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OTTO REDLICH: CHEMIST AND GENTLEMAN FROM THE "OLD SCHOOL"

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The name of Otto Redlich is generally remembered as co-author of one of most used equations of state for the calculation of volumetric and thermodynamic properties of pure substances and their mixtures. Nevertheless, he made also important contributions in different areas of chemistry and chemical engineering. Pursuits of race and religious order forced him and his family to leave his native Austria and emigrate to the United States. His professional career included both academic and industrial research achievements.

Keywords: Physical chemistry; thermodynamics of solutions; Raman spectroscopy.

INTRODUCTION

Practically every day students and professionals of all disciplines in science and engineering use theories, equations, and rules, baptized with the names of those scientists who derived them. A quick survey among a representative sample of such students and professionals very surely would reveal that, with few exceptions, not many would have at least a minimum knowledge about the persons who are responsible for these scientific contributions. The Redlich-Kwong equation of state has been used for several generations of chemists and chemical engineers for the calculation of volumetric and thermodynamic properties of pure substances and their mixtures, being preferred nowadays because of its simplicity and acceptable degree of accuracy in most situations. Nevertheless, little is commonly known about each of its authors.

The purpose of this article is to reveal the more fascinating and important facts about the life and scientific achievements of Otto Redlich, a man who stands out because of a great devotion to rigor in his scientific investigative approach and his teaching. Although he is mainly remembered because his works on equations of state and non-electrolyte solutions, he was also internationally recognized for his investigations into thermodynamics, molecular vibrations, Raman spectroscopy, studies of deuterium compounds, and the ionization constant of heavy water.

THE MAKING OF A CHEMIST

Otto Redlich (Figure 1) was born in Vienna, Austria, on November 4, 1896. The home, of Jewish tradition, was additionally made up of his father Hugo (1862-1942), who was a physician, his mother, Ana Donath (1873-1956) and an older brother, Frederick (1894-1962), who would later study medicine.[1] After attending primary school, Redlich enrolled in 1907 at a Gymnasium, the state college of the Döbling district, located in the north-western part of the city, studies that he completed with honours. In these eight years he acquired his basic academic training, including studies of Greek, Latin, and Music, subjects in which he maintained a profound interest throughout his full life. In that time, the Jews were only a small minority of the total population of Vienna (fewer than ten percent).[2] It was an exciting and creative period in the city's intellectual and cultural life, in which the schoolchildren practiced to study hard and where, in accord with the educational reforms introduced half a century before, the Gymnasium teachers tried to prepare their students for researching at the university in accord with their own specialty. There was a movement that tried to change an older and deeper culture based on the senses rather than on the mind, the new trend being that social progress comes through science, education and hard work. Toward the end of his years at the Gymnasium he began to show an interest in chemistry, thanks to some informal courses and conferences which he assisted, yet the Gymnasium placed little importance on the science of chemistry.[3]

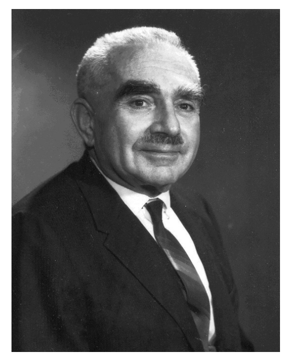

Figure 1. *Otto Redlich. Courtesy of his son, Martin Redlich*

In the spring of 1915, after finishing his secondary school examinations, Redlich joined the Austro-Hungarian Army. He spent 4 years serving as an artillery officer, mainly on the Italian front. His instructor in ballistics was the famous physicist Karl Ferdinand Herzfeld (1892-1978). Redlich's best-known student in the United States, Dr. Jacob Bigeleisen, Distinguished Professor Emeritus of the Department of Chemistry at Stony Brook University, remembers that Redlich always said about his experience in the artillery, that if "*your first shot is below the mark, your second shot must be above the mark; otherwise you have revealed your position to the enemy*".[4] At the beginning of 1918 he was allowed to attend a semester in chemistry at the Vienna Institute of Technology. In August of 1918 he was slightly wounded, and on the fourth of November he was taken prisoner by the Italian army in the South-Tirol (Südtirol) province that, until the following year, was part of the Austro-Hungarian Empire.

In August 1919 Redlich returned to Austria, and in the following autumn he began formally studying chemistry at the University of Vienna (although he also did take some courses at Vienna Institute of Technology). His main professors there were the physicist and chemist Emil Abel (1875-1958), the chemists Franz Halla (1884-1971),

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Rudolf Wegscheider (1859-1935) and Adolf Franke (1874-1964), and the physicists Gustav Jäger (1865-1938) and Ludwig Flamm (1885-1964), who decisively influenced his taste for analytical methods related to what was understood as physical chemistry. In the beginning of 1921, Otto Redlich started to work on his doctoral thesis focussed on the experimental investigation of the chemical equilibrium between the nitric acid and the nitrous and nitric oxides, with the purpose of filling the voids in the corresponding current theories. The research included the design and set up of complex equipment, and required the adaptation of different analytical methods for numerous measurements involved, mainly of electrical conductivity and electrical potential. The excessive numerical calculations required for the interpretation of the results caused him great stress and visual fatigue, yet his thesis was defended in March 1922, and he received his doctoral degree unanimously with honours.

Otto Redlich initially worked for two years in industry, at the "Cooper and Tin Foundry". In 1923 he published his first article, which dealt with strong electrolytes.[5] This subject was recurrent in his research efforts for more than four decades, through publications that, without doubt, contributed in some way to the evolution of the actual related theories. After two years, he joined the Vienna Institute of Technology as Assistant at the Chair of Emil Abel. In 1929 he was *Privatdozent* (lecturer) and in 1937 he received the title of *Ausserordentlicher Professor* (extraordinary professor). The years of working together strengthened a profound friendship between Redlich, Abel and Franz Halla, which remained as long as they lived (Figure 2). Among the students that took courses under the direction of Abel and Redlich was the Rumanian scientist Emilian Bratu (1904-1991), considered to be the forerunner of chemical engineering in his country, and founder there of the school of industrial chemistry. In 1929 Bratu was trained in the areas of physical chemistry and industrial electrochemistry, carrying out studies of evaluation of thermodynamic functions and of electrolytic dissociation of heavy water.[6]

In 1927 Redlich translated into German the book on thermodynamics that Gilbert Newton Lewis (1875-1946) and Merle Randall (1888-1950) had published in the United States four years before, and that would enjoy an immense popularity and success that still remains today.[7] The translated edition,[8] that has also been recognized as a classic in that language, would include two additional chapters, as Redlich's original contribution, the first one about the modern theory of electrolytes and the other about modern investigations of entropy at low temperatures.[9] Lewis' contributions to thermodynamics over the previous decades had been explicit with the introduction of new quantities such as fugacity, activity, and ionic strength, which boosted the use of several thermodynamic functions in chemistry, and the publication jointly with his collaborators, two years prior the appearance of the book, of a series of contributions leading to the understanding of the behavior of strong electrolytes,[10] later completed with the publication of the theory of Debye and Huckel in 1923.[11] Undoubtedly, these events significantly impacted and inspired, together with the translation of the book itself, Redlich's interest in thermodynamics from the early beginning of his career. Some later comments by Redlich, in which he shared the feelings expressed by Lewis on the subject, are conclusive at this point:[12]

> *"The fascination of a growing science lies in the work of the pioneers at the very borderland of the unknown, but to reach this frontier one must pass over well traveled roads; of those one of the safest and surest is the broad highway of thermodynamics."*

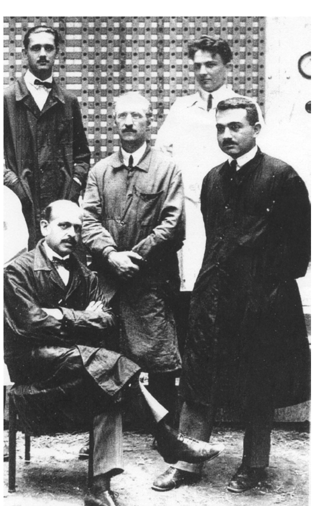

Figure 2. *Otto Redlich (right), with Emil Abel (seated) and Franz Halla (in the center) at the Vienna University of Technology in the 1920's. (The two men in the top row are unidentified). Courtesy of Martin Redlich*

The period between 1925 and 1938 was very productive for Redlich. As lecturer at the Vienna University of Technology, he shared his knowledge on a wide variety of chemical and technical subjects, on thermodynamics, analytical techniques, industrial measurements, automatic measuring, and controlling devices to be used in chemical plants, to name a few (Figure 1S – Supplementary Material). Besides that, he acted as consultant chemist for various manufacturing companies and published 52 papers on different subjects, such as thermometry, electrical conductivity and electrolyte solutions,[13] most of them in German journals. In 1932 he began to use the Raman effect as a new technique in his research.[14] The effect, that had been discovered four years before by the Indian physicist Chandrasekhara Venkata Raman (1888-1970) as a part of his work on the scattering of light,[15] had received greater attention in Germany and Austria than in any other country.[16] The use of the new technique is made evident in at least fourteen of Redlich's publications, as much in the subject of molecular structure as that of strong electrolytes. It appears that a personal relationship existed between these two scientists, since Redlich included Raman's name as a personal reference in the curriculum vitae he sent to different institutions for the first years after his arrival to the United States.[17] His two first patents, related to process instruments and control, were granted during this period.[18]

The recognition of the Redlich's scientific activity in those years is evident when on June 1, 1932 he received the Haitinger Prize for his investigations on the constitution of water and aqueous solutions,[19] a distinction conferred by the Austrian Academy of Sciences to those studies in chemistry and physics that proved to be of great practical use for industrial applications (Figures 2S and 3S). The same year he was nominated for the Ignaz L. Lieben Prize, the most important scientific award of the Imperial Academy of Sciences of Vienna, but instead it was awarded to the Austrian Chemists Georg Koller and Alois Zinke.[20] The reason may be because he had been simultaneously nominated for both prizes; it is very probable that members of both committees responsible for the two prizes were in contact with each other regarding their decisions, and it was decided not to award both prizes to one person.[21]

As a member of the Austrian Chemical Society, Redlich was very proud to have collaborated in raising money and arranging the transfer, in 1929, of Ludwig Boltzmann's mortal remains from his original burial place at the Cemetery of Döbling to an honorary grave at the Central Cemetery of Vienna. He always carried a picture of the tombstone with him.

FORCED EMIGRATION

By the early 1930's, Redlich had built a very good scientific reputation in several fields of scientific research interest. In addition to the above aforementioned work, he had also derived relations for the concentration dependence of the partial and apparent molal volume of strong electrolytes based on the Debye-Hückel theory of ionic interaction. In 1935 he had also formulated the widely used so-called "product-rule", or "Teller-Redlich" product rule, a general relation between the vibration frequencies of two isotopic molecules, by which it is established that the geometrical structure of the molecule and the masses of the atoms, and not the potential constants, are what determine the product of the frequency ratio values of all vibrations of a given symmetry type.[22] Edward Teller (1908-2003) wrote down the result, which was reported in one of his papers, but it seems that he never published its derivation. Apparently both men never met. Nevertheless, Redlich wanted to give him credit when he discovered that their work may have overlapped. It was during this period that Redlich first met some American professors, the chemists Joel Hildebrand (1881-1983), William C. Bray (1879-1959), and others from the University of California at Berkeley, when they visited Vienna. These chemists became visiting lecturers at meetings of the Chemical-Physical Society in Vienna, of which Redlich was a concurrent member.

Around this same time, Germany and nearby countries such as Austria, Russia, and Czechoslovakia, among others, began to feel a marked strong increase in organized anti-Semitism, which had begun fostering after the founding of the German nation in 1871. Some felt that this trend was justified, at least in its beginnings, because the important role played by the Jewish people in contemporary economic developments. This anti-Semitism, although with a still religious base, expanded into increasingly secular, cultural and intellectual circles. This fact, together with the belief in a supposed German cultural and racial superiority, quickly affected the scientists and academics. The 'Law for the Restoration of the German Civil Service' passed in Germany on April 7, 1933, which authorized the release or premature retirement from government services by persons who were Jews, people with Jewish origin and politically undesirable persons, came in force with even greater number of dismissals in a far briefer period at Austrian universities after the invasion of this country by the German armed forces in March 1938, because this law and similar Nuremberg Laws of 1935 were executed at once. The laws and decrees aimed at the expulsion of scientists clearly defined who had to go and who could stay on. Some of them went voluntarily and decided to emigrate because, under those circumstances, they saw no possibility to continue their scientific work in that region of Europe. The physicist and Nobel prize-winner Erwin Schrödinger (1887-1961), the physicist Hans Thirring (1888-1976), and the mathematician Kurt Gödel (1906-1978), to cite only a few names, were removed from office during the period of National Socialist rule. With very few exceptions, the silence and absence of solidarity by their non-Jewish colleagues, together with the actions of many students as carriers of the National Socialist 'revolution' at universities, contributed to making the emigration of scientists and scholars a mass phenomenon unprecedented in the modern history of academic life.[23]

At the Vienna Institute of Technology, approximately a third of the full staff of professors and lecturers of the faculty of philosophy (to which Chemistry was became a part of in 1849) were dismissed, becoming the worst loss of genius in its history. Chemistry and physical chemistry, and medical biochemistry in particular, were the most affected disciplines at academic institutions in Vienna by the Nazi policy of expulsion.[24] Among the names of professors that were expelled are those of the world-renowned polymer researcher Hermann Mark (1895-1992), the colloid-chemist Wolfgang Pauli (1869-1955) and the analytical chemist Fritz Feigl (1891-1971) for the University of Vienna, and Abel and Redlich himself, for the Technical University. Redlich lost his right to teach at the Vienna University of Technology in April 1938, was dismissed the following month, and emigrated on July 18, 1938, accompanied by his parents, his wife Marianne Loewy (1896-1972), whom he married twelve years earlier, and their only child Martin (1928-2005), first to London, and subsequently to the Unites States.[25]

THE EARLY YEARS IN AMERICA

For as long as the immigration process lasted, and some years that went ahead after it, Otto Redlich received the collaboration and support of the Emergency Committee in Aid of Displaced Foreign Scholars. This Committee helped scholars and intellectuals who were fleeing the racist and totalitarian regimes in Europe and seeking refuge abroad, relocating them in other academic institutions.[26] His name, as well that of Abel, was suggested in April 13, 1938 to the Committee by professor Bray. Because of unfavorable economic conditions at that time, there were almost no openings in the United States in the fields of inorganic chemistry, chemical kinetics and physical-chemistry. Abel got a job in the United Kingdom, and Redlich traveled to the United States.

Redlich arrived in New York on December 6, 1938. Until he got a stable job, he went on a lecture tour, giving twenty-five lectures on several subjects mainly related with theory of electrolytes, thermodynamics and Raman effect, in Boston, Minneapolis, Chicago, Seattle, Los Angeles, and other major cities, where he was always delighted by the kind reception. He had a four month stay at Berkeley, where he was the guest of Gilbert N. Lewis at the University of California, with whom he was acquainted professionally in connection with the translation of Lewis' book, but whom Redlich had never met personally before the stay. At Lewis's suggestion, he was also invited by Linus Pauling to speak before his seminar at the California Institute of Technology on April 19, 1939 on "Molecular vibrations and Raman effect of deuterium compounds".[27]

On September 16, 1939, thanks to the collaboration of the Nobel Prize winner in Chemistry and Lewis's former student, Harold Clayton Urey (1893-1981), who was very active in securing posts for refugee scientists, Redlich received an appointment as a Research Associate at Washington State College (now Washington State University). His entire time was given to research in chemistry with no teaching activities, and an initial annual salary of US $2400.[28] The position was very probably below his achievements and involved laboratory work with projects of many types (Figure 3). He was assigned some graduate students who were master's or doctoral degree candidates. The Chemistry Department at the time had little, if any, technical support facilities, no machine shop, nor a glass shop. However, these challenging circumstances allowed Redlich to utilize his wide talents. He re-purified the distilled water and numerous chemical reagents, helped with mechanical work, designed ingenious measuring instruments and control strategies for achieving the necessary high precision in techniques of analytical chemistry. In the course of this work he did also all the calculations with the experimental data, with the primary goal to verify or modify his own theories. Bigeleisen's thesis for the degree of Master in Science in chemistry, for example, confirmed Redlich's results from the early thirties and showed the dependence of the molal volume for solutions of strong electrolytes on the ionic strength.

Besides Bigeleisen, his other graduate students in 1940 were Lawrence F. Maranville and Lawrence E. Nielsen. Edward Kemp Holt joined the group later. Together they carried out research on

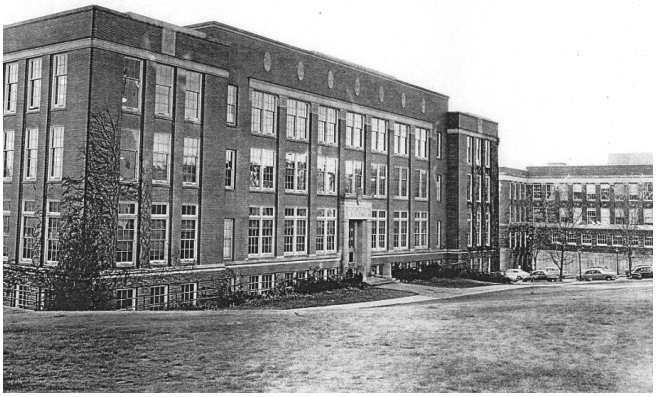

Figure 3. *Fulmer Hall building at the Washington State University, in whose top floor Otto Redlich had a laboratory. Courtesy of the Archives of the Washington State University*

solutions of strong electrolytes, structural properties of acetic, nitric and deuteron-nitric acids, and the Raman spectra and ionization of perchloric acid. When the equipment was not ready for any of his research projects, he would take advantage of the situation and undertake other research projects. For example, he continued with a series of seminars at which he, together with his students, presented topics not covered in the conventional graduate curricula in physical chemistry. These include lectures on group theory and its applications to molecular spectroscopy and the axiomatic formulation of the second law of thermodynamics according to Carathéodory.[29] This last subject was one of fundamental interest for Redlich, because, in accord with his thought, it demonstrated "*the importance of a through mathematical basis in the presentation of thermodynamics*".[30]

In the 1940's, a significant number of other faculty members in the Chemistry Department at Washington State College left to participate in various war research projects. Redlich, as a non-citizen from an enemy alien country, was not called upon for such projects, and took on some of the responsibilities of the research of those who left for war work. From early 1942 until the time that he left Washington State College, he was director of a project to investigate known processes for the extraction of alumina from residual kaolin type clays and to develop a new and economical process adapted to the economy of the Pacific Northwest and to the nation as a whole. This was one of many projects in the United States established to study the production of alumina from raw materials other than bauxite, and was justified by the need to materially assist in the nation's welfare, since deposits of high-alumina in that area, which contained sufficient alumina to supply the aluminum industry for many years, had been blocked. The project was completed in 1949, some years after Redlich had left the College, and the newly developed process involved the combined use of both sulfurous and sulfuric acid, and allowed also the production of iron-free aluminum sulfate.[31] The same project led to the development of a patent two years later.[32]

A good number of Redlich's publications are dated in that period. Some of them dealt with his favorite subjects, such as strong electrolytes,[33] Raman spectroscopy and molecular vibrations.[34] It was also during that period that the research of a young faculty member, Philip William Schutz (1908-1947), on azeotropic solutions, stimulated Redlich's interest in the thermodynamics of non-ideal solutions of non-electrolytes.[35] He then began to work out the general theory of such mixtures.

According to available information, it seems that Redlich didn't feel fully satisfied with his appointment at Washington State College for two reasons. On the one hand, he was not ever given a regular faculty appointment in spite of all the additional responsibilities he had acquired in connection with his colleagues' participation in the war research projects. This is clearly shown in the correspondence he kept with the Assistant Secretary of the Emergency Committee, where he expressed the idea that he didn't feel that he was on a 'regular track' with his position and continuously asked the Committee to remember his name if the Committee learned of an academic position or an industrial research position that he was able to fill[27]. He also let it be known that he was bitter about the lack of recognition he received from the Chairman of the Chemistry Department there.[4]

AN OUTSTANDING INDUSTRIAL RESEARCH CAREER

In 1945 Redlich worked at Washington State College as Research Associate of the Division of Industrial Research and Extension, with an annual salary of US $4200.[28] He resigned that position in October 1, 1945, and accepted a new one with Shell Development Co. in Emeryville, California. The paper on azeotropic solutions as an example of his wide recognition of his deep understanding of the thermodynamics of non-ideal solutions, and a recommendation of Redlich's friend and colleague, Dr. Fred Stross, whom he met in 1932 when Stross was studying in Vienna for a couple of years, were two important elements that collaborated to that recruitment.

His work there is probably what he is best known and recognized for. Some of his publications correspond to the extensive research he carried out on methods for determining some properties of solid complexes of urea and thiourea with hydrocarbons, mainly *n*-paraffins and their derivates, This subject included the compilation of the required technical information for the determination of composition and necessary thermodynamic properties that helped to get a general description of the complex formation,[36] and a systematic description of the sorption of normal paraffins as a function of temperature, concentration, and molecular size.[37] Others articles dealt with theory of electrolytes and different particular subjects,[38] but surely the main activity in those times was on thermodynamics of solutions, with great implications in followed developments in the next decades.[39] Jointly with A.T. Kister, Redlich made important contributions to the measurement, evaluation, and correlation of liquid phase activity coefficient data, leading to the development of methods for their determination in multicomponent mixtures, both of electrolytes and non-electrolytes, whose accuracy is especially good for most of the binary systems.[40] They worked also on the development of group solutions models, based on interactions between groups rather than those molecular incorporating heat and entropic effects,[41] and on the measurement of vapor-liquid equilibrium data for different binary hydrocarbon mixtures.[42] Seven of his 10 patents are related to his work during this period.[43]

In 1949, jointly with Joseph Neng Shun Kwong (1916-1998), Redlich proposed the world-known equation of state that bears both names:[44]

$$P = \frac{RT}{v-b} - \frac{a}{T^{0.5} v(v+b)} \tag{1}$$

where P, v, and T, represent the pressure, molar volume, and temperature, respectively, while a and b are specific parameters for each fluid. Kwong was a Chinese chemist who had become a naturalized American, with master's and doctorate's degrees in chemical engineering at American universities, who worked at Shell between 1944 and 1951 as a Research Engineer.[45] The subject of equations of state was then little developed, and the restricted computing tools existing in previous decades had led to the necessary use of simple models strongly represented by the van der Waals equation of state, or a second-order virial equation. With the arrival of the computer era, the tendency was towards increasing both the algebraic complexity and the number of the adjustable parameters.

Although in accord with the Redlich's interests in those times regarding the treatment of gas mixtures, it was not initially conceived to treat liquid phases, the Redlich-Kwong equation of state represented in those times a great advance and improvement, if it is compared with other existing equations, including that of Beattie-Bridgeman[46] and those multi-term virial equations in series of volume and pressure by Heike Kamerlingh Onnes, and by Ludwig Holborn and Joseph Otto, respectively.[47] The success of the Redlich-Kwong equation, even over a so well-received equation as that contemporary one by Manson Benedict, George B. Webb, and Louis C. Rubin,[48] laid in the fact that it was the only available equation of state that combined ease of mathematical treatment with accuracy. It was good in that it was of cubic nature, very practical to use, and with a potential application to calculations in the liquid phase also (as it was showed some years later). As John M. Prausnitz writes with the most appropriate words "*Redlich's [and Kwong's] great contribution was to revive the spirit of van der Waals*", that in accord with his own opinion is "*… to construct a useful equation of state, it is not only wise but essential to adopt a simple physical picture, to use simple algebraic functions and, very important, to pay attention to the boundary conditions*." He concludes with " *… while van der Waals was, of course, the founder of van der Waals theory, it was Redlich who led to its mass production*".[49]

In short, the modification by Redlich and Kwong to the van der Waals equation was to establish that whereas the first term in the original equation was a reasonable assessment of the repulsive forces, the second one, the attractive-force term, needed temperature dependence to reproduce a large quantity of experimental data more precisely. At expressing the equation in a reduced form,

$$P_r = \frac{T_r}{v_r - \Omega_b} - \frac{\Omega_a}{T^{0.5}_r v_r \left(v_r + \Omega_b\right)} \qquad (2)$$

where Ω_a and Ω_b are numerical constants, it was clear that the equation obeyed the two-parameter corresponding states principles, and because that all the calculated properties with it were functions of the reduced temperature only, while they should also depend on the substance considered. It was then necessary to find more accurate expressions for the dependence of parameters on the temperature, which, in turn, implied the introduction of at least one additional parameter in addition to the two critical constants involved in the evaluation of *a* and *b*.

The main modifications to the original expression showed that the temperature dependence of the attractive term could be modified to fit the experimental fact that the logarithm of the reduced vapor pressure of the substances is almost a rectilinear function of the inverse of the reduced temperature, thus improving the general prediction of pure fluid properties and phase equilibria of mixtures. The more appropriate new parameter that satisfied this relationship turned out to be the acentric factor, ω, introduced by Kenneth S. Pitzer (1914-1997) in 1957 for improve the treatment of mainly non-polar substances. The original equation turned into

$$P_r = \frac{T_r}{v_r - \Omega_b} - \frac{\Omega_a \alpha}{v_r \left(v_r + \Omega_b\right)} \qquad (3)$$

where the correction factor that replaced the term of reduced temperature was then function of this variable and acentric factor.

The first modification, proposed by Grant M. Wilson in 1964, was successfully applied with greater accuracy both to the gas and liquid phases, although with great divergences at low temperatures.[50] A subsequent modification, introduced in 1972 by the Italian chemical engineer, Giorgio S. Soave (1938-), express the α function as

$$\alpha = \left[1 + m\left(1 - \sqrt{T_r}\right)\right]^2 \qquad (4)$$

where *m* is function of the acentric factor only, and then a characteristic constant for each fluid. The new equation proved particularly successful and rapidly gained widespread acceptance, became the first generalized, wide-range predictive version of the original equation, and opened the subject to a great number of posterior variations.[51] In spite of the posterior development of numerous similar expressions, this modification of the original Redlich-Kwong two-parameters equation of state is still considered as a very good option to perform multiple volumetric and thermodynamic calculations, both for pure substances and their mixtures, because its combination of low complexity and good accuracy.

Through the next three decades, Redlich himself proposed modifications to improve the accuracy in wider ranges of pressure and temperature of the results obtained using his equation of state, both in gaseous and liquid states, mainly involving the use of additional parameters.[52] There are some published articles that survey in different periods of time the numerous reported modifications to the original equation.[53]

The company's policies at Shell Development then admitted a dual track by which an outstanding scientist could advance in the organization with identical compensation and benefits in both: promotion to management or promotion to a senior scientific research position. Redlich choose the last one. Although the normal rules at Shell Development at that time fixed the retirement age at 60, his personality and many contributions, most of them of immediate practical interest for the company, managed him to win everyone's respect by both the management and scientific staff, and allowed him to stay there five years more.[4]

RETURN TO ACADEMY

The next and the last stage of Redlich's professional activity was his return to academia. In 1962, after his retirement from Shell, Redlich was appointed Lecturer in Chemical Engineering at the College of Chemistry in the University of California at Berkeley and, as many members of the College, also investigator at Lawrence Radiation Laboratory (Lawrence Berkeley National Laboratory as it is now known), positions that he held until his definitive retirement in 1978 (Figure 4). The undergraduate program in chemical engineering at Berkeley started in 1946, and was one of the very few cases in the United States where it was not offered by a department of the college of engineering, separated from that of chemistry. It was not until 1957 that a Department of Chemical Engineering was created, but still within the College of Chemistry.[54] Redlich's joining the department occurred as part of a second great step of recruitment of professors in the course of that decade that included twelve more faculty.

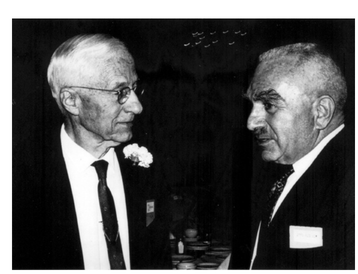

Figure 4. *Joel Hildebrand and Otto Redlich at Berkeley in the 1970's. Courtesy of Martin Redlich*

Redlich's son, Martin, related that his father said that *this period was the happiest of his career.*[12] He taught a well established course in the area of thermodynamics, and continued work on solutions and strong electrolytes. At the Lawrence Radiation Laboratory, an institution that contributed a part of his salary, he received significant research support and had several graduate students, with whom he continued his work on the ionization constants of several compounds, such nitric and perchloric acids, in normal, heavy, or deuterated water, using nuclear magnetic resonance techniques.

In the field of publications, this one was a especially important period for Redlich, in particular in the area of thermodynamics. Aware of his special fondness for this field, and faithful to a very deep-seated philosophy of thought that characterized him all the life about the fact that the precision in the language constituted an essential tool for the accuracy in analysis, Redlich began in 1962 the publication of a series of articles devoted to the clarification of some basic concepts in thermodynamics, and their relationship with other sciences.[30,55] The publication of his book *Thermodynamics: Fundamentals, applications*, just before his 80th birthday, constituted, without doubt the culminating moment of a full life devoted to the study of this science.[56] In its 10 chapters, Redlich develops the traditional content of books on this same subject, but within a general framework of basic concepts derived from epistemological principles, and with a structure that permit its application to any branch of science.

A excerpt from his publication reflects his strong conviction about the great role of this subject in a general context, and the appropriate understanding of the basic concepts. Referring to its position in the whole of science he said: "*Thermodynamics covers, indeed, our entire knowledge of equilibrium and process occurring near equilibrium in all fields of physical sciences. In this sense* ***thermodynamics may be called the root of all sciences***" (the emphasis is mine).[30] Some few months before his death on August 14, 1978, he showed once more his deep feelings about this area of knowledge, and referred again to the conceptual strength of the subject in what would be his last article, with the following conclusive words: "*For a long time, the beautiful thermodynamic edifice has stood on shaky grounds*". Then, he added: "*If we take the trouble to clarify our concepts and to eradicate the errors that have accumulated, the haze disappears and we realize that thermodynamic is more than a bunch of incoherent rules. In the end we may be surprised by the discovery that thermodynamics makes sense*".[57]

THE MAN BEHIND THE EQUATION

Otto Redlich was a kind man. This is the first sentence that it is listened when you ask to the people that met him. He was a very prudent and humble man, modest about his own achievements, and extremely easy to work with. Although generally he was sticked to old customs and traditions, was also receptive to listen new ideas. He preferred go for a postprandial walk than drive a car, an activity that learned later in his life, and that privately he considered as one of his greatest accomplishments. He successfully detached his personal life from his professional activities, and always showed an impartial attitude toward his surroundings and events. His hospitality was highly reputed.[58,59]

Lover of nature and outdoor vistas, he was owner of a superior classical education and a broad interest in history of all periods. He played the piano very well, and liked to take time to listen to music on his phonograph, in accord with his personal and particular tastes. He liked composers such as his compatriots Anton Bruckner and Gustav Mahler, and the Czech Antonin Dvoeák, but disliked the baroque music from Bach, and specially the impressionist style of the Frenchmen Claude Debussy and J. Maurice Ravel. His presentations of complicated subjects was clear, and he was a well regarded teacher, liked by his students, to whom he had a lot of fun to teach and lead as researchers without any sort of personal interests.[58,59]

About the Redlich's interests on thermodynamics and measuring techniques, Abel wrote in a letter of recommendation in 1938:[26]

> *Right at the beginning of our joint activity I learned to appreciate in you one of those men to whom the enthusiasm for the study on thermodynamics has paved the way to a profound knowledge of this science. You have successfully set out to achieve an extraordinary refinement in physical-chemical measuring methods at the time when the importance of this task had been perceived by only very few scientists in Europe.*

In the same letter, Abel refers so to Redlich's abilities and qualities for the technical and teaching work with the following words:

> *I have had an opportunity to observe your ability in organizing and in dealing with staff: these qualities, combined with your faculty to adapt yourself to varying circumstances and tasks and your resoluteness have often made me realize that you have the making of an excellent technical manager.*
>
> *The clearness of your thought, your unusual mathematical talent, your comprehensive knowledge, the certainty of your judgement, enabled you always to rise above the subject you were treating; these advantages showed themselves on many occasions during your lectures. In the University you proved yourself an eminent teacher, in the lecture room as well as in the laboratory.*

Abel, one of the men who probably knew better Redlich as consequence of nearly 16 years of close cooperation in work, emphasized some of his human qualities:

> *Loyally fulfilling your duties, kind and considerate, always ready to help, you have gained the esteem and friendship of all with whom you came into contact, your colleagues as well as your superiors and inferiors. From the moment when our joint activity started, you have gained my confidence in always increasing measure. You are one of those few men whom we learn to appreciate more the longer we know them.*

John M. Prausnitz, who was a Redlich's colleague at Berkeley, at referring him says:[49]

> *Perhaps his most valuable contribution to the education of both graduate students and faculty was his benevolent but firm reluctance to be impressed. If I, or anyone else, had some new result that I found exciting, he would smile pleasantly and say, 'That's very nice but essentially, we knew that already in Vienna many years ago, see article so-and-so published in 1927*'.

CONCLUSIONS

Otto Redlich made significant contributions to different fields in physical chemistry and thermodynamics. As a whole, his work contributed to the strengthening of the conceptual basis of thermodynamics and the evolution of our present theories of solutions. On specific practical subjects, some of his main developments still apply. The Redlich-Kwong equation is still being considered as one of the more accurate two-parameter equations of state proposed until now. Its ability to reproduce quite exactly the volumetric and thermodynamic properties of pure components and mixtures in gas phase, as well as its simplicity in comparison with other equations with more parameters made it widely favourite for calculation of densities, enthalpies and vapour-liquid equilibrium data in the design of chemical, petrochemical and related processes. By other side, the algebraic representation of the excess properties of non-electrolyte solutions, represented by the Redlich-Kister expansion, is still used to represent the activity coefficient of some systems for which the so-called local composition models later proposed don't apply. Otto Redlich has a primordial place in the recent evolution of thermodynamics, and his work still has a direct impact on the development of models for the correlation and prediction of different volumetric and thermodynamic properties of both electrolyte and non-electrolyte mixtures.

SUPPLEMENTARY MATERIAL

The Figures 1S, 2S, and 3S, showing different announcement cards for Otto Redlich's early seminars in Vienna, and the correspondence between the Austrian Academy of Sciences and Redlich on the awarding of the Haitinger Prize, respectively, are available on http://quimicanova.sbq.org.br.

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organized in New York City in May 1933 by American academicians to serve the needs of the university professors who had been dismissed from German universities because of political opinions or anti-Semitic legislation, and to preserve their attainments for the benefit of scholarship in the United States. With the outbreak of Nazi aggression, the Committee necessarily revised its mission, so as to include refugee professors from all countries in Western Europe overrun by the Nazi armies. During the 1930's and 1940's, this Committee helped bring more than 330 scholars, most of them Jewish, from Nazi Germany, Austria and Czechoslovakia to the United States. It included not only chemists, but also renowned physicists as the Nobel Prize winner in 1938, Enrico Fermi (1901-1954) and Fritz London (1900-1954), philosophers as Martin Buber (1878-1965) and novelists as the Nobel Prize winner in literature in 1929, Thomas Mann (1878-1955). The Committee was dismantled after the war in June 1945. For an account of this Committee, see Stephen Duggan and Betty Drury, *The rescue of science and learning : the story of the Emergency Committee in Aid of Displaced Foreign Scholars*, MacMillan: New York, 1948. Personal information about Redlich and his correspondence with the Committee is found in the Emergency Committee's archives in the Manuscripts and Archives Division of The New York Library, Astor, Lenox and Tilden Foundations, box 88.

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28. Manuscripts, Archives, and Special Collections, Homer Jackson Dana Papers, 1910-1968, Washington State University Libraries, Washington, boxes 39 and 47.
29. Constantin Carathéodory (1873-1950) was a skilled German mathematician, renowned for being one of the founders of the generalized metric geometry and for his deep works on the general theory of functions and the algebraic basis of the concept of integral, but mainly distinguished because of his studies on thermodynamics, which, in some way, put the full subject in a new perspective. He published in 1909 and 1925 two seminal explanatory and conclusive papers on an axiomatic approach to thermodynamics, with which, through a mathematical formulation, he was able to obtain in a formal way the first and second laws without the need for to turn to traditional artificial devices as the Carnot cycle or a periodically operating machine. With the aid of two axioms, he developed a new methodology which introduced the concepts of entropy and thermodynamic absolute temperature and allowed a rigorous formulation of the consequences of the second law. For an account on Carathéodory's work, see Pogliani, L.; Berberan-Santos, M. N.; *J. Math. Chem.* **2000**, *28*, 313.
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