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GILBERT NEWTON LEWIS: HIS INFLUENCE ON PHYSICAL-ORGANIC CHEMISTS AT BERKELEY

Melvin Calvin

March 1982



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BERKELEY

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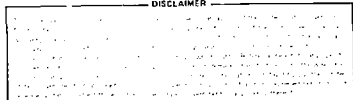
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Abstract

There will be a review of the historical contributions of Gilbert N. Lewis to science and a discussion of the influence of Lewis on the research of the members of the physical-organic staff at Berkeley, including Melvin Calvin, during the twenties, thirties and forties. Some specific examples will be discussed. Also, the effect of Lewis, his science and administrative concepts in the creation of excellence in a department of chemistry will be reviewed.

Presented at 183rd National Meeting of American Chemical Society, Las Vegas, Nevada, March 30-31, 1981, at a symposium of the Division of Chemical Education on "Gilbert Newton Lewis".

DISCLAIMER



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INTRODUCTION

My task is to describe the influence of Gilbert Newton Lewis on my own research and how Lewis influenced physical-organic chemistry research at Berkeley. In order to do that, I thought it would be useful to give you some sense of the background of Lewis at the time I encountered him in 1937. I was the last of the non-Berkeley faculty members whom Lewis brought to the Berkeley Campus and the first one in twenty-two years.

When Lewis (Figure 1) came to Berkeley in 1912, he brought with him three people. There were already four members of the chemistry faculty at Berkeley in 1912 (Booth, Blasdale, O'Neill and Biddle), and of the four who were here in 1912 I knew only Blasdale. Lewis brought with him: Merle Randall, Richard C. Tolman and William C. Bray and I knew all of them. Therefore, of the original group of chemistry faculty in Berkeley in 1912 I was acquainted with Lewis, Randall, Tolman and Blasdale. From 1913, when Joel Hildebrand and Ernest Gibson were added to the chemistry faculty, until 1937, when I came, every other addition to the/faculty was a graduate of the University of California, Berkeley. These were: Hildebrand (1913), Gibson (1913), Branch (1915), Porter (1917), Eastman (1917), Latimer (1917), Stewart (1917), Olson (1919), Hogness (1921), Giauque (1922), Rollefson (1923), Libby (1933), Pitzer (1937), Ruben (1938) and Seaborg (1939) (see Table I). The most important addition to the faculty for the subject I am discussing is "Jerry" Branch (Gerald E.K. Branch) who first took his degree in 1911 in Liverpool, England. He came to Berkeley in 1912 at the advice of his professor (F. G. Donnan) in Liverpool because at that time Lewis' reputation had already spread to Europe. Branch took his PhD in 1915 with Lewis on "The Free Energy of Formic Acid". The chemistry department in Berkeley as it appeared in 1916 and 1918 is shown in Figures 2 and 3.

TABLE I

University of California, Berkeley -- Chemistry Faculty

| Year Joined | Name | Degree Date | Where Taken/With Whom |
|-------------|--|-------------|---|
| 1912 | Faculty on hand at time Gilbert N. Lewis arrived in Berkeley | | |
| | Booth, Edward | 1877 | UC Berkeley |
| | O'Neill Edmond | 1879 | UC Berkeley |
| | Blasdale, Walter C. | 1892 | UC Berkeley |
| | Biddle, Henry C. | 1900 | University of Chicago |
| 1912 | Lewis, Gilbert Newton | 1899 | Harvard, T. W. Richards |
| | Tolman, Richard C. | 1910 | MIT |
| | Bray, William C. | 1905 | Leipzig, Luther |
| | Randall, Merle | 1912 | MIT, G. N. Lewis |
| 1913 | Hildebrand, Joel C. | 1906 | Pennsylvania, Edgar Fahs Smith |
| | Gibson, G. Ernest | 1911 | Breslau, Lummer |
| 1915 | Branch, Gerald E. K. | 1915 | UC Berkeley, Lewis |
| 1917 | Porter, C. Walter | 1917 | UC Berkeley, Biddle |
| | Eastman, Ermon D. | 1917 | UC Berkeley, Lewis |
| | Latimer, Wendell M. | 1917 | UC Berkeley, Gibson |
| | Stewart, T. Dale | 1913 | UC Berkeley, Tolman |
| 1919 | Olson, Axel R. | 1918 | UC Berkeley, Lewis |
| 1921 | Hogness, Thorfin R. | 1921 | UC Berkeley, Hildebrand |
| 1922 | Giauque, William F. | 1922 | UC Berkeley, Gibson |
| | NOBEL PRIZE, 1949 | | |
| 1923 | Rollefson, Gerhard K. | 1923 | UC Berkeley, Lewis |
| 1933 | Libby, Willard F. | 1933 | UC Berkeley, Latimer |
| | NOBEL PRIZE, 1960 | | |
| 1937 | Pitzer, Kenneth S. | 1937 | UC Berkeley, Latimer |
| | Calvin, Melvin | 1935 | Minnesota, Glockler (UC Berkeley, 1923, Gibson) |
| | NOBEL PRIZE, 1961 | | |
| 1938 | Ruben, Samuel C. | 1935 | UC Berkeley, Latimer/Libby |
| 1939 | Seaborg, Glenn T. | 1937 | UC Berkeley, Gibson |
| | NOBEL PRIZE 1951 | | |

LEWIS' ELECTRON PAIR BOND CONCEPT INITIATES THEORETICAL ORGANIC CHEMISTRY
AT BERKELEY

While Gerald Branch was a graduate student in 1913 he published a paper with Bray entitled "Valence and Tautomerism" (1) in which he introduced the idea, obviously as a result of discussions with Lewis (and remember, Branch had only been here one year as a graduate student). . There was a bit of confusion in 1913 as to what the term "tautomerism" meant. At that time, the precise definition was not as clear as it is today, but one of the concepts that Bray and Branch introduced (and Lewis refined) was the way of describing how atoms were held together. The terms in common use then were "valence", with no sign, only a number. Lewis objected to this lack and Bray and Branch introduced the terms, distinguishable from each other, valence number and coordination number. When Bray and Branch discussed these terms, they referred to them as "maximum" valence number and "maximum" coordination number, prompting Lewis to publish a paper immediately adjacent to theirs (2), pointing out that the term "maximum" was inappropriate. Those two terms finally evolved into "oxidation number", as actually defined by Wendell Latimer, and "coordination number". The oxidation number was the number of electrons removed from (or added to) the atom in the particular compound, and this number had either a plus or minus sign attached to it. The coordination number was defined simply as an integer, the total number of atoms or groups bound to a center. The introduction of these two terms and their precise definition by Bray and Branch in 1913, and confirmed by Lewis using the term "polar number" as having to do with the electrons taken away or given to an atom, was a "breakthrough" which led Lewis to the notion that the way in which atoms were actually held together was by virtue of the electrons between

them. Of course, there were extreme cases in which the electrons were transferred completely from one atom to another, as in lithium chloride. There were also other cases in which electrons were shared between two atoms, to fulfill the orbital requirements of both atoms between which those electrons lay, giving rise to the concept of the shared electron pair bond. These two papers (one by Branch and Bray and the other by Lewis) brought together in one pattern the ideas of inorganic chemistry (lithium chloride) and the ideas of organic chemistry (methane).

Shared Electron Pair Bond. In the early days probably the most important concept that Lewis developed was that of the shared electron pair bond, which eventually gave rise to some coherence to the theory of organic chemistry. I would like to quote from Robert Kohler (3) concerning the significance of that particular development:

"The first satisfactory picture of the chemical bond was proposed in early 1916 by Gilbert Newton Lewis. His book, Valence and the Structure of Atoms and Molecules published in 1923 (4), which elaborated the picture of the bond and its shared pair of electrons, was the textbook of the new generation of organic chemists. Without Lewis' conception of the shared pair bond the interpretation of reaction mechanisms already begun by the British school of Lapworth, Lowry, Ingold and Robinson, would not have gotten very far. The shared electron pair concept was really the foundation of physical-organic chemistry. Likewise, without the idea of the shared pair bond, then being used with increasing competence and success by organic chemists, the application of quantum mechanics to the chemical bond in the late 1920's by London, Schroedinger and Pauling, would have begun on far less certain grounds."

The organic chemists were trying to do mechanistic studies with a bond represented by a line, which was perfectly adequate for structural chemistry but was not at all adequate when chemical change discussions were started. The English chemists (Lapworth, Lowry, Ingold and Robinson) were trying to understand organic reaction mechanisms, and the first theoretical organic chemist in the United States (actually an Englishman!) trying to understand these reactions was Gerald Branch here in Berkeley. In this connection, I'd like to read you from a letter from Col. A. H. Foster, Air Force (Ret.) who was a student in the College of Chemistry from 1913-1917. In my correspondence with Col. Foster some years ago, he listed the faculty he knew here in Berkeley. He talked about Lewis, O'Neill, Tolman, Blasdale, Hildebrand, Randall, and then he came down to "Jerry" Branch, "whose status I never understood, although he and his lovely wife were both evident in the laboratories. Dr. Branch had one of the quickest minds of any person I ever knew, and I have often wondered what he made of his life".

Professor Branch wrote an essay on Gilbert Lewis in 1951 and presented it at the History of Science Dinner Club in Berkeley in 1953 (for text of this essay, see Appendix). I'd like to quote from that document, because it shows how close Branch and Lewis were, and how conscious they were of the transition that chemistry was undergoing under the influence of the electron pair bond:

"Lewis became interested in the nature of the atom very early in his career. His notebook of 1902 contains the first formulation of his theory, but he published nothing in this field until his interest was revived by the publication of a short article on the nature of bonds by W. C. Bray and the author (G.E.K. Branch) in 1913. This was followed almost immediately by a publication on the

same subject by Lewis, and the revival of his ideas of 1902, which were thrown to that den of lions, the research conference. The theory was not published until three years afterwards (in 1916, under the title of The Atom and the Molecule (5))."

Another essay (6) many years later, also confirmed the long term interest of Lewis in the nature of the atom.

The reference to the research conference by Branch was very significant. It was the way Lewis tried out his ideas. The research conference, when I came to Berkeley in 1937, was held on Thursday afternoons. Lewis would either try out his own ideas, or look around at the faculty members and graduate students and ask "What are you doing?", and that was the start of the discussion. This is where the research ideas of the faculty and students were honed, and where the inconsistencies of the theories were thrashed out (6). This is the reason why Bray and Branch could write an article for *J. Amer. Chem. Soc.* in 1913 (1) defining the oxidation number and coordination number and have it followed immediately in the same issue by an article by Gilbert Lewis on the same subject (2). The research conference had been the arena, so to speak, to clear up any arguments which might have existed, and by the time the papers were published, the various inconsistencies had been overcome.

The 1913 papers (one by Bray and Branch and the other by Lewis), for example, gave rise to the possibilities of describing the mechanistic way in which atoms change their bonds. The whole basis for theoretical organic chemistry was laid during those years when Branch and Lewis were working so closely together.

Tautomerism. There is another idea expressed in the 1916 paper by Lewis on The Atom and the Molecule where he used the terms valence and tautomerism in a special way. In order for you to understand the importance, I'd like to quote directly from the paper (5). This shows how far Lewis had gone in his thinking about how it is possible to describe the electron dis-

tribution in the molecule:

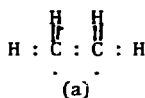
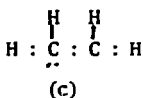
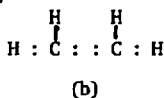
"I wish to emphasize once more the meaning that must be ascribed to the term tautomerism. In the simplest case where we deal with a single tautomeric change we speak of the two tautomers and sometimes write definite formulae to express the two. But we must not assume that all of the molecules of the substance possess either one structure or the other, but rather that these forms represent the two limiting types, and that the individual molecules range all the way from one limit to the other. In certain cases, where the majority of molecules lie very near to one limit or to the other, it is very convenient and desirable to attempt to express the percentage of the molecules belonging to the one or to the other tautomeric form; but in a case where the majority of molecules lie in the intermediate range and relatively few in the immediate neighborhood of the two limiting forms, such^a calculation loses most of its significance".

What Lewis is describing here is what Branch called "mesomerism" and what Pauling later termed resonance hybrids. Lewis recognized that in a paper published in 1933 (7) and by that time he called the phenomenon resonance.

Lewis uses, in the 1933 paper, the term tautomerism in the way we use the words mesomers or resonance hybrids. The English school from which Branch came didn't use the term resonance; they used mesomerism, that is, Lowry, Lapworth, Ingold and Robinson developed a whole theory of electron reaction mechanisms and their terms were mesomeric effects and electro-meric effects, polar effects which is an unfamiliar language today. However, this was the language of organic chemistry in the middle 1930's.

One last comment about Lewis' way of describing this, to show you he really understood what mesomerism, or resonance hybrids, were. He was trying to draw the structure of ethylene.

"Now if we have two formulae such as (b) and (c) which differ only in their electronic arrangement and are of the same "spectral type"



(he means multiplicity, he recognized the difference between these two and (a) and same gauge, then the two formulae ((b) and (c)) cannot be regarded as two possible separate states of the molecule. They must rather be regarded as two different representations of a single structural state, which ordinarily has lower energy than would be predicted if we should assume that there are two separate states and that some of the molecules are in one state and some in the other. This is the phenomenon which has been called by Ingold the T-effect, and which has been studied recently by Pauling under the name of resonance. We must not think that this phenomenon is due to the fact that we can write two or more different formulae to represent the same structure, but rather we must think that we can write two or more formulae because there has been a considerable loosening, without fracture, of the skeletal structure, so that within the skeletal structure there is far more freedom from the effects of quantum restrictions than in a saturated molecule such as ethane". (7)

COLOR OF SUBSTANCES

It turns out that Lewis arrived at his concept of the color of substances by constantly asking the rather obvious question from 1902 to 1933. He asks, for example, why is a substance colored, what causes the color. And the attempt to answer the question of why a substance is colored gave rise to the concept of the loose electron, and what we now call polarizability. Lewis recognized that those molecules which had deep color, such as dye-stuffs, had many loose electrons and these electrons can be distributed over a rather large skeleton of atoms. That idea gave rise to his notion of what he called tautomerism, but what we now recognize as mesomerism or resonance. Those two things taken together -- questions about the color of molecules, questions of polarity of molecules vs. nonpolarity (sodium chloride vs. ethane, for example) seem like rather small questions. Lewis, however, was able to put them into the context of the large question of all of chemistry.

This is what Lewis was able to do, which most people could not do, then or now. Most people can ask a small question and answer a small question, but they are not able to ask a small question and from that answer derive a larger idea. When Lewis asks a question, he asks a question that has some depth to it and that has relevance to the entire structure of chemistry and of science. For example, when Lewis asked the question about color he got into the subject of the nature of light, which has many other permutations. Lewis was not limited in his scope of thoughts to the particular question, but he always had in the back of his mind the relevance of the question to science.

I made my first contact with Gilbert Lewis when I arrived in Berkeley in 1937, and I was not aware of the things that we have been discussing about Lewis and his work. In this context, you should realize that I was the

last of Lewis' non-Berkeley faculty members, or, in another way of looking at it, I was the first non-Berkeley faculty member from outside since 1913. Lewis naturally had a little bit of uncertainty about me, and in retrospect I am able to see what he did. (Incidentally, it was Joel Hildebrand who was responsible for my coming to Berkeley. We had met in the laboratory of Professor Michael Polanyi at the University of Manchester, England, where I was a postdoctoral fellow working on porphyrin chemistry. Hildebrand discussed with me the possibility of coming to Berkeley, and when he returned to Berkeley he "convinced" Gilbert Lewis to hire me.) Lewis (shown in his Gilman Hall Office, Figure 4) was looking for a common interest between us, to see if he could get me closer to him and his research, to find out whether I could stay at Berkeley or not. He found a way through his favorite subject, namely, why are molecules colored, the question he had repeatedly asked from 1902 to 1933. Almost every one of his papers which were concerned with valence and structure of molecules had a little section in it on the color of organic molecules.

We began with that -- let's write a paper on the color of organic substances, he said. In order to do that, we had to review the subject of color, which meant finding out what the status of the knowledge of this field was at the time (1937). This meant laying out in the seminar room in Gilman Hall all the references to the color of organic substances which were available. The best reference was the large dyestuff compendium, the dye index, which listed thousands of dyestuffs made throughout the world. It contained paragraphs on the properties of the various dyestuffs. Lewis read the references, which took several months, and it was also necessary to have the structural formulas. This searching and literature examination went on for months, perhaps a year. Lewis examined the material and absorbed

it and then, in his usual characteristic fashion, felt it was time to write. (Other people in the program today have had experiences of writing with Lewis -- Seaborg, Kasha, Bigeleisen -- and they can confirm the method I will describe to you.) Lewis would walk around the table in the seminar room and dictate, and I would write. It went quite smoothly and very, very nicely. Lewis dictated very carefully, and little revision was necessary. One time he spoke his sentence and I didn't write it down. Lewis looked over my shoulder to see what happened, and it was already written down! That wasn't difficult, because we had discussed the subject so much, and I had heard that sentence before. Having finished the paper on The Color of Organic Substances (8) it was published in Chemical Reviews in 1939. Lewis had decided by that time that I could stay in Berkeley.

Phosphorescent State and Paramagnetism. As a result of writing the color paper we did another experiment (which you will hear more about from other participants). We found in examining some of the dyestuff information that we assembled for the color paper that characteristics such as fluorescence were described. Lewis got the idea to find out why all dyes did not fluoresce, why didn't dyes re-emit the light they absorbed. He recognized the reason for this was that a molecule in an excited electronic state can either emit the light directly (fluorescence) or transfer that electronic energy into vibrational-rotational modes (energy loss as heat). If such is the case, it should be possible to reduce that energy loss by freezing the molecule in a rigid medium so it cannot rotate and vibrate. Then it would have to emit light.

We made up a mixture of ether-pentane-alcohol, which could be frozen in liquid nitrogen where it makes a glass. If the dyestuffs are dissolved in this mixture and frozen in liquid air, they all fluoresce. In fact,

some of them phosphoresce, which means some emit light long after the exciting light is turned off. Lewis wanted to know the reason for that and also wanted to know the reason for the color change of the emitted light. When the emission lasted for a long time, it was of a different color from when it lasted a short time. In other words, the phosphorescence and fluorescence were different colors; usually the phosphorescence had a redder color.

Lewis didn't really know much quantum theory (he didn't really want to know very much), but he did know one selection rule, i.e., you cannot have a radiation transition between two states of different momentum. It is necessary to provide for the change in angle of momentum. He knew that if you have an excited state that has two electrons each with spin (by that time, spin had been discovered, even though Lewis had only postulated it in 1916), perhaps the excited state that normally fluoresces had a way to get into another kind of excited state, by thermal degradation, in which the two electrons are parallel instead of antiparallel as they normally are. This means the spin angular momentum will be different in the two states. The only way for the excited electron to get back down again is by vibrational interaction, which would allow the net momentum to return to zero, or it would have to wait in the new excited state for a long time. This is the prohibition of the singlet-triplet transition. Lewis surmised that the long-lived state is a triplet state, which is the reason it lasted so long.

Magnetism and color were his two "bugs". In all his papers, Lewis always asks about the color and what are the magnetic consequences of the color. That is the reason Lewis felt, for example, that ethylene was less diamagnetic than it should be, or a little more paramagnetic than it should be, due to the mobility of the electrons. He used Pascal's magnetism con-

stantly to confirm theoretical notions that he had evolved when he asked the question concerning color. The same ideas were recurring in 1938, twenty years after he had first proposed them. If the material is a triplet it has to be magnetic since the electrons are unpaired. If it is this kind of thing, then the molecule should be paramagnetic and it should be possible to find that answer. This was an elegant experiment which has been described in detail elsewhere (6). After Lewis was satisfied that the results were correct, the paper on the paramagnetism of the phosphorescent state was published (9). Lewis has satisfied himself once more that this "outlander" from the Midwest who had come to Berkeley was satisfactory.

GERALD BRANCH AND ORGANIC CHEMISTRY AT BERKELEY

At that stage (about 1939), Lewis gave me the clear impression that I should work with Gerald Branch (Figure 5) and collaborate with him in the preparation of a book on theoretical organic chemistry. Lewis told me that Jerry Branch had it all in his head, but he couldn't seem to get it written down. Therefore, it was my task, according to Lewis, to write down what was in Branch's head. He also must have said something to Branch as well, because he (Branch) invited me to come and work with him in this area.

In any case, we worked out an arrangement by which I would come to Branch's house for dinner once or twice a week, and Esther Branch, his wife (also a chemist with a PhD from Berkeley) would cook dinner, and we would then go to work. Branch would have written some material during the week, and I was supposed to write some more. We would rewrite and then plan next week's task. That's how the book was written. I must have had fifty dinners at the Branch's house, and the book was written and finally published in 1941 (10). It was the first book on theoretical organic

chemistry that had ever been written in the United States that contained quantum mechanical language in it (those were the chapters I wrote). The chapters written by Branch were detailed analysis of the effect of structure on the acidity of hundreds of organic (and inorganic) acids. (Acidity was another recurrent Lewis theme: Color, magnetism and acidity. The theory of acids and bases had been formulated much earlier (2,4), but Lewis expanded these notions, with Glenn Seaborg as his personal assistant to do this.) Branch had already in his mind gone over the effect of structure on acid strength, as measured by pH and pK_a and he used that as the foundation of the analysis of mesomeric(resonance, R_α) effects and inductive (polar, I_α) effects. These concepts are used to describe the effects of substituents and structure on acidity of any given series. The total effect is given in terms of a property of the substituent ($I_\alpha + R_\alpha$) and a property of the skeleton upon which the substituent is placed (A_α). (This same type of explanation was used later by Hammett (σ and ρ) at Columbia.) One constant had to do with the effect of substituent (σ) and the other constant had to do with the effect of structure (ρ) upon which the substituent was placed.

The publication of The Theory of Organic Chemistry in 1941 by Branch and Calvin was the beginning of theoretical organic chemistry in the United States, and it is related not only to the work of Gilbert Lewis but to the English school which began with Lapworth, Lowry, Ingold and Robinson. Branch, because of his English background as an undergraduate at Liverpool, had brought the seeds of these ideas to Berkeley. These concepts evolved under the stimulus of Lewis' ideas of the electron pair bond into resonance, coordination theory, etc.

We can trace the effect of Lewis' style and his thinking, not only on chemistry as a whole, but particularly on theoretical organic chemistry

really
which was/founded on the Lewis electron pair bond and all of the things that flowed from it, with the advent of Pauling's resonance quantum theory and modern molecular orbital theory. The initiation of this new phase of dynamic organic chemistry began with Lewis' invention, or recognition, of the electron pair bond and how it could be modified and used.

Branch's anticipation of this phenomenon has never been properly realized or acknowledged. Branch came to Berkeley in 1912 as a graduate student, the year that Lewis came to Berkeley, at the very beginning of the seminar discussions on the subject of how the molecule is put together. Branch participated in the discussions and was influential in the development of Lewis' ideas. They interacted with each other. When Lewis got through with me on the color and magnetism efforts, he thought that the next proper place for my efforts would be with Gerald Branch in the preparation of the treatise on theoretical organic chemistry. This book, in effect, organized all of organic chemistry in terms of electron theory. Physical-organic chemistry at Berkeley and in the United States depends upon Lewis' initial stimulus in the concept of the electron pair bond and Branch's evolution and development of that concept and stimulus.

LEWIS' ADMINISTRATIVE CONCEPTS AT BERKELEY

I'd like to touch briefly on some of the particular methods that Gilbert Lewis used in establishing the department at Berkeley, shown here in 1951 (Figure 6) on the occasion of Joel Hildebrand's seventieth birthday. (Note that even though Lewis himself is not in this photograph -- he died in 1946 -- Latimer, Stewart, Hildebrand, Giauque and Branch are evident, as well as Pitzer, Calvin and Seaborg, of the original group mentioned earlier in this paper.)

The department of chemistry at Berkeley was (and is) one of the most highly regarded in the United States and worldwide. The reason for this

is perhaps best expressed by Gerald Branch in his essay on Gilbert Lewis to which I referred earlier and which is reproduced in its entirety in Appendix I. As a teacher and administrator, Lewis opposed the tendency of specialization in curriculum and he kept the number of undergraduate courses to a minimum with the purpose of preparing students with a thorough grasp of fundamentals rather than a mass of facts. Also, segregation of students was made early, in the freshman year, and the better students, were assigned the better instructors. To quote Branch:

"Lewis believed that for a chemist to be useful to the world he should have a superior mind. In consequence, he preached that the department use its time and energy on good rather than average students. This somewhat undemocratic principle was often harshly criticized".

For graduate students, Lewis' methods were also not orthodox, but successful, the general principle being to allow the graduate student the greatest possible latitude. The students acquired initiative, morale and a fine cooperation among themselves and the faculty. The weekly research conference (described in reference (6) and Appendix I) was the most important medium through which Lewis educated the graduate students, staff and himself.

EVALUATION OF GILBERT LEWIS AS A SCIENTIST

The success of Lewis and his education of graduate students and faculty is mirrored in the worldwide achievements of those students and staff. As an example, the following people, either students of Lewis or faculty members at Berkeley under Lewis, have received the Nobel Prize in Chemistry: Harold C. Urey (1933), William F. Giaque (1949), Glenn T. Seaborg (1951), Willard F. Libby (1960) and Melvin Calvin (1961). No other single teacher has such a record of students.

There is no doubt that the pre-eminence of the department of chemistry at Berkeley was largely due to the fact of Gilbert Lewis' direction for twenty-nine years. The Lewis school of chemistry, with its concepts of intellectual vigor and excellence, has spread throughout the world, as his students, and their students, have created ever widening circles of teaching and research.

The following quotation concerning the effect that Gilbert Lewis had on chemistry (and science) is taken from the resolution read before the Academic Senate of the University of California, Berkeley, after the death of Lewis (Figure 7) in 1946; it was written by Professor G. E. Gibson (11):

"The half-century which terminated with the death of Gilbert Newton Lewis will always be regarded as one of the most brilliant in the history of scientific discovery, and his name ranks among the highest in the roster of those that made it great. The electron theory of chemical valence, the advance of chemical thermodynamics, the separation of isotopes....., the unravelling of the complex phenomena of the adsorption, fluorescence and phosphorescence of the organic dyes are among the achievements which will always be associated with his name.

The methods he chose were always simple and to the point. He was impatient of unnecessary elaboration....When the point at issue seemed to him sufficiently important, he would not hesitate to employ apparatus requiring skill and delicacy of manipulation, as in the beautiful but difficult experiment by which he and Calvin demonstrated the paramagnetism of the phosphorescent triplet state (9).

As a man he was a great soul whose inspiration will never be forgotten by those who knew and loved him. He was one of those rare

scientists...who are also great teachers and leaders of a school, so that their influence is multiplied by the many they have inspired".

CONCLUSION

Let the final words by Lewis' own, delivered on Charter Day, University of California, Los Angeles, March 22, 1935 (12):

"Society is becoming increasingly aware of the power of science, to bring weal or woe to mankind. But now when it is seen that the same science that brings prosperity and comfort may lead to depression and discomfort, men are beginning to look with mixed feelings at this monster which society may exalt or persecute, but cannot view with indifference. Perhaps my topic today should have read 'Ought Scientists to be Burnt at the Stake?' I shall not attempt to decide this question, but only to present in a cursory way some of the pros and cons...But if scientists are to be destroyed, let them not alone by the victims; every creative thought must be extirpated. A philosopher's epigram may kindle a world war. So scientist, inventor, artist, poet and every sort of troublesome enthusiast must together be brought before the bar of the new inquisition".

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APPENDIX

GILBERT NEWTON LEWIS, 1875-1946*

by

Gerald E. K. Branch

* From History of Science Dinner Club Papers, July 13, 1953. This essay was written in March 1951.

Gilbert Newton Lewis was born in Massachusetts on the 25th of October, 1875, but migrated to Lincoln, Nebraska while still a child. When thirteen years old he went to the preparatory school for the University of Nebraska. On graduating from this school he went to the University of Nebraska for two years and then to Harvard College, where he obtained a B.A. in 1896.

After graduation he taught for a year at Phillips Academy in Andover. He then returned to Harvard and obtained the M.A. and Ph.D. degrees in 1898 and 1899 respectively. After obtaining his doctorate he was appointed an instructor in Harvard. With a break of a year of study in Germany, he remained an instructor at Harvard until 1904.

Why Lewis' career at Harvard came to an end is not clear. In later life he boasted that he was fired. As an instructor he published three papers of high quality in quick succession, and then published nothing for three years. For one who published, on an average, four papers a year for the rest of his life, this lack of publications suggests a serious maladjustment to his environment. The break with Harvard may well have been fortunate for science, for on leaving that college Lewis resumed publication, although his next job might have been expected to quench the fire of his budding genius, as it took him far from any center of research. The position was Superintendent of Weights and Measures in the Philippine Islands and Chemist of the Bureau of Science at Manila. Somehow he found time for research in pure chemistry and published three papers while at Manila.

After a year in the Philippines he returned to Massachusetts as a member of the staff of the Institute of Technology, which at that time was a lively center of scientific thought. He remained at M.I.T. from 1905 to 1912, during which time he started his great contributions to thermodynamics. He also worked in other fields including relativity. The article he published with E. B. Wilson (1) was an outstanding contribution to the theory of relativity.

In the seven years Lewis was at M.I.T. he became famous both at home and abroad. In 1912 F. G. Donnan of Liverpool advised the author of this article to accept an opportunity to go to the University of California for graduate study, because he thought that Lewis was the most brilliant young physical chemist at that time.

In 1912 Lewis was appointed Chairman of the Department of Chemistry and Dean of the College of Chemistry at the University of California. These

positions he held until he was sixty-five. He continued as professor and professor emeritus until he died suddenly while working in his laboratory on the 23rd of March, 1946.

Just before coming to California, Lewis was married to Mary Sheldon, the daughter of a Harvard professor. Three children, Margery, Richard and Edward, were born to this marriage. Mrs. Lewis and the children are still alive. The two boys followed their father's footsteps in becoming chemists.

Lewis' work in California was interrupted by World War I. Shortly after America's entry into the war, he received a commission. He arrived in France in January 1918, and after a visit to the front during the German offensive of that year, he was appointed Chief of the Defense Division of the Chemical Warfare Service. For his work in this capacity he received the Distinguished Service Medal from his own country and the Cross of the Legion of Honor from France.

During the nineteenth century America was definitely inferior to Western Europe in pure science. At the turn of the century a wave of progress in science started in this country and after fifty years the United States leads the world in pure science. These great movements are common in the histories of civilizations. In such a movement, a man may be a contributor because of his scientific research and its consequent inspiration to others or because of his teaching. We do not limit the latter to conducting classes, but include the organization and leadership which enable others to be successful teachers. In both respects Lewis was a historical figure in the great movement that brought America to the foremost place in chemistry.

The effect of a man's research is not restricted to his own country; it is primarily an advance of science throughout the world. But the effect of his teaching is to a large extent localized. Further, a man's scientific discoveries are widely known, but his teaching is known only to a few. For these reasons we shall first consider Lewis as a teacher, and the founder of a great department of chemistry in Berkeley.

Lewis felt that a chemistry department should both teach the science and advance it. But in both functions the emphasis should be on the fundamental principles of the subject rather than on its industrial applications. There is always some conflict between pure and applied chemistry, and the proper balance between them in a university varies with the conditions in the country. In a young and rapidly developing country the danger is that the applied chemistry will devour the pure one, and will in its turn lose originality from being out of contact with fundamental principles. Thus Lewis' emphasis on the pure science was fortunate for California, even though it might have been deleterious to a more developed country.

Lewis believed that for a chemist to be useful to the world he should have a superior mind. In consequence he preached that the department use its time and energy on good rather than average students. This somewhat undemocratic principle was often harshly criticized. That some success was achieved in obtaining a more than average ability amongst the students in chemistry is shown by the unduly high percentage of chemists among the valedictorians.

Chemistry is a subject where specialization is rife, with the result that a curriculum is apt to become gargantuan. Lewis strongly opposed such tendencies, and kept the number of undergraduate courses in chemistry to a minimum. These courses were aimed to give a thorough grasp of the fundamentals rather than a mass of facts. Thus at a time when many chemistry departments in the country had no undergraduate courses in thermodynamics, California had two. On the other hand there was no course on petroleum chemistry in the department, although oil was a major industry in the state.

The aim of getting the student to think for himself was attained by free discussion between student and teacher and the large use of problems. The value of the former was probably increased by the circumstance that the teacher was often a graduate student. The graduate student is usually not as learned as the professor, but the student is less shy with his instructor when the latter is a graduate student. It may be noted that such contact between undergraduate and graduate student is educational to both parties.

Lewis' belief in the effectiveness of problem sets spread even to so factual a subject as organic chemistry. At the present time many textbooks in organic chemistry contain problem sets that challenge the ingenuity of both student and instructor. This might well be traced back to Lewis' influence.

Segregation of students was made as early as the freshman year. The freshman laboratory was divided into rooms that could hold no more than twenty-five. Efforts were made to assign rooms to the better students and the better instructors to these rooms. Many of the upper division courses were restricted to the better students. At one time undergraduate courses were divided into three classes, courses restricted to honor students, those attended by the better students but not restricted to honor students, and finally unrestricted courses. All honor students were required to do some research. The laboratory course in physical chemistry was divided into two sections, one of which was restricted to honor students. These complicated rules aimed at giving the better teaching to the better students.

In training graduate students Lewis' methods were unorthodox, but on the whole successful. The general principle was to allow the graduate student the greatest possible latitude. They were given the run of the store-rooms and laboratory facilities. They chose their own instructors for their research work, and could change horses in the middle of the stream. Lewis was careful to limit the number of graduate students working for him, thus preventing his eminence in chemistry from depriving the other members of the staff of assistance in their work.

The freedom given to the graduate students to obtain apparatus and materials no doubt led to some unwarranted expenditure, but the avoidance of delays and red tape more than compensated for this. Further, the students acquired initiative, morale and a fine cooperation amongst themselves. This spirit of cooperation was not limited to the graduate students, but included the staff. No one was ever too busy to help the research of another. No one, not even Lewis, was too proud to seek assistance.

The choice of graduate students was carefully made by Lewis, with the assistance of members of the staff. In this choice natural gifts were considered more important than knowledge of chemistry. In many cases the lacunae in the students' education had to be filled by undergraduate courses. Generally the deficiency was readily made up. Thus Herman C. Ramsberger took junior year courses in chemistry, yet he readily obtained his doctorate, and went on to contribute much to the advancement of chemistry, until an untimely death put an end to what promised to be a brilliant career.

Having chosen a candidate for the Ph.D., the next problem was to make sure that the choice was a happy one. The aim was to obtain this knowledge early. Constant oral examinations were used. As the number of graduate students increased, the burden of the examinations to the staff became serious. The problem was never solved satisfactorily. In some cases it was doubtful whether the recipient of the degree was really worthy. Still worse, some failed to get the degree yet in later life contributed much to chemistry and became eminent scientists.

One good result of Lewis' efforts to ascertain the worth of the student as soon as possible was the abolition of the final examination for the Ph.D. This examination was either a farce or an evil.

The weekly research conference was the most important medium through which Lewis educated the graduate students, the staff and himself. In this meeting papers were presented by graduate students, staff and distinguished visitors. These papers included work in progress in the laboratory, work planned and excerpts from the current literature. Each paper was followed by an active discussion, in which everyone could have his say, irrespective of his rank or the orthodoxy of his ideas.

In these discussions Lewis displayed a depth of insight and brilliance of thought that were an education to all who heard him. He accepted with good grace the harsh criticisms that were characteristic of the conferences. Needless to say his remarks were enlivened by his wit. This wit often pierced to the heart of a problem. One of the old graduate students has said that he never fully understood entropy until he heard Lewis define the entropy of a system as to what we do not know about it. Before publishing Lewis frequently submitted his ideas to the criticism of the research conference, or to similarly conducted special seminars.

The success of Lewis' education of graduate students is mirrored in the success of the students. Fourteen of those who obtained their doctorate during his regime are or have been heads of departments or deans in reputable universities. Three have received the Nobel prize, namely, W. F. Giaouque, G. T. Seaborg and H. C. Urey. Strange to say, many have achieved outstanding success in industrial chemistry.

Naturally Lewis owed much of his success in California to the other members of the department. Since it is at the beginning of things that contributions are most important, we shall mention William C. Bray, Joel H. Hildebrant, George E. Gibson and Richard C. Tolman as those to whom Lewis owed most. Of the later additions to the department, Wendell M. Latimer contributed most and succeeded to the deanship.

If Lewis had done nothing else but build a great department of chemistry in the University of California, he would have been an illustrious Californian; but from a wider viewpoint this achievement is trivial beside his contributions to science. These contributions covered a wide field, even extending beyond the confines of chemistry. Nevertheless, two-thirds of his published work relates to the application of thermodynamics to chemical equilibrium, a theory of atoms, molecules and chemical bonds, isotopes, especially deuterium, and the interaction of light with substances. In describing Lewis' contributions to science we shall limit ourselves to these four fields.

At the time when thermodynamics was first applied to chemical equilibrium in Europe, J. Willard Gibbs did the same thing in America independently. In a sense, at that time there were two chemical thermodynamics, European and American. In its second phase of development W. Nernst became the leading figure in Europe and Lewis in America, though somewhat later. Both men wrote textbooks that became standard works, one for Europe, the other for America.

Lewis was quite familiar with European thermodynamics. In fact, he had spent a semester at Göttingen with Nernst in 1901. Nevertheless he considered himself a disciple of Gibbs, whom he greatly admired. Fundamentally the two thermodynamics do not differ from each other. In the European system equilibrium is tied to the maximum work obtainable, which is usually represented by the symbol ΔA . In the American system it is tied to the maximum useful work obtainable, which is usually represented by the symbol ΔF . The ΔF and ΔA differ by work done against a uniform pressure by the change of volume resulting from the reaction. To some extent the use of ΔF is simpler than that of ΔA , but both are perfectly permissible. Had Lewis' contribution to thermodynamics merely been the rescue of ΔF , it would have had only academic value. But as we shall see it was much more than this.

Lewis was introduced to thermodynamics by T. W. Richards, with whom he published his first paper on the subject (2). Eight years later he published "The Outlines of a New System of Thermodynamics" (3) which contains the seeds of his later work. The fruition of his work on thermodynamics was reached in 1923 with the publication with Merle Randall of "Thermodynamics and the Free Energy of Chemical Substances" (4). This book is one of the masterpieces of chemical literature and has been translated into many languages, including Russian.

Thermodynamics is logically deduced from axioms concerning energy, work, heat and entropy. Corrected for the interchangeability of mass and energy, these axioms are exact as far as is known. Chemical equilibrium is measured by analysis. For thermodynamics to be applicable to chemical equilibrium it is necessary that some relationship be found between some quantity measured by analysis and some quantity related to work or energy. This connection was the empirical law of the proportionality between the partial pressure of a substance and its concentration. However, this law is not exact. Lewis' chief contribution to the application of thermody-

namics to chemical equilibrium was to use the proportionality between pressure and concentration only for infinitely dilute concentrations, where the proportionality is exact as far as is known. To do this he introduced two quantities, the fugacity and the activity. For infinitely dilute conditions these quantities are equal to the pressure and concentration, respectively.

In reality only one of these quantities is necessary. Although fugacity is perhaps the more fundamental, activity is the one that has survived. This is because it is more nearly related to the concentration of a solution as obtained by analysis, and the deviations between activity and concentration in solutions are normally greater than those between fugacities and pressures in gases.

The introduction of this more exact application of thermodynamics to chemical equilibrium necessitated a change in the equilibrium constant of the old mass law. These equilibrium constants are now expressed in activities instead of concentrations if the activities have been measured. The new equilibrium constants are really constant as long as the temperature is constant, whereas the older constants often varied greatly at the same temperature.

One can measure an extensive property, say volume, of a homogeneous mixture, but how much of this property is to be assigned to a particular component is indefinite. Yet for thermodynamic application it is sometimes necessary to assign a certain amount of this property to a mole of some component. Lewis surmounted this hurdle by using the increase of the property resulting from the addition of a mole of the substance to so large an amount of the mixture that the proportions of the components are not affected materially. These properties were called partial molal quantities.

Lewis introduced the term ionic strength into thermodynamics. In solutions of electrolytes the activities of electrolytes are greatly affected by the electrical charges of ions. So the effects of ions depend on their concentrations and charges. To combine the two effects Lewis introduced the quantity called by him the ionic strength, and defined as one-half the sum of all the products $Z_i^2 m_i$, where Z_i is the charge and m_i the molality, of the i th species of ion. He discovered the empirical law that the activity coefficient of a salt in dilute solution is fixed by the ionic strength. The dependence of the activities on the ionic strength was later deduced from electrostatics and statistical mechanics by Debye and Hückel, and gave rise to the famous Debye-Hückel equation.

Lewis' experimental work on thermodynamics consisted chiefly in the determination of standard free energies of substances and ions. More than half of the free energies determined were those of ions. In consequence his chief tool was electromotive force determinations.

Calorimetry at very low temperature is another powerful weapon in the determination of free energies. Lewis therefore instigated the development of low temperature calorimetric technique in Berkeley. Although he himself did not publish many papers involving low temperatures, the laboratory has become famous for this kind of investigation. The most shining worker in this field at the University of California has been W. F. Giauque, who lately has been awarded the Nobel Prize for his achievements.

Lewis became interested in the nature of the atom very early in his career. His notebook of 1902 contains his first formulation of his theory, but he published nothing in this field until his interest was revived by the publication of a short article on the nature of bonds by W. C. Bray and the author in 1913 (5). This was followed almost immediately by a publication on the same subject by Lewis (6) and the revival of his ideas of 1902, which were thrown to that den of lions, the research conference. The theory was not published until three years afterwards (7). In the same year, a very similar theory was published by W. Kossel in Germany (8).

Lewis' paper was entitled "The Atom and the Molecule". As its title suggests it was essentially two theories. The first part came immediately into conflict with the Bohr atomic theory, for in the Lewis theory the constituents of the atom were quiescent while in the Bohr theory there was constant motion of electrons within the atom. It was for this reason that the Lewis theory became known as that of the static atom. Actually the essence of both theories was the central location and greater mass of the positive nucleus of the atom, and the classification of its constituent electrons. In the Bohr theory the classification was by orbit, and it achieved the astounding success of predicting the spectrum of the hydrogen atom. In the Lewis theory the classification was by shell. It established the nature of the periodic table as based on the noble gases (helium, argon, etc.) and the dependence of the electropositivities and electro-negativities of the elements on their positions in the periodic table. At the present time the two theories have been reconciled, or if one prefers, superseded by quantum mechanics.

Actually neither Lewis' nor Bohr's theory conformed to the laws of physics as then known. For this reason Lewis sought for variation in these laws to fit his theory. In nothing that he published was there anything of this nature that was worthwhile, but actually he often tampered with thoughts that might have led to something similar to quantum mechanics.

Lewis' and Kossel's theory of the atom were essentially the same, but Lewis' theory of the molecule was not shared with anyone, nor was it contained in his early notebooks. The essence of this theory was that a bond between atoms was made by their sharing a pair of electrons of opposite magnetisms. Hence combination decreases the paramagnetism of atoms or radicals and their compounds are diamagnetic unless unpaired electrons lie below the valence shell of electrons.

This formation of bonds by sharing electrons and atoms was an anathema to the laws of physics of 1916. Lewis' attempts to explain it by magnetic forces inherent in the electrons were unsuccessful, although the inherent magnetism of electrons was shown shortly afterwards. Although the theory was very successful in organic chemistry, it almost received its death blow with the discovery of a molecule with the formula H_2^+ , for this molecule the hydrogen atoms cannot be bonded together by sharing a pair of electrons between them for the very good reason that it has only one electron. But the theory of the electron pair bond was saved by quantum mechanics, for from the postulates of the latter it was shown to be deducible (9) that two atoms or radicals would be bonded together by a pair of electrons of opposite magnetism. Further, quantum mechanics showed that two equivalent units, as for instance the two protons of H_2^+ , would be bonded by sharing a single electron. This limitation to the equivalence of the bonded particles makes the single electron bond a rarity.

Lewis' theory of the chemical bond is one of the most important contributions to structural theory and hence to organic chemistry. Linus Pauling in his book "The Nature of the Chemical Bond" (10) expresses a similar opinion. It is a striking coincidence that organic chemistry owes so much to the two physical chemists, Van't Hoff and Lewis.

Quantum mechanics not only freed Lewis' theory of the stigma of unorthodoxy which it had carried at first, but also increased its value by allowing electrons to be used in more than one way at a time. This made possible the ready explanation of the body of facts which in the older structural theory were explained by the somewhat unsatisfactory concept of residual affinity. It also solved the problem of the benzene ring, a problem that had led to the partition of books and courses on organic chemistry into aliphatic and aromatic sections.

Among Lewis' own applications of this theory of the electron pair bond to chemistry was his generalized concept of acids and bases. In this theory the base has a pair of electrons to share with the acid which has room for such a pair. Lewis' definition of an acid was therefore based on phenomena as well as on theory. Thus an acidic hydrogen compound was classed as an acid, not only for its ability to form an addition compound with a base by a hydrogen bond, but also because it gives its proton to a base in an almost instantaneous process.

Lewis also applied his theory to show the necessity of paramagnetism in free radicals. This has led to the *magnetic method* for analyzing for free radicals. It is somewhat amusing to note that this phase of the theory led Lewis to assign a new and indubitably correct formula to so simple a substance as oxygen.

In the early thirties Lewis started work on deuterium. The inspiration came from Harold C. Urey, one of the most famous of the men who have obtained the Ph.D. from Berkeley. During 1933 and 1934 Lewis published twenty-six papers on this subject. This phase of his work lasted a very short time, as nothing about deuterium appears in his writings after 1934.

He was the first to prepare pure deuterium and its compounds. Many of the chemical and physical properties of these were measured. One of the important discoveries made by Lewis was that the chemical properties of deuterium compounds differ from those of the corresponding hydrogen compounds. Theory requires that there should be a difference in the zero point energies of deuterium and hydrogen bonds, and hence difference in the chemical properties of their compounds.

To fully appreciate Lewis' work on deuterium one has to realize that at the time it was done deuterium compounds could not be obtained in bulk, and microchemical technique was in its infancy.

The last phase of Lewis' work was on the relationship between chemical constitution, absorption of light and its re-emission in fluorescence and phosphorescence. Actually this subject had interested him for many years. He had written a paper concerning the color and hydration of ions in 1906. In 1920 when he gave the Faculty Research Lecture of the Univer-

sity of California the subject was the relationship between color and chemical constitution.

His serious work on this subject began when he was sixty-four years old and continued until his death at the age of seventy-one. Leaving out posthumous work, he published eighteen papers in this field. The first of these papers and the last before his death were in collaboration with Melvin Calvin. It is fitting that this last publication was about an outstanding piece of work, in which it was experimentally shown that the phosphorescent state is paramagnetic.

Needless to say, Lewis was showered with honors at home and abroad. The most appropriate of these was the Richards Medal, for he was the most famous of Richards' students. To the regret of all his friends he was not awarded the Nobel prize. His contributions to thermodynamics and his theory of the chemical bond deserved this honor.

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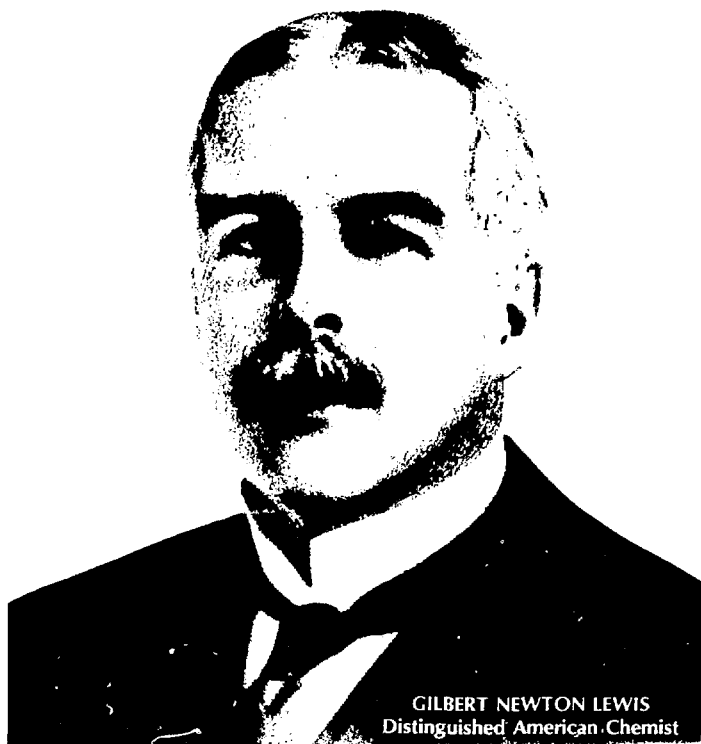


Figure 1. Gilbert N. Lewis, 1912

XBB 822-1711



Figure 2. Chemistry Department, Berkeley, 1916.
G.N. Lewis, 3rd from left, back row.

XBB 759-7206

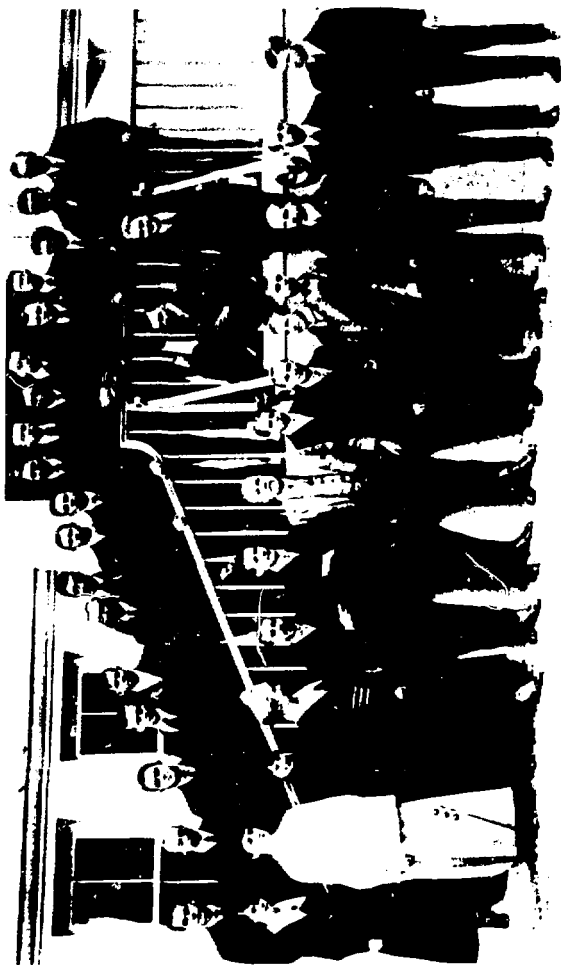


Figure 3.
*University of Cambridge officials and Cambridge staff members. Shown in this US photograph included from the
lower row: J. P. Foy, Mitchell Walker, Richard Wilham, Bill Mark Randall, G. Tim Gibson, Walter Porter,
I. Park Stewart, Edmund Cox, J. P. Foy, and Gilbert N. Lewis.*

XBB 822-1710

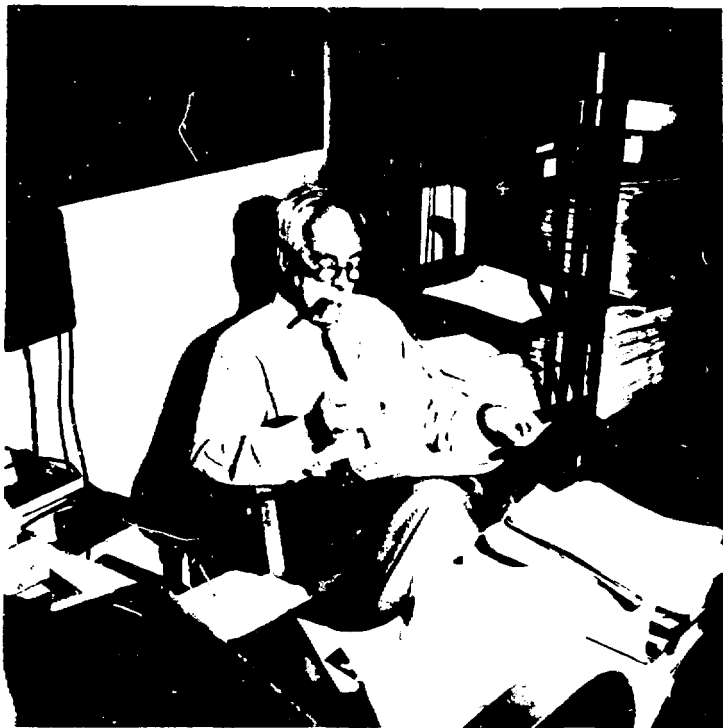


Figure 4. Gilbert N. Lewis in his Gilman Hall office, about 1940.

XBB 821-98



Figure 5. Gerald E.K. Branch, 1936

XBB 822-1104



XBB 821-874

Figure 6. Chemistry Department 1951

Front row, left to right: Latimer, Stewart, Blasdale, Hildebrand,
Gibson, Ciauaque, Branch, Rollefson. (Pitzer and Calvin immediately
behind.)

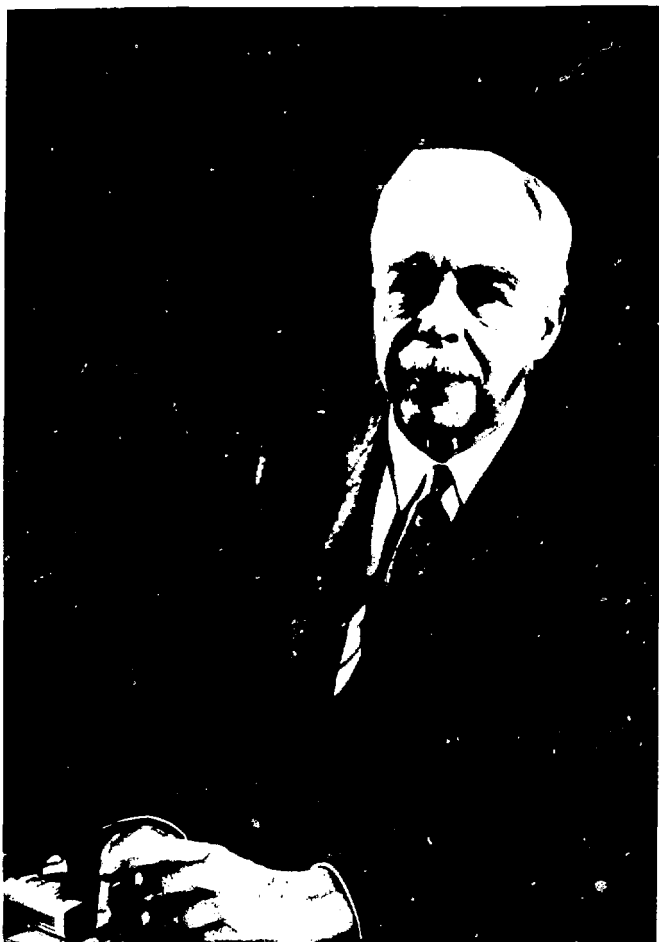


Figure 7. Gilbert N. Lewis, 1945
(photograph by Michael Kasha)

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