The Journal of Physical Chemistry

VOLUME 90, NUMBER 20 SEPTEMBER 25, 1986

Paul H. Emmett (1900–1985): Six Decades of Catalysis

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Dr. Paul H. Emmett contributed to the literature of physical chemistry, and especially heterogeneous catalysis, during six decades. The quality, even more than the quantity, of these publications will make them cornerstones of catalysis science for decades to come. In 1951, Sir Hugh S. Taylor,[1] a U.S. pioneer in heterogeneous catalysis, evaluated Emmett's contributions in ammonia synthesis: "These authors [Emmett, Brunauer and co-workers] have given us the most detailed kinetic study ever made of a single reaction, with all aspects of the reaction studied, adsorptions, kinetics, influence of reactant concentrations, surface areas as a function of composition and mode of preparations. The treatment is so comprehensive that it is possible to present an almost complete account of the phenomenon of surface catalysis by reference to this one example alone." This statement is all the more remarkable when one considers that Taylor made it in a review of the progress of catalysis during a 30-year period between the World Wars, not in a testimonial to Dr. Emmett. It is amazing that a person could publish such a broad spectrum of original, fundamental research papers in a field as multidisciplinary as heterogeneous catalysis. These papers are even more impressive when one considers that they were written, not in the early stages of development of the research area, but 150 years after Davy's observations on catalysis and more than 100 years after Berzelius provided a definition of catalysis. Even more astounding is the fact that, while he had essentially completed his experimental studies on ammonia catalysis, Dr. Emmett had published less than half of his scientific papers when Sir Hugh Taylor wrote the above assessment.

Paul Hugh Emmett was born in Portland, Oregon on September 22, 1900. His interest in science was developed by his high school chemistry teacher and with the encouragement of a high school advisor; in fact, he did not take science courses until his last year of high school.[2] He completed his undergraduate studies at Oregon Agriculture College and started graduate studies at California Institute of Technology. This school had, at that time, just started emphasizing graduate studies and had granted only six Ph.D.'s in chemistry prior to Emmett's class.[3] At Cal Tech his research with Dr. A. F. Benton, a recent graduate under Sir H. S. Taylor, set him onto a path toward catalysis research. His work with Professor Benton included investigations on autocatalysis of metal oxide reduction.

Dr. Benton was an excellent experimentalist and his influence is evident in the experimental design and attention to detail that characterized Dr. Emmett's studies throughout his career. However, this experimental skill apparently did not extend to the actual hands-on manipulation of equipment. Dr. Joe Kummer, an associate of Dr. Emmett for several years, and shown with Dr. Emmett in the accompanying photograph (Figure 1), constructed an elaborate adsorption apparatus. Dr. Emmett, impatient to obtain needed data, decided to help with the measurements on a Friday afternoon. The end result of his help was that a large, essential vacuum stopcock ended up on the floor shattered into many pieces and Joe had a few days of repair work. Also, even an outstanding experimental planner sometimes overlooks an important detail. Dr. Kummer, at 6 foot 6 inches or more, placed the stopcocks on his adsorption apparatus at a height that was convenient for him. Dr. Kummer's replacement, at a height barely over 5 feet, found most of the stopcocks out of reach; however, a stepladder provided a workable, if not convenient, solution to this particular experimental difficulty.[4]

(1) Taylor, H. S. *Am. Sci.* **1946** (October), 553.

(2) Davis, B. H. (interview with P. H. Emmett) *J. Chem. Educ.* **1978**, *55*, 248.

(3) Pauling, L. *Annu. Rev. Phys. Chem.* **1965**, *16*, 1.

(4) Garten, R. L. In *Heterogeneous Catalysis: Selected American Histories*, Davis, B. H., Hettinger, W. P., Jr., Eds.; American Chemical Society: Washington, DC, 1984; Symp. Ser. No. 222.

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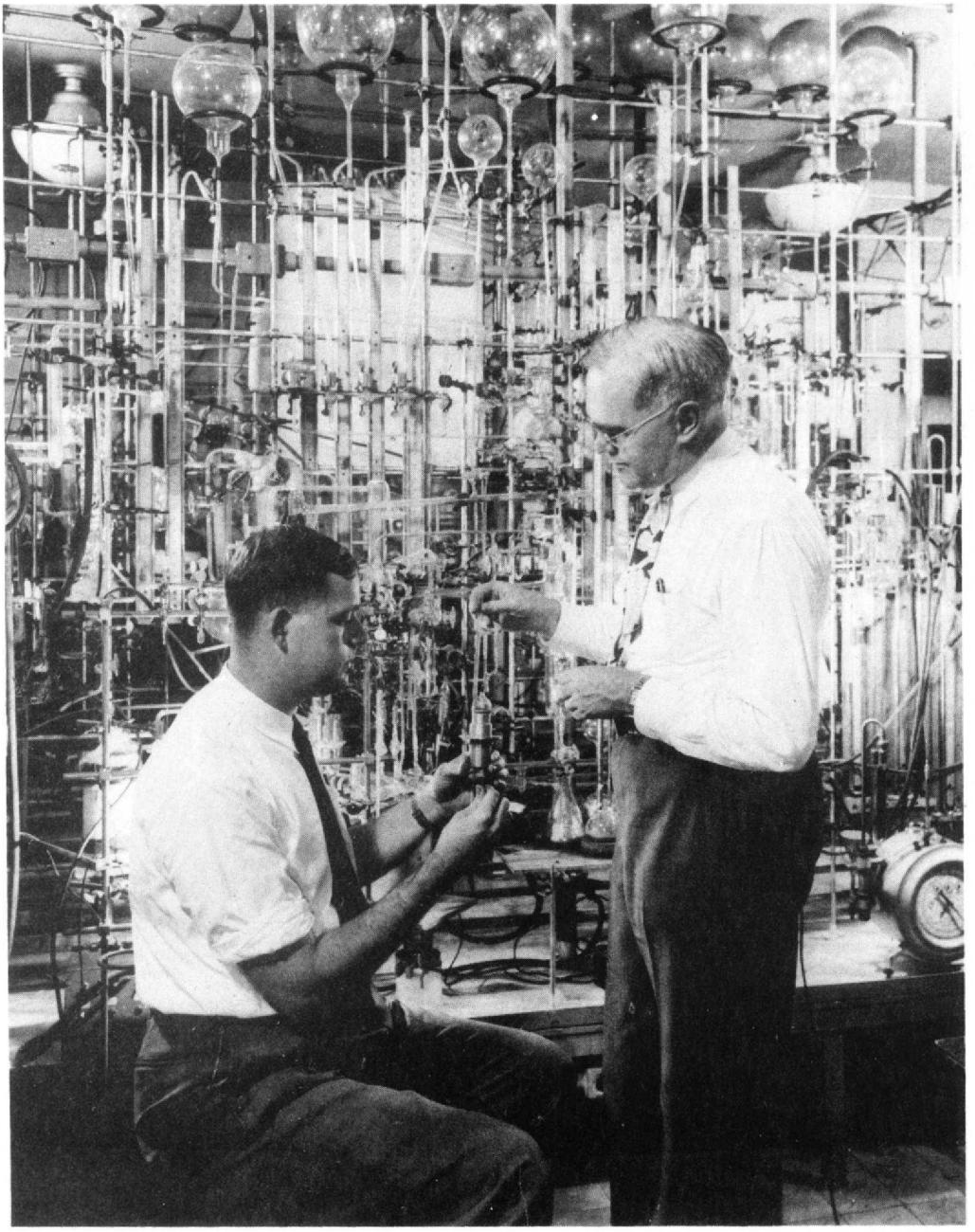

Figure 1. Dr. Joseph Kummer (seated) and Professor Emmett at the Mellon Institute. (Reprinted from ref 4.)

After teaching for 1 year at Oregon State College (his alma mater with a name change), he resumed his research in catalysis at the Fixed Nitrogen Research Laboratory in Washington, DC.[5] This was an especially productive 11-year period for him. Included among his accomplishments during this period was the development of methods for surface area measurements. Benton, Emmett's research professor, had published nitrogen adsorption isotherms that contained two "kinks" that Benton suggested might correspond to completion of monolayers of adsorbed nitrogen. Brunauer and Emmett performed gas adsorption studies which showed that the "kinks" disappeared when one made corrections for the imperfection of nitrogen at its boiling point. However, the points corresponding to the beginning of the long straight portion of the isotherms for several gases (which they designated point B) agreed with each other nicely and were considered to be the gas volume corresponding to monolayer coverage. Brunauer believes that, without the point B method, they would not have been able to progress to the development of the Brunauer–Emmett–Teller (BET) equation for calculating the surface area of a material from the amount of gas it adsorbs. This was an epoch paper that became a classic citation and still receives numerous citations after 50 years.[6] Dr. Brunauer reports that one place alone, the Institute of Catalysis of USSR Academy of Sciences, makes *ten thousand* B.E.T. surface area measurements in 1 yr.[7] The present day reader should recall that I. Langmuir had, in the 1910's, developed an important equation to describe gas adsorption.[8] In Langmuir's view, adsorption was limited to a monolayer of localized atoms; he was an effective advocate, and defender,[9] of this view. Langmuir had, in 1932, received a Nobel Prize based on his accomplishments in surface science. Thus, in the 1930's, Brunauer and Emmett's multilayer view of adsorption was not the widely accepted view. The BET paper, and the experimental and theoretical studies that followed it, altered this situation. Its importance can hardly be overestimated since it has provided a

(5) Emmett, P. H. In *The Physical Basis for Heterogeneous Catalysis*, Drauglis, E., Jaffee, R. I., Eds.; Plenum: New York, 1984; xi–xxi.

(6) Garfield, E. *Current Contents*, **1984** (No. 29), 3.
(7) Brunauer, S. *J. Colloid. Interface Sci.* **1985**, *107*, 593.
(8) Langmuir, I. *J. Am. Chem. Soc.* **1916**, *38*, 2221.
(9) Langmuir, I. *J. Am. Chem. Soc.* **1917**, *39*, 1848.

universal unifying parameter for the development and understanding of heterogeneous catalysts and their practical applications for nearly 50 years.

In addition to the extensive studies on physical adsorption, a method was worked out for measuring the extent of surface coverage of the iron catalysts by the alumina and alkali oxide promoters that are deliberately added to the iron catalyst. The method involved measuring the extent of the chemisorption of carbon monoxide on the iron portion of the catalyst surface. The method they developed is the same one used today to measure metallic surface area. This technique has become a classic method for catalyst characterization and, with the more frequently utilized BET method, provides the basis for comparing results of catalysis studies for different laboratories.

Emmett and Brunauer measured the free energies of formation of the nitrides of iron for the first time and found that it required thousands of atmospheres of nitrogen pressure to form a compound between nitrogen and iron. An iron nitride could not therefore be an intermediate in the ammonia synthesis. Nevertheless, it was obvious that nitrogen was taken up by iron at a fraction of an atmosphere pressure. From this they concluded that a surface nitride or, what is the same, chemisorbed nitrogen, may be an intermediate in ammonia synthesis. Adsorption studies showed that two types of hydrogen chemisorption occurred on the doubly promoted iron catalysts in the temperature range from −195 to 450 °C; on the singly promoted catalyst containing only alumina as a promoter three types of hydrogen chemisorption were shown to exist. Now hundreds of papers dealing with the different types of chemisorbed materials are published yearly; these confirm the pioneering work of the 1930's. Emmett also studied the chemisorption of nitrogen on iron catalysts and showed that the slow step in ammonia synthesis was the adsorption of nitrogen on the catalyst surface. This conclusion is still generally accepted. In addition to the many kinetic measurements Emmett and coworkers made on the synthesis of ammonia, Love and Emmett carried out a classical study on the decomposition of ammonia. The mechanistic details of the ammonia synthesis reaction that Emmett and co-workers developed remain essentially unchanged to this day and, in fact, now have a firmer basis due to the extensive surface science investigation of recent years.[10]

Rowland Hansford became Dr. Emmett's first graduate student. At that time, Dr. Emmett was an adjunct professor of chemistry at George Washington University while he was employed with the Fixed Nitrogen Laboratories in Washington, DC. The discovery that 1–2% of a promoter could cover a large fraction of the ammonia catalyst surface caused Dr. Emmett to wonder how this might affect the activity of an iron catalyst for the hydrogenation of other molecules such as olefins and aromatics. Briefly, it was found that unpromoted iron was unexpectedly active at dry ice temperature. The reaction involved type A activated hydrogen adsorption, never before observed to participate in a chemical reaction, and physically adsorbed ethylene. A promoted catalyst was much less active on a surface basis; i.e., the turnover number was of the order of 1% of that of the unpromoted iron.

But what followed illustrates the thorough and scrupulously critical attention to the interpretation of research data that characterized Paul Emmett. The hydrogenation had been studied in a constant-volume system, with the pressure drop being the indicator of reaction. The research was completed, the thesis accepted, and Hansford was employed in industry. Prior to completing the manuscript for journal publication, Dr. Emmett read that nickel catalyzed the dimerization of ethylene under some conditions. Even though the final pressure correponded to that expected for the hydrogenation reaction, Dr. Emmett wanted to be certain dimerization of ethylene did not occur with an iron catalyst under the experimental conditions used in the thesis research. Thus, Dr. Emmett contacted his former student and requested additional work. Hansford borrowed gas analysis equipment from his employer and returned to Washington on the train from Philadelphia. In those days gas chromatography, mass spectrometers, or other instruments were not available for gas analysis. Thus, Hansford took a rather bulky glass apparatus from Philadelphia to Washington, DC. Nevertheless, a replicate experiment was performed in the original apparatus and the equilibrated product analyzed for butenes with 85% H_2SO_4 saturated with silver sulfate in a then (1937) "state-of-the-art" conventional Orsat bubbler-burett gas analyzer. The results eliminated the possibility of dimerization and the data were soon published.

For a student to go to such lengths for his research professor tells much about both men; indeed this devotion lasted through the years and was especially evident at the Gordon Research Conferences on Catalysis, where one could be sure to see Emmett, Hansford, and their wives in friendly discussions, seated in chairs in the shade of a tree. Dr. Emmett enjoyed these annual meetings and was proud to observe that his annual attendance dated to the first Gibson Island catalysis meeting that evolved to the Gordon Research Conference on Catalysis. He maintained the tradition of these conferences not only by his bearing, lively discussions, and penetrating questions but also by participation in the annual swim in Sunapee Lake. I vividly remember him, now in his 80s, entering the water in spite of exceptionally cool temperatures and the pleas of his devoted wife, Pauline, while I remained on the lake bank, *wearing a jacket*.

In 1937 he became chairman of the Chemical Engineering Department at The Johns Hopkins University; within 3 years this newly formed department received accreditation. The stay at Hopkins brought him into contact with Dr. E. Emmett Reid, a catalysis scientist perhaps better known for having his autobiography published on his 100th birthday. Dr. Reid lived up to his saying, "The way to be active is to be active". I remember being in Dr. Emmett's office when Dr. Reid telephoned him. Dr. Reid was calling to give Dr. Emmett, then 65, some research suggestions that Emmett, some 25 years younger than Dr. Reid and still an active researcher, could work on.

Walter Frankenburg, who was forced to leave Germany during the 1930's, joined Dr. Emmett at The Johns Hopkins University as an associate in research on the adsorption of hydrogen and nitrogen on tungsten. Limited space for the new department necessitated that Frankenburg's adsorption apparatus be housed in a small, detached, one-room building that could have, at one time, served as a "springhouse". Nevertheless, important adsorption measurements were carried out in this setting. Frankenburg was noted for his extremely careful experimental work and precision in gathering his data. Legend has it that Frankenburg would build a complex glass adsorption apparatus, replete with many mercury manometers and transfer cutoffs, and make an extensive set of measurements. He would then completely dismantle and reassemble the apparatus in order to repeat the data collection to be certain that there were no systematic errors in the data. Dr. Emmett approached science in a like manner, constantly dismantling scientific concepts and then reassembling them, usually in an improved form.

He left The Johns Hopkins University to join the Manhattan Project staff in 1943. His work at the Manhattan Project was largely administrative. One of his accomplishments was to participate in the development of a fluorocarbon polymeric barrier material suitable for the separation of uranium-235 and -238 isotopes by diffusion. At the time a decision had to be made whether it would be possible to scale-up to a viable production plant, only a tiny piece of product had been synthesized and this required very high pressure. The decision to proceed to production paid off handsomely. Dr. Emmett states that he was proud to have been associated with the project[5] and his frequent comments during the ensuing years about this period of his career certainly show that this was the case.

He left the Manhattan Project in 1944 to accept a position at the Mellon Institute of Industrial Research on a Multiple Fellowship sponsored by Gulf Oil Company. During this period he pioneered the use of radioisotopes in mechanism of catalytic reaction studies. One of his early applications of the use of radioactive carbon was an attempt to learn whether the surface

(10) Ertl, G. *Catal. Rev.-Surf. Sci.* **1980**, *21*, 201.

of an iron catalyst was heterogeneous or homogeneous. First ^{14}CO was adsorbed to cover approximately 50% of the surface and this was followed by completing the surface coverage with nonradioactive CO. If the surface was heterogeneous the CO added first should be adsorbed on the higher energy sites and, thus, be bonded more strongly to the iron surface than the CO adsorbed in later exposures. Upon evacuation CO would be expected to desorb from a heterogeneous surface in the reverse order of addition. The experiment was only partially successful and showed that the surface behaved as though it was partly heterogeneous and partly homogeneous. This was another pioneering study by Dr. Emmett.

He also utilized ^{14}C-labeled hydrocarbons to study catalytic cracking, a process for making gasoline. These studies introduced him to the "real-world" of newspaper reporting since the *Wall Street Journal's* headline for a story describing his tracer studies on the mechanism of catalytic cracking read "Gulf Chemist Makes Radioactive Gasoline". During this research a new microcatalytic-chromatographic technique was developed and it has become a widely accepted method in catalysis studies.

His work with ^{14}C added much to our understanding of Fischer–Tropsch catalysis. He prepared ^{14}C-labeled iron carbide and showed that bulk carbide, proposed in the mechanism widely accepted at that time, could not be an intermediate in the synthesis; this finding drastically altered the mechanistic view that had been accepted since the discovery of the synthesis reaction. He also pointed out that the results did not eliminate *surface* carbide as a reaction intermediate; however, many who quoted his work failed to include this detail. This demonstrates a trait that was characteristic of his work; he extracted the maximum information from his data but he was careful to identify important points where his data were inadequate to make a definitive conclusion.

Dr. Emmett returned to Hopkins as W. R. Grace Professor in the Chemistry Department in 1955 and remained here until 1971. A postdoctorate from Mellon Institute, Dr. Richard Kokes, eventually joined him at Hopkins and the two of them were actively engaged in catalysis research during this period. Emmett continued research in the same general areas that he was pursuing at the Mellon Institute. These studies solidified the concepts generated by his earlier tracer studies, and provided some surprises. For example, it turned out that most of the aromatics formed in cracking cetane are formed, as secondary products, from the polymerization of low molecular weight alkenes. This finding was nearly 30 years ahead of the recent discovery of Mobil Oil researchers on the conversion of methanol, through alkenes, to highly aromatic gasoline-range hydrocarbons.[11]

One research activity that appears, at first glance, to be out of place for Dr. Emmett was an investigation of the reaction of xenon and fluorine to form xenon fluoride. While the unexpected news that xenon and fluorine react to form stable compounds would certainly attract his attention, it was a report by Baker and Fox[12] that the reaction was catalytic that prompted him to undertake research on this topic. The thing that intrigued him was not so much that it was a catalytic reaction, rather, it was a sentence stating that the reaction was zero order in both xenon and fluorine. This was a unique feature that would require additional thought since it implied that both of the reactants were strongly chemisorbed. Up to this time, for bimolecular catalytic reactions, only one of two reactants had been observed to be so strongly chemisorbed that the reaction was zero order in that reactant's concentration. A reaction that was zero order in both reactants was novel; that one of the strongly chemisorbed reactants was xenon, until 2 years earlier considered an unreactive inert gas, was astounding. His work[13] along with others[14,15] confirmed the catalytic action of metal fluorides, the zero-order kinetics, and a reasonable explanation for this. Something that was new to Dr. Emmett, especially when it disturbed the order of established concepts, had to be pursued, both experimentally and intellectually, with vigor. My rough draft of our paper on the reaction of xenon and fluorine underwent, in his hands, a remarkable metamorphosis with his numerous hand calculations being carried out at home on a variety of scratch paper, including his wife's new stationery.

This same drive toward understanding is shown by the following question and his reply:[2]

Davis: Do you consider the Brunauer–Emmett–Teller equation for physical adsorption your major contribution or was there some other work you consider more important?

Emmett: Probably the BET work will remain as the major contribution though one of the most intriguing and satisfying results that I obtained was in a research to ascertain why the water gas constant for the water-gas-shift reaction $H_2O + CO = CO_2 + H_2$ had two different experimental values. As of 1930 if one measured this equilibrium directly, one obtained one set of values. On the other hand, if one measured the equilibrium value for the reactions $Fe + H_2O = H_2 + FeO$ and $CO + FeO = CO_2 + Fe$ and combined these two equations, one should obtain the equilibrium constant for the water-gas-shift reactions. The water-gas-shift equilibrium constant obtained indirectly was about 40% smaller than the direct value. In view of the fact that the water-gas-shift reaction was important in ammonia syntheses on which I worked after I went to the Fixed Nitrogen Laboraory, I designed some experiments to try to find out the cause of this 40% discrepancy. This turned out to be a very surprising and interesting series of experiments. Entering into the success of our efforts was the fact that we did not have a piece of quartz long enough to go through a furnace in which we wanted to carry out the experiments so we had the glassblower seal a quartz tube 2 mm in diameter onto a quartz tube 2 cm in diameter. We then inserted a boat of iron oxide at the junction of these two tubes and arranged to circulate water vapor–hydrogen in both directions through this tube. To our surprise we found that when the circulation was in the direction *going out the small tube, the ratio of water vapor to hydrogen* for the equilibrium reaction was 40% different from that obtained when the reaction mixture was circulated out through the 2-cm tube. We suggested that the result was due to thermal diffusion. According to this, if one end of a tube containing a mixture of gases of different molecular weights is heated, the light gas will tend to concentrate in the hot end of the tube; the heavy gas will gravitate toward the cold end. With a temperature gradient of 1000 °C in an ordinary tube, a mixture of helium–carbon dioxide would tend to separate so that the helium would become more concentrated in the hot end of the tube and the CO_2 in the cold end of the tube. So in these experiments in the water vapor–hydrogen mixture the water vapor tended to go toward the room temperature end of the tube and the hydrogen tended to stay in the hot part of the tube. This thermal diffusion effect amounted to about 40% for water vapor–hydrogen at a temperature of about 1000 °C compared to room temperature and accounted for the odd experimental results. This result was probably the most satisfying of my career, but I suspect that the BET work will be longer remembered as a very useful tool for catalytic chemists.

Thus, it was not fame, but the solution to a particularly demanding scientific puzzle, that was his satisfaction. And, satisfaction for him did not require that he be credited with the solution. He derived great pleasure in helping others solve problems. Many, many papers have acknowledgments to the helpful discussions or critique that Dr. Emmett provided. On several occasions, his helpful discussion approached, or even exceeded, the length of the paper he was reviewing. And in reviewing a paper, he believed that he should detail those points that should be modified, improved, or validated but that he should never reject a manuscript. He genuinely enjoyed helping others in their work and would seek out, and engage, younger scientists in discussions.

Even with this modest, kind, and friendly nature, Professor Emmett did have a competitive spirit. He repeated, on numerous occasions, that he had only been "scooped" once. One of his students was very successful in rapidly developing a gas chro-

(11) Chang, C. D.; Silvestri, A. J. *J. Catal.* **1977**, *47*, 249.
(12) Baker, B. G.; Fox, P. G. *Nature (London)* **1969**, *204*, 466.
(13) Davis, B. H.; Wishlade, J. L.; Emmett, P. H. *J. Catal.* **1968**, *10*, 266.
(14) Weaver, C. F. Thesis, University of California, Berkeley, CA, 1966.
(15) Baker, B. G.; Fox, P. G. *J. Catal.* **1970**, *16*, 102, 108.

matographic method to analyze a mixture of H_2, HD, and D_2, an analysis that is very useful in catalysis research. Professor Emmett was tardy in getting a paper describing this research written with the result that someone else published similar work. From his description of this incident, it was apparent that, while he gracefully accepted it one time, it was not something that he intended to repeat.

In 1971, he retired from Hopkins, 5 years later than originally planned. His "retirement rocking chair" was an appointment as research professor in the Chemistry Department at Portland State University, undertaking new research areas such as surface area of soils and the porosity of coals, presenting seminar talks, offering advanced courses in catalysis, consulting at W. R. Grace, Oak Ridge, and Mobil, reviewing manuscripts, publishing 12 papers with students and co-workers, frequent travel to national and international scientific meetings, receiving seven of his distinguished achievement awards, etc. Throughout his 14 years at Portland State University he always had time to talk to younger faculty members and graduate students, frequently during lunch. He continued to be active in scientific endeavors until a few months prior to his death on April 22, 1985.

Dr. Emmett was renowed among his colleagues for his encyclopedic knowledge of catalysis literature. After his retirement, Professor Emmett was a consultant for a group working on fuel cells at United Technologies. During one of his visits, he suggested that an investigator look up a paper in a certain journal that had been published shortly after the war since it should be very helpful in solving a problem that they had been discussing. The investigator did not easily find the paper and, knowing Professor Emmett's reputation for recall, went through, paper by paper, a number of volumes of the journal that had been published following WWII. Even though the investigator repeated the search, he was unable to find the paper. During Dr. Emmett's next visit the investigator mentioned his failure to find the paper even though he carefully searched each volume published during a 10-year period following 1945. It is easy to imagine the slightly amused, and very "sheepish", grin as Dr. Emmett apologetically replied, "Oh!!! I forgot to mention that it was WWI, not WWII." Needless to say, the investigator found the paper just where Dr. Emmett had remembered it to be.

Included among Dr. Emmett's awards and honors are the following: Pittsburgh Award, Pittsburgh Section of the American Chemical Society, 1953; election to the National Academy of Science, 1955; Kendall Award, American Chemical Society, 1958; Highest Award for Scientific Achievement, Government of Spain, 1964; Catalyst Club of Philadelphia Award, 1970; Distinguished Service Award, Oregon State University, 1974; Distinguished Alumni Award, California Institute of Technology, 1977; Howard Vollum Award for Science and Technology, Reed College, 1980; Pioneer in Chemistry Award, American Institute of Chemistry, 1980; establishment of the Paul Emmett Award by the Catalysis Society of North America, 1972; establishment of the Paul Emmett Lecture on Catalysis, University of Utah, 1983; establishment of the Paul H. Emmett Scholarship in Chemistry, Portland State University Foundation, 1985; and honorary degrees from Oregon State College, 1939; University of Lyons, 1964; Clarkson College, 1969; University of Wisconsin, Milwaukee, 1971; and University of Hokkaido, 1976.

To write about Dr. Emmett's scientific accomplishments is easy because there are so many and because he was able to present them both in "oversimplified" intellectual models as well as with the necessary experimental and theoretical vigor. To write about Dr. Emmett's human qualities is an impossible task because he possessed, and continuously demonstrated, them to such a degree that one had to experience them to even begin to appreciate them. Chemist A. W. Hofmann wrote,[16] over 100 years ago, of Martin Klaproth: "Possessed of a modesty devoid of all conceit, filled with appreciation of the merits of others, though mindful of their weakness, but implacably strict in the appraisal of his own work, Klaproth has given us for all times the model of a true scientist." Changing Klaproth to Emmett in the above quote provides a fitting description of Dr. Emmett that is difficult to improve. And yet, in spite of the elegance of this quote, these words cannot begin to describe him. Dr. Emmett was an omnipotent scientist but it is safe to say that those of us who were fortunate enough to have known him will recall him, first, for his human qualities and, secondly, for his scientific accomplishments.

A pupil and admirer of Dr. Emmett wrote "Paul has been a friend and personal benefactor ever since I met him in 1942. There does not exist a man more kind or more thoughtful than Paul Emmett, and no one has done more in developing the science of catalysis. I count it a lifelong privilege to have known him." Dr. Stephen Brunauer, a collaborator during Emmett's early professional years, wrote[6] that "He was truly an understanding example of a scholar and a gentleman, and everybody who knew him loved him and admired him." All of us would express, perhaps in different words, this view of Emmett, the man. His response, to one of my questions:[2] "I was inclined to think the best of people and to be optimistic about them..." states very simply his approach to life. And in thinking the best of all people and in being optimistic about them, he was equally successful as he was in his in catalysis research.

The assistance of a number of people, especially Stephen Brunauer, Roland Hansford, George Young, Clyde Brooks, and Sol Weller, has greatly improved and added to my original manuscript.

(16) Buage, G. *Buch der Grossen Chemiker*, 1929, Vol. I, pp 334–341; translated by Olsper, R. E. In *Great Chemists*, Farber, E., Ed.; Interscience: New York, 1961; p 295.