

Instead, Henry ended up at Berkeley where chemists such as Lewis, W. F. Giauque, J. H. Hildebrand, and W. M. Latimer, as well as his Ph.D. mentor, W. C. Bray, were on the faculty and where he earned his Ph.D. in 1940.

The environment for Henry at Berkeley was exactly what he needed. He noted that the faculty shared their

> *enthusiasm for their subject matter and their frankness in admitting their own limitations and their willingness to learn from others.*

There was little pretense for impressing others. The caliber of the people and the place spilled over into Henry and the way he conducted his own research. That became clear in later conversations with his former students and colleagues. With the benefit of a drink or two, Henry occasionally talked about his student days at Berkeley, where he became deeply interested in chemistry, in part, because of the stimulating environment created by its faculty.

With his Ph.D. in hand, Henry joined the chemistry faculty at Cornell University in 1941. His early research at Cornell and throughout the 1940s was characterized by investigations on the kinetics and mechanisms of oxidation-reduction reactions, initially those of the main group nonmetals. In the beginning, hydrogen peroxide and ozone were key substrates.[4] In his research, a key and characteristic feature was the identification of important issues needing to be resolved that could be dealt with by using dynamical studies to elucidate the mechanistic details about how they occurred.

Henry departed Cornell in 1946 to take a post at the University of Chicago, where he turned his skills and capabilities toward a new, long-term theme, the chemistry of transition metal complexes, including a significant and long-term interest in redox reactions. One of his first interests was in the details of how ligand substitution reactions occurred in metal complexes. With that theme in mind, he spent the large part of a Guggenheim Fellowship during the late 1940s in a return to the chemistry library at Berkeley, reviewing all that was known about substitution reactions. The result was a memorable 1952 article in *Chemical Reviews* in which he highlighted the details of metal complex, ligand substitution reactions. This article provided a framework for interpreting changes in rates and showed that they were induced by changes in the electronic configuration at the metal.[5] As a follow-up, his analysis of the data provided a direct lead into the design of a series of truly elegant experiments that elucidated, in detail, how electron transfer reactions occur between transition metal complexes in solution.

In one experiment, which helped define the area, he investigated the mechanistic details of how electron transfer occurs. The initial phases were telling in their simplicity. The strongly reducing, aquated chromium(II) complex $Cr^{2+}aq$, which was known to undergo rapid ligand exchange, was mixed with a solution of the substitution-inert, hexa-coordinate cobalt(III) complex $Co(NH_3)_5Cl^{2+}$. A key to the experiment was that upon oxidation to Cr(III), the Cr(III) product is unreactive toward loss of ligands while the Co(II) product rapidly loses its ligands to the surrounding solvent, giving an aquated Co(II) ion. In Henry's test tube experiment, the deep green of the chromium(III) complex $Cr(H_2O)_5Cl^{2+}$ appeared in solution immediately after mixing. Therefore, since $Cr^{2+}aq$ is "labile" and $Co(NH_3)_5Cl^{2+}$ and $Cr(H_2O)_5Cl^{2+}$ are "inert" to ligand substitution, the chloride ion must have been transferred from cobalt(III) to chromium(II) during the redox step. As shown in equation 1, this result demonstrated that the electron transfer reaction must have occurred when the chloride ligand was bridging the two metal centers. Henry called this process "inner sphere electron transfer."[6-8]

$$Co^{III}(NH_3)_5Cl^{2+} + Cr^{2+}aq \rightarrow \{(NH_3)_5Co^{III}\text{-}Cl\text{-}Cr^{II}aq\}^{4+} \rightarrow \{(NH_3)_5Co^{II}\text{-}Cl\text{-}Cr^{III}aq\}^{4+} \quad (1)$$

Elucidating the details of electron transfer was a major accomplishment, but Henry's contributions to mechanistic inorganic chemistry were far more broadly based. The list of his many findings is both significant and extensive. Early in his career, he and his group were the first to determine coordination numbers, geometries, and stabilities of solvated metal ions, all of which provided a critical understanding of the solution chemistry of transition metal ions.[9] He and his group were also the first to utilize paramagnetic metal complexes as NMR shift-reagents.[10] And they made some of the first observations on the importance of ion-pairing in the reactivity of metal complexes.[11] They carried out studies on photochemical reactions in solution[12] and were among the earliest users of isotopes in the elucidation of reaction mechanisms.[13] Of particular importance was Henry's use of oxygen-18 to establish that oxidation-reduction reactions could occur by oxygen-atom transfer.[14]

Henry took his last academic posting at Stanford University in 1962 and remained there till his retirement in 1986. In the late ‘60s he conducted a series of experiments with his late student, Carol Creutz, focusing on the chemistry and properties of the soon to be famous, “Creutz-Taube” ion, $\{(NH_3)_5Ru(pz)Ru(NH_3)_5\}^{5+}$, with pyrazine (pz) as a bridging ligand between the two ruthenium centers. Given the mixed-valence form of the dimer (a molecule made up of two smaller, identical molecules), and electronic coupling across the bridge, a major question was, how do you describe the electronic distribution in the mixed-valence complex? Was it localized, delocalized, or something in between?[8, 15] Henry’s experiments in this area were part of a large and extensive study on the redox and spectroscopic properties of ruthenium ammine complexes, and later of osmium ammine complexes. He and his group explored a number of important issues, including the existence of an extended reactivity involving osmium ammines and unsaturated organic ligands, including N_2 as a coordinated ligand.[16, 17] He and his student Dean Harmon demonstrated the existence of an extensive reactivity between osmium amines and a variety of unsaturated organic ligands in a series of experiments that lie at the interface between traditional coordination chemistry and organometallic chemistry.[18]

Henry’s major contributions to research have been summarized elsewhere, including an extensive 2006 retrospective published in the journal *Inorganic Chemistry*.[1] In looking back, it is clear that his contributions were fundamental to the development of Inorganic Chemistry as an important field of study. His timing and his ability to investigate and understand issues of importance for chemical reactivity of transition metal complexes in solution were extraordinary. When he began, little was known in quantitative detail about the area, but he became the master. He had the creativity and quantitative insight needed to help build it into the discipline that it is today. With the importance of transition metal complexes, and their reactivity in chemical systems, which range from biological oxidation-reduction mechanisms to industrial catalysis, the broadly based impact of his studies was enormous.

The importance of his contributions was highlighted in a news release in 1983, upon his winning the Nobel Prize in Chemistry, which said, “There is no question that Henry Taube has been one of the most creative research workers of our age in the field of coordination chemistry throughout its extent.”[19] The implications of what he accomplished continue today. Looking toward the future, he anticipated molecular-level electronic devices, the exploitation of the redox properties of excited states, and the utilization of electrochemical processes at the device level.

Henry was the recipient of nearly every major award in chemistry. President Jimmy Carter bestowed on him the 1977 National Medal of Science «in recognition of contributions to the understanding of reactivity and reaction mechanisms in inorganic chemistry.»[20] In addition to the Nobel Prize, 1983 was an especially good year for Henry; he collected the National Academy of Science Award in Chemistry and the Robert Welch Award in Chemistry. He was also a recipient of the American Chemical Society (ACS) Award in Inorganic Chemistry (1981), the Award for Distinguished Service in the Advancement of Inorganic Chemistry (1967), and in 1985, the Priestley Medal, the highest award of the ACS.

His list of scientific accomplishments was truly impressive, but there were other things that characterized Henry, which were relished by those of us who knew him well. It was abundantly clear to his many friends and colleagues, and especially to his many students and postdoctoral fellows, that he was a genuinely warm and interesting person. As a mentor, he made chemistry and the discovery of new phenomena not only challenging and stimulating, but fun. He created a research environment in which the key was the identification of issues of importance and how to solve them. He was even known to bet a bottle of wine or whiskey on the potential outcome of an experiment, and pay off when he, rarely, was the loser.

Henry's impact on his students and research collaborators was an important part of what he did, and how he did it. When asked in the interview described above[2] about his secret for inspiring students, his response was,

> *I think students feel that I really want to understand. They like to participate in the generation of new knowledge. I tried to introduce them to questions not answers. I was willing to learn from the facts.*

Many of his students and research collaborators, including both of the authors of this account, were strongly influenced by his role as a research advisor. All benefitted from their interactions with him and the research environment that he created. His laboratory was a magnet for talented graduate students and postdoctoral research fellows. It attracted senior visitors from around the USA and other countries, especially from Brazil, Australia, and Denmark, and those interactions led to some very significant, interactive science.

As a person, Henry was many things. It is clear that he was a world-class scientist, but he was also very much at home talking about gardening, opera, politics, mystery novels, baseball, and even tennis. He was also an avid collector, and loved to rummage through thrift shops for possible items of interest. At one point, he had a world-class collection of old phonograph records, specializing in tenors. He also enjoyed collecting obscure items. His list of collectibles included espresso coffee pots, antique blowtorches, and old bottles.

Henry took great delight in sharing his enthusiasms with others. Socially, he loved a good martini. Once, with one of the authors was a visitor at the Taube's, he overfilled his glass. He then dropped to his knee for a drink which brought his lovely wife, Mary, to exclaim, "Henry Taube! what in the world are you doing?" When the visitor commented that he saw that the glass was overfilled, Mary commented "Oh, I thought he was genuflecting to his martini." In response, Henry said, "it was a bit of both."

Henry was an infinite source of chemical wisdom and of continued advice for his colleagues and collaborators. When once asked what advice would he give to young scientists, he offered the following: "My advice is to keep your eye on the ball and don't be persuaded by your wishes. We all have ideas that we would like to prove to be correct, but we have to learn that we are limited."[2]

Henry published more than 80 research papers in refereed journals after his retirement and consulted with the company Catalytica, where he contributed to seminal studies on the catalytic activation of methane.[3] His final paper, number 412, appeared in 2003, when he was 87 years old. He died on November 16, 2005.

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