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Research Article

Friedlieb Ferdinand Runge (1794-1867) – An Unusual Chemist

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Abstract. This paper presents life and work of Friedlieb Ferdinand Runge, an unusual chemist, ignored for a long time. There are discussed his researches in natural products and coal tar, the discovery of caffeine and priority claims on the discovery of quinine. Important industrial activities at the Oranienburg factory are described, including the 'German guano'. Important issues are the discussion of alternative chemical theories by the anti-atomist Runge, as well as his occupation with the relation Chemistry-Art, expressed in his *Musterbilder*, also a forerunner of chromatography.

Keywords: History of Chemistry – Friedlieb F. Runge – Phytochemistry – Coal tar – Chemical Industry – Alternative chemical theories - Chemistry and Arts

Chemistry is the science of the components of our Earth
(F. F. Runge)

INTRODUCTION

Friedlieb Ferdinand Runge (1794-1867) was certainly one of the chemists with the most informal and unusual personality and behaviour of the 19th century, which gained him great popularity. Coming from Medicine to Chemistry, he worked and researched in several areas, from phytochemistry and tar, a productive industrial activity, the first data on what we now call chromatography. He wrote books in a simple but chemically correct language, to reach a wider audience and disseminate chemical knowledge, an activity that contributed to his popularity during his lifetime. Interestingly, soon after his death in 1867, he quickly fell into oblivion, despite the efforts of August Wilhelm Hofmann (1818-1892), then president of the German Chemical Society, to preserve his memory. What could have led to such a rapid decline? Informality and unorthodox behaviour for a member of the Academy certainly contributed, but I believe that his adherence to *Naturphilosophie*, useful in phytochemistry but detrimental to the development of Organic Chemistry as a whole, and his staunch anti-atomism, visible in his presentation of General Chemistry and Inorganic Chemistry, made it

difficult to work on an equal footing with the academic community of the time, represented for example by Liebig, Wöhler or Bunsen.



Figure 1 Friedlieb Ferdinand Runge (1794-1867), holding a glass of wine (perhaps his ‘synthetic wine’). Photo by an unknown author, the only real portrait of Runge, ca.1860. (Stiftung Preussischer Kulturbesitz, image in public domain).

In any case, given the important contributions to phytochemistry and the study of tar, the forgetfulness is surprising. Runge had few collaborators, and according to August Wilhelm Hofmann there seemed to be “something that put people off” about him, perhaps his idiosyncratic behaviour. In the international context, silence was established around Runge. For example, James R. Partington (1886-1965), in volume IV of his “History of Chemistry” (1962), dedicated a few lines to him, which we transcribe here:

“F. F. Runge was at first a pharmacist, then associate professor in Breslau (after a long residence in Paris), then in the Prussian Marine Service in Berlin and Oranienburg. He published several technological and other papers, also on the motion of electrically polarized mercury, and books. Runge rediscovered aniline in coal tar oil and called it cyanol, since it gave a blue color with bleaching powder, he found that it stained pine wood and elder pith yellow, and gave a greenish-black color when oxidized by a cupric salt (aniline black). In the

same research he discovered in coal tar oil another base leukol (quinoline), an acidic substance which he called carbolic acid (*Karbolsäure*, phenol), pyrrol (*pyrró* = fiery red), also rosolic acid, and three other bases. He did not analyse any of these substances”¹.

Aaron Ihde (1909-2000), in “Development of Modern Chemistry” (1984), gives him some space, related to research with atropine, tar and as a remote precursor of chromatography. More recent historians, or historians belonging to the Anglo-Saxon cultural context, such as William Brock (*1936), in “The Fontana History of Chemistry” (London, 1992), or Eduard Farber (1892-1969), in “Great Chemists” (New York, 1961), don’t even mention him.



Figure 2 Plaque allusive to Runge’s birthplace, 1936, on the Rectory building (Courtesy Billwälder Verein and Katja Haack, Hamburg)

Finally in 1994, at the bicentenary of Runge’s birth, the city of Oranienburg, where Runge carried out much of his activity as a chemist and where he lived and died, decided to pay him a double tribute. In front of the building attached to the Oranienburg Palace, where Runge had his laboratory, was erected a bronze statue, designed by Stephan Möller (*1954); and the *Stiftung Preussischer Seehandel* established the Friedlieb Ferdinand Runge Prize, for “Unconventional Forms of Art Transmission” (very much to Runge’s taste...). In fact, in 1863, through August Wilhelm Hofmann, the London Industrial Exhibition awarded Runge a medal. To the messenger who brought it to his home, Runge responded with Friedrich Schiller’s quote, “You arrive late – but you arrive!” (from the play “Wallenstein”).

Runge’s basic biographies are those by Hermann Schelenz (1848-1922), pharmacy historian, “Friedlieb Ferdinand Runge” (1907); by Max Rehberg (1882-1945),

“Friedlieb Ferdinand Runge, Entdecker der Teerfarben” (1935), both hagiographic, to be consulted *cum grano salis*, and by Berthold Anft, “Friedlieb Ferdinand Runge – sein Leben und Werk” (1937, reprint 1977). A recent biography is “Friedlieb Ferdinand Runge, sein Leben, sein Werk und die Chemische Produkten-Fabrik Oranienburg”, by Christa Niedobitek and Fred Niedobitek (2011).



Figure 3 Parish Rectory in Billwärder, Hamburg, erected in 1840, in substitution of the old Rectory, where Runge was born in 1794 (Courtesy Billwärder Verein and Katja Haack, Hamburg)

ORIGIN AND STUDIES

Friedlieb Ferdinand Runge was born on February 8, 1794 (not 1795, as it is sometimes read; the origin of the confusion is the baptismal certificate) in Billwerder, a village southeast of Hamburg, the third of the seven children of the Lutheran pastor Johann Georg Runge. Billwerder was an ancient rural community, known since 1150; incorporated into Hamburg, it maintained its rural character. During the Napoleonic Wars, Hamburg was occupied by the French (1806/1811), and during the administration of Marshal Louis-Nicolas Davout (1770-1823) the city's population decreased from 100,000 to 55,000 residents. Pastor Runge's family also experienced difficulties, and Friedlieb was unable to attend a renowned school, such as the famous *Johanneum* in Hamburg, founded in 1529 by the reformer Johannes Bugenhagen (1485-1558). So, he went to study at Schiffbeck elementary school. The Billwerder rectory was demolished in 1840 and replaced by a more solid building, where in 1936 a bronze plaque was placed commemorating the birth there of the chemist Runge (figures 2 and 3).

From 1810 to 1812 he learned pharmacy from an uncle at the *Ratsapotheke* in Lübeck, the oldest municipi-

pal pharmacy in Germany (founded in 1412). There he carried out his first experiments and had his first contact with henbane (*Hyoscyamus niger* L.), which among other alkaloids contains atropine, so important to him in his future activities. The obvious path for Friedlieb was the study of Medicine, from 1816 to 1818 at the University of Berlin. In 1818 he transferred to the University of Göttingen, where Friedrich Stromeyer (1776-1836), professor of Chemistry, convinced him to study Chemistry. A new transfer, still in 1818, to the University of Jena, continued his interest in Chemistry, taught there by Johann Wolfgang Döbereiner (1780-1849), Goethe's scientific advisor. He also attended lectures on *Naturphilosophie* by Lorenz Oken (1779-1851), a disciple of Friedrich Wilhelm von Schelling (1775-1854). He received his doctorate in 1819 and returned to Berlin for a second doctorate, aiming to work as a *Privatdozent* (1822), being approved by a panel composed of Hermbstädt, Weiss and Hegel.

After a short stay at university, he traveled in Europe from 1823 to 1826: in Paris he met Alexander von Humboldt and Liebig, in France and Switzerland he visited industrial facilities, in England and the Netherlands agricultural activities. Coming back from the trip, his *peregrinatio academica*, he returned to Breslau (since 1945 the Polish city of Wrocław), where he had already settled in 1823. He served there as *Privatdozent*, and in 1828 was appointed permanent professor of Technological Chemistry at the University of Breslau, where he remained until 1831, when he returned to Berlin. In Breslau he met again his friend, the poet and professor August Heinrich Hoffmann von Fallersleben (1798-1874), who had founded the “*Zwecklose Gesellschaft*” (Society without Purpose), and joined the “*Schlesische Gesellschaft für Vaterländische Kultur*” (Silesian Society for National Culture), founded in 1803, where he gave lectures on chemical and technological topics for pharmacists, students and workers. In the spirit of the intellectual formation in force in the 18th and early 19th centuries, Runge had his formal education at the university and his *peregrinatio academica*.

RUNGE AND PHYTOCHEMISTRY

In 1820 Runge published the book “*Neueste phytochemische Entdeckungen zur Begründung einer wissenschaftlichen Phytochemie*”² (Newest phytochemical discoveries for a scientific Phytochemistry), in which he presented the observations (but not yet analyses) on alkaloids, that he had made in Jena, and in which he also presented the criteria that he believed were neces-

sary for a “scientific phytochemistry” (*phytos* = plant, the chemistry of plant constituents). It is difficult to say who was the first phytochemist, it is a subjective question that depends on who makes the choice, and on the criteria chosen for defining “phytochemistry”. There were Nicolas de Saussure (1767-1846), from Geneva, for the discovery of photosynthesis, the “mother reaction” of the entire plant universe; Friedrich Wilhelm Sertürner (1783-1841), for the first isolation of an alkaloid (morphine, in 1805); Pierre Joseph Pelletier (1788-1842), for the introduction of new solvent extraction methods, for the analysis of the “green matter” of plants (1817), and for the isolation, with Jean Bienaimé Caventou (1795-1877), of quinine (1820). Others prefer to wait for analyses and studies of groups of plant substances to be carried out, as those done by Friedrich Rochleder (1819-1874), a student of Liebig, or by Heinrich Hlasiwetz (1825-1875). But there is also the earlier study of extracts from medicinal plants, such as those by Johann Friedrich Cartheuser (1704-1777) at the University of Frankfurt/Oder, and even the isolation of thymol (1719) and camphor (1725) by Caspar Neumann (1683-1737).

Let's go back to Runge in Jena. In Döbereiner's laboratory he studied the extracts of the “three narcotics”³, the henbane (*Hyoscyamus niger* L.), the native *Bilsenkraut* (a variety of *Hyoscyamus*); the *Tollkirsche* (nightshade), the *Erba Belladonna* of the Venetians or the *Atropa belladonna* L. of Carl von Linné (1707-1778), whose toxic effect was already known by botanist Leonhart Fuchs (1501-1566), the reformer of Botany during the Scientific Revolution; and the thorn apple, devil's trumpet, or “witch fig”, *Datura stramonium* L., the *Gemeiner Stechapfel*. [the three plants contain, among others, the alkaloids atropine and hyoscyamine]. Henbane was already familiar to Runge, from the Lübeck pharmacy: when preparing a medicine, part of the plant's extract splashed into Runge's eyes, causing intense and long-lasting mydriasis, a dilation of the pupil caused by some drugs. In Jena, he repeated these experiments, now including *Belladonna*. Mydriasis caused by the extracts was tested on Runge's cat. (The curious effect has even had forensic use: if a suspicious liquid causes mydriasis in an animal, it contains the toxic alkaloid). Runge dealt with toxic substances so much that he was nicknamed *Dr. Gift*, poison doctor, a source of pride for him.

Goethe, who was interested in chemistry and science in general, had heard about mydriasis, and through Döbereiner, invited Runge to demonstrate the experiment at his home, in what would be one of the most delicious anecdotes in the history of Chemistry, mainly as narrated by Runge himself⁴. So, on October 3, 1819,

wearing borrowed clothes a little too big for him (which later became fashionable in Jena), with the cat under his arm, he set out on his way to Goethe's house (Goethe lived in Weimar, capital of the grand duchy, but in September and October 1819, returning from Karlsbad, he remained for some time in Jena). The demonstration and conversation were quite informal, and at the end Goethe gave the young chemist a box of coffee beans, perhaps he could find there the principle responsible for his insomnia. In fact, Runge isolated caffeine from coffee beans in 1819. Nervous when leaving Goethe's house, Runge forgot the cat: Goethe called him back, saying “you forgot your assistant”⁵. The visit was important for both: Goethe began to become interested in alkaloids, and Runge began his important and largely forgotten contributions to Chemistry. But the alkaloids atropine and hyoscyamine were not isolated by Runge, but, still in 1819, by the pharmacist Rudolph Brandes (1795-1842), also active in Döbereiner's laboratory.

From coffee beans, Runge extracted in 1819 the “*Kaffeebase*”, the “base of coffee”, that is, the alkaloid caffeine⁶, a discovery also claimed (1821), without reason, by Pelletier, Caventou and Pierre Jean Robiquet (1780-1840). Runge published the discovery of caffeine in 1819, in Lorenz Oken's (1779-1851) journal *Isis*⁷, and in 1820 in his own book on phytochemistry⁸. There is no doubt about the priority of Runge's discovery of caffeine. The formula for caffeine, $C_8H_{10}N_4O_2$, [in our notation] was only established in 1833 by Christoph Heinrich Pfaff (1773-1852) and Justus von Liebig (1803-1873).

Still in Jena, and still in Döbereiner's laboratory, Runge continued his phytochemical research, now with the bark of *Cinchona* from South America, *Cortex Peruvianum*, from which he extracted “*Chinabase*”, the “base of cinchona” (“base” is a substance of a basic, or alkaline, nature), that is, the alkaloid quinine, in 1819 (actually a mixture of alkaloids). The results were published in the same article in the journal *Isis*, still in 1819⁹. *Isis* was a respected scientific journal at the time, published from 1816 to 1848, first in Jena, then in Rudolstadt; the high standard of the journal is demonstrated by its collaborators, like Alexander von Humboldt, Georges Cuvier, Mme. De Staël; the excuse that Runge published in unknown or less qualified journals does not apply. The histories of Chemistry are practically unanimous in attributing the discovery of quinine to Pelletier and Caventou, in 1820¹⁰. Runge's *Chinabase* and the compound isolated by the French were later analysed and compared, finding that they were the same substance, and therefore, the priority in the discovery of quinine clearly lies with Runge, whose work had been published a year earlier than that of Pelletier and Caventou¹¹. Histo-

riographical revisionism? No, just the consistent application of a lesson of the great historian Marc Bloch (1886-1944): “The past is, by definition, a given that nothing can change. But knowledge of the past is something in progress, which is continually transformed and perfected”¹². Is Runge undoubtedly the ignored discoverer of quinine? The priority discussion about the discovery of quinine is indeed complex. Runge was one year in advance in the research on china bark. Using acetic acid as solvent, he obtained a basic extract, which was actually a mixture of alkaloids, which he was unable to separate, so he could not study its properties. He considered the extract useless for medical purposes, and went to search for the ‘true’ effective component of the bark. These results were published in Runge’s *Isis* paper (1819) and in his book on Phytochemistry (1820). At the same time, unaware of Runge’s work, Pelletier and Caventou obtained from Peruvian bark the same alkaloid, in a more pure form, the quinine. They were able to study its properties and its medical uses, and published their results in 1820. Pelletier was unaware of Runge’s researches, but historians from later times were not, and several authors suggested a more rigorous investigation about the priority of the discovery of quinine. In 1970, Wolfgang Schneider (1912-2007), professor in Braunschweig, and his coworker Horst Real, repeated Runge’s experiments, following exactly Runge’s recipe, obtaining the same results¹³, so that the priority question is not yet solved. In my opinion, Runge deserves the priority, using as criterium the first (later confirmed) published results. This is in accordance with Marc Bloch’s proposal mentioned above.

Another matter of priorities is the isolation of the dye glitter (*Krapp-purpur*), a less common component of madder (*Rubia tinctorium L.*), a discovery generally attributed to Robiquet and Jean-Jacques Colin (1784-1865), as *purpurin*, in 1826, along with alizarin¹⁴. Runge had already dealt with madder before. Unfortunately for him – academically and financially – the discovery of alizarin escaped him, but there is no doubt about the discovery of glitter, for which he applied for a patent in 1822, granted in 1823. But the official historiography is irreducible.

In Anft’s opinion, Runge’s experiments on natural products strictly followed the precepts of *Naturphilosophie*, but Runge also considered the purely empirical data of his experiments. Adherence to *Naturphilosophie*, the “pest of the sciences” in Liebig’s words, contributed to Runge’s oblivion. Another issue to determine is the almost systematic refusal to recognize its proven priority in some discoveries, such as quinine and purpurin. Many of his articles received heavy criticism at the time of their publication.

At the beginning of his treatise on phytochemistry as a new discipline¹⁵ Runge initially addresses mineral chemistry and divides it into an empirical part (essentially analysis), a mathematical part (stoichiometry) and a speculative part, yet to be studied in detail. He compares phytochemistry with mineral chemistry, saying that phytochemistry, as it deals with living matter, is mineral chemistry at a higher level. Until now, there was little knowledge of phytochemistry; mineral chemistry is taught in universities, and textbooks also deal with mineral chemistry. Phytochemistry is mentioned at a glance, mentioning extracts of some plants, generally still problematic. For its evolution, the publication of the work “The Development of Vegetable Substances” by Nees von Esenbeck [Theodor Nees von Esenbeck (1787-1837), professor in Bonn] was important. Next, considering the genesis of the plant as also the genesis of phytochemistry, it sets out a series of rules and principles to arrive at a “scientific” phytochemistry, discussing obtaining (extraction with solvents, precipitation), properties, reactions, etc.

For my part, I add that Organic Chemistry at the end of the 18th century and beginning of the 19th century is essentially chemistry of vegetable extracts, that is, phytochemistry: see for example the obtaining of carboxylic acids by extraction or precipitation, by Carl Wilhelm Scheele (1742-1786), or the quantity of compounds obtained from alcohol, such as the obtaining of ether described by Valerius Cordus (1515-1544), perhaps the first organic chemist.

RUNGE AND THE COAL TAR

After leaving his studies and Jena, Runge dedicated himself to a new field of research, in Berlin, Breslau and Oranienburg: coal tar. Some of the discoveries are pioneering, others are expansions of what was already known. The increasing use of mineral coal from 1830 onwards, for coking and obtaining lighting gas, left ammoniacal waters and coal tar as by-products, both sources of obtaining various substances. Several researchers began to focus on tar, including Runge, and this simultaneity would lead to controversies and disputes over priorities. It is discussed, for example, whether we can consider Runge as the first chemist to obtain artificial dyes, as early as 1834, which in the light of the writings we have is not the case¹⁶, although it cannot be denied that some of his pioneering experiments with tar provided elements for future dye syntheses. Judging by Runge’s own later writings (1866), he thought that with good will a dye industry based on his data would have

been possible. Biographer B. Anft (1957) is also of the opinion that Runge should be credited with starting an artificial coloring industry¹⁷. The obtaining by Runge, from coal tar, of phenol (carbolic acid, 1833), aniline (*Kyanol*, 1834), pyrrole (1834) and quinoline (*Leukol*, 1834) are undisputed facts. Runge's experimental procedures were systematic, involving, in addition to distillations, also extractions with acidic and basic solutions. Directly attacking the tar with solvents (water, alcohol, ethyl ether) and acid and basic solutions did not produce any results, which led Runge to move on to more drastic methods¹⁸. Dry distillation in a sand bath provided a distillate, which was then broken down into two fractions: a volatile liquid and a thick brown oil. The volatile liquid was subjected to new procedures: with $\text{Ca}(\text{OH})_2$ it forms a clear solution (a sign that the liquid was acidic), which when treated with HCl forms "carbolic acid" (our phenol); heating the clear solution formed by the addition of $\text{Ca}(\text{OH})_2$ gives rise to a dark red powder, from which Runge extracted rosolic acid (a synthetic dye, methylaurine) and "brunolic acid", a substance that even today chemists were unable to identify. Treating the volatile liquid itself with H_2SO_4 and then with NaOH leads to obtaining "*leukol*" (our quinoline, actually a mixture that Runge was unable to separate); The addition of calcium chloride (CaCl_2) to the volatile liquid gives rise to a blue solution, which with chlorinated lime forms "*Kyanol*" (our aniline), and by oxidation with dichromate a black compound (aniline black). The above description is very simplified, Runge's experimental procedure is extremely complicated, which in the opinion of August Wilhelm Hofmann, who subsequently dealt with this subject, discouraged other chemists from dealing with this topic. Runge did not analyse these compounds, which is why he is criticized, but at the time no one analysed the compounds he obtained, as analysis and laboratory methods in general were still poorly developed. The analysis of organic compounds became routine after the techniques introduced by Jean-Baptiste Dumas (1800-1884) and mainly by Justus von Liebig (1803-1873). The chemist Carl von Reichenbach (1788-1869), discoverer of creosote (1833) and paraffin (1830), harshly criticized Runge's discoveries, denying the existence of the four announced substances, to which Runge responded with elegance and conviction.

The four compounds obtained by Runge from coal tar were of great importance in the evolution of Organic Chemistry. In coal tar Runge rediscovered aniline, discovered in 1826 by Otto Unverdorben (1806-1873), a pharmacist in Dahme, near Berlin, by sublimation of indigo. Unverdorben called the compound "*Kristallin*"¹⁹. Tar was a much more abundant raw material, a reason

why Runge's rediscovery is important. Other chemists obtained aniline: Carl Julius von Fritzsche (1798-1871) in 1840, by heating indigo with alkali, naming the product "aniline" (from the Portuguese "*anil*"). In 1843, Russian chemist Nikolai Zinin (1812-1880) obtained aniline (which he called "*benzidam*") by reduction of nitrobenzene, a compound obtained by nitration of benzene, by Eilhard Mitscherlich (1796-1863) in 1835.



Figure 4 New building of the Preussische Seehandlung, 1904, on the Gendarmenmarkt, Berlin, by an unknown photographer. From "Atlas zur Zeitschrift für Bauwesen", 1904 (image in public domain)



Figure 5 Oranienburg Palace, in Oranienburg, built in the 17th century for the Great Elector's wife, Louise Henriette. In a lateral wing was installed Runge's chemical industry (Courtesy City Archive and Municipality of Oranienburg)

This whole subject was studied again in more detail by August Wilhelm von Hofmann (1818-1892), initially in his doctoral thesis (1841) with Justus von Liebig (1803-1873) in Giessen, a thesis whose theme was precisely aniline. As Liebig's assistant in 1843, Hofmann found that Unverdorben's *Kristallin*, Fritzsche's aniline

and Zinin's *benzidam* were the same compound, for which Hofmann chose the name "aniline". Hofmann himself developed a more effective method of obtaining aniline, by distillation. He also discovered that Runge's "*leukol*" was a mixture of quinoline, isoquinoline and quinaldine. In 1845 Hofmann published all these details in an article in the *Annalen*²⁰. From aniline he prepared some coloring principles, which leads most historians to consider Hofmann, and not Runge, as the precursor of the chemistry of synthetic dyes. It was Hofmann, then at the *Royal College of Chemistry* in London, who suggested to his student William Perkin (1838-1907) to try to synthesize quinine from aniline: the result was mauvein, frequently considered as the first artificial dye (1856).

In 1866, ten years after Perkin's discovery, Runge commented on the discovery of artificial dyes: "Finally Dr. A. W. Hofmann arrived and showed in his article [...] that all my data on this dye were absolutely true. With that, I once again dedicated myself to this almost forgotten subject..." Runge proposed in 1847 to the state factory in Oranienburg (see below) the manufacture of artificial dyes, "but my efforts failed because of the opinion of an official unfamiliar with the subject"²¹ (financial director E. Cochius).

RUNGE AND THE CHEMICAL INDUSTRY

Runge carried out his experiments on coal tar while he was chemical director (1832/1852) of the chemical factory in Oranienburg, a city located northeast of Berlin. The factory was part of the *Preussische Seehandlung* (figure 4), a state-owned company founded in 1772 by King Frederick II the Great (1712-1786), to develop industrial and commercial activities. The chemical factory was housed in Oranienburg Palace (figure 5), built in 1651 by Frederick William the Great Elector (1620-1688) for his wife Louise Henriette (1627-1667), a princess of the house of Orange-Nassau (hence Oranienburg), expanded by King Frederick I (1657-1713), but later decommissioned as a royal residence and sold in 1802 to pharmacist Johann Gottfried Hempel (1752-1817). Hempel and his son Georg established a weaving factory there and in 1814 the chemical factory, producing mainly sulfuric acid using the lead chamber process, transferred in 1848 to a new location²². In the 19th century, the palace was partially restored as a royal residence by King Frederick William IV (1795-1861) and is now a museum. The poet and novelist Theodor Fontane (1819-1898) wrote about the factory and the palace: "The vapors of sulfuric acid corroded and deteriorated and

swept away the last traces of the past beauty. I remember when I was a child, when I came this way, and from the square and the bridge I looked with fear at the old and terrifying building, immersed in smoke and ash"²³.

The *Preussische Seehandlung* went through difficulties in the Napoleonic period, but recovered with the efficient administration of Christian von Rother (1778-1848), maintaining textile factories (Breslau, Glatz), metallurgical factories (Berlin, Breslau, Dirschau), paper factories (Bromberg), zinc (Ohlau), and since 1841 the Oranienburg factory; the *Seehandlung* was one of the first European companies to trade with independent South America: in 1822 a ship left for Rio de Janeiro, taking textile products and returning with coffee, sugar and cotton.

The period of Rother's administration corresponds to the period in which Runge was chemical director, from 1832 to 1852. The factory began to produce, in addition to sulfuric acid and *oleum*, stearin (1833) and paraffin (1834) candles, soaps, soda, ammonium salts, Glauber's salt, Prussian blue. Runge's proposal to attempt the manufacture of dyes (1847) was not accepted by the factory management. For Andreas, the factory did not have the necessary equipment. In the opinion of Richard Anschütz (1852-1937), Kekulé's successor and biographer, the initiative would have been possible²⁴.

Table 1 Runge's table of 'Mischgewichte'. Illustration from 'Einleitung in die Technische Chemie für Jedermann', Berlin, 1836, on page 16

1	Wasserstoff.	32,2	Zink.
6	Kohlenstoff.	35,4	Chlor.
8	Sauerstoff.	37,6	Arsenik.
8	Silicium.	39,2	Kalium.
9	Alumium.	40	Selen.
12,5	Magnium.	44	Strontium.
14	Stickstoff.	48	Platin.
16	Schwefel.	56	Kadmium.
16	Phosphor.	59	Zinn.
18,6	Fluor.	64,5	Antimon.
20,5	Calcium.	66	Gold.
21	Bor.	68,6	Barium.
23,3	Natrium.	71	Wismuth.
27	Eisen.	78,4	Brom.
28	Chrom.	101	Quecksilber.
28,5	Mangan.	104	Blei.
29,5	Nickel.	108	Silber.
29,5	Kobalt.	217	Uran.
32	Kupfer.		

From 1844 onwards the company experienced financial difficulties, and Runge feared the privatization of the factory in the course of the liberal movements of 1848. In fact, the *Seehandlung* sold the factory in 1850 to Ernst Eduard Cochius, who dismissed Runge in 1852, but was unable to avoid the bankruptcy of the enterprise. Cochius committed suicide in 1856. Even without an official connection with the factory, Runge continued to provide his assistance, maintaining his salary and a modest home.

In 1853/1855 Runge invented a substitute for saltpeter from Chile and guano from Peru, the “German guano”, rejected “because it was not natural” (in the words of Cochius, it is “a criminal arrogance to want to produce bird excrement in the laboratory”)²⁵ which led Runge to sell the process to the ministry of war. Runge explicitly refers to competition with England in the trade of Peruvian guano. He used recycled products to obtain the fertilizer, which in essence was ammonium phosphate combined with ammonium sulfate. A friend of Runge said: “In Oranienburg there must be a guano island!”²⁶ Offended by Runge’s initiative, Cochius’s widow expelled him from his palace residence and cut off the pension he received. Runge went to live in Oranienburg in a more than modest way.

RUNGE AND THEORY²⁷

It is a widespread opinion that Runge had no theoretical interests, which is not correct. We will look at his publications later, for now let us limit ourselves to analysing his theoretical points of view expressed in the “*Einleitung in die Technische Chemie*”, from 1836 (Introduction to Technological Chemistry) (figures 6 and 9). It is likely that Runge’s theoretical work did not have greater repercussion because of his anti-atomism and his adherence to *Naturphilosophie*, already rejected by the majority of chemists. Runge does not consider his conceptions as “theory”, but as the result of his experiments. He prefers a “dynamic chemistry” to atomism, and chemical compounds would be formed by mutual “interpenetration” (*Durchdringung*) of substances with opposite properties, which presupposes the divisibility of matter “to infinity”. From this *Durchdringung* emerges a “third compound”.

Runge knew 54 elements (Table 1), which he classified into three groups: the elements of the “oxygen series” (O, S, Se, Te, Cl, Br, I, P), roughly our non-metals; the metals; the metalloids (H, C, B, N). In compounds, metals can be replaced by others, which is a criterion for including them in a group. Runge classifies metals as

light (Na, K, Mg, Ca, Sr, Ba, Al, etc.) and heavy (Fe, Ni, Co, Ag, Au, Tt, Mn, U etc.) and observes that in many aspects H is close to metals. It is also true in Chemistry that different attract each other and similars repel each other, but this is in the presence of a metal. Without the metal, elements from the oxygen group can also combine with each other. The concept of metalloid used by Runge differs from the concept of metalloid common at his time²⁸.

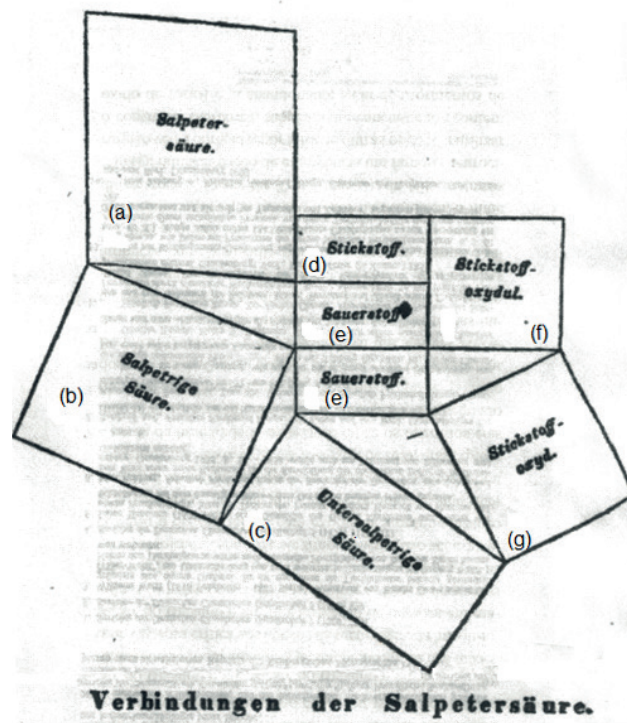


Figure 6 Compounds derived from Nitric Acid. Illustration from “*Einleitung in die Technische Chemie*”, Berlin, 1836, on page 338. (a) nitric acid; (b) nitrous acid; (c) hyponitrous acid $H_2N_2O_2$; (d) nitrogen (e) oxygen; (f) N_2O ; (g) NO_2 .

Klaus Röker observes that with his “dynamic chemistry” Runge independently arrived at the laws of constant proportions (Proust) and multiple proportions. To obtain a chemical compound by the interpenetration of substances with opposite properties, a certain “*quantum*” [term used by Runge] of properties of one of the substances must be canceled by an equal “*quantum*” of opposite properties of the other substance. Substances therefore combine according to numerical proportions determined by their “chemical activity” (*Chemische Wirksamkeit*). The greater the “chemical activity”, the smaller the numerical value involved in these proportions, and the weaker the activity, the greater the value

of this number: hydrogen, the lightest substance, therefore has stronger chemical activity (*Chemische Wirksamkeit*), uranium, the heaviest substance then known, the weakest activity.

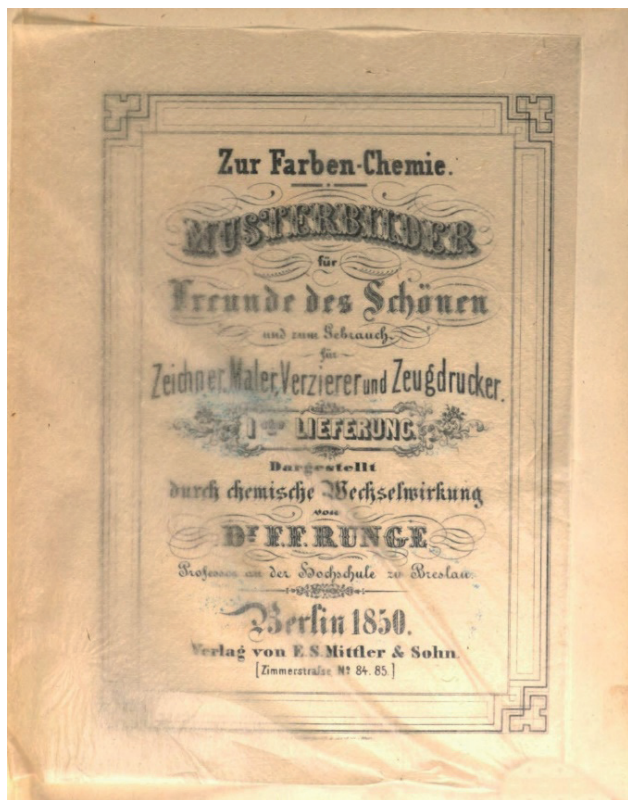


Figure 7 F. F. Runge, “Musterbilder”, cover, F. Mittler & Son, Berlin, 1850. (image in public domain).

Thus, to reach balance in the interpenetration of two substances, numerical values must be assigned, the *Mischungsgewichte* (= something like interaction weights), or *Chemischer Wert* (= chemical value), and Runge assigned numerical values to the 54 simple substances (= elements) then known. Hydrogen has the value 1, uranium has the value 217. In this system, the values 6 were assigned to carbon, 8 to oxygen, 14 to nitrogen. Therefore, to completely cancel out the opposing proportions of hydrogen and oxygen, 8 pounds [unit used by Runge] of oxygen interact with 1 pound of hydrogen, forming 9 pounds of water or ice²⁹. Runge created a table of *Mischungsgewichte*³⁰, and, if he were an atomist, we would say that he created a table of atomic weights (see Table 1). He wrote about combinations (Runge used the expressions ‘mixture’ and ‘combination’ interchangeably): “In words, this relationship means nothing more than saying that the number that precedes

each substance represents the numerical relationship with which it participates in a chemical combination. It is unalterable and can only vary in the sense of doubling, tripling, etc. Because of this, all oxygenated substances contain oxygen in proportions of 8 or 16 or 24 or 32 or 40 and so on”³¹. A comment not only historiographical, but even epistemological is in order here: being anti-atomist, Runge arrived, based only on empirical data and concepts consistent with *Naturphilosophie*, and without making use of the concept of quantitative atom, to conclusions equal to those reached by Dalton and others with the application of atomic theory and its consequences. The evolution of the theory is later than Dalton’s, and much more complicated than it would be with the use of the concept of ‘atom’. In any case, there are often several conceptually different ways to explain certain theoretical situations. In Runge’s case, as the specific situations to be explained become more complicated, the theory cannot satisfactorily explain what happens, which led to its abandonment and oblivion. But its role in the evolution of theories involving formulas, equivalents, atomic weights, ‘chemical value’ and so on, remains important. I believe it can be said that there is a certain analogy with the abandonment of the phlogiston theory: there was a need for more and more *ad hoc* hypotheses, so that the system became impractical, and it was abandoned. In any case, with respect to atoms, molecules, equivalents etc., there would only be some order after the Karlsruhe Congress in 1860.

Runge also presented some other theoretical ideas. Combinations can be of various “orders”. “First order” combinations involve only two substances, but there can be different stoichiometries, as in the combinations between N and O (N_2O , NO, NO_2 , N_2O_3 , N_2O_5). In “higher order” combinations, with three or more substances, the ‘construction’ of substances occurs in stages³².

He also distinguishes three classes of combined compounds³³:

- true acids, which always behave like acids (sulfuric acid, nitric acid, hydrochloric acid).
- true bases, which always behave like bases (soda, potash, ammonia).
- “acidic bases” (our amphoteric acids), which behave either as an acid, or as a base, depending on the other substance present (alumina, zinc oxide, tin oxide, lead oxide).

With an ingenious graphical representation method Runge represented the “oxygenation stages”. Figure 6 shows the case of nitrogen³⁴, and Runge developed such graphical representations of the “oxygenation stages” of several other elements, like, manganese, iron, lead, cop-

per, tin and others. Substances (or elements) are represented by rectangles on the right of the diagram, compounds by rectangles arranged at an angle, around internal triangles.

Other theoretical subjects were addressed by Runge, such as Elective Affinities, but nothing new or different was added.

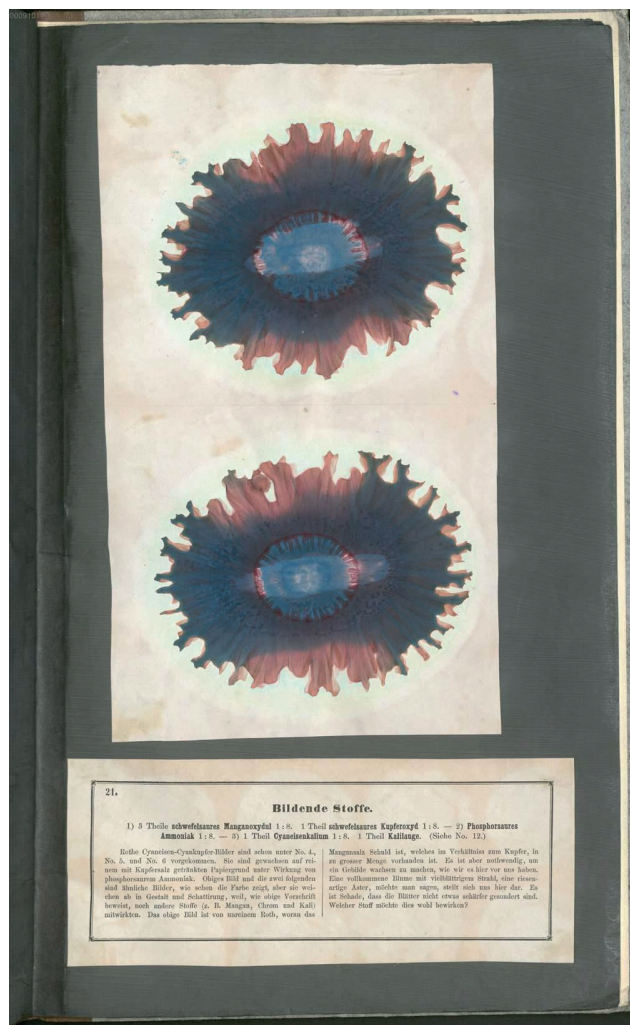


Figure 8 An example of a 'Musterbild', page 21. (1) reaction of manganese sulfate with copper sulfate. (2) reaction of ammonium phosphate with potassium ferrocyanide (State Library of Bavaria, Munich) (image in public domain)

RUNGE, THE MUSTERBILDER, AND THE BEGINNINGS OF CHROMATOGRAPHY

That Science creates Art is nothing new, and Runge's "Musterbilder" or *Professorenklexe* are an eloquent example. The absorption on paper of the product of two react-

ing solutions forms "self-forming" designs, which resulted in Runge's famous book (figure 8), and the procedure can be considered a precursor to chromatography. In an 1836 book by the Englishman George Field (1777-1854) the word "chromatography" already appears. And there is a record from 1731, in "A treatise on colors", by a certain Taylor (1685-1731), watercolourist, which contains the word "chromatography", from the Latinized Greek words 'chromato' (= color) and 'graphein' (= to write).

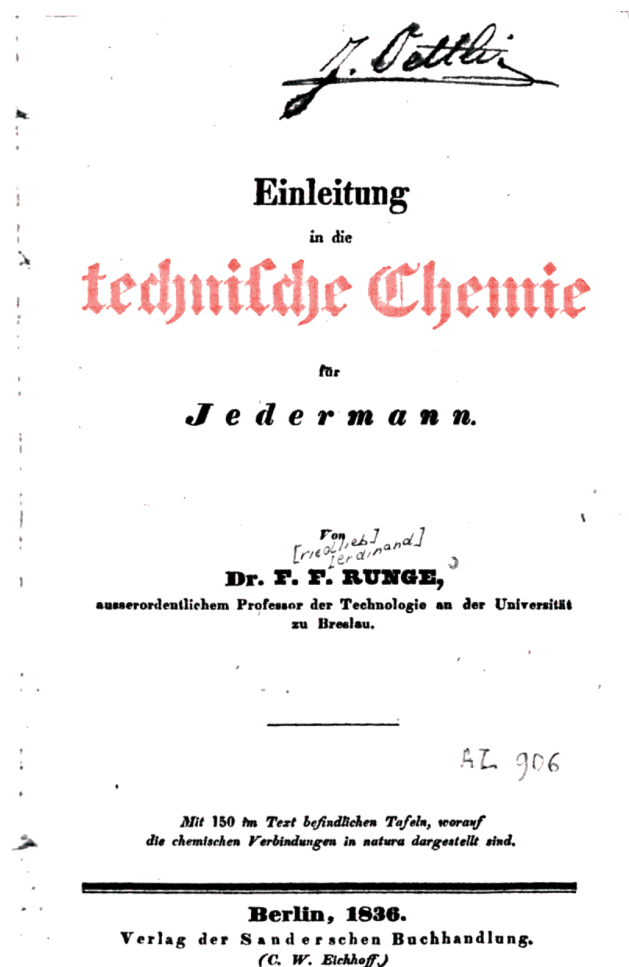


Figure 9 Cover of Runge's "Einleitung zur Technischen Chemie für Jedermann", published by Sandersche Buchhandlung, Berlin, 1836. (public domain)

There is more or less consensus that the invention of absorption chromatography should be credited (there are dissenting voices) to the Russian botanist Michail Tswett (1872-1919). For many historians there are precursors,

such as the Englishman Lester Reed (1860-1926), in Beneke's opinion, or the American David Talbot Day (1889-1925), for Aaron Ihde³⁵. But both Lester and Day limited themselves to describing some simple cases of separation, they never worried about the theoretical aspects of this separation technique. The 'capillary analysis' of the Swiss Friedrich Goppelsröder (1837-1919) can also be considered a precursor to chromatography. And there are historians who go back even further, considering that the *spot tests* that Runge carried out in 1822 on fabrics and on paper, with the purpose of observing the behavior of dyes, would be the beginnings of empirical chromatography³⁶.



Figure 10 Runge's house in Oranienburg. He lived there from 1856 until his death. The house was demolished in the thirties (Courtesy City Archives and the Municipality of Oranienburg)

Runge's "natural creations" and his "*Musterbilder*" resemble a chromatogram *avant la lettre*, they had nothing to do with processes of separation, but rather with an incursion into aesthetics, into works of art "created by nature", spontaneously, and by the self-organizing force of Nature. Runge wrote about it: "What, then, are these figures? They are natural creations, which arise as a result of chemical reactions. The simple narrative of how I arrived at them will clarify to the reader the story of their creation"³⁷. These spontaneous "natural creations" are formed as follows: paper impregnated with copper sulfate (blue) and potassium ferrocyanide solution ($K_4[Fe(CN)_6]$), mixed with $(NH_3)_2H_2PO_4$ as a developer, form a stain with a red core and green edges; a solution of manganese sulfate (pink) reacting with a solution of potassium chromate ($K_2Cr_2O_4$), and ammonia as a developer, give rise to a blue stain with a brown border³⁸. Runge explains that by repeating these experiments exactly, using the same type of paper, the same salts in the same concentrations, and dripping the solutions at the same speed and in the same sequence,

any researcher will obtain as many images as he wants. "Natural creation" is really science: the different speed of adsorption of the different colored solutions by the paper corresponds to what happens in chromatography, in the solid stationary phase.

Incredible as it may seem, Runge's interest in the spontaneous formation of these figures is not the chemical aspects involved, but the chemical-aesthetic aspects (although the experiments originate from his occupation with dyes). Something difficult to imagine at a time when Chemistry more and more takes on the characteristics of an exact science, but Runge's personality was eccentric enough to get involved with such experiments that for him were more aesthetic than chemical. In the year 1850, Runge chose 126 of these colorful figures and published the book "*Zur Farbenchemie. Musterbilder für Freunde des Schönen und zum Gebrauch für Zeichner, Maler, Verzierer und Zeugdrucker*", (figure 7) dedicated to the King of Prussia, Frederick William IV (1795-1861). The king personally thanked his somewhat exotic subject. In 1855, Runge published a second volume on the subject, "*Der Bildungstrieb der Stoffe. Veranschaulicht in selbständig gewachsenen Bildern*"³⁹.

Runge highlights the aesthetic aspect of these figures, their importance for the arts. He was convinced that his *Musterbilder* would be useful to painters and designers, for example, with new colors and new color combinations. Runge himself said: "Would we condemn Chemistry, if it, with more pride than Michelangelo, would exclaim: also *io sono pittore!*, why Chemistry is a painter, without a brush?". Today's idea of self-organization was expressed early on by Runge, when he stated that his still wet drawings are alive, as they still grow!⁴⁰

Let's see what Runge himself writes about his *Musterbilder*: "Whoever carefully observes the various figures in this book will soon clearly perceive that they could not have been painted with a brush. The so peculiar blurring and shading show that there cannot be here the arbitrariness practiced by the brush. The same goes for different colors, which can be produced by some arbitrary combination [...] The colors here are separate and not separate; they interpenetrate equally in separation and interpenetration. Something like this can only develop naturally and from within. What are these figures? They are natural formations, which are formed by chemical interactions"⁴¹. And further on: "Here a new world of formations, structures and color combinations suddenly appears, which I would never have imagined and which were not predictable, which surprised me even more. I soon learned the conditions for reproducing them in any quantity. Determining these aspects was particularly important for me, because this discovery, in

addition to its chemical value, acquires importance for the plastic arts [...]”⁴². Without a doubt, a subject suitable for an eccentric and unorthodox chemist.



Figure 11 Friedlieb Ferdinand Runge (1794-1867), bronze statue by Stephan Möller (*1956), erected 1994 in Oranienburg, in front of Runge’s laboratory, near the Palace (Courtesy City Archives and the Municipality of Oranienburg)

This item cannot be concluded without referring to an analogy between the *Bildungstrieb*, the driving force for the formation of substances, proposed by Runge, and the *Od* proposed by Karl von Reichenbach (1799-1869), a “hypothetical force that permeates all of Nature”, responsible for phenomena such as hypnotism, magnetism, light and others. This very little known analogy was suggested by Leslie Etre (1922-2010) and H. Bussemas, historians of chromatography. Runge wrote on the subject “*Das Od als Bildungstrieb der Stoffe*” (Oranienburg, 1866), a copy of which survives in the Yale University Library⁴³. The peculiarity of this book also suggests a philosophical basis common to Runge and Reichenbach, in this case, linked to *Naturphilosophie*. Interestingly, Runge and Reichenbach had a strong personal friction in the 1830s, when both were researching coal tar, with

dissenting points of view⁴⁴. The Yale book may contain other secrets.

The subject of the *Musterbilder*, however, soon fell into oblivion, but recently, a more open view of Chemistry has returned to value these curious experiments by an unique chemist. Runge already valued non-chemical subjects from the beginning of his professional activity. In 1826, in Breslau, where he was a professor, he participated in the *Zwecklose Gesellschaft*, ‘Society without Purpose’, with his friend the poet August Heinrich Hoffmann von Fallersleben (1798-1874), who visited him twice in Oranienburg, to which also belonged painters (Carl Bräuer [1798-1874]), musicians (Immanuel Sauer- mann, Carl Schwindt), sculptors (Mächtigt), the philologist Wilhelm Wackernagel (1806-1869), the industrialist Karl August Milde (1805-1851), in whose fabrics factory in Breslau Runge tested his dyes, and with whom he traveled through France, Switzerland and Great Britain, to visit industrial facilities. The Society met on Saturday nights, to read poems and other texts, to discuss cultural aspects, and ephemerides⁴⁵.

PUBLICATIONS

Unlike most chemists of his time, Runge published little in the scientific journals then in vogue, mainly his research on phytochemistry and coal tar derivatives, in *Isis*, in Liebig’s *Annalen*, in *Poggendorffs Annalen*, all of them journals with wide circulation at the time. It cannot, therefore, be said that Runge was and is little cited and remembered because he published in little-known journals.

On the other hand, Runge published many books on Chemistry, both theoretical (as we have seen, without considering atomic theory) and practical. They were aimed not at the scientist, but rather at the common reader, the industrialist, the merchant, the practical chemist, which is why the practical chemist, which is why the most important series of these texts was called “*Jedermann-Chemie*”, something like “Chemistry for everyone” (the translation of the German word *Jedermann*, literally ‘anyone’, presents many subtleties). Runge himself explained his objective as follows: “[it is] a Chemistry of general understanding, but rigorously scientific, intended mainly for the trader, the manufacturer, the practical chemist, to be at their disposal as a faithful advisor”. He intentionally gives up many chemical facts, which are less cited, but which can be found in Chemistry treatises. He also finds it unnecessary to present the physical properties of chemical compounds in great detail⁴⁶.

The first book published by Runge with this conception was “*Grundlagen der Chemie für Jedermann*” (Fundamentals of Chemistry for everyone), in 1830. We have already seen the theoretical bases on which Runge based his book. This was followed by “*Einleitung in die Technische Chemie für Jedermann*” (Introduction to the Technical Chemistry for everyone) in 1836, and “*Technische Chemie der Nützlichen Metalle für Jedermann*” (Technical Chemistry of Useful Metals for everyone), in 1838/1839. Unlike the first, these last two books are abundantly illustrated. In 1844, the crown prince and future King Maximilian II of Bavaria (1811-1864) ordered from Runge more than 7000 copies of a book in the “*Jedermann*” style, the “*Grundriss der Chemie für Jedermann*” (1846, Outline of Chemistry for everyone), to be distributed in all schools in Bavaria. He also published, in 1834, “*Farbenchemie. Die Kunst zu Färben*” (Chemistry of Colors. The Art of dyeing), supplemented in 1842 by “*Farbenchemie. Die Kunst zu Drucken*” (Chemistry of colors; The Art of printing). In 1839, he published the translation into German of “Conversations on Chemistry”, by Jane Marcet (1769-1858), a popular text much to his liking⁴⁷.

In addition to the aforementioned “*Zur Farbenchemie. Musterbilder für Freunde des Schönen und zum Gebrauch für Zeichner, Maler, Verzierer und Zeugdrucker*” (1850), and a second volume on the subject, “*Der Bildungstrieb der Stoffe*” (1855), and “*Deutscher Guano*” (1858), Runge published in 1866 “*Hauswirtschaftliche Briefe*” (Domestic Letters), in which he addressed several issues, including those relating to his personal life (such as the visit made to Goethe on October 3, 1819, commented above) and the recipe for making his “synthetic wine”. This “synthetic wine” was an alcoholic solution of citric acid, with the addition of various flavoring ingredients, which he kept secret.

With his *Jedermann* books, we can conclude today, Runge had in mind a new educational proposal. In the Preface on his “*Einleitung in die Technische Chemie*” (1836) he wrote: “The reglements of the *Schulmeister* (school teacher) are past. Even this submission no more exists in our times, which became free. To be educated is no more the same as to know the old classics. Finally we all are convinced that the knowledge of God’s creations is more important than the knowledge of the human creations; that it is better to learn about Nature and all its connections with life, than to learn the dead languages of former times, related to our times only through the thread of History”⁴⁸. Thus, Runge proposes an education system in which Mathematics and Sciences exert a central role, and should be the nucleus of an educational system; even at the universities, Medicine

and Law students should be confronted with physical and chemical lectures. Almost every activity in modern times has a physical and chemical basis. The variety and quantity of chemical discoveries and researches call the attention of lay people, so that “confusion turns to clarity and fright turns into surprise and joy”. Doubtless Runge’s educational ideas contributed to his oblivion.

FINALE IN ORANIENBURG

After Runge’s friction with the managers of the Oranienburg factory, Cochiuss’s widow expelled him in 1856 from his lodgings in Oranienburg Palace. He then rented a more modest one-story house from a friend (figure 10), compatible with his income, which no longer exists (demolished in the 1930s). Also the commemorative plaque placed there in 1923 no longer exists. Single, he lived there writing the “*Hauswirtschaftliche Briefe*”, or notices for newspapers and journals, but without carrying out any formal activity: King Frederick William IV granted him an annual pension. He met up with some friends, demonstrating his skills as a cook and enjoying his “synthetic wine”. He also manufactured other “wines” and prepared various types of preserves. He lived simply, advised professionals in all areas, and also the common people who showed an interest in certain subjects. He died at his home on March 26, 1867, and was buried at the *Nikolaifriedhof*, where his tomb was a victim of vandals in 1923, but restored shortly afterwards. In 1949 the cemetery was closed, and the tomb was transferred to the municipal cemetery⁴⁹.

As we saw, belatedly, in 1994, a bronze statue of Runge was erected in front of his laboratory, attached to the Oranienburg Palace, designed by Stephan Möller (figure 11). The statue, at ground level, shows Runge at his work table, with various equipments. In 2017, on the 150th anniversary of his death, there were several celebrations in Oranienburg.

CONCLUSION

Runge was certainly an unusual, unorthodox chemist, which brought him a certain popularity during his lifetime, but not recognition among his peers. Why did he not find recognition from his colleagues, despite some undoubtedly important discoveries in the field of phytochemistry and coal tar? Perhaps for leaving academic activities and research to dedicate himself

to industry, where his industrialization proposals were also not well received? Or perhaps because of his exotic and unusual behaviour? Perhaps for remaining faithful to *Naturphilosophie*, and because he did not accept the atomic theory? If Runge enjoyed some popularity during his lifetime, after his death he fell into almost total oblivion. A first writing lamenting the neglect of Runge, "Contribution to the history of scientific discoveries", is anonymous, and was published in 1863 in the popular (382.000 copies in 1875) weekly magazine *Die Gartenlaube*, founded by Ernst Keil (1816-1878) and published in Leipzig since 1853⁵⁰. In 1869 the German Chemical Society created a commission (Hofmann, Baeyer, Magnus, Graebe) to honor Runge with a representative tomb, which only occurred in 1872⁵¹. The biographies of Schelenz (1907) and Rehberg (1936) clearly exaggerate the qualities attributed to Runge and his achievements, and the presentation of a "typical German researcher", in science and in personal life, was a big deal for Nazi-fascist ideology from the 1930s, even distorting Runge's real personality. All of this did not contribute to keeping Runge's name visible in historiography. The novelist Karl Aloys Schenzinger (1886-1962), a supporter of Nazi ideology but without being a member of the party, found in Runge, by disfiguring his character and personality, the first "hero" of his novel "Anilin" (1936), the "novel of a dye", nationalist and full of errors and omissions, but very popular (almost a million copies sold until 1945)⁵². In the 1950s, after the compromising passages from the first editions had been expunged, new versions of the novel were published (last edition in 1973)⁵³. Clearly, the idealized chemist in "Anilin" is not Runge. In any case, one should not confuse character and author/work: with the growing importance of chromatography, Runge and the "Musterbilder" gained prominence, and with the interaction that exists today between Chemistry and other human activities, Runge's role draws attention, for example, in the Runge Prize for unconventional artistic creations, awarded since 1994. A new Runge emerges, closer to historical reality.

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