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Reviews of the Recent Literature

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CHAPTER 6

Physical Chemistry

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6.1 General

Several historical studies, both by professional historians and by scientists, address general aspects of the development of physical chemistry.[1] A dissertation discussing the rise of physical chemistry[2] examines the work of the three 'founders', Wilhelm Friedrich Ostwald (1853–1932), J. H. van't Hoff (1852–1911), and Svante Arrhenius (1859–1927). Labelling them 'the ionists', it treats especially their thermodynamic theory of solutions based upon ionic dissociation in electrolytes. Max Planck's (1858–1947) own theory of dissociation, first published in 1887 and largely ignored, is also discussed. The analysis revolves around four themes: the different (national) research traditions of the ionists and Planck; the particularity of each investigator in the context of these traditions; the reception of the innovations they introduced, which depended on the compatibility of their own styles with the styles of other scientists; the character of the ionists' research which, in contrast to Planck whose work was at the forefront of a specialized field, resulted from their mixing diverse, old traditions and problems with new styles and techniques.

Apart from his very influential textbooks, Partington wrote extensively on the history of chemistry.[3] The fourth volume[4] includes a detailed narrative of the various developments in physical chemistry to the 1920s. Partington's references to the original papers are still an invaluable asset of an otherwise descriptive history.

A recent book on physical chemistry,[5] written by a scientist[6] and aimed primarily at other scientists, contains substantial historical information on the beginnings of physical chemistry and on various topics, such as chemical spectroscopy, electrochemistry, chemical kinetics, colloid and surface chemistry, and quantum chemistry. The book also discusses more general topics, such as the development of the physical sciences and the role of scientific journals in scientific communication. The same author has written a brief account of the development of physical chemistry after 1937,[7] emphasizing the application of quantum theory and the invention of new experimental methods: stopped-flow techniques (1940), nuclear magnetic resonance

(1946), flash photolysis (1950–52), crossed molecular beams (1954), temperature jump (1954), laser spectroscopy (1957), and Mössbauer spectroscopy (1958). He has also written about the history of transition-state theory.[8]

There have also been brief, general surveys of the development of physical chemistry,[9–13] a discussion of its early phase in Canada,[14] and in Japan,[15] and an analysis of physical chemistry in higher education in the inter-war period in Poland,[16] as well as more specific studies on its applications to other areas.[17,18]

A book[19] attempts to present the history of an ambivalent attitude concerning the identity of chemistry during the last two centuries and to articulate the methodological complexity of such a reading. The book addresses directly the question of whether chemistry is reducible to physics, but it also examines this issue by studying the formation of the boundaries of physical chemistry, chemical physics, theoretical chemistry and/or quantum chemistry. The author argues that the legitimacy of theoretical chemistry and the drawing of disciplinary boundaries was the result of a whole network of factors involved in the construction of the *identity* of a discipline. She examines that identity by discussing systematically, in various cases, six issues: the genealogy and historical mythology of heroic origins in the initial period of the formation of each discipline; a core literature defining archetypal language and imagery; the practices and rituals that are codified and performed; a physical homeland, including institutions based on citizenship rights and responsibilities; and finally, external recognition and shared values together with unsolved problems. The author traces these elements in the Paris School and in the London–Manchester School of theoretical organic chemistry, both for the period between 1880 and 1930; in Christopher Ingold's (1893–1970) attempt to integrate physical and organic chemistry; and in the development of quantum chemistry in the USA and Britain until the end of the 1940s.

In a previous article,[20] the same author discussed two approaches to chemistry in the 1920s, a British and a French, which shared the same objective, namely the solution of chemical problems by means of concepts borrowed from physics. The former was based on ions and electric charges and employed visual representations and non-mathematical language; the latter employed the concepts of energy and radiation and aimed at constructing a deductive chemistry. Resistance, from the end of the 19th century to the 1930s, to applying the electronic theory to reactive chemistry in France is the subject of a study.[21] Polanyi's work is, also, examined in two articles.[22,23]

Another study presents the emergence of physical chemistry in the USA,[24] by concentrating on a number of institutions and persons who played a pivotal role: the Massachusetts Institute of Technology and A. A. Noyes' initiatives there; the College of Chemistry at Berkeley and G. N. Lewis' (1875–1946) unquestionable dominance in the formation of its academic culture; Cornell and Wilder Bancroft's insistence on the utility of the phase rule, and his influence through his editorship of the *Journal of Physical Chemistry*; and the California Institute of Technology, where Linus Pauling (1901–1994) had his entire professional career from its start to the end of his life. Of particular interest are the discussion of the 'migration' of physical chemistry from Europe to the USA and the analysis of Ostwald's relationship with the many American students who spent some time at his laboratory.

The professionalization of physical chemists at the end of the 19th century and, more generally, the institutional aspects of the history of physical chemistry have

been the subject of several articles.[25–33] Finally, there are several studies on topics that were either directly related to physical chemistry, or implicated with its development.[34–38] Physical oganic chemistry is discussed in detail in Chapter 5.

6.2 Biographies

There have been numerous biographies of chemists or other scientists with considerable contributions to physical chemistry. Some of these works are full-fledged biographies, others are popular accounts, and others are papers with short biographical sketches. Among the more substantial works is a full-length study of Edward Frankland's (1825–1899) early years and his work on organic acids, the nature of flame, his founding of organometallic chemistry, his theory of valency and his coining of the term 'bond.'[39] The same author has, also, written a comprehensive biography of Frankland.[40] Svante Arrhenius and his various contributions, ranging from the ionic theory of solutions to environmental science, have been investigated.[41,42] There is also a biography of Alfred Werner (1866–1919) examining his foundational work in coordination chemistry.[43] Walther Nernst (1864–1941) has been studied in relation to science in Germany[44] and his work in physical chemistry.[45] Giorgio Piccardi (1895–1972) and his work on the chemistry of the sun have been investigated,[46,47] as have Kasimir Fajans (1887–1975), his work in radiochemistry,[48] and his idiosyncratic proposal of quanticule theory.[49] Frederic Soddy's (1877–1956) baffling life[50] and his contributions to radiochemistry have been written about and reprints of some of his fundamental papers have been published.[51] There have been numerous studies of Linus Pauling and his many scientific and political activities.[52–58] A selection of Pauling's papers is also available.[59]

William Prout (1785–1850) and his pioneering attempts to understand the nature of matter[60] have been studied, as has Ladislaus Farkas' (1904–1948) role in the establishment of physical chemistry in Israel.[61] Some protagonists of physical chemistry in The Netherlands, J. H. Van't Hoff[62] and Johannes van Laar (1860–1938),[63] have been investigated, as has Johannes Diderik van der Waals (1837–1923) and his contributions to molecular science.[64] Van't Hoff's decisive role in the development of chemical thermodynamics has also been examined.[65]

There have been studies of Chaim Weizmann (1874–1952) and his many activities, both in chemistry and in politics;[66] Jean Perrin (1870–1942) and his intensive work in Brownian motion, which consolidated his view about the reality of atoms and molecules;[67] and of The Svedberg (1884–1971) and his related work.[68] There have been contributions on various aspects of the works of William Harkins (1873–1951),[69] Morris Loeb (1863–1912),[70] Neville Sidgwick (1873–1952),[71] Viacheslav Vasil'evich Lebedinskii (1888–1956),[72] Robert Havemann (1910–1982),[73] Glenn Seaborg (1912–1999),[74] August Horstmann (1842–1929),[75] Frantisek Wald (1861–1930),[76] Elliot Alexander,[77] and Charles Hurd.[78] A dissertation discusses the role of Josiah Willard Gibbs (1839–1903) in the founding of physical chemistry.[79] There is a brief comparative analysis of Gibbs and Ostwald.[80] Fritz London (1900–1954) and his work on the covalent bond has had substantial treatment.[81] There is a study of Alfonso Cossa (1833–1902) and his work in coordination chemistry[82] and also of Wilder Bancroft's (1867–1953) attempts to found the

Journal of Physical Chemistry,[83] as well as of Charles Bury's (1890–1968) contributions to physical chemistry.[84,85]

6.3 Electrochemistry

Ever since the discovery of the wet battery by A. Volta (1745–1827) there have been attempts to understand chemical change by means of electricity. Discussions about electrochemical processes started gaining momentum at the beginning of the 19th century with the discovery of electrolysis – the decomposition of compounds by an electric current – by W. Nicholson (1753–1827) and A. Carlisle (1768–1840). But it was Humphry Davy (1778–1829) whose investigations led him to suggest that chemical affinity was due to electrical mechanisms. J. J. Berzelius (1779–1848) attempted to classify the elements according to their electrochemical properties and considered the compounds as a union of elements with specific electronegativity and positivity. However, his dualistic theory could not explain the formation of the diatomic molecules of elements. Michael Faraday (1791–1867), H. Helmholtz (1821–1894), J. H. van't Hoff and Svante Arrhenius all played important roles in attempting to systematize within their theoretical schemata the vast data that was being collected about the electrical properties of elements and compounds. The advent of the electron, of course, redefined the whole subject of electrochemistry.

Ostwald's classic book on electrochemistry has been translated into English.[86] In addition to the theory itself, the reader is presented with a host of historical details relating to the 18th and 19th centuries as well as a discussion of Ostwald's empiricist views. Another work, arising from a conference held under the auspices of the American Chemical Society, analyses the major developments and 'technologies of … electrochemistry, electrosynthesis, electroanalytical chemistry, industrial electrochemistry, electrode systems, and pH measurement.'[87]

One of the most intriguing achievements of the 19th century was James Joule's (1818–1889) determination of the mechanical equivalent of heat and, therefore, the first law of thermodynamics. There is a study of the background to this paper, through the analysis of Joule's work in electrochemistry.[88]

The German industrialist Emil Rathenau (1838–1915) recognized the industrial potential of electrochemistry. In 1887 he founded the firm Allgemeine Elektrizitätsgesellschaft (A. E. G.) and in 1893 he started a new company, named Elektrochemische Werke, which was headed by his son, Walther (1867–1922). The two men together led the expansion of these companies into the field of electrochemistry.[89]

At the end of the 19th century, the theory of electrolytic dissociation became an important part of physical chemistry. Wilhelm Ostwald, Svante Arrhenius, and Walther Nernst were among the most vigorous supporters of that theory, which also had some severe critics. The ensuing debate has been discussed in a paper, which analyses the arguments on both sides and shows how the proponents of the theory attempted to resolve its difficulties.[90]

The discovery of the piezoelectric effect (the appearance of electrical charges on different surfaces of crystals under mechanical stress) in 1880 by Pierre Curie (1859–1906) and his brother Jacques (1855–1941) is discussed in a brief paper.[91]

J. J. Thomson (1856–1940) performed extensive work in electrochemistry, atomic structure, and valency and his interest in chemistry led him to the construction of a series of models of the chemical atom.[92]

Several works discuss institutional aspects of electrochemistry in Germany.[93–97] The history of the Kaiser Wilhelm Institute for Physics and Electrochemistry, the role of its first director Fritz Haber[98] (1868–1934), and the war work carried on there have been discussed.[99] There are also papers on the researches related to electrochemistry during the Napoleonic period in Tuscany,[100] on the work of Kohlrausch in electrolytic conductivity,[101] on Humphry Davy's work for the Royal Navy on the protection of a ship's body by electrochemical processes,[102] on aspects of silver-based coulometry,[103] on Eben Horsford's (1818–1893) contributions in measuring electrolytic resistance,[104] on the role of Oliver Wolcott Gibbs (1822–1908) and C. Luckow in the early development of electrogravimetry,[105] on the electrochemical school of Edgar Fahs Smith[106] (1878–1913), and on some aspects of the work in electrochemistry of R. Behrend.[107]

6.4 Thermochemistry

The development of thermochemistry and, especially, its use for the experimental study of chemical affinity has been the subject of a number of works. Among them is an analysis of Julius Thomsen's (1826–1909) early work in thermochemistry (1852–1854) and the incorporation of the newly formulated principle of energy conservation into chemical theory.[108] Thomsen attempted to quantify chemical affinity by specifying the magnitude of chemical forces on the basis of thermochemical measurements. The attempt, however, to develop thermochemistry as a programme for reducing chemistry, *via* the mechanical theory of heat, to Newtonian mechanics became increasingly inadequate and in the 1880s was "overtaken" by the development of chemical thermodynamics. By 1884, van't Hoff and Arrhenius were able to relate the effect of temperature increases on reaction velocities.

There has been a discussion of some aspects of the history of thermochemistry, considered as a precursor of the development of chemical thermodynamics.[109] There is also a study of C. M. Guldberg (1836–1902) and P. Waage's (1833–1900) attempts to formulate an equation for the temperature dependence of the rates of chemical reactions.[110] In 1878, Paul Vieille (1854–1934), who did important work in thermochemistry, invented the calorimetric bomb. In 1884, Vieille suggested that this instrument could be used to determine, in a novel and accurate manner, the heats of combustion of carbon and organic compounds.[111]

There is also a paper on the use of platinum in high temperature gas thermometry during the 19th century[112] and there are brief discussions of some aspects of the early developments of microcalorimetry in France,[113] and the thermal dissociation of water.[114]

6.5 Chemical Statics and Dynamics

Between 1864 and 1879, two Norwegians, C. Guldberg and P. Waage,[115] studied heterogeneous systems containing solids in contact with solutions, demonstrating

experimentally that an equilibrium is reached in incomplete reactions. They also found that such an equilibrium could be approached from either direction. The driving force for a substitution was mathematically expressed as being directly proportional to the product of the masses, each raised to some definite power. Equilibrium conditions at a given temperature were expressed in terms of molecular concentration, called 'active mass.' In 1877, an equivalent mathematical derivation was reached through thermodynamic considerations by van't Hoff, who suggested that equilibrium is reached when the velocities of opposing reactions become equal and that this dynamic state is related to the concentrations or active masses. There followed a claim of priority by Guldberg and Waage who had written their original papers in Norwegian.

J. H. van't Hoff's classic text on chemical dynamics has been republished[116] and his work on chemical dynamics,[117] as well as that of Ostwald[118] and Arrhenius,[119] has been briefly discussed. There is a comprehensive narrative of the development of chemical kinetics from about the middle of the 19th century, which reconstructs Guldberg and Waage's work on chemical kinetics and compares their efforts to simultaneous work at Oxford by A. G. V. Harcourt (1834–1919) and W. Esson (1839–1916).[120]

Oxford University's College Laboratories played a very important role in the revival of the sciences at Oxford and the work done there in chemistry, and especially chemical kinetics, comprised a rather significant part of their activities. There is a study that provides information about the number of students, the kinds of institutional changes at Oxford and lists the various courses taught there.[121] There is also a history of science, including chemistry, at Oxford in the interwar years.[122]

The role of the early work on chemical kinetics in the evolution of physical chemistry has been examined with reference to van't Hoff's, Ostwald's, and Harcourt's researches prior to the 1880s.[123] There is also a discussion of chemical kinetics and thermodynamics during the 19th century,[124] and an analysis of the relation of chemical kinetics and physical chemistry up to the early part of the 20th century.[125] Studies have been made of the role of instruments and the specific laboratory locales for chemical kinetics in the interwar years,[126] and the work of H. Eyring[127] and J.-A. Muller[128] in chemical kinetics has been analysed.

The proposal, elaboration, and eventual demise in the late 1920s (after considerable controversy) of the 'Radiation Hypothesis', which was introduced in the first decade of the 20th century to account for chemical reactions that were indirectly caused by radiation, has been discussed.[129] There is a book on the history of radical chemistry[130] and also a book co-authored by one of the participants about the development of free radical chemistry during the half century from about the end of World War II.[131] The Dutch School of Catalysis,[132] P. Sabatier's (1854–1941) role in the discovery of catalysis,[133] and the establishment and development of the Ipatieff Laboratory at Northwestern University[134] have also been presented.

6.6 Valency and Structural Chemistry

Edward Frankland's suggestion, in 1852, that an element always appears to combine with the same number of atoms of any given kind started systematic investigations

on valence, one of the most fundamental concepts of chemistry. The development of the notion of valency has been systematically discussed in two older books.[135,136] The history of the electron theory of valence has been reconstructed, from the electrostatic models of Thomson and Abegg to G. N. Lewis' proposal and elaboration of the electron pair as the basis of the chemical bond to the development of quantum mechanics.[137] Two further papers examine the 'chemists' electron' for the same period.[138,139] There is also a discussion of the concept of the chemical bond, with a chapter on its prehistory from antiquity to Kekulé, ending with post-World War II developments.[140] Early developments in quantum chemistry [141] and the differences between the various approaches have been analysed.[142,143] There is a special issue of a journal focusing on historical and philosophical issues concerning quantum and/or theoretical chemistry.[144] The controversy between Linus Pauling and the Soviet Chemists concerning resonance theory[145,146] has also been discussed. The work of C. Coulson,[147] R. Fowler,[148] E. Hückel,[149,150] and Pauling and Wheland[151] in quantum chemistry has been systematically examined. Two papers provide an overview of quantum chemistry.[152,153] Finally, there is a paper on some aspects of valence theory, which could be useful in avoiding confusions in teaching.[154]

Several works give an overview the development of the concept of affinity.[155–158] There is an autobiographical narrative by Robert Mulliken (1896–1986), who contributed so decisively to the development of molecular orbital theory.[159] There are also studies on a number of more specific bonding-mechanisms[160–162] and on the reaction of the chemical community to hydrogen bonding.[163]

There is a systematic study of the origins of structural theory in organic chemistry, where the emphasis is on the period from about 1830 to 1861.[164] The same author has examined Kekulé's attempt to account for valency in terms of the internal structure of polyvalent atoms.[165] Robinson's work on strychnine has also been studied.[166] Examples from structural chemistry have been used for illuminating a number of epistemological issues.[167,168]

Furthermore, we have studies on M. Berthelot's (1897–1907) contributions to the development of the notion of isomerism;[169] the errors of Alfred Werner in his account of spontaneous resolution, mainly due to his neglect of the work of others;[170] Adolphe Wurtz's insistence on atomism and its cultural milieu;[171,172] and the early applications of infra-red spectroscopy to chemistry.[173]

6.7 Stereochemistry

J. B. Biot's (1774–1862) discovery that a number of organic compounds and their solutions could rotate the plane of polarized light suggested that optical activity might not be due only to crystal form, but that it might be an inherent property of compounds not, necessarily, related to their crystalline form. The extensive crystallographic studies of Louis Pasteur (1822–1895) pointed to the asymmetrical character of numerous compounds. However, it was J. H. van't Hoff and J. Le Bel (1847–1930) who proposed independently in 1874 the asymmetrical structure of the carbon atom. They observed that when a carbon atom is attached to four different atoms or atomic groups the four substituents can be arranged in two different ways so that the resulting molecules will be mirror images of one another. Van't Hoff

talked of tetrahedral arrangements, while Le Bel argued about systems that permitted two arrangements of different substituents around an asymmetrical carbon atom.

The early history of stereochemistry has been explored.[174] An account of stereochemistry from the mid-19th century to 1960 surveys the proposals of van't Hoff and Le Bel as well as those of later workers in the field, such as Odd Hassel and D. H. R. Barton (1918–1998), and J. W. Cornforth and V. Prelog (1906–1998).[175] G. J. W. Bremer's experiments, which gave the initial experimental support to van't Hoff's stereochemical views, have been re-assessed through an analysis of the correspondence between the two chemists.[176] There is a paper arguing that, in effect, van't Hoff and Le Bel produced two different theories[177] and another one suggesting that John Dalton (1766–1844) was the first stereochemist.[178] There is a brief analysis of the independent contributions of Alfred Werner and William Jackson Pope (1870–1939) to stereochemistry.[179]

Discussions concerning the origins of mirror-image isomers, first discovered in the mid-19th century, are the subject of a study examining the causes of this phenomenon, variously attributed to light, magnetism, heat, vitalism, pure chance and Darwinian evolution.[180] The work of the American chemist Arthur Michael (1853–1945), whose discovery that the multiple bonds of halogens exhibit an axial symmetry led him to a critique of stereochemistry, has also been studied. His views, which were difficult to visualize and did not account for axial symmetry as a process, were not preferred over Johannes Wislicenus' more visualizable approach.[181,182] Finally, Charlotte Thomas' early text-book on stereochemistry has been commented upon.[183] See also Chapters, 4 and 5.

6.8 Solutions

There is a brief description of the development of the physical chemistry of non-aqueous solutions from 1920 to 1985[184] and an analysis of van't Hoff's theory of diluted solutions.[185,186] The role of the chemist Paul Walden (1863–1957) in the development of theories about chemical solutions has been discussed.[187] Amedeo Avogadro's (1776–1856) research on the nature of metal salts has been examined.[188] There is, also, a discussion of Thomas Graham's (1805-1869) work on the diffusion of gases and liquids.[189]

6.9 Colloid Chemistry

In 1861, Graham introduced the term "colloid" to distinguish those solutions he called 'crystalloids,' which could pass through membranes, and those which could not go through these membranes, his 'colloids.'[190] Systematic studies of colloids, especially of the conditions of coagulation of colloids by salts, were undertaken before the end of the 19th century, when it was also noticed that colloids migrate in an electric field – a technique used for their purification. Developments in microscopy made possible the counting of colloid particles and the estimation of their size.

Some aspects of the history of colloid chemistry have been explored. There is a study of Isidor Traube (1860–1943), highlighting his work on molecular volumes of pure liquids and particularly on the apparent molar volumes of dissolved molecules

in various solvents. It discusses some of the controversies in which he was involved.[191] There is also a systematic treatment of the developments of colloid chemistry in the USA and Canada from the beginning of the 20th century,[192] and of the specific role of laboratory equipment in colloid chemistry laboratories.[193,194] Finally, there is a brief discussion of Ostwald's work on colloids.[195]

6.10 Coordination Chemistry

Coordination chemistry deals with compounds containing a central atom or ion to which are bonded molecules or ions, whose number usually exceeds the value corresponding to the valence of the central atom. The molecules are called ligands and the bonding consists usually of a covalent bond, formed by the donation of a pair of electrons from the ligand (donor) to the central atom or ion (acceptor). The structure and properties of coordination compounds were explained by the coordination theory of Alfred Werner in 1893.[196] An American Chemical Society Symposium marked the centenary of the founding of coordination chemistry.[197] A book and a paper discuss the early developments of coordination chemistry and especially Werner's contributions.[198,199] There is also a historical narrative of the evolution of coordination chemistry to 1930.[200] Two further studies discuss various aspects of coordination chemistry in the USA[201] and Australia.[202] Finally, the origin and the dissemination of the term 'ligand' in chemistry have been discussed.[203–205]

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