NATIONAL ACADEMY OF SCIENCES

MELVIN CALVIN

1911—1996

A Biographical Memoir by

GLENN T. SEABORG AND ANDREW A. BENSON

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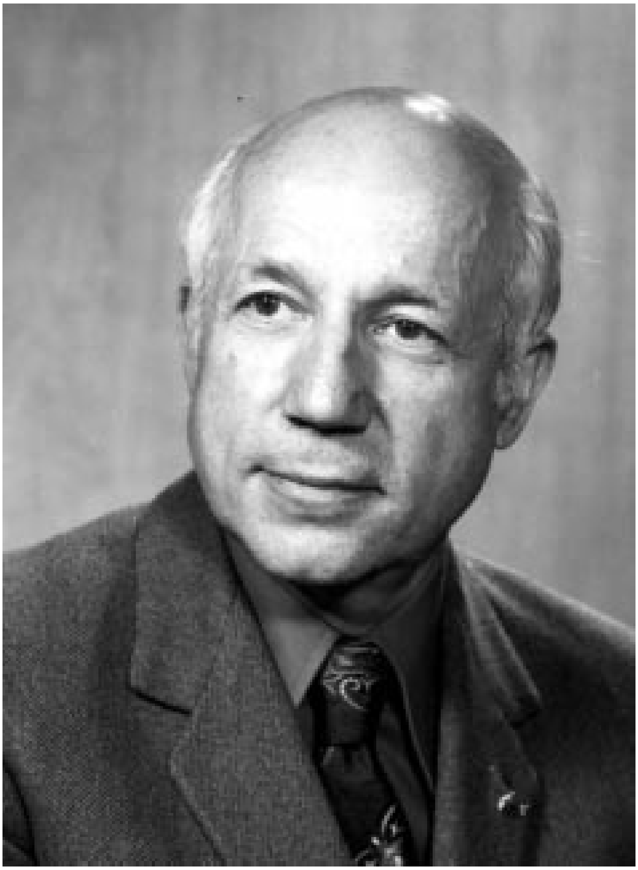

MELVIN CALVIN

April 8, 1911–January 8, 1997

BY GLENN T. SEABORG AND ANDREW A. BENSON

MELVIN CALVIN DIED IN Berkeley on January 8, 1997, at the age of eighty-five from a heart attack following years of declining health. He was widely known for his mental intensity, skill in asking questions, and impressive presentation of his research and ideas.

During the period 1946-57 Calvin directed laboratories utilizing radiocarbon-14 and other radioisotopes in the University of California's Radiation Laboratory, founded by Ernest Orlando Lawrence. Among his achievements was the delineation of the path of carbon in photosynthesis, for which he was awarded a Nobel Prize in 1961. He was elected to the National Academy of Sciences in 1954. Among his many honors were the Priestley Medal of the American Chemical Society in 1978, the U.S. National Medal of Science in 1989, and the Davy Medal of the Royal Society in 1964.

Born in St. Paul, Minnesota, Melvin Calvin was the son of a Lithuanian immigrant father and a mother from Russian Georgia; they ran a small grocery store in Detroit, where young Melvin helped while going to Central High School. He became intrigued by the products on sale in the store, began to wonder what they were made of, and early on recognized the importance of chemistry in their makeup. Deciding to be a chemist, Melvin received his B.S. in 1931 at the Michigan College of Science and Technology (now Michigan Technological University), where he was the college's first chemistry major. He went on to take a Ph.D. with George C. Glockler at the University of Minnesota in 1935.

The next two years were spent working with the intellectual giant Michael Polanyi in Manchester, England, where Melvin became interested in phthalocyanines. This first incursion into photochemistry was later to lead to chlorophyll, photosynthesis, and artificial photosynthetic membrane models. A chance visit with Joel Hildebrand at Manchester resulted in an invitation from Gilbert N. Lewis to join the chemistry faculty at the University of California. So, in 1937 Calvin arrived as an instructor at Berkeley, where he was to remain for the rest of his life. He was the first non-Berkeley graduate to be hired by the Department of Chemistry in more than a quarter century. His work with G. N. Lewis developed the photochemistry of colored porphyrin analogs and their coordination of iron and other central metals. Melvin was at home in discussions of the excited triplet states of chlorophyll and intermediates in the energy transfer processes of photosynthesis, subjects that clearly passed over the heads of most plant biologists of that period. Such discussion later involved James Franck, A. A. Krasnovsky, A. Terenin, George Porter, R. G. W. Norrish, Bill Arnold, Sterling Hendricks, and G. O. Schenk.

At Lewis's chemistry departmental research conferences in 1937, he was soon known for his skill in asking important questions. Throughout his career, Melvin asked questions, seeking to know and understand his surroundings and the research being done in his laboratories. As the radioautographs of the paper chromatograms (Radiograms) were developed, he tried to memorize their information, as well as the information related by the people doing the laboratory work. Melvin usually finished his lectures and office and committee work about 5:30 p.m. and stopped in at the lab to ask his signature question: "What's new?" The next morning when Melvin came in at eight o'clock or before (he was an early riser), it was also his first cheery comment of the day. There was no letup. His coworkers had to keep some tidbits in reserve so they could always have something interesting to report. When important chromatograms were exposed on X-ray film, they used two sheets, one to develop too early to appease Melvin's insatiable curiosity and one for proper documentation.

Among his skills were effective management of personnel, budgets, publications, consultancies, and presentations at important scientific conferences. Such skills engendered productive laboratories and enthusiastic collaboration of their scientists. Over the years Calvin's ever widening activities and responsibilities were efficiently managed through his office. Selection of the 230 to 250 visiting scientists and students, arranging for their schedules and research, handling the dozens of distinguished lectureships, and acceptances of honorary degrees from thirteen institutions consumed time, but he was a skilled planner and remarkably quick at making decisions.

Melvin Calvin was a fearless scientist, totally unafraid to venture into new fields like hot atom chemistry, carcinogenesis, chemical evolution and the origin of life, organic geochemistry, immunochemistry, petroleum production from plants, farming, moon rock analysis, and development of novel synthetic biomembrane models for plant photosystems. By asking questions and quickly reading some books, he felt comfortable in many fields of endeavor. He stimulated and supported interdisciplinary thought and research among his colleagues. The circular Melvin Calvin Laboratory, first occupied in 1964, was designed to promote continuous communication for students and visiting scientists working on a broad range of subjects and technologies (and was thus nicknamed “the round house” and “Calvin carousel”). The breadth of his interests and experience in chemical science served him well in his interactions with his colleagues, and he was clearly an awesome figure for visitors and students, whose experience was generally limited in scope. It seemed that he had few peers who could criticize or analyze the impact of his ideas and projects. The primary limitation of his ideas stemmed from his limited experience in mechanics and practical laboratory techniques. His energetic curiosity, however, overcame such limitations as he probed the experience of experts and soon became conversant with a new subject or the operating principle of a new instrument.

Melvin was equally fearless in pursuing novel ideas. He had no qualms about suggesting experiments or publishing papers that further research soon undermined. The isolation of radioactive succinate from algae fed radioactive CO_2 in the dark, conditions where succinate accumulates, led to the suggestion for a C-4 cycle of photosynthetic carbon reduction. The fact that such a cycle provided no useful information on photosynthetic carbon dioxide reduction didn’t bother him a bit. The most exciting was his thioctic acid theory of photosynthesis, which consumed over a year’s work by the group in the Old Radiation Laboratory. As the several pillars of evidence began to fall, Melvin was unperturbed and went on to a new approach.

Melvin’s marvelous technique for delivering a scientific lecture was unique. His mind must have roamed constantly, especially in planning lectures. His remarkable memory enabled him to formulate a lecture or manuscript with no breaks in the sequence of his thoughts. His lectures usually began hesitatingly, as if he had little idea of how to begin or what to say. This completely disarmed his audiences, who would try to guess what he might have to say. Soon enough, however, his ideas would coalesce, to be delivered like an approaching freight train, reaching a crescendo of information at breakneck speed and leaving his rapt audience nearly overwhelmed. One evening at a meeting of the American Association for the Advancement of Science, his novel theory of photosynthesis was so impressive that, when he finished, the great C. B. van Niel jumped up from his seat in the front row and, with tears in his eyes, congratulated Melvin for making the ultimate discovery of the mechanism of photosynthesis. It was a great theory, indeed.

Melvin Calvin was a survivor. Against great odds, he enjoyed nearly fifty productive years after his frightening experience of a coronary during an Atomic Energy Commission site visit in 1949. He was thirty-eight years old; his father and uncle had succumbed to coronaries at age thirty-seven. It was a sobering thought, indeed. With his wife Genevieve's determined efforts and his own, he lost 50 pounds, quit smoking, and after a year's convalescence continued the work he had set about to accomplish. Like so many aspects of his research, it was sheer good fortune that he should suffer a coronary in a room with a distinguished physician, Nathaniel Berlin, and next door to Jack Gofman, a physician and father of LDL and HDL serum lipoproteins and the consequent modern dietary concerns.

Clearly, Melvin's agile mind was working day and night, continuously churning through his data, plans, and deductions. Even the writing of a paper for publication was clearly the result of intense planning. His coauthor would meet him and his secretary at eight o'clock in the morning to find him pacing around the table, ready to dictate the manuscript, the technique he had learned from G. N. Lewis. In an hour his part was finished, and he went off to deliver his lecture, while his coauthor would add the numbers and the secretary would transfer it all to paper and after review by members of the laboratory ship it to a journal.

THE PATH OF CARBON IN PHOTOSYNTHESIS, I-XXIV

Melvin Calvin was recruited by College of Chemistry and Chemical Engineering dean Wendell Latimer and Radiation Laboratory director Ernest Lawrence in 1945 to lead biological chemical efforts utilizing the newly available radioisotopes from Hanford and Oak Ridge National Laboratory in medical and photosynthesis research. This followed the pioneering works of chemistry faculty member Sam Ruben and Radiation Laboratory staff chemist and physicist Martin Kamen. That work had ended with the accidental death of Sam Ruben and Martin Kamen's security problems envisioned by the FBI and perpetuated by the U.S. Department of State. There is no doubt that Ruben and Kamen unequivocally earned a Nobel Prize for their discovery of long-lived radioactive carbon C-14, which engendered a revolution in humanity's understanding of biology and medicine. Lawrence was understandably proud of his contribution to that discovery and, with his brother John Lawrence, was eager to continue support once the war was over. In Ernest's words recollected by Melvin Calvin, "It's time to do something worthwhile."

The first programs centered on applications of C-14 in medicine and synthesis of radio-labeled amino acids and biological metabolites for medical research in John Lawrence's Donner Laboratory and other laboratories across the country. Calvin assembled a strong group of excellent chemists with biological interests. Then, with the 60-inch Crocker Laboratory cyclotron available for biomedical research, the original 37-inch cyclotron across the alley was donated to "Goodwill" and transported to the UCLA Department of Physics. Its wooden building, the Old Radiation Laboratory, was released stepwise for Calvin's use for the photosynthesis research program, which interested Ernest Lawrence especially. To provide continuity with the Ruben-Kamen research efforts, Calvin invited Benson, then at Caltech to return to Berkeley to establish the laboratory and direct the photosynthesis research. Benson had begun work on the path of carbon in photosynthesis as an instructor in the Department of Chemistry with Sam Ruben in 1942-43, and research with carbon-14 was well under way. Separation of the products by partition between immiscible solvents showed promise and, in the subsequent application of filter paper chromatography, involved the same principle. Solvent partition was already a standard procedure in studies of the transuranium elements and their compounds.

MAJOR STEPS IN ESTABLISHING THE PATH OF CARBON IN PHOTOSYNTHESIS

In spite of the advanced concepts of Ruben and Kamen, who brought the path of carbon into modern biochemistry, a major tenet of contemporary leaders, such as James Franck and Farrington Daniels, involved absorption of carbon dioxide and chlorophyll within some protein complex whereby the energy of a photoexcited state of chlorophyll could result in transfer of hydrogens from water to carbon dioxide. The product (formaldehyde) could polymerize to sugar and release the oxygen as molecular oxygen gas. Thus, photosynthetic production of sugars and other reduced carbon compounds was considered a "light" (photochemical) reaction. Even the products of dark fixation of $^{14}CO_2$ had not clearly dispelled such theory. Benson continued his isolation of the product of dark $^{14}CO_2$ fixation and, for a time, with the near daily assistance of Edwin McMillan, crystallized the radioactive product soon identified as succinic acid. The unequivocal demonstration of non-photochemical reduction of CO_2, however, involved illumination of algae in the absence of CO_2 instantly followed by transfer of the algae to black flasks containing $^{14}CO_2$. Analysis of the products formed revealed production of radioactive sucrose at rates approaching those in the light. Clearly, the energy absorbed by chlorophyll was used for production of phosphorylating and reducing agents capable of driving the conversion of CO_2 to sugar. Following "The Path of Carbon in Photosynthesis," then, involved metabolism independent of chlorophyll and the far more complex reactions leading to liberation of oxygen. That phrase, coined by Melvin Calvin, was used in the title of twenty-four publications.

THE FIRST PRODUCT OF CO_2 FIXATION

As designed by Sam Ruben, experiments to discern the first product of CO_2 fixation necessarily required examination of products of shorter and shorter times of exposure to $^{14}CO_2$. Such experiments were not easily accomplished with the low-specific activity C-14 (2.4×10^5 cpm/millimole) available from the wall of tanks of saturated ammonium nitrate exposed to the slow neutron clouds emerging from the 60-inch cyclotron. The far higher specific activities available in 1946 from Hanford and Oak Ridge reactions, however, rendered such experiments feasible.

With the introduction to the Old Radiation Laboratory of paper chromatographic techniques by W. A. Stepka from the Department of Plant Nutrition, short-time (5 seconds) $^{14}CO_2$ fixations could be resolved and appeared to produce only one major labeled product. Melvin Calvin had introduced the new Duolite A-3 anion exchange resin to the project. They found that the labeled compound eluted far less readily than sugar monophosphates from the resin on the column. Logically, Calvin attributed this to its having more than a single anionic group, one being a carboxyl group. Such a category included phosphoglyceric acid with its phosphate and carboxylate anionic groups. One may get the impression that Melvin was never a hands-on chemist. Not true! When they were down to the wire with the first product of CO_2 fixation, it was Melvin with the latest information on anion exchange resin from Dow Chemical who pushed identification of the unknown compound. He collected the fractions from the ion exchange column and prepared them for counting in order to get the answer to the $64,000 question.

Benson removed the phosphate by hydrolysis and produced the phenacyl ester of the resultant glyceric acid in presence of authentic Kahlbaum glyzerinsaüre from the chemistry department's great chemical storeroom. Recrystallization failed to remove its radioactivity. The first product of CO_2 fixation, then, was three-carbon phosphoglyceric acid (PGA), a long-known product of glucose fermentation with its carboxyl group containing the radioactivity, following the general reaction outlined six years earlier by Ruben and Kamen.

The competing laboratory of James Franck, Hans Gaffron, and colleagues at the University of Chicago failed to confirm this discovery and engendered a polemic in the literature greatly enhancing the inner tensions of Melvin Calvin. A symposium planned by Farrington Daniels to decide which laboratory was following the correct path was held in Chicago under auspices of the American Association for the Advancement of Science. Calvin and Benson traveled by train, as was Calvin's practice at that time, to present their procedures and results before a huge audience. At first, the Chicago group refused to acquiesce, but it soon recognized its failure and the polemic was defused.

INTERMEDIATES OF SUCROSE SYNTHESIS

With the first product identified as a known metabolite, the remaining members of the glycolytic sequence were identified by their chemical behavior. ^{14}C in the two hexoses of sucrose was measured. The prior labeling of fructose confirmed the predicted sequence of reactions.

THE "UNKNOWN" SUGARS

Two sugars separated on the paper chromatograms intrigued Benson, who examined their reactivities and recognized them as ketoses. With the able collaboration of James A. Bassham the compounds were subjected to periodate degradation. In one of them, Bassham found 14% of the activity in the carbonyl carbon, immediately turning the group's attention to the seven-carbon sugars. Benson doubly labeled the smaller of the sugars with ^{32}P and ^{14}C and measured their ratio, 2 phosphorus atoms to 5 carbon atoms. The pentose diphosphate could have few possible structures and they were identified radiochromatographically. Still, there was no indication of the two-carbon precursor of phosphoglycerate. The list of conceivable two-carbon CO_2 acceptors was exhausted. Nature had securely camouflaged its mechanism of the carboxylation process.

RIBULOSE BIS-PHOSPHATE ACCUMULATES IN THE ABSENCE OF CO2

Experiments restricting uptake of CO_2 led to increased levels of the five-carbon ribulose bis-phosphate, a logical indication that it could be the CO_2 acceptor molecule. Though they recognized this fact, it was not immediately obvious that nature had chosen to add CO_2 to a five-carbon acceptor and cleave it to produce two molecules of phosphoglycerate. Melvin Calvin's mind, constantly on the move, recognized the relationship and explained a possible mechanism for dismutation, simultaneous oxidation of one carbon and reduction of another, followed by cleavage of a six-carbon intermediate, each of the individual processes being energetically favorable. Nature had devised an efficient process that organic chemistry had failed to match.

THE PHOTOSYNTHETIC CARBON REDUCTION CYCLE

Cyclic regeneration of the CO_2 acceptor was basic to the thought of Ruben and Kamen and others concerned with photosynthesis. Recognition of the novel carboxylation mechanism closed the cyclic sequence since all the necessary intermediates were then known. With paths XXII and XXIII, the series was complete in 1958.

VISITORS TO THE LABORATORY

Isolation of the product of dark fixation of $^{14}CO_2$ attracted the interest of Edwin McMillan. For some time in 1947 he stopped in at the Old Radiation Laboratory (ORL) daily to assist with crystallization of the "dark product."

The ORL and its work on photosynthesis attracted countless scholars from abroad. Perhaps the most illustrious but least hospitably received was Irène Joliot-Curie, who with her husband Frederic Joliot had discovered artificial radioactivity. She was allowed to visit the laboratory in ORL, but she was not allowed into the restricted areas of the nuclear physics research efforts.

In 1948 Professor Hiroshi Tamiya of the University of Tokyo and Mrs. Tamiya visited Berkeley to do C-14 experiments to demonstrate photorespiration, the effects of oxygen in inhibiting plant productivity, which he had recognized during the war from his kinetic data. Tamiya's C-11 experiments with the Riken cyclotron had been terminated by the war. Tamiya was the chief guide for Vannevar Bush, Harry Kelly, and a host of scientist visitors during the American occupation. Skillful in English, trained in Germany and

France, and a scholar of the classic culture of Japan, Tamiya gained access to the upper echelons of American science and photosynthesis research in particular. He was singularly effective in bringing the finest of young Japanese scientists to American laboratories. As international relationships were restored, Tamiya was an international leader for two decades. The experiments he attempted with Benson in ORL were the first steps toward recognition of a major aspect of photosynthetic metabolism and its relationship to agricultural productivity.

For a few weeks in 1953 Ernest Lawrence and Philo Farmsworth came through the laboratory almost daily on their way to Harry Powell's glassblower's shop, carrying their prototype television tube model, which later appeared as the Sony Trinitron.

PUBLIC SERVICE

Professor Calvin performed major services for four of the nation's most distinguished scientific societies: the American Chemical Society, American Association for the Advancement of Science, American Society of Plant Physiology, and the National Academy of Sciences. For two of these societies (American Chemical Society and American Society of Plant Physiology) he served as president. He also served as president of the Pacific Division of the American Association for the Advancement of Science and on important committees of the National Academy of Sciences (including the chairmanship of the Committee on Science and Public Policy). All of this service constituted an admirable contribution to the functioning of these important scientific organizations and to the scientific and scholarly progress of the nation.

Calvin's simultaneous service to the U.S. government was equally outstanding. Of the many advisory posts listed in the record, particular attention should be drawn to his roles with the National Aeronautics and Space Administration (NASA), the Executive Office of the President, and the Department of Energy.

The entire effort that NASA mounted to search for life in extraterrestrial space was greatly influenced by Professor Calvin's participation and advice. His efforts included (1) plans to protect the Moon against biological contamination from the Earth during the first lunar landing (Apollo), (2) procedures to protect the Earth from possible lunar pathogens on and in the returning Apollo spacecraft, (3) strategies for the search for organic and biological compounds in returned lunar samples, and (4) plans for the search for biological compounds for life on other planets.

Professor Calvin served the Executive Office of the President in two ways. From 1963 to 1966 he was a member of the President's Science Advisory Committee. In 1975 Professor Calvin served on the President's Advisory Group on Major Advances in Science and Technology. At the conclusion of that service, President Ford wrote to Calvin:

> Throughout the past nine months, while we were awaiting creation of the Office of Science and Technology Policy, you and your colleagues were of great assistance to me and to our country in focusing attention on issues vital to our Nation which have involved science and technology. Your advice and counsel have helped give the new Office of Science and Technology and the president's Committee on Science and Technology a head start in carrying out their responsibilities.[1]

From 1981 to 1985 he served on the top advisory body of the Department of Energy, the Energy Research Advisory Board.

Professor Calvin also served on a number of international groups dedicated to the progress of world science. He served on the Joint Commission on Applied Radioactivity of the International Union of Pure and Applied Chemistry, the

U.S. Committee of the International Union of Biochemistry, the Commission on Molecular Biophysics of the International Organization for Pure and Applied Biophysics, and as U.S. chairman of the joint U.S.-U.S.S.R. editorial board for the four-volume summary entitled *Foundations of Space Biology and Medicine*. Concerning the latter, NASA administrator James C. Fletcher wrote to Calvin:

> I want especially to thank you as Chairman of the Editorial Board who worked so diligently toward the completion of these volumes in a most challenging and unusual context requiring the greatest tact and persistency. The result is a truly comprehensive and systematic treatise dealing with the problems of space biology and medicine. Congratulations.[2]

At the memorial service for Melvin Calvin held on January 25, 1997, in Hertz Hall on the Berkeley campus, Glenn Seaborg, one of the authors of this memoir, made the following remarks:

> I met Melvin Calvin 60 years ago when he arrived at Berkeley in 1937 to assume his instructorship in the Department of Chemistry. At that time I was serving as the personal research assistant of the famous Gilbert Newton Lewis. Melvin soon began to work on the theory of the color of organic substances.
>
> Melvin and I and a number of our bachelor friends lived in the Faculty Club, he in the Tower Room and the rest of us in the second story wing above the present Heyns Room. I recall that Joe Kennedy and I used to join Melvin in his room to imbibe alcoholic drinks which put us in a good mood for flirting during dinner with the girl waitresses in the Club's main dining room, which is now the Kerr Room. Melvin was more successful at this than Joe and I.
>
> Melvin and I were especially close and I have much to thank him for. I remember that he was responsible for breaking the ice to enable me to start a serious courtship of my wife, Helen. I had been trying unsuccessfully to date Helen but Melvin found a way. He induced her to accompany him to the Oakland Airport to meet me as I arrived home from a trip East in August of 1941. He put his Oldsmobile convertible at my disposal and after delivering Melvin home I brought Helen home along a long circuitous route that enabled me to get much better acquainted with her.

I took advantage of Melvin's Oldsmobile to get even better acquainted with Helen during the ensuing months and we became engaged in March 1942, just before I left for Chicago to work on the Plutonium Project of the Manhattan Engineer District, culminating in our marriage in Nevada in June during a return visit to the West Coast.

A couple of months later, upon a return trip, I found a telegram from Melvin inviting me to be his best man at his wedding, scheduled for October 4. I traveled to Berkeley on the streamliner "City of San Francisco," arriving the morning of October 4. Later that morning I served as best man at Melvin's marriage to Genevieve Jemtegaard. I congratulated him heartily for having the good taste to marry a Scandinavian. The ceremony was followed by a lunch for all the guests at the Claremont Hotel in Berkeley. Later Melvin and Genevieve left for their honeymoon and I don't have any details to present regarding their activities on this occasion. (Their marriage led to three children, daughter Elin (Mrs. Sowle) and Karole (Mrs. Campbell) and son Noel, six grandchildren and two great-grandchildren. Genevieve died of cancer in 1987.)

Melvin conceived and led the development of the thenoyltrifluoroacetone (TTA) solvent extraction process for the separation and decontamination of plutonium. Although this process was not developed in time to be used in the plutonium production plant at the Hanford Engineer Works during the war, the use of TTA in separation processes proved very useful in laboratory work during the ensuing years.

Right after the war Melvin began his seminal research on photosynthesis using carbon-14 as a tracer, which led to his brilliant elucidation of this vital process. This culminated with his receipt in 1961 of the coveted Nobel Prize in Chemistry.

The following April, President John F. Kennedy had his remarkable dinner at the White House for 49 Nobel Prize winners. This was the occasion when Kennedy made his famous ad lib remark approximately as follows: "This is the greatest gathering of brains in the White House since Thomas Jefferson dined here alone." I recall that at the picture-taking session Melvin occupied the position of honor next to Jacqueline Kennedy while I found myself standing in the back row nearly out of sight.

I won't try to speak of Melvin's many other outstanding scientific discoveries and achievements. Let me just conclude by reiterating that I have been close to Melvin during these 60 years. I regard him as one of the best friends I ever had.[3]

In 1980, after stepping down from the directorship of the Laboratory of Chemical Biodynamics/Chemical Biodynamics Division, Calvin maintained a small research group in one of the chemistry department buildings until 1996. Two areas of research were emphasized: (1) artificial photosynthesis, the concept of using solar energy conversion and modeled after natural photosynthesis and (2) the determination of whether or not certain plants could produce hydrocarbon-like materials that could be used as substitutes for fuel and chemical feedstocks.

Melvin Calvin engaged in an extraordinarily broad range of significant scientific activity, of which his role as "Mr. Photosynthesis" was the most outstanding.

Melvin's alma mater Michigan Technological University established in 1997 the Melvin Calvin Nobel Laureate Lecture, for which Glenn Seaborg was scheduled to give the first lecture in June 1998.

THE AUTHORS GRATEFULLY acknowledge the helpful assistance of J. A. Bassham and Marilyn Taylor, dedicated secretary to Melvin Calvin from 1946 to the present.

NOTES

1. President Gerald R. Ford, personal communication, August 13, 1976.

2. James C. Fletcher, personal communication, October 5, 1976.

3. Glenn T. Seaborg, remarks at Calvin memorial, University of California, Berkeley, January 25, 1997.

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