



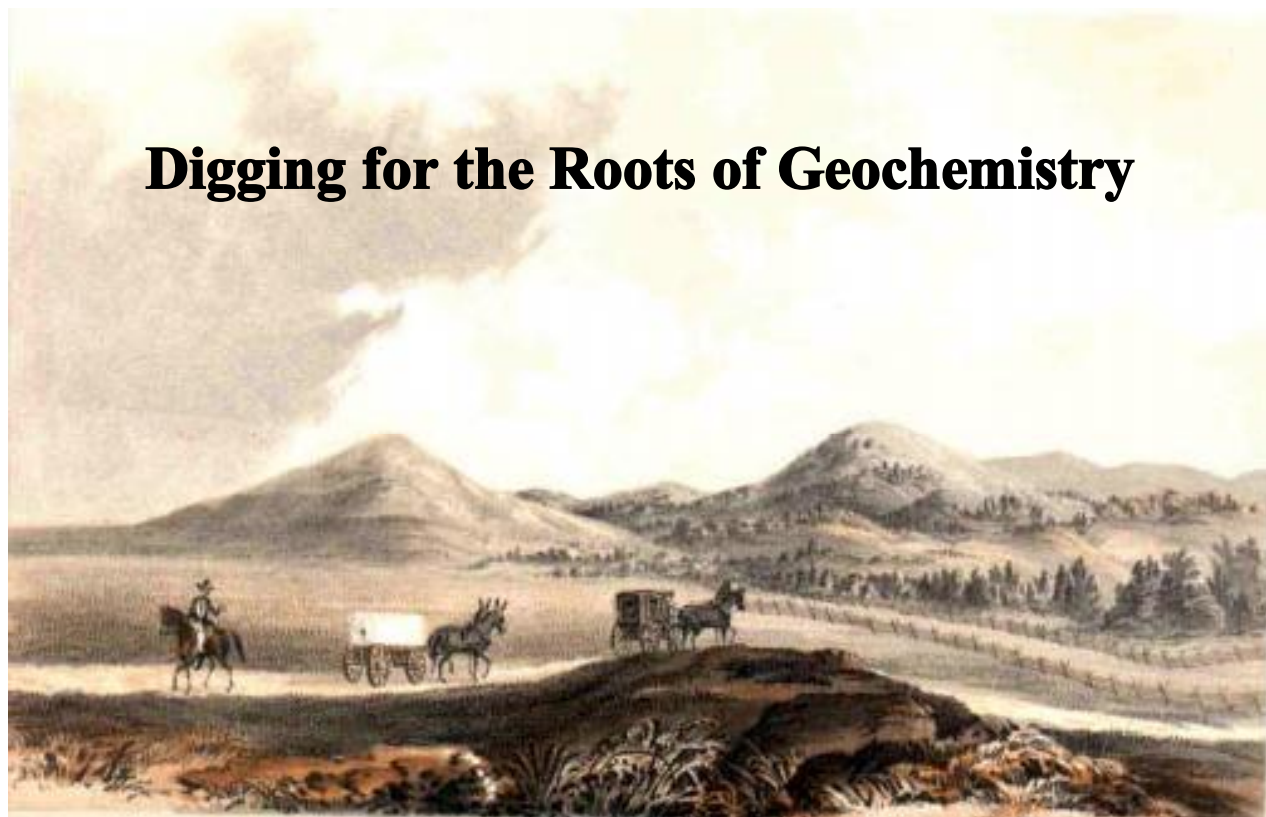
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Digging for the Roots of Geochemistry

In this issue:

Van der Weijden Retires.....	7
Digging for the Roots of Geochemistry.....	10
In Memoriam - Werner Stumm.....	16
When Did The Earth's Atmosphere Become Oxic? A Reply.....	20
Meetings Calendar	23
GS Special Publications.....	26
GS Membership Application	27

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(see page 5 for more information)

Digging for the roots of geochemistry

One could say that the history of geochemistry goes back to the times of Thales, Aristotle and Empedocles. One could even go further and point to our ancestors, who knew how to make iron and bronze tools. Geochemistry as we know it is however a fairly recent development. Geology didn't become a science of its own until the 19th century and the inclusion of chemistry in geology seems to have sprung from agricultural and economic concerns. Aqueous geochemistry of freshwater appears to have been spawned mainly by environmental and health concerns. Both were boosted to a much higher level by the input from physical chemistry, the discipline exploring the borderland between physics and chemistry, created about a century ago. Physical chemistry also influenced chemical oceanography, the foundations of which were laid by Gay-Lussac, Boyle and Lavoisier, but chemical oceanography has always been fairly close to physical chemistry.

Aqueous and environmental geochemistry

Environmental concerns may appear typical of our era, but a look at the history of the city of Amsterdam reveals that they already cropped up in the 15th century. In 1394, the water from the Amstel river and the canals of Amsterdam was still clean enough to be used in beer brewing.



The quality of freshwater is essential for the quality of beer.

Eighty years later, pollution had forced the breweries to obtain their water from elsewhere. The city council now also routinely tested the environmental quality of the ice that was used in a form of winter entertainment called 'ice biting'. In 1530, professions that were known to pollute heavily, such as the tanning of leather and dyeing of cloth, became subject to regulations. More regulations, prohibitions, as well as a system of fines attempted to get a grip on pollution during the following centuries, without a great deal of success. In 1794, the citizens of Amsterdam suffered a bout of lead poisoning. This was caused by the use of peat

as fuel, in turn leading to a form of acid rain which dissolved the lead from paints and gutters. The lead subsequently got into the basins near the churches from where drinking water was distributed to the population. The Clean Freshwater Society had been founded in 1786, but didn't start publishing chemical analyses of drinking water until 1825. One can assume that they also checked the levels of lead. Of course, oxygen still had a molecular weight of 8 in those days.

Geology and geochemistry

By this time, the science of geology was well on its way, as a result of the work of James Hutton (1726-1797), the so-called 'father of geology'. Until the times of Hutton, the earth sciences did not have a domain of their own, but were part of other studies. In countries with a history of mining, such as Germany and France, geochemistry had stronger roots than in countries without ore mines. The French École des Mines already built an analytical chemistry laboratory in as early as 1838.

North America had only just started to develop, so Europe was still ahead in most aspects. The first American professor in science was Benjamin Silliman. Silliman, a 22-year old law student became professor of chemistry at Yale in 1802. Although American geology picked up surprisingly rapidly during first half of the 19th century, early American geology was dedicated to stratigraphy and paleontology. Thomas Jefferson's strong interest in vertebrate paleontology played a role in both the rapid development of geology as in its bias toward the non-chemical aspects.

Nevertheless, some remarkable geochemical feats were accomplished as well. In 1827, Lardner Vanuxem drew attention to the chemical interaction between the atmosphere and the earth's crust and in 1844, Henry D. Rogers estimated how much carbon was taken from the air and stored in rocks. If only they knew how important these topics are today! James Dana introduced the concept of metamorphism in 1843. Although the discussion of these ideas may have been limited to academic circles, chemistry certainly was a normal part of 'ordinary' geological studies. This becomes evident upon browsing through some of the geological reports of that period.

'As a measure of economy, as far as this state is concerned, I was pleased to learn that the chemical work could all be done in your laboratory at New Harmony, and would be pleased to learn what annual expense this state will incur under such an arrangement, and for all instruments which I presume you have and can use in the prosecution of the work, including office-rent and fuel, whilst doing the office-work at New Harmony.

As you know best the kind of wagons and camp equipage you will require to suit you, I presume it would be better for you to procure and ship them to Jacksonport, Arkansas, than to obtain them in this state. The horses and mules which you would require, could, perhaps be obtained in Arkansas, as well as common laborers.

We shall have to be confined to the amount of appropriation by the law, and that is so small for such an important work, we will have to use economy to accomplish much good, and I shall depend greatly on your experience and good management in the whole matter.'

Ignore the part about the horses and the mules and these words sound almost awkwardly familiar. They are part of a letter that appoints David Dale Owen, M.D. as State Geologist for Arkansas. The letter was published in Little Rock in 1858, as part of the "First report of a geological reconnaissance of the northern counties of Arkansas, made during the years 1857 and 1858..."

William Elderhorst, M.D. was the State Geologist's Chemical Assistant. He wrote the chapter titled 'Chemical Report of the Ores, Rocks, and Mineral Waters of Arkansas'. The samples of well and river waters were analyzed in the field 'with comparatively limited means', but all other analyses were performed at the laboratory. Elderhorst describes the ores, gives the analyses and includes a methods section for each type of analysis. The State Geologist also had a Geological Assistant. This was a certain Edward T. Cox, whose part of the report also includes chemical compositions, albeit mainly of waters. Cox often added remarks such as 'It is, therefore, a weak saline sulphuretted water, and its medicinal properties will be that of a mild laxative, it will also be found beneficial in all cutaneous diseases.'

Apart from the fact that chemistry was clearly a well-established aspect of field geology, two other issues jump out: the State Geologist and his Chemical Assistant were both medical doctors and the water analyses are seen in terms of health implications.



Sugar Loaf Mountain, Marion County, Arkansas, in the middle of the 19th century. This is how the State Geologist traveled in those days, if he got lucky. He describes some of the roads in Marion County as 'very rough and disagreeable to travel over'. The prevailing rocks of this county are 'varieties of magnesian limestone, with some intercalated sandstones'.

Another excerpt, from the "Report on the agriculture and geology of Mississippi embracing a sketch of the social and natural history of the State" further illustrates the practical use of chemistry in geology and shows aspects of the ongoing discussions at that time. The report was written by B.L.C. Wailes, 'geologist of Mississippi; member of the American Association for the Promotion of Science; corresponding member of the National Institute, and of the Boston Society of Natural History, etc. etc.' and published in 1854 by Lippincott, Grambo and Co. for E. Barksdale, State Printer.

'It is well known that some soils are, by nature, unsuited to the production of particular plants, even where climate and

other conditions would favor their cultivation, and that from other soils, originally prolific, the productive elements are continually abstracted in the course of tillage until, in the end, they become exhausted and sterile. It is the province of Agricultural Chemistry, therefore, to determine the elements of plants derived from the earth in which they grow, and the presence or deficiency of those elements in the soils in which they are cultivated, in such manner as to make this knowledge available to the husbandman, and to instruct him also in the chemical composition of the manures, animal or mineral, proper to be applied to supply the exhaustion, or to fit the otherwise unfruitful soil for his purpose. As to the practical value of analysis of soils as usually conducted, or the ability, in the present state of chemical knowledge, of determining those minute constituents in a soil - such as alkali or potash and phosphoric acid - generally regarded as the greatest cause of fertility, eminent chemists are at issue, and, in our own country, many distinguished for high scientific attainments are found to agree with Boussingault, that we are much less interested in the chemical composition of the soil than in its mechanical mixture.

But, whatever may be the differences of opinion as to the value of analysis of soils, and whether the benefits would justify the expense attending the minute and multiplied chemical examinations required to impart a useful knowledge of their properties, there can be none as to those of the marls or mineral fertilizers employed, as no one would be willing to apply an ingredient to his land, which a simple test might prove to be not only unsuitable, but absolutely pernicious. In view of the general use into which they must come eventually, when their existence and value shall be better known, adequate analyses of all the varieties of marls which abound in the State are highly desirable.'

These words are the beginning of the chapter 'Analysis' and are followed by analyses of marls, mineral waters, cotton, cottonseed and other materials.

Analysis of the Water of Ocean Springs, near Bahuci, in Jackson County, Miss. By J. LAWRENCE SMITH, M. D.

The water colorless, even when kept in bottles for a length of time, provided the bottles be well corked; as soon as opened, the water begins to blacken, from a deposit of sulphur of iron; the odor of the water is that of sulphuretted hydrogen, which the water contains in considerable quantity; the taste, that known to belong to this class of waters.

Specific gravity, 1.00682.

Gaseous contents in one gallon in grains:—	
Carbonic acid	4.632
Sulphuretted hydrogen	481
Solid contents of one gallon in grains:—	
Chloride of sodium	47.770
Chloride of calcium	3.882
Chloride of magnesia	4.989
Protoside of iron	4.712
Iodine, a strong trace.	
Chloride of potassium, a trace.	
Organic matter, a trace.	
Alumina, a trace.	

The iron doubtless in combination with both the sulphuretted and carbonic acid gases, the excess of the carbonic acid holding both these combinations in solution.

'Analysis of the Water of Ocean Springs, near Bahuci, in Jackson County, Miss. .'

Continued on page 12

Continued from page 11

Impact of physical chemistry

Chemistry had gathered momentum in the 18th century and developed more or less independently of physics during the 19th century. Although the physical aspects of chemistry had already drawn attention, it was the 'ionists', Svante Arrhenius, Jacobus Henricus van 't Hoff and particularly Wilhelm Ostwald, who really catapulted physical chemistry into orbit at the end of the 19th century (<http://www.nidlink.com/~jfromm/history/physical.htm>). They wanted chemistry to study the conditions under which compounds formed. They wanted to look at reactions, kinetics, equilibrium and chemical affinities and not focus only on the compounds themselves. In the United States, physical chemistry became quite 'hot' in the beginning of the 20th century, where it strongly impacted many disciplines, notably igneous and metamorphic petrology and geochemistry.

It seems only natural that physical chemistry should become so crucial to petrology. In 1851, Robert Bunsen had already noted that magmas were in fact solutions and that they were probably ruled by the same principles that applied to solutions. In 1857, the French petrographer J. Durocher realized that magmas and alloys also shared similarities. These ideas remained undeveloped until the 1890s, when Arrhenius (<http://nobel.sdsc.edu/laureates/chemistry--1903-1-bio.html>) and Van 't Hoff (<http://nobel.sdsc.edu/laureates/chemistry-1901-1-bio.html>) started applying their ionist ideas to rocks. Arrhenius also designed a theory of the importance of the CO₂-content of the atmosphere for the climate and Van't Hoff tackled marine chemistry issues.

However, physical chemistry wasn't exactly received with open arms in Europe and European geologists were also reluctant to embrace the new ideas. Although the competition from organic chemistry in Germany was fierce, physical chemistry managed to flourish at Germany's fast-growing Technische Hochschulen. For instance at Leipzig, where Ostwald (<http://nobel.sdsc.edu/laureates/chemistry-1909-1-bio.html>) continued to promote physical chemistry as a science of processes as opposed to structures. In Britain, inorganic chemists strongly opposed the ionists' views and physical chemistry didn't even get a foothold in Britain until after World War I. This was largely owing to William Ramsay, a chemist at University College London, and to the University of Liverpool, not to Oxford and Cambridge. Ramsay opened his lab to many of the ionists' followers, such as Alexander Findlay, whose name would later become strongly associated with the application of the phase rule in Europe. In France, physical chemistry found homes at Nancy, Toulouse and Bordeaux but neither at the Sorbonne nor at any of the other Paris institutions. Arrhenius, Van 't Hoff and their students worked at the new Nobel Institute and at the relatively young University of Amsterdam. Americans did not have to fight their way into existing labs; they were still building them. It is easy to see the advantages and disadvantages of that situation. The advantages won. The American situation proved highly beneficial to both the progress of physical chemistry and its incorporation into petrology and geochemistry.



The first Stanford Geological Survey, Summer, 1893, worked along rivers near the present location of Shasta Lake in northern California. Pictured are (left to right) Henry Cory, E.B. Kimball, Frank L. Cole, Jack Sheehan, Professor James Perrin Smith and William Stuart.

Physical chemistry wasn't the only science to expand rapidly at the turn of the century. So did geology. Many American chemists, who went to Germany to study chemistry, acquired a working knowledge of rocks. American geology students on the other hand could hardly avoid picking up some of the new chemical know-how in Germany. Back in America, these chemists and geologists discovered that they had more in common besides having been trained at the same German institutions. They compared notes and were struck by the similarities between geology and physical chemistry. Important geological questions were postulated in the 1880s and 1890s. How could a single body of magma lead to various rock types? Did it have to do with assimilation of country rock, was it mainly a result of fractional crystallization or should the origin be sought in the liquid itself? Could the order in which minerals crystallized in magmas be influenced by pressure, magma composition and the rate of cooling, as was the case for aqueous solutions? Did pressure have a similar effect on the fusibility of minerals as it had on the solubility of salts in water? And what about supersaturation and the effect of the presence of other minerals?

The phase rule and the Phase Ruler

The phase rule, that familiar petrologists' tool, was formulated in 1876 by Josiah Willard Gibbs (<http://www-groups.dcs.st-and.ac.uk/history/Mathematicians/Gibbs.html>) and then forgotten. Gibbs was a solution chemist and physicist at Yale, where he laid the foundations of chemical thermodynamics. Later, Hendrik W. Bakhuis Roozeboom and Van

der Waals, a chemist and a physicist respectively at the University of Leiden in The Netherlands (<http://ci.mond.org/9707/970715.html>), rediscovered Gibbs's work on the phase rule. They induced a lively interest for it, particularly among scientists in the Low Countries but also elsewhere. Wilder Dwight Bancroft reintroduced the phase rule in America. Bancroft went to Leipzig to study with Ostwald, who happened to be preparing a translation of Gibbs's papers. After Bancroft received his Ph.D. in 1892, he first went to Berlin to work with Helmholtz. Then he traveled to Amsterdam where he continued his potentiometric studies in Van 't Hoff's lab and where he became further acquainted with the phase rule. Bancroft initially wasn't impressed. He thought that "people made a good deal of unnecessary fuss over the phase rule", which seemed to him "an interesting mathematical relation but nothing more". Back at Harvard, Bancroft studied solubility relations in ternary systems and had his work severely criticized several times. It was then that he started looking at these systems with the phase rule in mind. He later joined the chemistry department at Cornell and went on to become such an advocate and authority on the phase rule that he was nicknamed the 'Phase Ruler'.

Incidentally, Bancroft may have been quite a character. It is said that he ingested 1,000 mg of thiocyanate a day for a period of 23 years in the process of studying the cumulative properties of this chemical. He reported only positive effects from the experiment.

The phase rule was so attractive, because it finally enabled physical chemists to deal with problems in geology, medicine and industry, that usually could not be handled by conventional mathematics. In fact, Bancroft saw little promise and practical use in mathematical chemistry and he overestimated the realm within which the phase rule could be applied.

Bancroft's work however was considered too 'practical' for academia and too abstract for industry. Most physical chemists were not concerned with the phase rule and Bancroft moved on to other topics, although he never lost interest in the phase rule. Wilder Bancroft is also known as a pioneer in the field of colloid chemistry which he summed up as follows: "essential to anyone who really wishes to understand oils, greases, soaps, glue, starch, adhesives, paints, varnishes, lacquers, cream, butter; cheese, cooking, washing, dyeing, colloid chemistry is the chemistry of life."

Tying the knot between the two disciplines

Joseph Paxson Iddings and Charles R. Van Hise, two petrographers, were instrumental in securing the link between physical chemistry and geology in America. Iddings was actually the first American geologist who drew attention to the implications of the ionists' work for geology. He incorporated the works of Arrhenius and Van 't Hoff into his own papers. He also conducted experiments on the electrical resistance of molten rocks, in an attempt to substantiate the analogy with aqueous salt solutions. A magma turned out to be composed not of minerals but of ions, a long-awaited follow-up of the early observation of Bunsen.

Iddings tried to explain magmatic differentiation with a combination of Van 't Hoff's theory of osmotic pressure and C.

Soret's empirical findings that, given a temperature gradient, solute molecules tend to concentrate in the cooler portion of a solution. Although this wasn't correct, Iddings's approach nudged more geologists toward physical chemistry.



Mr and Mrs Charles Van Hise. Original owned by the University of Wisconsin Archives.

Charles Van Hise was driven beyond his duties as a geologist within the USGS, to theories about ore deposition and metamorphism. Blessed with an unusual discipline and tenacity, he pushed himself forward to make up for the gaps in his education and also arrived at the conclusion that physical chemistry was vital to petrology. Van Hise focused on the study of metamorphic rocks, but geology wasn't the only topic that caught his attention. Charles Van Hise's speech on "The regulation of competition versus the regulation of monopoly", which he gave at the Economic Club of New York in 1912, can be perused at <http://library.wisc.edu/etext/WIRReader/WER0134.html>.

Both Iddings and Van Hise were very eager to start laboratory work in which physical chemistry could join forces with geology. Such a project, where high-temperature and high-pressure work could be done and for which also a fair amount of platinum would be needed, for its crucibles, was going to be expensive and also somewhat esoteric. It was not easy to get such a lab off the ground. Van Hise kept pushing the idea within the USGS and at the turn of the century, the USGS launched a laboratory in which geology finally would team up with chemistry and physics. This was the big turning point. The lights went on green for the study of the physicochemical aspects of geology.

Two other important names in the history of geochemistry are those of Arthur L. Day and E.T. Allen, a physicist and a chemist. Day, Allen and Iddings did a lot of work on the thermal properties of the albite-anorthite solid solution series. They tried, in vain, to explain their experimental results in terms of Van 't Hoff's theories. They then turned to Bakhuis Roozeboom's work and discovered that the plagioclases already fit into one of the

Continued on page 14

Continued from page 13

Dutchman's system categories.

In 1905, Day and Allen published the fish-shaped equilibrium diagram, which has become such a fixture on the menu of today's petrology courses. This concept, with compositions shifting along the solidus and liquidus through the (P)T domain, helped explain many phenomena, such as the hitherto puzzling compositional zoning of feldspar crystals.

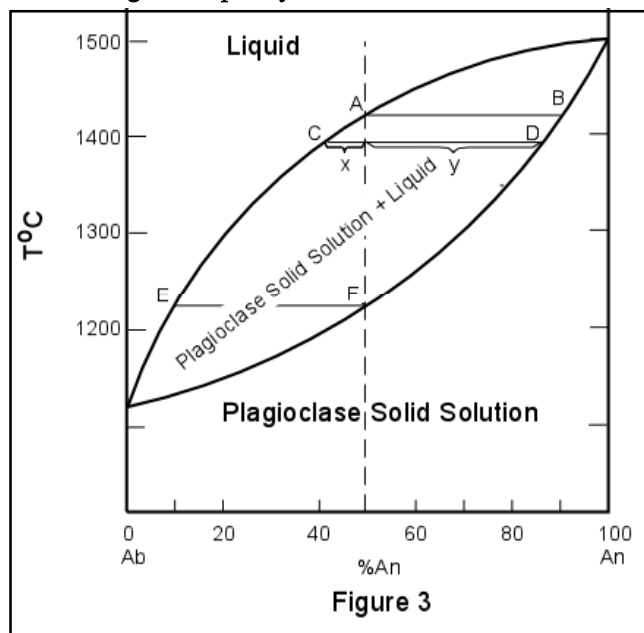


Figure 3

The plagioclase diagram.

For an extensive explanation of the plagioclase diagram, other binary phase diagrams and the phase rule, see <http://www.tulane.edu/~sanelson/geol212/2compphasdiag.html>. For definitions and interpretations of the phase rule, see http://jwgibbs.cchem.berkeley.edu/phase_rule.html, <http://chemengineer.miningco.com/library/weekly/aa022398.htm> and <http://clayir.gly.uga.edu/schroeder/geol3010/phaserule.html>.

The Geophysical Laboratory

The research started by Allen and Day at the USGS blossomed. In 1907, the Carnegie Institution in Washington opened its Geophysical Laboratory, which rapidly became a leader in petrological and mineralogical research. Van Hise and Iddings also played a crucial role in the conception of the Geophysical Laboratory and more particularly, in pushing the physical chemistry of petrogenesis as its main research topic.

Day and Allen became its first director and chief chemist. The Geophysical Laboratory employed many chemists who had studied with Gilbert Newton Lewis and Arthur Amos Noyes at MIT, renowned physical chemists who both had trained with Ostwald. Lewis did a lot of frontier work on chemical bonds but also on chemical thermodynamics, so essential to for instance geothermobarometry. Noyes undertook some of the earliest work in X-ray crystallography. Norman Levi Bowen was of those MIT-trained scientists. The Day and Allen group carried on with the

hunt for the data, necessary to construct the phase diagrams for a host of binary and ternary mineral systems. These diagrams fit in very well with Bowen's theory of the evolution of igneous rocks. Put forward in 1922, it became another of those obligatory petrology course ingredients. In the meantime, Wilder Bancroft was very pleased with these developments, of course, and he dubbed the Geophysical Laboratory the "Phase Rule Laboratory" but it was much more than just that. It was truly a lab of applied physical chemistry, in the broadest sense.

Geochemistry on its way to the 21st century

Both geologists and physical chemists benefited enormously from the co-operation. There were several industrial spin-offs, for instance for the production of quartz glass and ceramics. During World War I, companies like Bausch and Lomb opened the doors for scientists from the Geophysical Laboratory. This trend continued after the war, when the industry had realized what physical chemistry had to offer. A phase diagram can also describe Portland cement, for instance, and phase diagrams became widely applied in many areas. In the early 1920s, physical chemists were 'hot' and enjoyed a multitude of widely varying opportunities, such as in metallurgy and pharmacology.

Hard-rock petrology and geochemistry had received an enormous boost and would never be the same. Nuclear chemistry and analytical developments further advanced the field. Tools such as EPMA and XRF were introduced and a progression of mass spectrometers and computers continues to march into the labs. More hyphenated techniques, the cluster concept and computational chemistry will be new fuels to keep the ovens of geochemical research going.



Stanford enters field of isotope geology. Stanford Geology Department's mass spectrometry lab circa 1961. Graduate student William Taft (seated) operates the Consolidated 21-401 Mass Spectrometer under supervision of Professor John W. Harbaugh. Vacuum system for preparation of gas samples for mass spectrometric analysis is in the background.

Coincidentally, as we move into another century, yet another discipline is being added to petrology and geochemistry: biology. Ten, twenty years ago, the role of microorganisms in ore formation wasn't even mentioned in ore geology courses. Today, most geologists are well aware of the role of bacteria in the redox

reactions that can lead to the precipitation of ore minerals. Chemical oceanographers are talking with marine biologists and actually enjoying it. What is chemically unlikely, such as trivalent manganese remaining in solution or celestite not dissolving instantaneously in seawater, can be mediated by biology. Fungi have been observed to head straight for apatite inclusions in feldspars in order to procure their phosphate. They secrete organic acids and dissolve the apatite and who knows which implications this has had for some whole-rock analyses in the past. Bacteria are found in hard-rock cores obtained from great depths. Geochemists now also find themselves studying apatite aggregates that enclose the fossilized remnants of early life forms and ponder Archean nitrogen geochemistry. As geochemistry moves into the next millennium, it also moves toward more and more multidisciplinary. Biophysicogeochemistry?

Note: This article does not pretend to be an extensive and complete overview of the history of geochemistry. For a timetable of petrology for the years 1800-1899, see <http://www.gl.ciw.edu/TofP-1899.html>. Except the geological reports quoted in the article, several other publications were used for its preparation, notably:

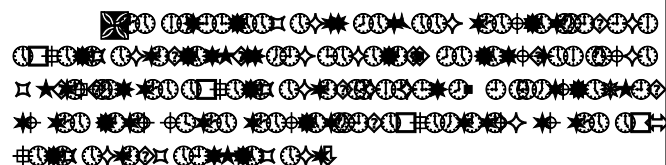
Bensaude-Vincent B. and Stengers I. (1996) *A history of chemistry*. Translated by Deborah van Dam. Harvard University Press, Cambridge, Massachusetts. 305 pp.

Hogener, K. (1997) *Costelijck Stadswater. Geschiedenis van de Amsterdamse waterhuishouding in vogelvlucht*. Stadsuitgeverij/Dienst Riolering en Waterhuishouding Amsterdam. Amsterdam, The Netherlands. 112 pp.

Nye, M.J. (1996) *Before big science. The pursuit of modern chemistry and physics 1800-1940*. Twayne Publishers/New York and Prentice Hall International, London, England. 282 pp.

Servos, J.W. (1990) *Physical chemistry from Ostwald to Pauling. The making of a science in America*. Princeton University Press, Princeton, New Jersey. 402 pp.

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Wilder Bancroft

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Geochemistry and the Origin of Life. P. Maurice, Dept. of Geology, Kent State University, Kent, OH 44242, email: pmaurice@kent.edu; D. Deamer, Department of Chemistry, University of California, Santa Cruz, Santa Cruz, CA 95064, email: deamer@hydrogen.ucsc.edu.

Characterization of High Maturity Fluids. J. M. Moldowan, Dept of Geological and Environmental Sciences, Stanford University, Stanford, CA 94305-2115, email: moldowan@pangea.stanford.edu; J. Dahl, Dept of Geological and Environmental Sciences, Stanford University, Stanford, CA 94305-2115, email: dahl@pangea.stanford.edu.

Geochemistry and Deepwater Petroleum Exploration. A. N. Bishop, Texaco UTD, 3901 Briarpark, Houston, TX 77042, email: bishoan@texaco.com; Paul Taylor, Unocal, 14141 Southwest Freeway, SugarLand, TX 77478, email: paul.taylor@unocal.com.

The Integration of Organic Geochemistry and PVT Studies in Petroleum Exploration and Production. G. Macleod, Shell E&P tech Co., 3737 Bellaire Blvd., Houston, TX, 77025, email: gmac@shellus.com; P. Meulbroek, Woods Hole Oceanographic Institute, 360 Woods Hole Rd. MS#4, Woods Hole, MA 02543, email: pmeulbroek@whoi.edu.

New Insights into the Petroleum Geochemistry of NSO Compounds. H. Wilkes, Institute for Petroleum and Organic Geochemistry (ICG-4), Research Center Jülich, D52425 Jülich, Germany, email: h.wilkes@fz-juelich.de; P. Taylor, Unocal, 14141 Southwest Freeway, SugarLand, TX 77478, email: paul.taylor@unocal.com.

Organic Solids in Petroleum Production. B. A. Stankiewicz, Shell E&P TechCo., 3737 Bellaire Blvd., Houston, TX 77025, email: artur@shellus.com; E. Tegelaar, Baseline Resolution, Inc., Suite 810, 2000 Avenue G, Plano, TX 75074, email: ETegelaar@brilabs.com.

Geochemical Analysis in the New Millennium. L. W. Elrod, Geotechnology Research Inst., Houston Advanced Research Ctr., 4800 Research Forest Dr., The Woodlands, TX 77381, email: lwe@gtri.harc.edu; M. A. Engel, University of Oklahoma, School of Geology & Geophysics, 100 East Boyd St., Norman, OK 73019, email: ab1635@ou.edu.