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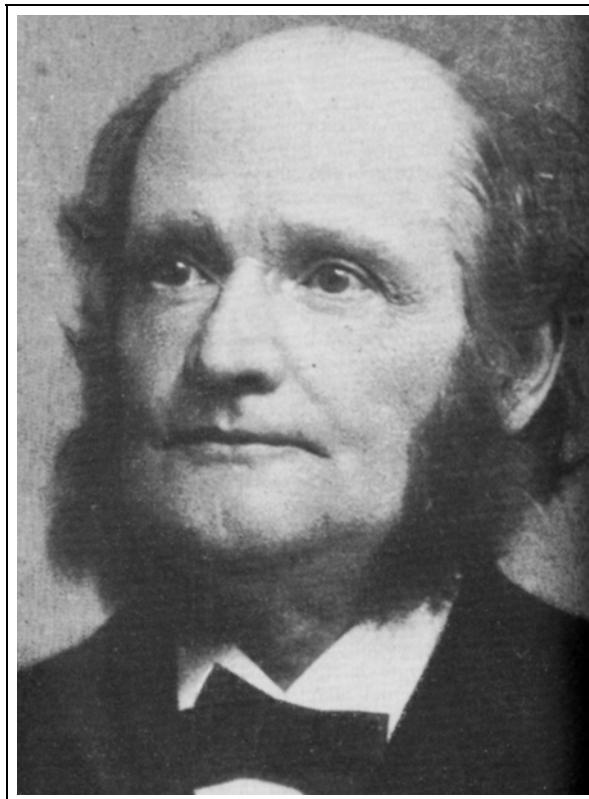
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Jakob Maarten van Bemmelen

(November 3, 1830 Amelo – March 13, 1911 Leiden)

and the history of the theory of adsorption from solution

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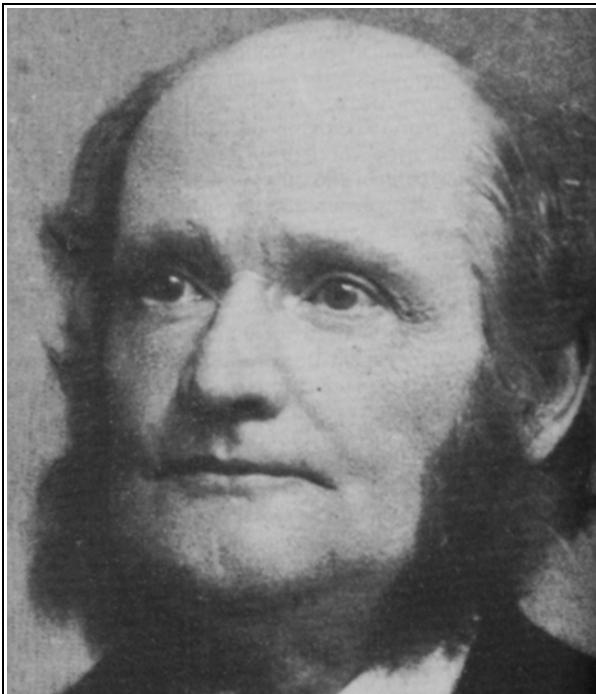


**February 2005,
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Jakob Maarten van Bemmelen

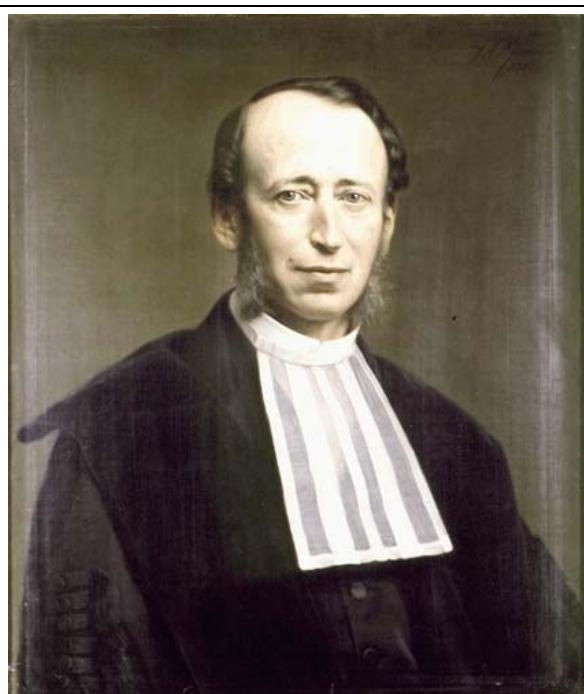
(November 3, 1830 Amelo – March 13, 1911 Leiden)

and the history of the theory of adsorption from solution



Jakob Maarten van Bemmelen

Van Bemmelen was descended from a family, many of whose members were distinguished in education, law, and statesmanship. Jakob Maarten van Bemmelen was born on November 3, 1830 in Amelo where his father Jan Frans van Bemmelen was headmaster of the grammar school. His mother was Antoinetta Adriana de Kempenaer. His father died in the same year (December 26, 1830) and his family moved to Leiden. On September 13, 1847 he matriculated there as a student. He studied inorganic- and organic chemistry also pharmacy under Professor Antonius Henrik van der Boon Mesch (15.04.1804 Delft - 12.08.1874 Leiden), who had only a primitive laboratory at his disposal, where van Bemmelen made some quantitative inorganic analysis. Van der Boon Mesch studied fermentation, mineralogy, industrial chemistry, construction of voltaic batteries and boiling points of water/alcohol mixtures.



Petrus Jan van Kerckhoff

In 1852, van Bemmelen was appointed as an assistant in the laboratory of Professor Petrus Jan van Kerckhoff (1813 - 1876) in Groningen. Van Kerckhoff was head of the chemistry institute since 1851. The houses of the colleges were just finished, the laboratories conditions in Groningen were excellent for the time. The academic library for students was very well

developed. In Groningen van Bemmelen started his research with chemical reactions of acids on glycerine and glucose (BEMMELEN, 1856, 1859). He also carried out an investigation on the composition of infertile types of soil in Groningen province. The thesis which gained him his doctorate at Leiden University in 1854 (October 16, 1854 by A. H. van Boon Mesch with 'summa cum laude') was written in Groningen and summarised his results of investigations on substances which can be extracted by different solvents from the fibres and wood of *Cibutium cumingii*, a Sumatra herb known as a styptic agent (BEMMELEN, 1854).

Two years later (September 1856), J. M. van Bemmelen was appointed lecturer in chemistry and physics at the "Akademie Minerva", an industrial school at Groningen, while remaining an assistant at the University. However, he was soon appointed to the teaching staff of the agricultural school in Groningen and this brought him into close contact with agriculture. Shortly later he married Miss Maria Boeke (August 31, 1858). They had two sons and three daughters (JORISSEN, 1910, SNELDERS, 1979).

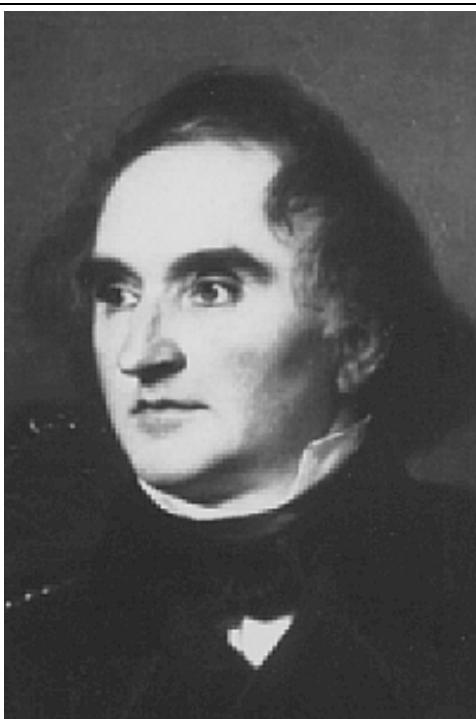


Heike Kamerlingh Onnes

Work done at the request of the committee for Statistics of the Province was published in 1863 as "Contributions to the knowledge of clay soils in Groningen Province" (BEMMELEN, 1863a). In the meantime, he had also acquired a high reputation as a teacher and was appointed headmaster (1864) of the State "Rijkshoogereburgerschool" (a type of reformed secondary school without classical languages) in Groningen, a post he held for five years. One of the pupils at this school was Heike Kamerlingh Onnes (21.09.1853 Groningen - 21.02.1926 Leiden), the Nobel prize winner in Physics 1913.

During this time, van Bemmelen conducted an investigation on exhaustive farming in the Netherlands, following the well-known views of Justus von Liebig (12.05.1803 Darmstadt - 18.04.1873 Munich) (BEMMELEN, 1863b). He also analysed a large number of soil specimens from the Zuider Zee in connexion with the plants to surround the southern part of this basin with dykes with the aim of pumping it dry, a plan which has only been fully implemented in recent years. At this time, he also planned his experiments on the

adsorption potential of agricultural soil, the first results he published 1877 (BEMMELLEN, 1877a, 1878).



Justus von Liebig

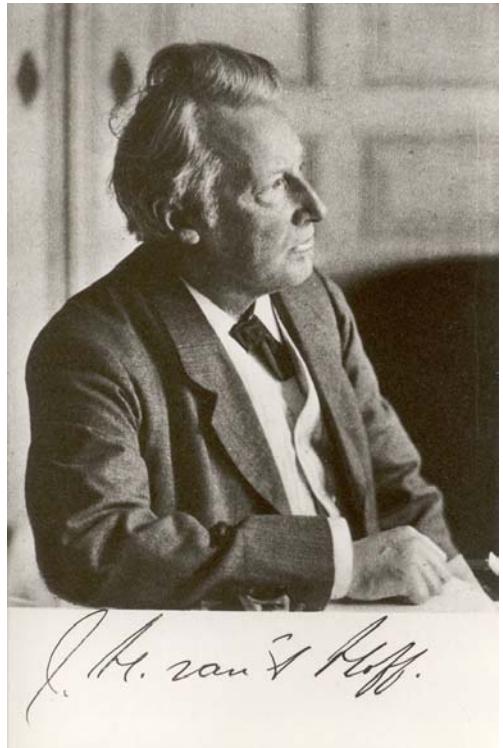


Hendrik Willem Bakhuis Roozeboom

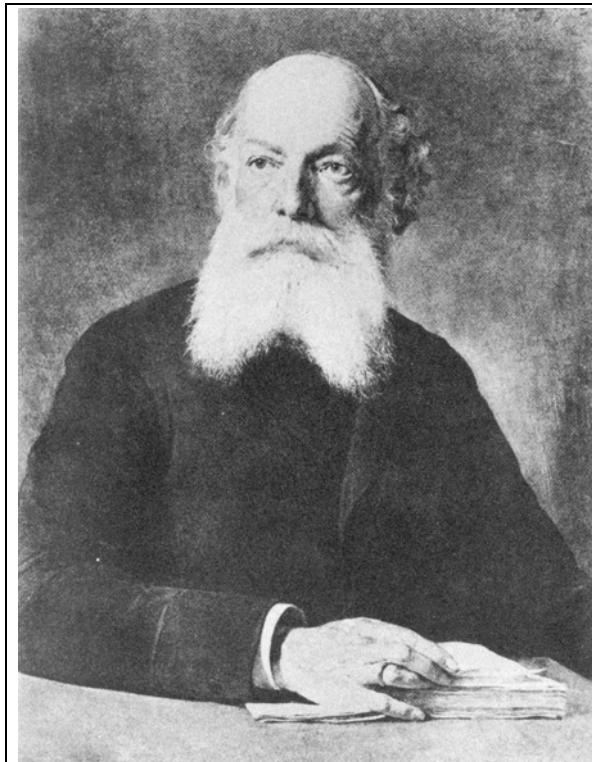
Van Bemmelen was appointed headmaster of the "Hoogereburgerschool" in Arnhem in 1869 where he remained for nearly five years. Here he devoted much interest to the conditions in the public and private schools in the town (BEMMELLEN, 1870, 1871). Here he worked an „change of air“ in schools so that pupils had pure air in schoolrooms (BEMMELLEN, 1874).

However, J. M. van Bemmelen's main work dealt with the analysis of natural waters and the investigation of types of soil. At this time (1873) his brother in law Dr J. D. Boeke, commended 1873 van Bemmelen a pupil of his school (Hoogereburgerschool in Alkmaar) to help van Bemmelen by ground analysis of the Ijerpolder (BEMMELLEN, 1872, 1873a,b). This man was Hendrik Willem Bakhuis Roozeboom (24.10.1854 Alkmaar - 08.02.1907 Amsterdam). He studied (1874) chemistry in Leiden but had not enough money for a longer study. Bakhuis Roozeboom worked in a food factory, but the factory burnt down in 1878. Then he started his chemistry study new in Leiden and graduated in 1884 under J. M. van Bemmelen. In 1896 Bakhuis Roozeboom became follower of Jacobus Henricus van't Hoff (30.08.1852 Rotterdam - 01.03.1911 Berlin; Nobel prize winner in chemistry 1901) at the University of Amsterdam. Bakhuis Roozeboom worked about the Gibbs

phase rule and the chemical composition of steel (BAKHUIS ROOZEBOOM, 1900, 1901-1918; BEMMELLEN, JORISSEN, RINGER, 1907).



Jacobus Henricus van't Hoff



Friedrich August Kekulé

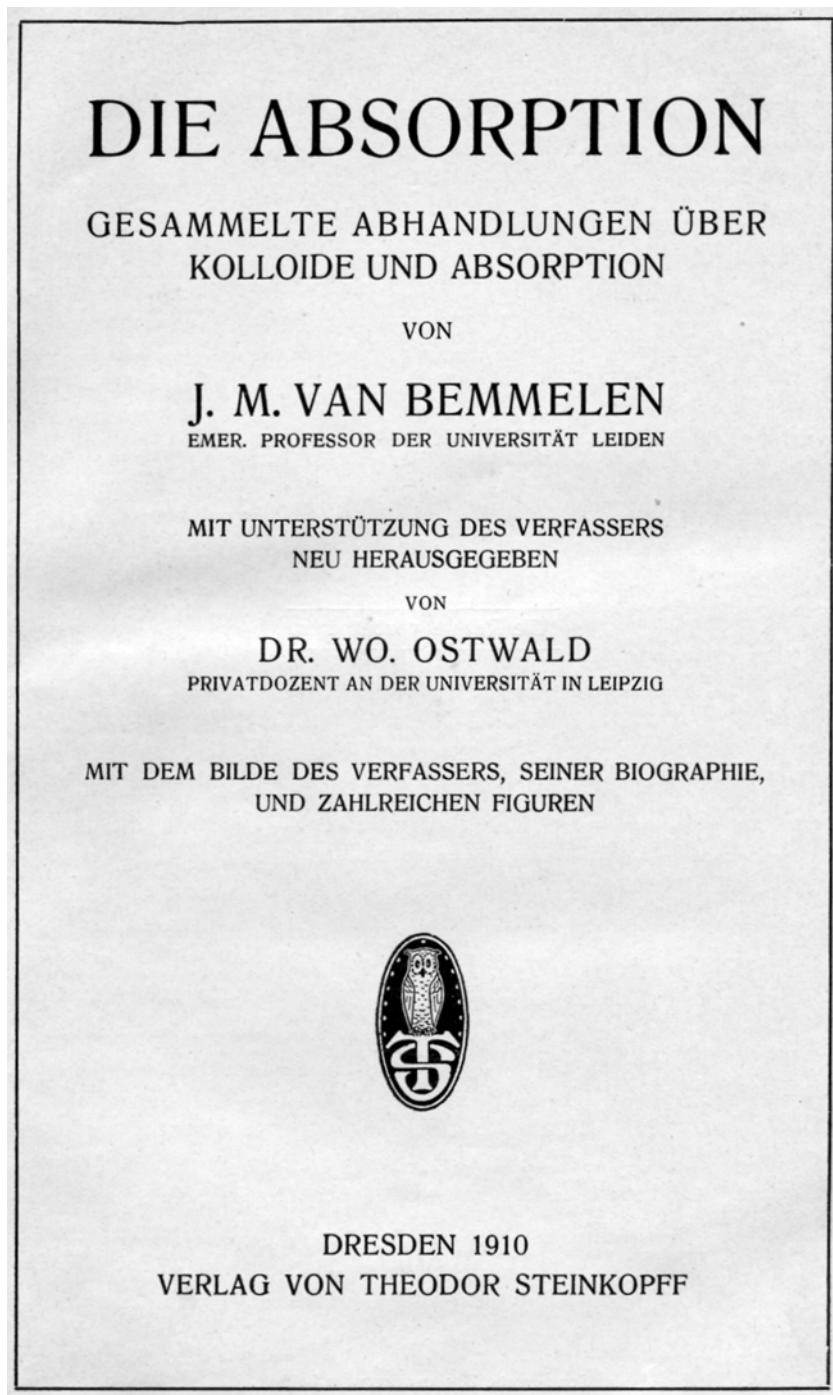


Charles Adolphe Wurtz

Van der Boon Mesch had a disease ridden in December 1873 and he must stop his academic career in Leiden, he died shortly later. In February 1874, van Bemmelen was appointed to the chair of inorganic chemistry in Leiden University, and in his lectures revitalised and brought up to date the teaching in this subject. The first chair of organic chemistry in Leiden occupied with Antoine Paul Nicolas Franchimont (10.05.1844 Leiden - 02.07.1919 Leiden) who had studied at Leiden University and worked under van der Boon Mesch. After his thesis (1871) Franchimont spent a short time as a private assistant to

Friedrich August Kekulé (07.09.1829 Darmstadt - 13.07.1896 Bonn) at the University Bonn and to Charles Adolphe Wurtz (26.11.1817 Wolfisheim near Strasbourg -

12.05.1884 Paris) in Paris (1872/73). In 1873/74 he was lecturer in chemistry at the Wageningen Agricultural College. He studied the acylation of sugars and cellulose, nitroamino compounds, urea, urethanes, nitric acid and oxalic acid. He retired in 1914 (JORISSEN, 1910, KERKWIJK, 1934, SNELDERS, 1979).



In April 25, 1874 J. M. van Bemmelen held his inaugural lecture in Leiden, the title "*Die Scheikunde als leer der stofwisseling*". His laboratory is now the Kammerlingh Onnes laboratory for low temperature research. He continued his

agricultural investigations and initially published several reports on many soil analyses which he carried out at the request of the Government and of some companies. These also dealt with soil specimens collected during drillings in the Zuider Zee and soil types of some dried-off polders (BEMMELLEN, 1877b,c 1880, 1886a,b). In addition, in 1877 and 1878 he published two important papers on the adsorption potential of agricultural soil. These investigations led him to conduct a detailed experimental with colloidal systems and to enter the field of research on which his reputation is based. The papers of colloidal science were reprinted in a book "*Die Absorption*" (BEMMELLEN, 1910).

J. M. van Bemmelen was in 1887 appointed a member of the Geological Committee of the Dutch Royal Academy of Science (Koninklijke Akademie van Wetenschappen), being elected secretary of this committee five years later. Another of his interests lay in instruction for practical work. Over 31 years he was also active (nine terms as chairman) in the "Society Mathesis Scientiarium Genitrix" established in 1785 for the purpose of intellectual education of the youth from the active classes of the bourgeoisie, artisans and labourers. Between 1882 and 1891, J. M. van Bemmelen was also chairman of the management of the "Praktische Ambachtsschule" and remained an honorary member of this body till his death. The last 31 years of his life he was a member of the Municipal School Board, too.

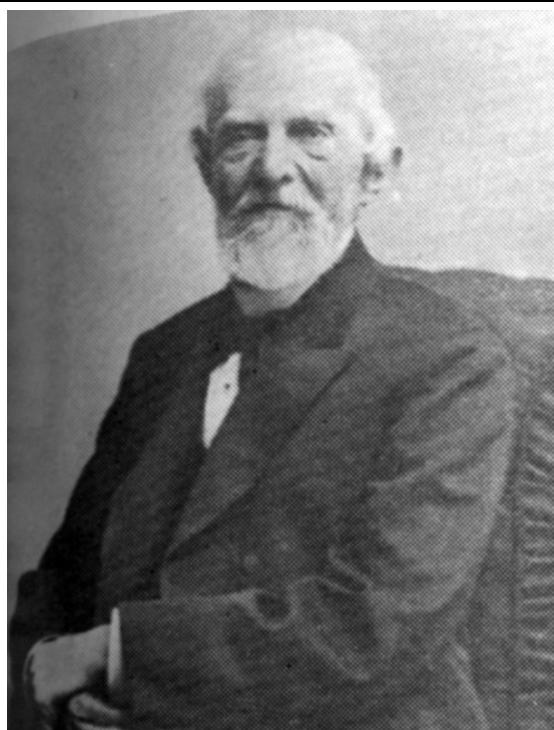


Wolfgang Ostwald

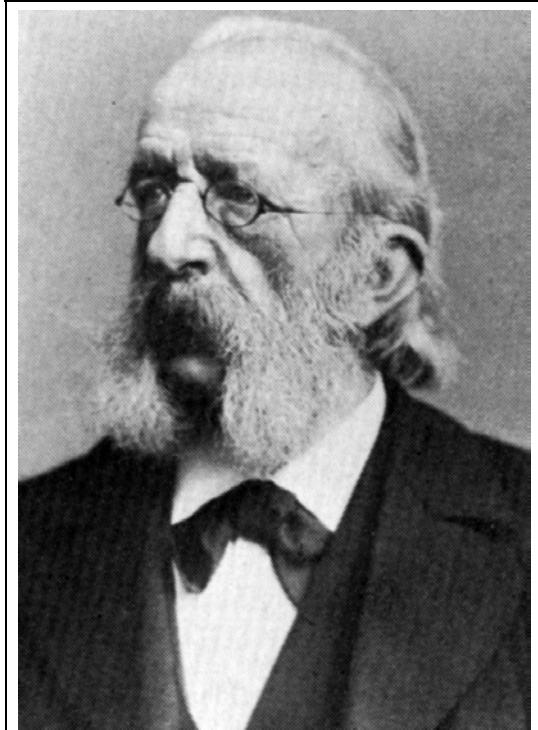
In 1889 van Bemmelen was rector of the University Leiden. In his speech as rector he demanded to raise a fund to have own money for the university. One year later the endowment fund "Leidsch Universiteitsfonds" was founded. This fund supported students and young scientists with money. On September 16, 1901 Jakob Maarten van Bemmelen became professor emeritus. He was one of the first in Holland to demand that his body be cremated, which necessitated its transport to Bremen in Germany, because there was then no crematorium in Holland. Jakob Maarten van Bemmelen died on March 13, 1911 in Leiden (FORRESTER AND GILES, 1971; FISCHER, 1989)

Carl Wilhelm Wolfgang Ostwald (27.05.1883 Riga - 22.11.1943 Dresden/ Bad Weißen Hirsch) wrote in the foreword of "*Die Absorption*" that Jakob Maarten van

Bemmelen was the founder of the theory of absorption (adsorption) from solution (BEMMELLEN, 1910). The history of solute-solid adsorption theory can be traced back to



Carl H. D. Boedeker



Wilhelm Henneberg



Thomas Graham

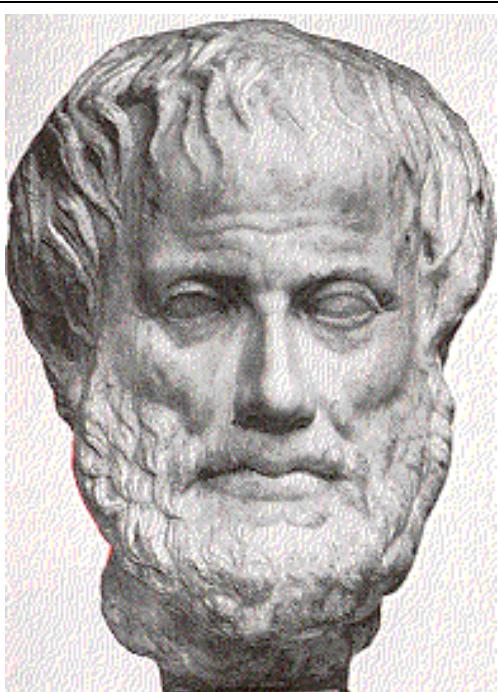


Friedrich Wöhler

the time of Carl H. D. Boedeker (1815 - 1895) who has given an equation of the adsorption of soil and shows that its development was entirely due to agricultural

chemists (BOEDEKER, 1859). The investigation of the adsorption of both basic and acidic radicals continued to attract the attention of soil chemists.

Similar results are reported by John Thomas Way (1820 Tunbridge Wells, Kent - 1883 Kensington) and Johann Wilhelm Julius Henneberg (10.09.1825 Wasserleben near Wernigerode - 22.09.1890 Göttingen) and Friedrich Carl Adolf Stohmann (25.04.1832 Bremen - 01.11.1897 Leipzig) (WAY, 1850; HENNEBERG AND STOHMANN, 1858). Way and Stohmann were students of Thomas Graham (21.12.1805 Glasgow - 16.09.1869 London), who was founder and first president of the Chemical Society of London, and who first named the term "colloid". Since 1851 Stohman studied under Friedrich Wöhler (31.07.1800 Eschersheim, now Frankfurt on Main - 23.09.1882 Göttingen) at the University Göttingen, than (1853) at the College of Chemistry in London and shortly later he became assistant of Thomas Graham shortly at the University of London, now University College. A short time he worked in the chemical factory of his father in Neusalzwerk (now Bad Oeynhausen) but with due to economic plight the family must give up the factory and lost their money. He became assistant at the Agriculturchemical Laboratory of the the Royal Agriculture Society (Agriculturnchemisches Laboratorium der Königlichen Landwirtschaftsgesellschaft) in Celle. Later he was Professor of Chemical Agriculture in Brunswick, Munich, Halle, and, 26 years until his death, in Leipzig (OSTWALD WI., 1897).



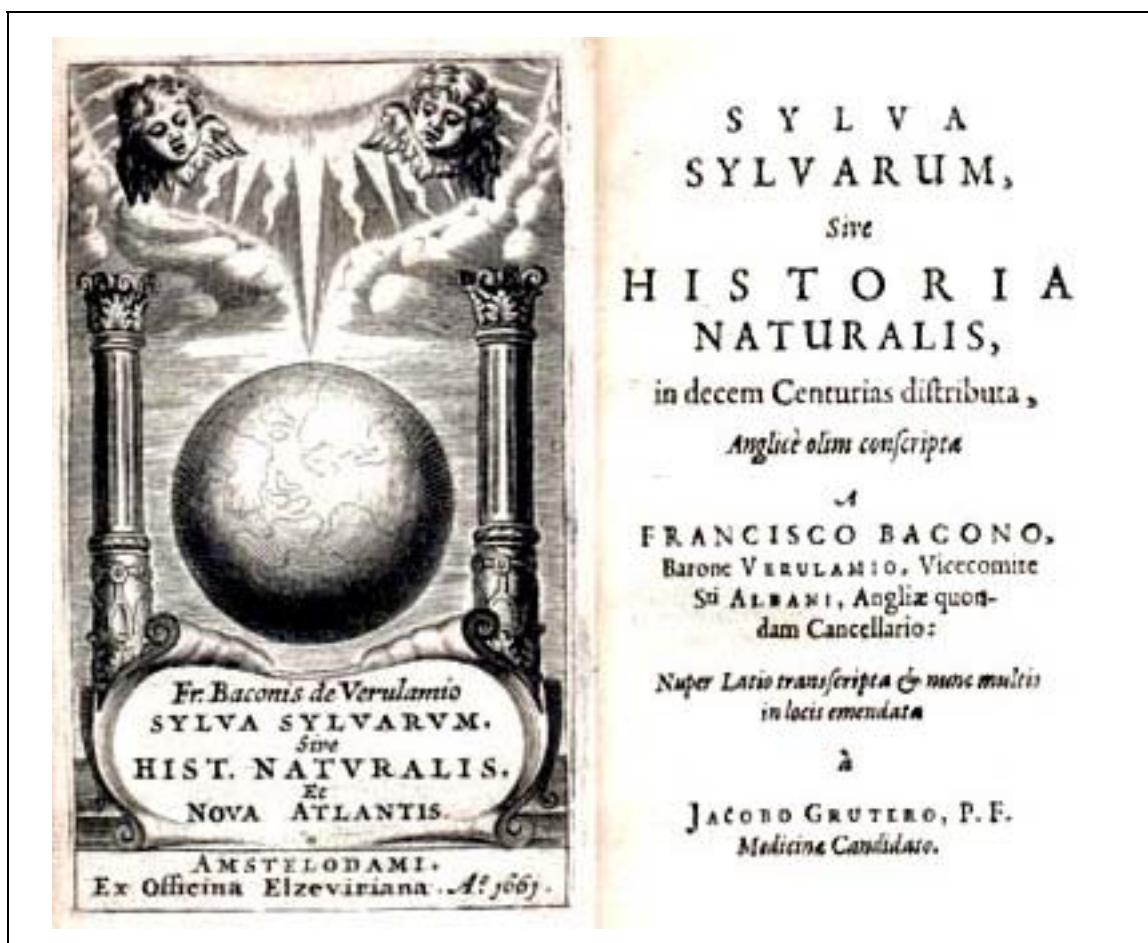
Aristoteles



Francis Bacon (Baron of Verulam)

The filtration of sea water through sand is known from the earliest times. Aristotele (384 B. C. Stagirus, Macedonia/Greece - 322 B. C. Chalcis, Euboea/

Greece) reported that sea water lost some of its taste by filtration through sand. Lord Francis Bacon (Baron of Verulam; 22.01.1561 London - 09.04.1626 Highgate near London) speaks of a method of obtaining fresh water which was practised on the coast of Barbary in his *Sylva Sylvarum*: “*Digge a hole on the sea shore somewhat above high water mark, and as deep as low water mark, which when the tide cometh will be filled with water fresh and potable*”. He also remembers: “*to have read that trial hath been made of salt water pressed through earth through ten vessels, one within another, and yet it hath not lost its saltiness as to become potable*”, but when “*drayned through twenty vessels hath become fresh...*” (BACON, 1627; FORRESTER AND GILES, 1971).



Johann Rudolph Glauber (1604 Karlstadt/Franconia - 10.03.1670 Amsterdam, other 16.03.1670 or 10.03.1668 Amsterdam) later an alchemist, applied chemist, and technical chemist, began his education at the Latin school in Karlstadt, but did not finish. Probably he also worked at the Mohren-Apotheke. He did not attend an university. Glauber settled his life through many towns. He visits laboratories in Paris, Basel, Salzburg (1626), and Vienna (1625/26). In 1635 he worked at the court apothecary in Giessen. From 1636 to 1639 he leaved Giessen and went to Frankfurt, and then probably went to work for the Landgraf von Hessen-Darmstadt in Bonn. In

1640 Glauber left Germany and settled in Amsterdam. In 1644 Glauber came back to Giessen as a court apothecary, 1646 he returned to Amsterdam and bought a large house for which he had to pay annual property tax of 1 000 Gulden which seems an enormous amount. He produced and sold drugs. He fled 1651 to Germany because he was bankrupt with 10 000 Gulden debts. He settled in Wertheim and began experiments in wine improvement, probably also continuing other alchemical experiments. In 1652 in Kitzingen he bought a large house with cash. Glauber maintained a medical practise of sorts, dispensing primarily antimony-containing medicines free of charge. He returned 1655 to Amsterdam where he died.



Johann Rudolph Glauber

In the *Glauberus concentratus* or *Kern der Glauberschen Schriften* written from his pupils (1668, and posthum 1715):

“Die beste Präcipitation geschiehet durch einen sonderlichen Sand, den mir nicht beliebt zu offenbaren, welcher alles Saltz, Schleim, Gestanck und Unreinigkeit aus dem Wasser, Mistlachen, die angebohrene Röte im rothen Wein, Bier etc. etc. in wenigen Stunden fällt, und aus rotem Wein weißer Wein wird. Wird doch das Meer-Wasser süß, wann es durch gemeinen Sand lauffet, und allda sein Saltz hinter sich lässt: geschieht nun solches natürlich, warum nicht durch die Kunst”. (“The best technology is given through an special sand, which I don not reveal. All salts, slime, maladour and uncleaness from water, liquid dung, the red of the red-wine, beer, etc., falling down in several hours, and the red wine will be white wine. When sea-water will be sweetly, when it was filtered through normaly sand and all the salt goes out of the sea-water: happened this naturally, why not by means of artistry”) (GLAUBER, 1715, ROBENS, 2000).



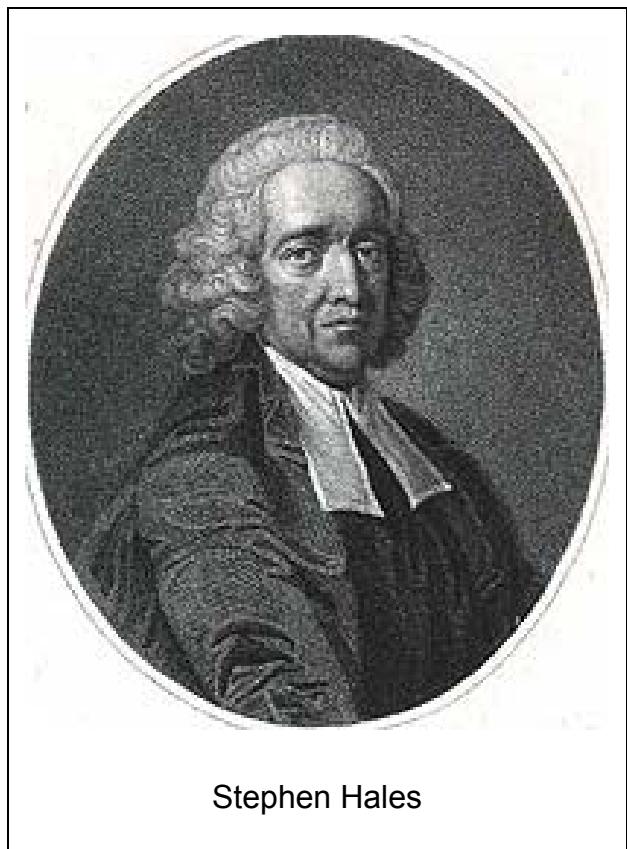
Luigi Ferdinando Marsili

Luigi Ferdinando Marsili (20.07.1658 Bologna - 1730 Bologna) in 1725 made quantitative experiments by filtering of sea

water through fifteen successive vessels of garden soil; a reduction of the salt content was proved by evaporation and by change in specific gravity (MARSILI, 1725).

Similar results were obtained with sand. Boyle Godfrey discussed the question of making sea water fit for use on ships and observed that if sea water be put into a stone straining cistern the first pint that runs through will be like pure water, having no taste of salt, but the next pint will be as usual (GODFREY, 1737).

Stephen Hales (17.09.1677 Bekesbourne/Kent - 04.01.1761 Teddington/Middlesex) worked in plant physiology. His book *Vegetable Statics* (1727) included his most important observations in plant physiology. Hales demonstrated that plant leaves adsorb air and that a portion of air is used for plant nutrition. In *Haemostatics* (1733) Hales made medical recommendations on therapeutic bleeding and surgery and practical medicinal applications. Hales dealing in *Philosophical experiments* (1739) with the same question as Godfrey, refers to make use of a soft stone as a filtering material, but he points out that this method is of no practical value as only the first portions of the filtrate are free from salt (FORRESTER AND GILES, 1971; BENEKE, 1995a).



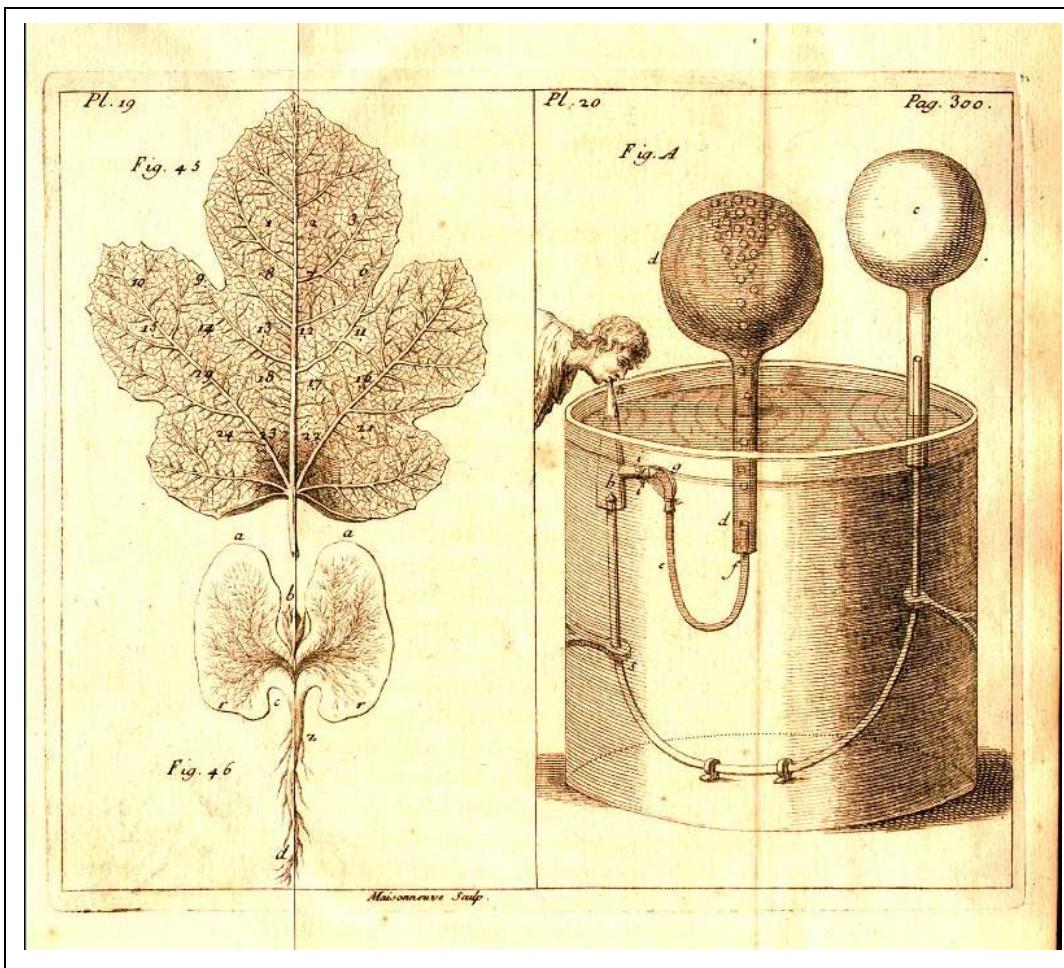
Stephen Hales



Jöns Jakob Berzelius

Jöns Jakob Berzelius (20.08.1779 Väversunda - 07.08.1848 Stockholm) reported when solutions of common salt filtered through sand, the first portions that passed are quite free from saline impregnation. Carlo Maria Giovanni Matteucci (21.06.1811 Forli/Romagna - 25.07.1868 Ardenza near Livorno) founded this observation to other salts, and found that the solutions when filtered through sand

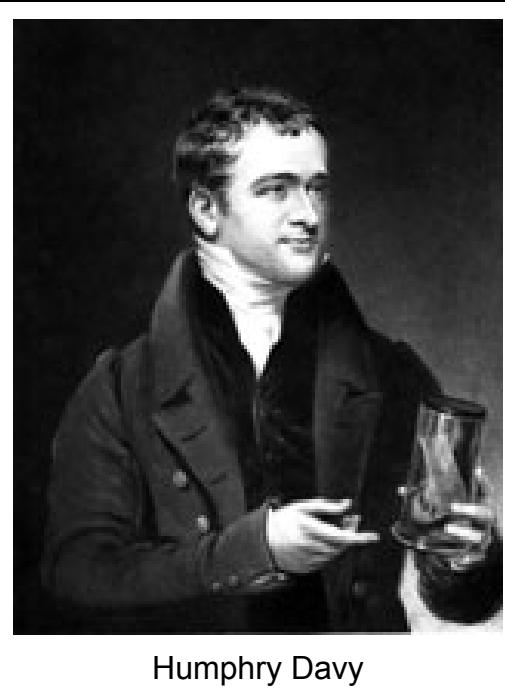
were diminished in density, thus indicating a retention by the sand of thin quantities of the sand used. Matteucci explains these phenomena on the principle of capillarity. The particles of the sand have a surface section for the water and the salt, but this is greater when the latter, and, therefore, the salt is concentrated and condensed on the sand, while the liquid is proportionally diminished in strength and density. When this attraction has been gratified the solution ceases to be affected, and goes through as it went in (FORRESTER AND GILES, 1971).



Stephen Hales: metabolism by plants and animals

F. H. Storer commented in 1887 that, naturally enough, practical men have long observed how readily barnyard liquor and other coloured waters are made clear and odourless when put in contact with fresh loam or clay. Several writers at one time or another have called attention to the fact, and have dealt upon its importance notably Sir Humphry Davy (17.12.1778 Penzance/Cornwall - 29.05.1829 Geneva/Switzerland) in 1813, Giuseppe Gazzeri (1771 Florence - 1847) in 1819 and Lambruschini in 1830. Gazzeri wrote as follows: "*Loam and especially clay take possession of soluble matters which are intrusted to the soil, and retain them in order*

to give them by degrees to plants, conformably with their needs...“. Lambruschini suggested that it might perhaps be well to apply the name “incorporation” to the act of combining dung liquor and soil. We can readily recognise, he said, that fertilising liquids and the constituents of a well prepared soil enter into a peculiar combination, by virtue of a special affinity. This combination is not weak enough to allow any very easy loss of the fertilising constituents, and to permit plants to consume them too rapidly, and yet the combination is not strong, but that the vital action of growing plants can gradually overcome it (GAZZERI, 1919; LAMBRUSCHINI, 1830; STORER, 1887; FORRESTER AND GILES, 1971).



Humphry Davy



Giuseppe Gazzeri

In 1836 Johann Philipp Bronner (11.02.1792 Neckargemünd, near Heidelberg - 04.12.1864 Wiesloch, near Heidelberg) who lived in Wiesloch (Germany) has written in his book about grape culture the following passages:

“Fill a bottle which has a small hole in its bottom with fine river sand or halfdry sifted garden earth. Pour gradually into the bottle thick and putrefied dung-liquor until its contents are saturated. The liquid that flows out at the lower opening appears almost odourless and colourness, and had entirely lost its original properties“.

After instancing the facts that wells situated near dung-pits are not spoiled by the latter, and that the foul water of the Seine at Paris becomes potable after filtering through porous sandstone, Bronner continues:



Johann Philipp Bronner

"These examples sufficiently prove that the soil, even sand, possesses the property of attracting and fully absorbing the extractive matters so that the water which subsequently passes is not able to remove them; even the soluble salts are absorbed, and are only washed out to a small extent by new quantitives of water" (BRONNER, 1836; FORRESTER AND GILES, 1971).

Johann Philipp Bronner was apothecary in Wiesloch (Stadt-apotheke) and in 1820 started practical experiments in his vineyard. Before this time he had traveled through Europe obtaining great experience in winegrowing. J. P. Bronner made a conditioning of vine cuttings, with later 400 breeds. He was a pioneer and one of the resourcefulliest and forward-looking vineculter explorer of the 19th century in Germany. He wrote eight books about oenology (BRONNER, 1833, 1834, 1835, 1836, 1837a,b, 1839, 1842). The pharmacy of Bronner (Stadt-apotheke) became later in August 8, 1888 the first fuel filling station of the world, when Bertha Benz (03.05.1849 Pforzheim - 05.05.1944 Ladenburg near Heidelberg), the woman of Carl Friedrich Benz (25.11.1844 Mühlburg near Karlsruhe - 05.04.1929 Ladenburg), drived - without the knowledge of her husband - with her sons Eugen and Richard with the first car "Benz Patent Motorwagen" 106 km from Mannheim to Pforzheim, to refuel in the pharmacy in Wiesloch now under apothecary Willi Ockel 3 liters of

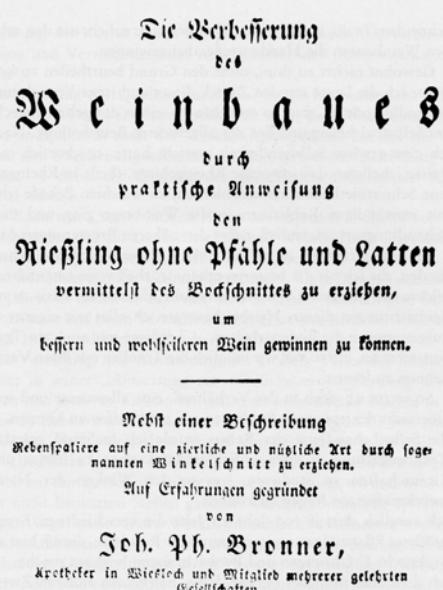


Abb. 4. Titelseite des ersten Buches von Bronner 1830

Ligorin. The “Benz Patent Motorwagen” was in January 29th 1886 patented under the German Patent-Number 37 435.



Bertha Benz



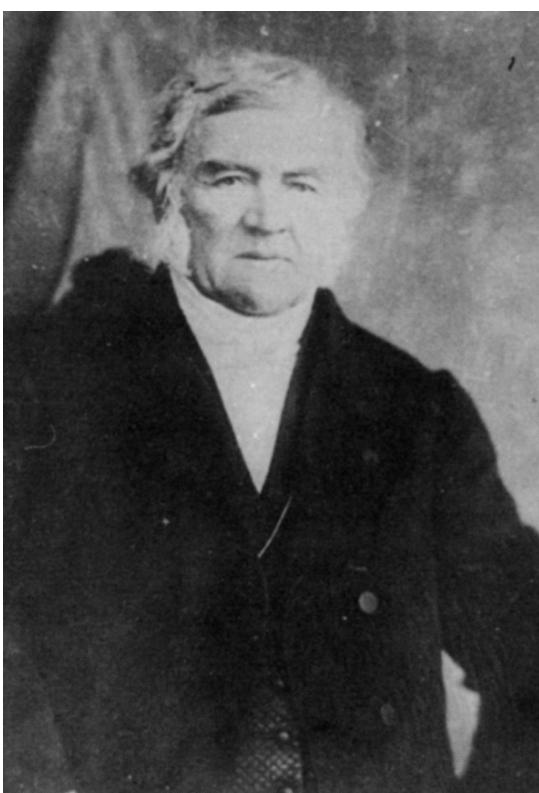
Carl Friedrich Benz



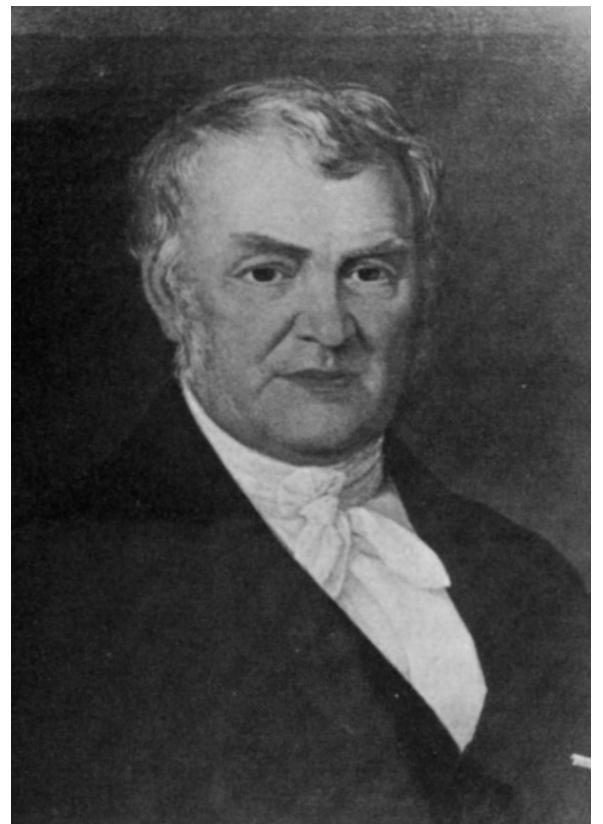
Old pharmacy of Johann Philipp Bronner in Wiesloch (2003)



Memorial tablet for Johann Philipp Bronner in Wiesloch



Johann Philipp Bronner



Johann Philipp Bronner



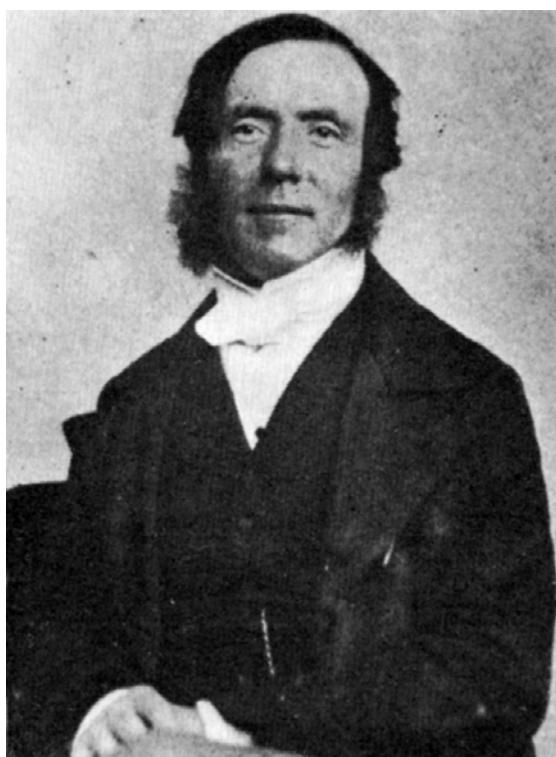
Memorial for Bertha Benz and her sons with the first car and the fuel filling station in Wiesloch



Harry S. Meysey Thompson

Similar observations as made by Bronner were reported by two members of the Royal Agricultural Society of England, the Reverend Canon Anthony Huxtable (30.11.1808 Somerset - 12.12.1883 St. Leonard's on Sea) (later Archdeacon) who farmed at Sutton Waldron, near Shaftesbury on the chalk hills of Dorset and Harry Stephen Thompson (11.08.1809 Moat Hill - 26.03.1874 Kirby Hill) (later Sir Harry S. Meysey Thompson of Kirby Hall), a Yorkshire farmer from Moat Hall, near York. Thompson was a founder member of the Agricultural Society and was very active in the society all his life. From 1855 till his death Harry S. Thompson was the second editor of the Journal, and for 1867 President of the Society.

C. A. Huxtable collected the liquid running from cowhouses, stables and piggeries into a tank and made an experiment in the filtration of the liquid manure in his tanks through a bed of an ordinary loamy soil. After the passage through the filter bed, the urine was found to be deprived of colour and smell. This was a singular and interesting observation, implying, the power of the soil to separate from solution those organic substances which give colour and offensive smell to putrid animal liquids.



Canon Anthony Huxtable

At about this time the practise of dosing tanks and manure heaps with sulphuric acid was widespread to prevent the escape of the useful volatile ammonia, whereby large quantities of ammonium sulphate were formed. Thompson records that he was aware that the soil had a certain power of retaining ammonia but he was anxious to list the extent of this power and to ascertain whether it also extended to the sulphate, as, if not, the use of sulphuric acid as a fixe, through preventing the escape of ammonia in a volatile form, would have been objectionable from its forming a highly soluble salt that would be readily washed down into the subsoil and carried off in the drains.

Harry S. Meysey Thompson made experiments and used cultivated soil from one of his fields, a light sandy loam of good quality and placed it in glass percolators. He weighted all substances and measured the liquids. Thompson must get the credit for the first quantitative studies on cation exchange. He discovered that the addition of $(\text{NH}_4)_2\text{SO}_4$ to a column of soil resulted in the appearance of CaSO_4 solution after leaching. Before Thompson published his results, he told 1848 a young consulting chemist to the Royal Agricultural Society, John Thomas Way, of his results (THOMPSON, 1850).

John Thomas Way was in 1846 appointed Professor of Chemistry of the Royal Agricultural Society of England. He was a member of the first (1847) and second (1864) Commission on River Pollution. 1864 health began to fail and he did little more scientific work.

Way started his experiments and used a variety of soils, pipe clay, and some home-made aluminosilicates. In addition to NH_4^+ , he studied the exchange of all cations (and anions) usually found in soils. Way worked four years and his results are summarized briefly as follow:

- 1) The common cations (Na^+ , K^+ , NH_4^+) added as salts of strong acids, are retained by the soil, and equivalent quantities of the same salts are replaced, (e.g $2 \text{ KCl} + \text{Soil} \rightarrow \text{CaCl}_2 + \text{K}_2\text{-soil}$)
- 2) Hydroxides and carbonates of cations are adsorbed completely with no replacement of Ca^{2+} or anions.
- 3) The strong acid salts (Cl^- , NO_3^- and SO_4^{2-}) of Ca^{2+} are not adsorbed by soils.
- 4) Clay is the material responsible for cation adsorption, organic matter and sand are unimportant.
- 5) Heating or acid treatment tend to destroy the adsorption.
- 6) The adsorption is very rapid, almost instantaneous.
- 7) Increasing the concentration of the added salt increases the amount of adsorption.
- 8) Cation adsorption is irreversible.
- 9) Phosphate is held by soils.

The conclusion about organic matter was wrong and so was the conclusion about irreversibility, which his own data disproved (WAY, 1852). In other respects, the conclusions were correct, given the Ca-saturation of his original soils. Conclusions 2 and 3 are only apparently correct because the analysis of leachates gave no clue to the actual reactions. Nevertheless the results were revolutionary in their implications that the great agricultural chemist Justus von Liebig commented: "*These experiments are very remarkable and should be opposed with might and main*" (WIEGNER, 1931, FORRESTER AND GILES, 1971; THOMAS, 1977).

Friedrich Stohmann and Wilhelm Henneberg met end of 1856 in the agricultural chemistry in Celle (Laboratorium der Königlich Hannoverschen Landwirtschaftsgesellschaft Celle, since 1857 Weende, now Göttingen), repeated Way's observations. They looked carefully at the effect of concentration on the quality of ammonium adsorbed by soils, and by doing so became the first to measure a solute-solid adsorption isotherm. They also showed that there were differences in adsorption of ammonium from different salts, in the order $\text{H}_2\text{PO}_4^- > \text{SO}_4^{2-} > \text{Cl}^- = \text{NO}_3^-$, an observation that took many years to understand. The Table in Henneberg and Stohmann's paper gives the first quantitative results published on adsorption from solution (HENNEBERG AND STOHMANN, 1858).

Table of Henneberg and Stohmann (1858)

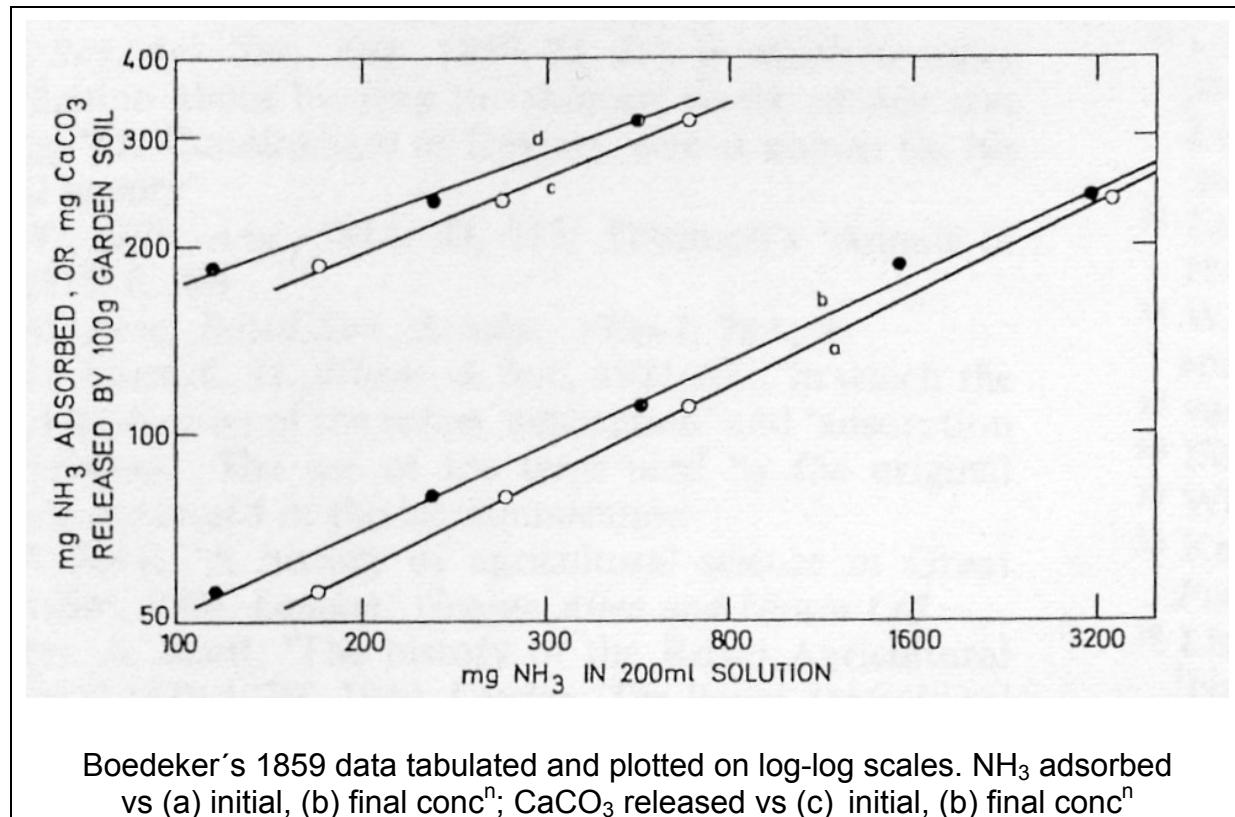
Ammoniakabsorption durch 100 Grm. Gartenerde aus 200 Cubikcentimetern Lösung in Gramm NH ₃ ausgedrückt :												
		Dauer der Berührung, Stunden	200 CC. Lösung enthielten NH ₃ :									
			0,170 Grm. 0,05 Atom im Liter		0,340 Grm. 0,1 Atom im Liter		0,680 Grm. 0,2 Atom im Liter		1,700 Grm. 0,5 Atom im Liter		3,400 Grm. 1 Atom im Liter	
Hbg.	St.	Hbg.	St.	Hbg.	St.	Hbg.	St.	Hbg.	St.	Hbg.	St.	
1) Aetzammoniak		4 24 168		— 0,056 0,052	— 0,058 0,052	— 0,067 0,067	0,086 0,120	— 0,122 0,174	— — —	— — —	— — —	
	Durchschnitt			0,058	0,095		0,149					
2) Chlorammonium		4 24 168		0,056 0,058 0,056	0,056 0,052 0,049	0,070 — 0,072	0,070 — 0,074	0,102 0,112 0,112	0,102 0,104 0,102	0,106 0,115 0,123	0,122 0,122 0,102	
	Durchschnitt			0,0545	0,071		0,107		0,118	0,136 0,162	0,206 0,238 0,130	
3) Salpetersaures Ammoniak		4 24 168		— 0,052 0,052	0,055 0,056 0,053	0,083 0,080 —	0,081 0,078 0,079	0,106 — —	0,106 0,105	0,154 — —	0,198 0,174 0,157	
	Durchschnitt			0,054	0,080		0,106					
4) Schwefelsaures Ammoniak		4 24 168		— — —	0,057 0,057 0,057	— 0,087 0,086	— 0,086 0,086	0,120 0,114 0,116	— 0,120 0,122	— — —	— — —	
	Durchschnitt			0,057	0,086		0,118					
5) Mischung von Aetzammoniak und Chlorammonium zu gleichen Atomen		4 24 168		— — —	— 0,089 —	— — —	— 0,118 —	— — —	— — —	— — —		
	Durchschnitt			—	0,089		0,118					
6) Phosphorsaures Ammoniak (2 NH ₄ O, HO, PO ₃)		4 24 168		— — —	0,140 0,144 —	0,136 0,136 —	0,136 0,150 0,144	0,188 0,202 —	— — —	— — —		
	Durchschnitt			—	0,141		0,206		— — —	— — —		
Durchschnitt von 2 bis 4				0,055	0,079		0,110		— — —	— — —		

Table from Henneberg and Stohman's paper. The first detailed quantitative results published on adsorption from solution.

der Ackerkrume gegen Ammoniak u. Ammoniakalze. 159

Table of Boedeker (1859): Ammonia adsorption and calcium release by soil

Ammonia in 200 cm ³ solution (mg)	Ammonia adsorbed by soil (mg/100 g)	Ammonia adsorbed by soil (mg/100 g)	'Carbonate of lime' released from soil (mg/100 g)	'Carbonate of lime' released from soil (mg/100 g)
	Calculated	Found	Calculated	Found
170		55	161.8	186
340	77.8	79	227.9	233.5
680	110	110	323.6	312.5
1700	173.9	188 (max.)		
3400	245.9	238 (max.)		



Henneberg and Stohmann made no attempt to express their findings by a law, but a few month later Carl Boedeker, a colleague, then head of the Physiological Institute in Göttingen, published his deductions from their results (BOEDEKER, 1859).

Samuel William Johnson (born 1830 in Kingsborough/New York) studied chemistry at the Yale Scientific School, and at the Universities in Leipzig and Munich, where he was a student of Justus von Liebig. Johnson reviewed the work of Way and corrected his two wrong conclusions. Johnson recognized the adsorption work of Henneberg and Stohmann and showed that nutrients can go into solution after they are adsorbed (exchange is reversible) and thus be taken up by plants. He found that Way was incorrect in starting that organic matter did not adsorb NH₄. He found it hold even more NH₄⁺ than clay did, and Johnson coined the term "exchange of bases" for Way's observation (JOHNSON, 1859). Johnson was later the first director of the first experimental station in Connecticut, elected President of the American Chemical Society (1878) and (1866) received an election to the National Academy of Sciences (FORRESTER AND GILES, 1971; THOMAS, 1977).

Most of the work between the late 1850's and 1900 was an extension of or elaboration on Way's original work. Important researchers were H. Eichhorn who

showed that crystalline zeolites could be changed easily from one to another merely by replacing the cation present (e.g. Ca^{2+} to Na^+) (EICHHORN, 1858); C. Peters who did detailed studies on concentration, anion and cation effects, amplifying Hennebergs and Stohmann's results (PETERS, 1869); and Jakob Maarten van Bemmelen who showed that cations other than Ca^{2+} could be replaced from soils (Bemmelen, 1877a).

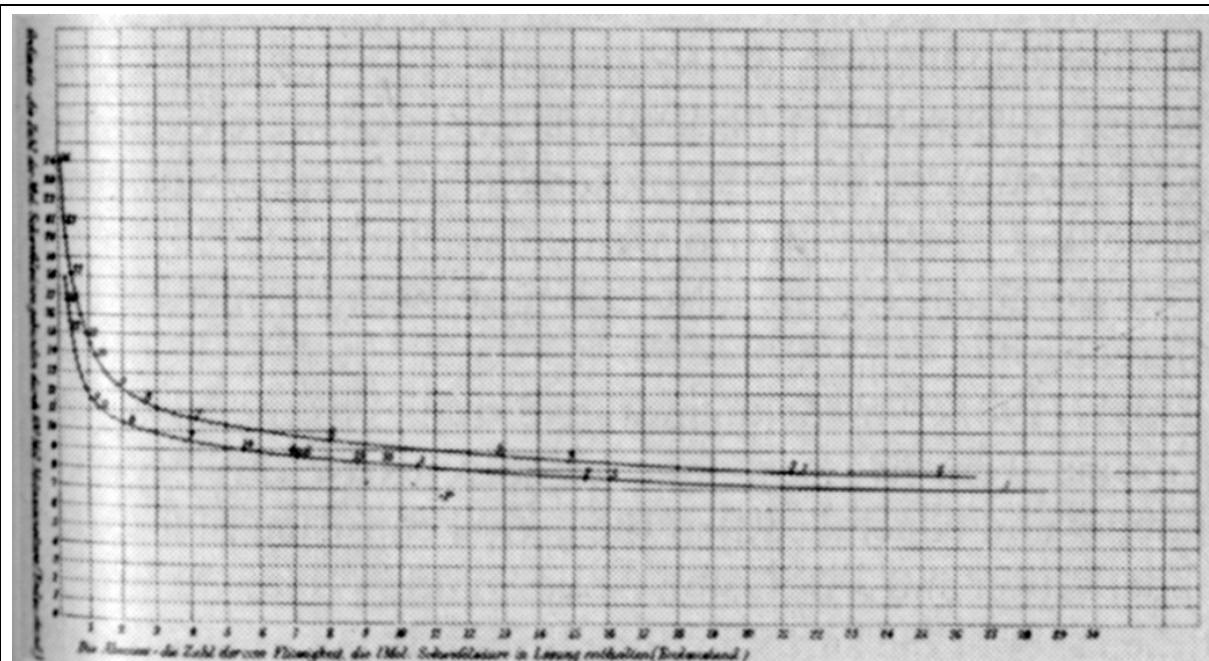


Figure 1 (BEMMELLEN, 1881):

The adsorptions isotherms of sulphuric acid by metastannic acid by Jakob Maarten van Bemmelen are probably the first published adsorption isotherms from solution

J. M. van Bemmelen made the greatest contributions. His data show conclusively that cation exchange is not restricted to calcium ions in the soil, as Way assumed, but works with other cations as well. He also formulated the first mental picture of cation exchange and the first published adsorption isotherm from solution in the form now used (BEMMELLEN, 1888). This paper is a classic one of a long line of publications on adsorption and was often quoted by agricultural and colloid scientists in the early year of the last century.

The isotherms of Boedeker (BOEDEKER, 1859) were interpreted in 1906/07 from Herbert Max Finlay Freundlich (28.01.1880 Charlottenburg, now Berlin - 31.03.1941 Minneapolis/ Minnesota) (*Freundlich-Isotherm*) (FREUNDLICH, 1906a,b).

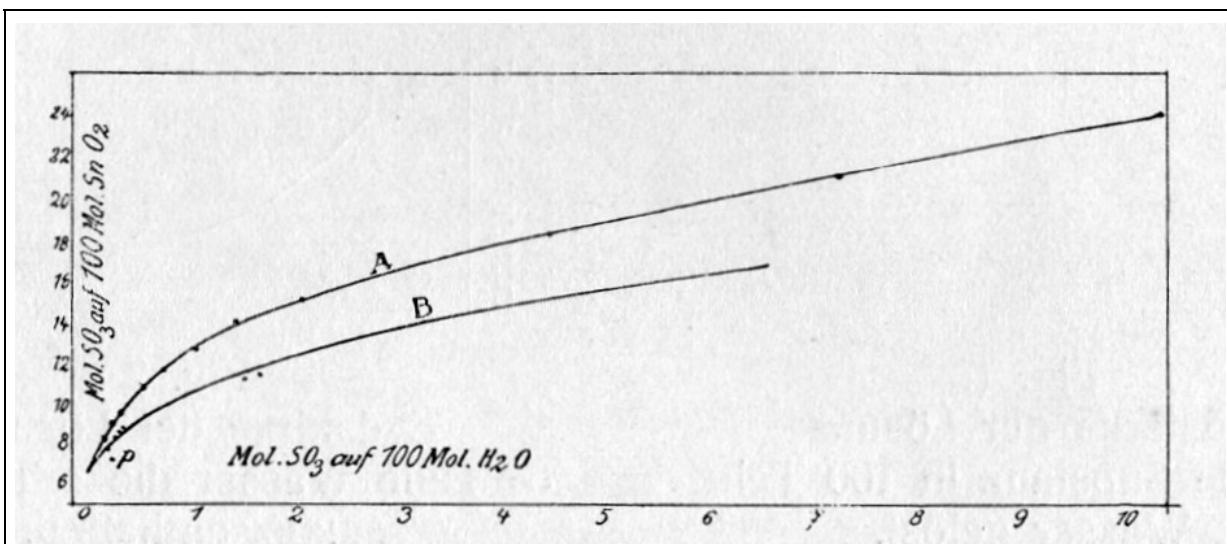


Figure 2 (BEMMELEN, 1881,1910):
The isotherms of sulphuric acid by metastannic acid by Jakob Maarten van Bemmelen from the original data of 1881 (Figure 1), published 1910, are probably the first complete adsorption isotherms

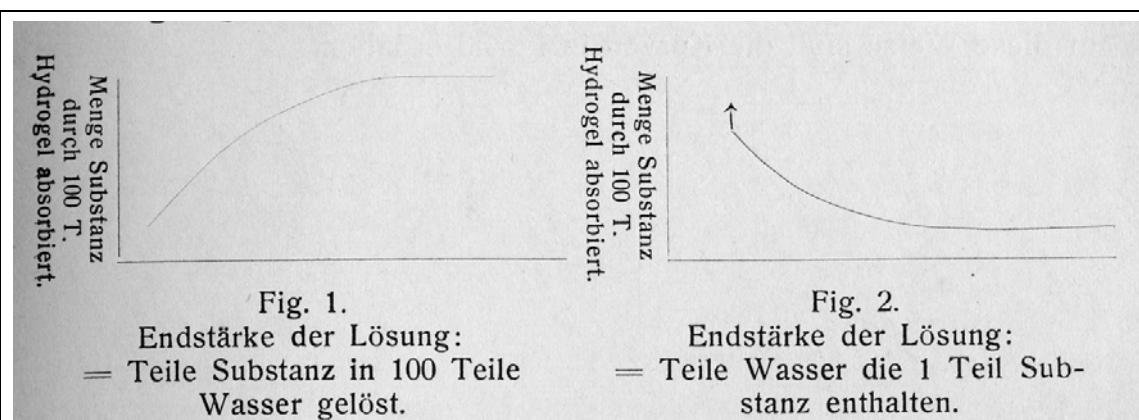
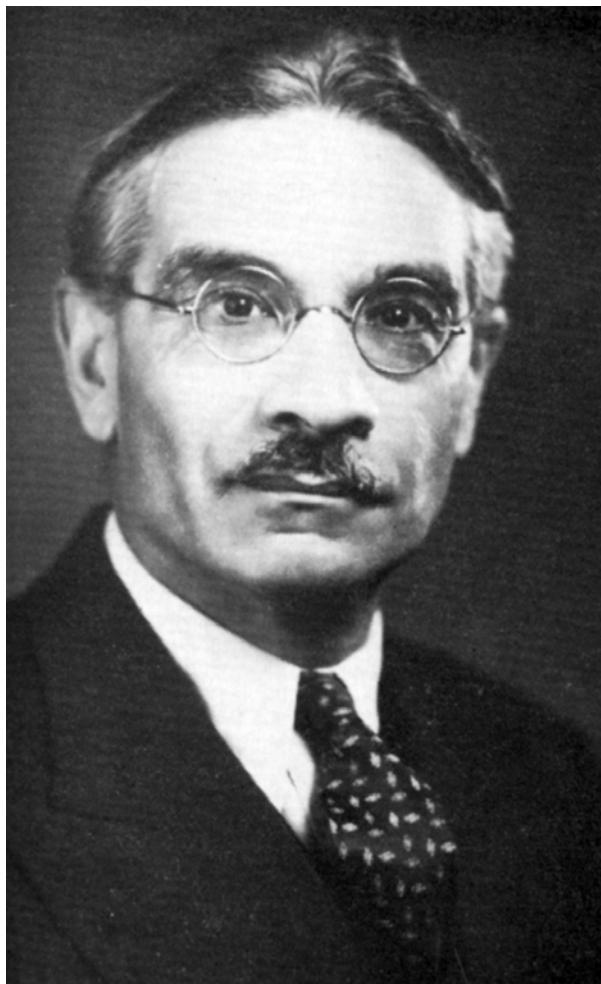


Figure 3 (BEMMELEN, 1888):
The diagram on the left is probably the first published adsorption isotherm in the form now used

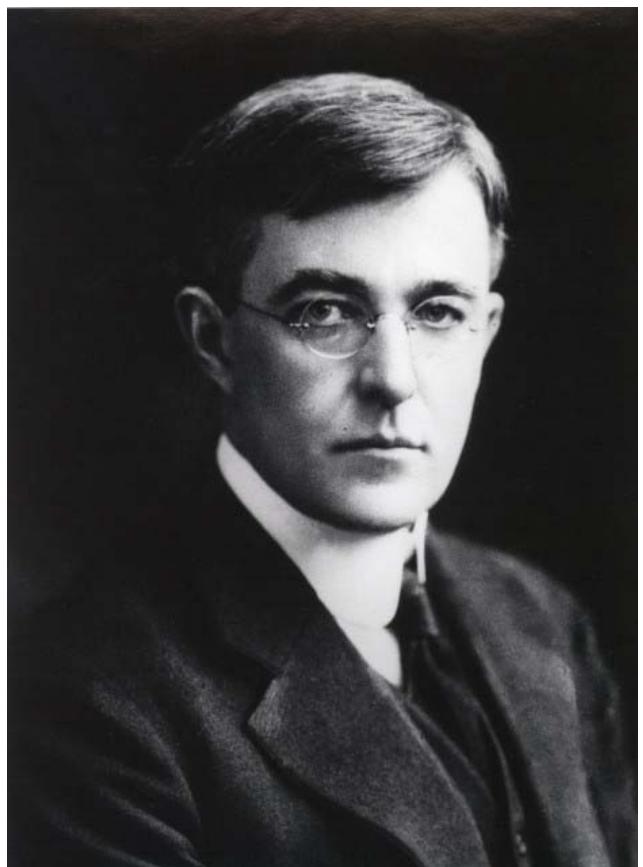
The famous *Langmuir-Isotherm* was published by Irving Langmuir (31.01.1881 Brooklyn - 16.08.1957 Falmouth/ Mass.) in 1918 (LANGMUIR, 1918).

In 1938 Stephen Brunauer (12.02.1903 Budapest - 06.07.1986 Potsdam/New York), Paul Hugh Emmett (22.09.1900 Portland/Oregon - 22.04.1985

Corvallis/Oregon) and Edward Teller (15.01.1908 Budapest - 09.09.2003 Stanford/California) published an adsorption isotherm for gases on solids (*BET-equation*) (BRUNAUER, EMMETT, TELLER, 1938).



Herbert Max Finlay Freundlich

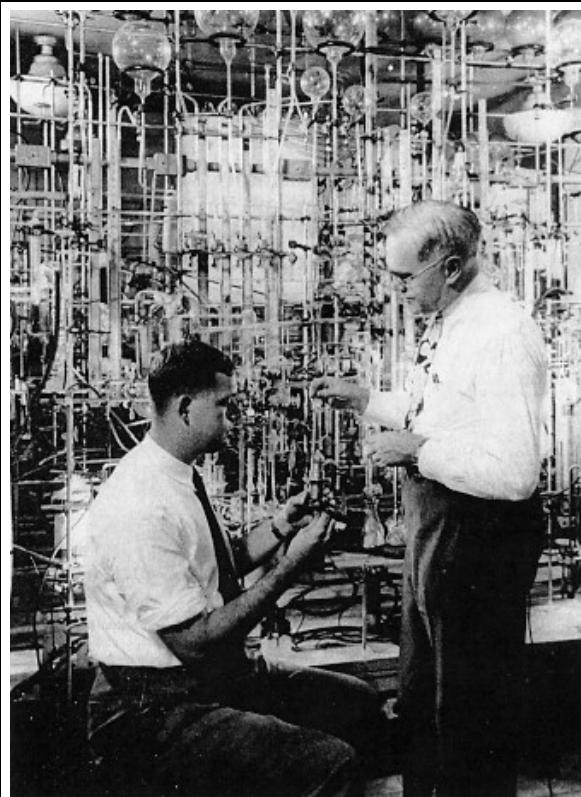


Irving Langmuir

The term “absorption” was used in the 19th century for all absorption and adsorption problems. Moritz Ludwig Frankenheim (29.06.1801 Brunswick - 14.01.1869 Dresden) in 1835 recognized that absorption in porous systems was different to homogeneous liquids. He elaborated the concepts *adsorption* and *insorption*. Frankenheim understood by adsorption not only the aggregation of gases in porous systems but also the liquid uptakes. Frankenheim called “insorption” what we today call absorption. This classification was not very fortunate because the adsorption of gases and the uptake of liquids are two different operations. The terms were not often used and other authors used further “absorption” (FRANKENHEIM, 1835, BENEKE, 1995b).



Stephen Brunauer

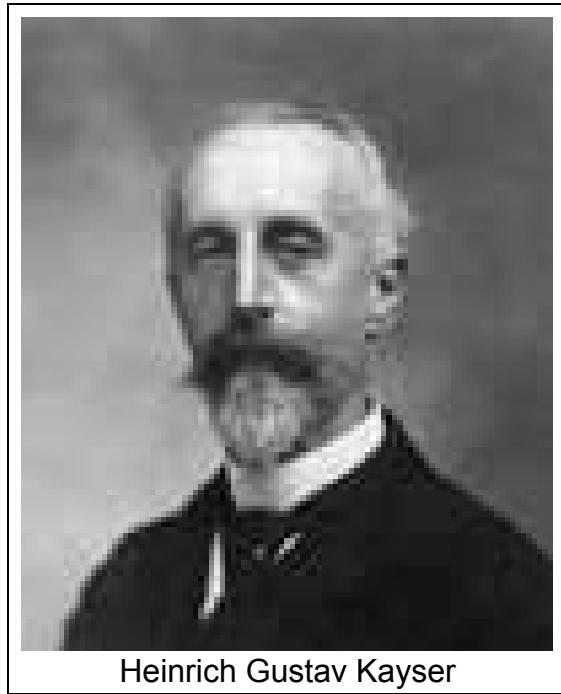


Paul Hugh Emmett (right) with a coworker in front of a gas adsorption apparatus (1953)



Edward Teller

(Johann) Heinrich Gustav Kayser (16.03.1853 Bingen at Rhine - 14.10.1940 Bonn) in 1881 used the term “adsorption” for the accumulation of gases on surfaces and the adsorption of molecules from solutions through porous substances (KAYSER, 1881a,b). The sorption of gases and the adsorption of solutes in gels were named “adsorption” but Jakob Maarten van Bemmelen avoided this word and spoke his long life from “absorption”. In the adsorption in gels he saw a special process; which was neither an adsorption from solution nor a formation of a chemical compound identic (BEMMELEN, 1888, BENEKE, 1995b).



Heinrich Gustav Kayser

Jakob Maarten van Bemmelen had a very independent mind and an excellent memory, and was small in figure. He held firm opinions, and had a boyish sense of humour. He was much interested in physics and theological research and devoted much time to study the genealogy of his own family. Once, he sketched out the plan of the Zuider Zee on the breakfast table - in treacle to instruct his family. Being admonished by his wife, he replied that they had to learn their geography! At his 80th birthday (1910) his grandson held a speech to congratulate his grandfather. Jakob Maarten van Bemmelen recited the speech in Greek which he had delivered at his own University Matriculation

examination (FORRESTER AND GILES, 1971; FISCHER, 1989).



Pierre Maurice Marie Duhem



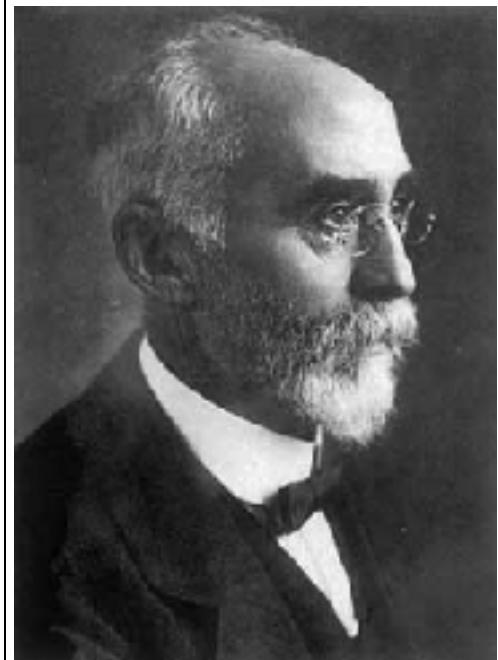
Henry Louis Le Chatelier

To his 80th birthday friends published a book in honor of Jakob Maarten van Bemmelen. In this book are contributions from Jacobus Henricus van't Hoff, Pierre Maurice Marie Duhem (10.06.1861 Paris - 14.09.1916 Cabrespine (Aude)), Heike Kammerlingh Onnes, Henry Louis Le Chatelier (08.10.1850 Paris - 17.09.1936

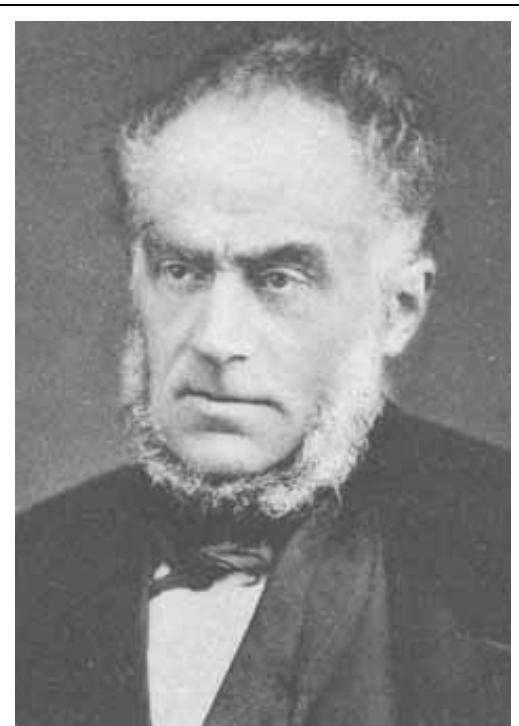
Miribel-les-Échelles (Isère)), Raphael Eduard Julius Liesegang (01.11.1869 Elberfeld, now Wuppertal - 13.11.1947 Bad Homburg vor der Höhe), Hendrik Antoon Lorentz (18.07.1853 Arnhem - 04.02.1928 Haarlem), Wolfgang Ostwald, Theodor Svedberg (30.08.1884 Valbo near Gävle - 26.02.1971 Kopparberg near Örebro) and others (HARDY, 1911).

The signature of Raphael Eduard Julius Liesegang, written in cursive script below his portrait.

Raphael Eduard Julius Liesegang



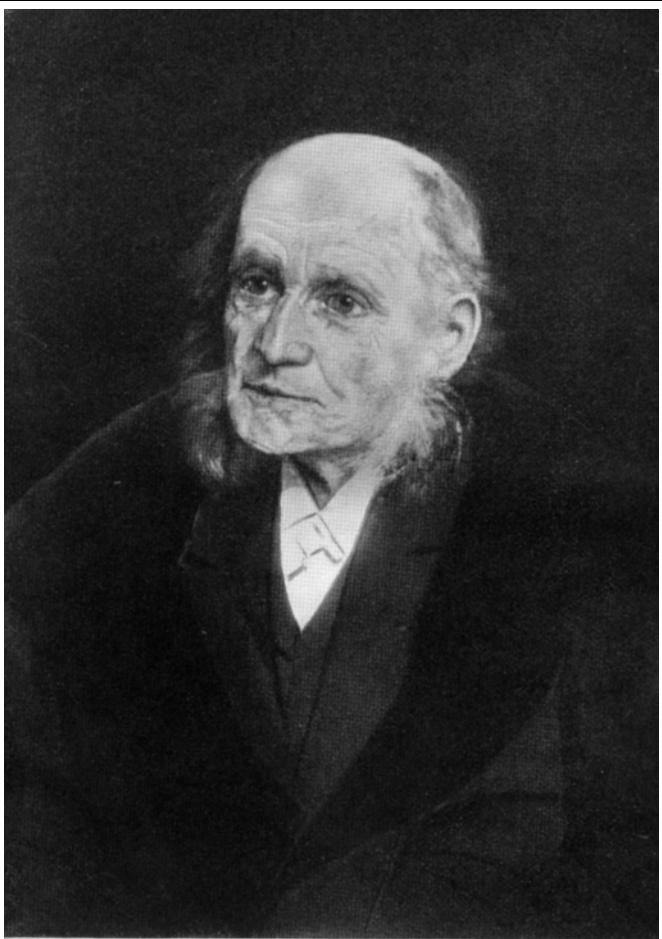
Hendrik Antoon Lorentz



Carlo Matteucci



The Svedberg (left) with mechanist



J.M.vanBemmelen

Jakob Maarten van Bemmelen

The eldest son of Jakob Maarten van Bemmelen, Johan Frans van Bemmelen (26.12.1859 Groningen - 06.08.1956 Leiden) married Adriana Jacoba Paulus in July 8, 1897. They had one son and two daughters. J. F. van Bemmelen studied biology and was later (since 1907) Professor of zoology in Groningen and he retired 1930 (VISSER, 2001).

The grandson of Jakob Maarten van Bemmelen and son of Johan Frans van Bemmelen, Jacob Maarten van Bemmelen (20.04.1898 's-Gravenhage - 12.01.1982 Dordrecht) was Professor for criminal law in Leiden. He wrote his thesis 1923 in Groningen "*Van zedelijke Vertreterin tot Reclasseering. Geschiedenis van het nederlandsch Genootschap tot zedelijke Verbetering der Gevangenenden 1823-1923*": He was married (1. Marriage July 12, 1922) with Clara Adriana Petronella Kluit, they

had four sons. Second marriage (May 23, 1966) with Johanna Maria Leendern, no children. Jacob Maarten van Bemmelen was sometimes editor of the journals "Tijdschrift voor Strafrecht", "Weekblad van het Recht" and "Nederlands Juristenblad". Later his interests were soldier laws. During the second world war he joined the Netherland Council of War in Den Haag. Later he became editor (1946-1955) of the journal "Militair-Rechtelijk Tijdschrift" and Vice-president of the Association Internationale de Droit Pénal and the International Society of Criminology (VEEN, 1994).

**Table of the chronology of the early experimental age
of adsorption science**

(ROBENS, 1992, BENEKE 1995c, DĄBROWSI, 2001)

Date	Explorer/Land	Significance
5000 BC	Cyprus	Washing of woollen cloth with fuller's earth was certainly practised in Cyprus (BENEKE AND LAGALY, 2002).
3750 BC	Egyptians and Sumerians	Use of charcoal for reduction of copper, zinc and tin ores for manufacture of bronze.
1550 BC	Egyptians	Application of charcoal for medicinal purposes to adsorb odorous vapours from putrefactive wounds and from intestine.
460 BC	Hippocrates (at 460 BC Kos-at 370 BC Larissa Pliny the elder (24 Comun - 79 Stabiae)	introduced the use of charcoal to treat a wide range of affections including epilepsy, chlorolysis and anthrax
460 BC	Phoenicians	First recorded application of charcoal filters for purification of drinking water.
157 AD	Claudius Galen (129 Pergamon - 199 Rom)	introduced the use of carbons of both vegetable and animal origin to treat a wide range of complaints.
17th century		Deposits of fuller's earth were founded in England. The fuller's earth was not only used for fulling cloths but also in pharmaceutical and cosmetic applications in form of powders and ointments (BENEKE AND LAGALY, 2002).
1773 1777	Carl Wilhelm Scheele (19.12.1742 Stralsund - 21.05. 1786 Köping) Félice Fontana (15.04.1730 Pomarolo - 10.03.1805 Florence)	reported some experiments of the uptake of gasses by charcoal and clays derived from various sources (SCHEELE, 1777; FONTANA, 1777).
1786, 1788	Johann Tobias Lowitz (25.04.1757 Göttingen- 07.12.1804 St. Petersburg)	awarded the antiseptic effect in powder form charcoal. Used charcoal for decolorization of tartaric acid solutions (LOWITZ, 1786).
1793	D. M. Kehl	discussed helpfulness of charcoal for removal of odours from gangrenous ulcer and applied carbons of animal origin for removal of colours from sugar (KEHL, 1793).
1794	England	Charcoal was used in the sugar industry in England as a decolorization agent of cane sugar syrups.

1808	France	Charcoal was used in the sugar industry in France as a decolorization agent of beet sugar syrups.
1814	Nicolas Théodore de Saussure (04.10.1767 Geneva - 18.04.1845 Geneva)	started systematic studies of adsorption of various gases by porous substances such as sea-foam, cork, charcoal, and asbestos. He discovered the exothermic character of adsorption processes (DE SAUSSURE 1814, 1815).
Since 1828		New methods were found for reactivation of required charcoal. Other commodities (blood, cereals, tar, turf, waste paper) were found to produce charcoal.
1850	Friedlieb Ferdinand Runge (08.02.1795 Billwärder, now Hamburg - 25.03.1867 Oranienburg near Berlin)	adsorptive separation of chemicals on filter paper by capillary force (Runge-pictures, later paper-chromatography) (RUNGE, 1850 a, b).
1865	J. Hunter	reported the high sorption capacity of charcoal made from coconuts (HUNTER, 1865).
1871	W. Thomson	reported by the adsorption of gases and vapours in alumina that the adsorption will be a factor of the pore volume and pore diameter (THOMSON, 1871).
1881	Heinrich Gustav Kayser (16.03.1853 Bingen - 14.10.1940 Bonn)	introduced the terms 'adsorption', 'isotherm' or 'isotherm curve'. He also developed some theoretical concepts that became basic to monomolecular adsorption theory (KAYSER, 1881a,b).
1879, 1883	P. Chappuis	made the first calorimetric measurements of the heat of wetting of various carbons by liquids (CHAPPUIS, 1879, 1881, 1883).
1878	USA	English fuller's earth was used for bleaching vegetable oils and became the most important fuller's earth on the European and American market. Some time before an American traveller in the Middle East had observed that people used clay to lighten olive oil (BENEKE AND LAGALY, 2002).
Since 1890	USA	Great deposits of fuller's earth were found in Arkansas, Florida, Montana, Wyoming (BENEKE AND LAGALY, 2002).
1901	R. von Ostreyko	set the basis for commercial development of activated carbons through processes that involve the incorporation of metallic chlorides with carbonaceous materials before carbonization and the mild oxidation

		of charred materials with carbon dioxide or steam at increased temperatures (Patented) (VON OSTREYKO, 1901.)
1903	Michail Semjonowitsch Tswett (19.05.1872 Asti - 26.06.1919 Woronesch)	discovered the phenomenon of selective adsorption during separation of chlorophyl and other plant pigments by means of silica materials. He introduced the term 'column solid-liquid adsorption chromatography'. This discovery was not only the beginning of a new analytical technique, but also the origin of a new field of surface science. TSWETT, 1903, 1906a,b).
1904	Sir James Dewar (20.09.1842 Kincardine on Forth/Scotland - 27.03.1923 London)	found selective adsorption of oxygen from its mixture with nitrogen, during air uptake by charcoal (DEWAR, 1904, a,b).
1906/07	Bavaria	The production of acid activated clays (bleaching earth) started in Bavaria (BENEKE AND LAGALY, 2002).
1909	James William McBain (22.03.1882 Chatham/New Brunswick - 1953)	proposed the term 'absorption' to determine a much slower uptake of hydrogen by carbon than adsorption. He also proposed the term 'sorption' for both adsorption and absorption (MCBAIN, 1909).
1911	Netherland	The NORIT factory in Amsterdam was founded, and in the beginning produced charcoal from turf. It is now one of the most advanced international manufactures of active carbons
1911	Poland	A wood distillation plant was built in Hanjówka (East Poland), initially manufacturing active carbons solely from wood materials.
1914		World War I introduced the problem of protecting humans respiratory tracts from toxic warfare agents.
1915	Nikolai Dimitrievic Zelinsky (06.02.1861 Tiraspol - 31.07.1953 Moscow)	the first who suggested and applied the use of active carbons as the adsorption medium in gas masks.
1941	Archer John Porter Martin (01.03.1910 London) Richard L. M. Synge (28.10.1914 Liverpool - 18.08.1994 Norwich)	introduced into laboratory practice the solid-liquid partition chromatography, both in column and planar form (Nobel-Prize of chemistry in 1952).
1956	Richard Maling Barrer (16.10.1910 New Zealand - 12.09.1996 Chislehurst/Kent)	invented methods of zeolite synthesis. In this year the North-American Linde Company started production of synthetic zeolites at a commercial scale.

Acknowledgement

We thank Marjo C. Mittelmeijer-Hazeleger, Van 't Hoff Institute for Molecular Sciences, Nieuwe Achtergracht 166, 1018 WV Amsterdam, The Netherlands for later corrections of the literature list.

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Gedenkboek aangeboden aan Jakob Maarten van Bemmelen (1830-1910). Uitgegeven bij C. de Boer jr. Te Helder (1910)

Vorwort: Aan Jakob Maarten van Bemmelen den scherpzinnigen en onvermoeiden onderzoeker, den Nestor der Nederlandsche Scheikundigen, wordt dit Gedenkboek op zijn tachtigsten Verjaardag aangeboden door zijne Vrienden.