

Felix-Henri Boudet

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Resumen

Félix-Henri Boudet (1806-1878) fue un farmacéutico francés que estudió la composición de la sangre, particularmente de su suero; la isomerización cis-trans de grasas y su fermentación; la preparación de aguas minerales artificiales; el uso de cianuro de potasio como medicamento; etc. Como resultado de su trabajo, descubrió dos tipos diferentes de oleínas, una presente en los aceites secantes, la otra en los aceites no secantes; determinó la composición de algunas aguas minerales francesas, desarrolló un nuevo instrumento para este propósito y procedimientos para hacer aguas minerales artificiales, en sustitución de las naturales.

Palabras clave: aceites, aguas naturales, aguas artificiales, fermentación, isomerización cis-trans, sangre, suero.

Abstract

Félix-Henri Boudet (1806-1878) was a French pharmacist who studied the composition of blood, particularly of its serum; the cis-trans isomerization of fats and their fermentation; the preparation of artificial mineral waters; the use of potassium cyanide as a medicine; etc. As a result of his work, he discovered two diverse types of oleins, one present in drying oils, the other in non-drying oils; determined the composition of some French mineral waters, developed a new instrument for this purpose and procedures for making artificial mineral waters, as substitute of the natural ones.

Keywords : blood, cis-trans isomerization, fats, fermentation, natural and artificial waters, serum.

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Life and career (Anonymous, 2024; Boudet, 1879; Sainte-Claire Deville, et al., 1878)



FIGURE 1: Felix-Henri Boudet (1806-1878)

Little information is available about the early life and education of Felix-Henri Bouquet (Figure 1). He was born in Paris, on May 22, 1806, the son of a pharmacist. After taking his basic education at the Massin institution and at the college Charlemagne and obtaining his diploma of bachelier ès-lettres (1823), he began to work as a pharmacist apprentice in a local pharmacy, while furthering his education in the École Supérieure de Pharmacie and in the Faculté des Sciences of Paris. In 1828 he obtained his diploma of bachelier ès-sciences and in 1831 his diploma of licencié ès-sciences physiques. In 1833 he graduated as pharmacien de 1^{er} classe and of docteur ès-sciences physiques after successfully defending theses about the action of hyponitric acid on vegetable oils (Boudet, 1832a), and a critical and experimental essay about blood (Boudet, 1833a). In addition to managing the family pharmacy, he served

as adjoint professor at the École Supérieure de Pharmacie (from 1845 on).

Bouquet died in Paris on August 8, 1878, as a result of a previous cerebral congestion.

Bouquet was a member of the Académie de Médecine (pharmacy section, 1856); a member of the Société de Pharmacie de Paris; an honorary member of the Conseil de d'Hygiène et de Salubrité of the Département de la Seine (1852); the general secretary of the Société de Secours des Amis des Sciences (1860), and honorary president of the Société Protectrice de l'Enfance. In 1846 he was nominated Chevalier de la Légion d'Honneur.

Scientific contribution

Félix-Henri Boudet conducted most of his research on the subjects of inorganic, organic, and physical chemistry, potable and mineral waters, and physiology. This work led to the publication of about twenty-five scientific papers and books (i.e., Boudet, 1833c, Boutron-Charlard & Boudet 1866). As customary to candidates to the Académie de Médecine, Boudet prepared a booklet describing his research activities and services (Boudet, 1855). In addition to the few subjects described below, Boudet made a critical study of the composition of blood (Boudet, 1833a); came against those who believed that gallbladder stones could be dissolved by drinking chemical solvents or mineral waters such as those from Vichy (Boudet, 1837); analyzed the brain substance of a fetus (Boudet, 1840); suggested new formulations for the use of ferrous iodide for the treatment of pulmonary phthisis (Boudet, 1841); determined the composition of the Liard oil used to lubricate machinery (Boudet, 1850); discussed the information regarding the controversy between Leblanc and Dizé about the paternity of the synthesis of sodium carbonate (Boudet, 1852); analyzed the discovery of aluminum and its uses (Boudet, 1858); discussed the composition and

preparation of the so-called Boutigny's salts (Boudet, 1859); reported to the Académie des Sciences some statistical data about the French population, such its growth according to the geographic location, public hygiene, mortality, regional movements, etc. (Boudet, 1866-1867); together Antoine-François Boutron-Chartard (1796-1879) reported the analysis of different parts of an Egyptian mummy (Boutron-Chartard & Boudet, 1833); alone or with Boutron-Chartard, discussed the situation and problematics of the Parisian potable water distribution network, including the use of lead pipes (Boudet, 1863, 1874ab; Boutron-Chartard & Boudet, 1854); the use of hydrometry as an analytical method of river waters (Boutron-Chartard & Boudet, 1855); etc.

Action of hyponitric acid on vegetable oils. Cis-trans isomerization

Boudet commented that olive oils were usually adulterated by addition of a grain oil such as poppy oil and that this action could be detected by the tests proposed by Jean-Joseph Étienne Poutet (1779-1859), and Rousseau (Boudet, 1832a). Poutet's test was based on the property of a nitric acid solution of mercuric nitrate of freezing and solidifying olive oil, after some hours, and leaving other grain oils in the liquid state. The reagent was prepared by mixing six parts of mercury with 7.5 parts of nitric acid of specific gravity 1.355, at room temperature (Poutet, 1819). Rousseau's method was based on the fact that olive oil conducted electricity 675 times better than other vegetable oil (Rousseau, 1823). This property was measured with a diaphragm constructed of a galvanometer connected with a dry battery. The oil was placed in a small cup and the current transmitted. This test was very sensitive but required an expensive apparatus. Poutet's process was within the reach of everyone but did not detect the presence of less than 10% of foreign oil. Boudet added that Poutet had not concerned himself with the mechanism of his process and this fact led him to look further into the matter (Boudet, 1832a).

A first look into the composition of the reagent showed that the reaction of mercury with nitric acid resulted mainly in the formation of mercurous and mercuric nitrate, accompanied by a small amount of mercuric nitrite, all dissolved in an excess of acid (Boudet, 1832a). Hence, Boudet submitted the olive oil to the separate action of mercuric oxide, mercurous nitrate, mercuric nitrate, and nitric acid, and was surprised to find that none of these reagents alone resulted in solidification of the oil. This result left mercuric nitrite as the only possible agent of the phenomenon and at being originated by the reaction of mercuric oxide and nitric acid. In other words, it was very probable that the solidification was induced by hyponitric acid (Boudet, 1832a).

To assess this assumption, Boudet carried the following experiment (Boudet, 1832a): A certain amount of olive oil was put in a bell jar containing mercury and then treated at room temperature with two hundred volumes of nitrogen dioxide, followed by 100 volumes of oxygen. The gas mixture instantly transformed into hyponitric acid, which reacted with the olive oil, releasing heat, and turning the oil green but initially not changing its consistency. As time went by, the oil became cloudy and at the end of about two hours it became solid and quite similar to the product of the reaction with Poutet's reagent. Repetition of this experiment with liquid hyponitric acid obtained by the distillation of lead nitrate, led to the same result. Hence, the mercury components of Poutet's reagent were unnecessary, hyponitric acid alone solidified the oil. This suggested that hyponitric acid could be a mixture of nitric and nitrous acids and that the

latter determined the solidification of olive oil. Further experiments showed that the best results were obtained with a mixture of one part of hyponitric acid with three parts of nitric acid of specific gravity 1.355 (Boudet, 1832a).

The next series of experiments studied the effect of the ratio olive oil/hyponitric acid on the length of time for achieving solidification at 16 °C. The results indicated that 5/1000 parts of hyponitric acid were enough to solidify the oil (Boudet, 1832a).

Boudet found that hyponitric acid was also capable of inducing the solidification of the oils of sweet and bitter almonds, hazelnuts, cashew nuts, castor oil, and rapeseed, but not those of linseed, hempseed, walnut, poppy, and beechnut. Treatment of 100 parts of oil with 12 parts of a mixture of nitric acid and hyponitric acid (representing 3 parts of anhydrous hyponitric acid) led to solidification of the pertinent oil in the following number of minutes: Olive (7.5), cashew (45), hazelnut (103), sweet almond (160), bitter almond (160), castor (603), and rapeseed (2400). Boudet remarked that oils were usually classified as drying and non-drying oils; his results indicated that the oils of the first group did not solidify with hyponitric acid while non-drying oils did (castor oil was an exception) (Boudet, 1832a).

The next stage was a study of the solidified state of the oils of olives, sweet almonds, hazelnuts, cashew nuts, and castor (Boudet, 1832a). According to Boudet, these solids were white or yellowish, according to i they were the product of hyponitric acid or mercury nitrate and were slightly soluble in alcohol of 36°. Solidified olive oil, heated with alcohol, lost its yellow color, and became white like lard; it could be further purified by pressing between unpasted paper. Boudet suggested naming it *elaidin* (not necessarily the pure trans isomer and being a generic name for all the pertinent solids) (Boudet, 1832a).

Boudet mentioned that pure elaidin melted at 36 °C, was totally soluble in ether and partially soluble in alcohol of specific gravity 0.8975, and was not colored by KOH, ammonia, or ammonium bisulfate, which proved that the color developed by these reagents in yellow elaidin did not belong to the fatty matter itself. Elaidin was easily saponified by KOH and NaOH yielding glycerin and an acid fat, which united with the alkali. It was hydrolyzed by hot HCl yielding a liquid acid, which then solidified. Boudet named the acid *elaidic acid* (Boudet, 1832a).

Elaidic acid melted at 44 °C, solidified in small pearly and shiny flakes like boric acid, and distilled without decomposition. At higher temperatures it decomposed partially into water, acetic acid, an odorous volatile oil, and an empyreumatic oily liquid, and left a carbonaceous residue. Elaidic was soluble in ether and in boiling alcohol. Boudet prepared several elaidates (sodium, potassium, ammonia, lead, mercury, and magnesium) and described their properties.

Boudet studied in detail the reaction of castor oil with acid mercuric nitrate and hyponitric acid. In both cases, the product was a solid mass with a wax-like appearance, which Boudet named *palmin*. The formation of castor palmin was eight times slower than that of olive oil placed in the same circumstances. Palmin was saponified by concentrated and boiling KOH and hydrolyzed by HCl into an acid fatty substance, which turned into a crystalline mass on cooling and which Boudet named *palmitic acid*. Palmitic acid melted at 50 °C and crystalized into white silky needles. It was soluble in ether and in concentrated alcohol. Boudet prepared various of its salts and determined their properties (Boudet, 1832a).

Boudet found that gaseous SO_2 was also capable of transforming castor oil into a solid mass, melting at 66°C and being completely soluble in alcohol of 36° . He believed that this reagent was able to solidify all the oils mentioned before (Boudet, 1832a).

Today we know that agents like nitrogen oxides, SO_2 , selenium, and catalytic hydrogenation, are able to convert unsaturated fatty materials into solid materials by geometrical isomerization of their double bonds. The reaction is thought to proceed by addition of the chemical agent to one of the double bonds, its subsequent split, and reformation of the double bond in the *trans* configuration, which is thermodynamically more stable than the *cis* one and has a much higher melting point.

Boudet and Théophile-Jules Pelouze (1807-1867) extended the research to the action of hyponitric acid on fatty bodies in general, starting from the assumed knowledge that most vegetable oils contained the two distinct immediate principles, olein and margarine (Pelouze & Boudet, 1838). They were unable to separate the two by all known procedures, nevertheless, they learned that there existed two essentially distinct oleins, one found in the non-drying oils of sweet almonds, olives, hazelnuts, human fat, pork fat, etc. (common olein); the other, as a constituent of the greater part of the drying oils of linseed, walnut, poppy, hempseed, the liquid oil of coconut butter, etc. (linolein). In addition, they found that both oleins held in solution the same margarine because they always succeeded in extracting margaric acid (then palmitic acid) melting at 60°C (today, 62.9°C). These two oleins differed by their different solubility in different vehicles; one was siccative (linoleic acid) while the other was not; the first always contained much less hydrogen than the second; and hyponitric acid transformed the one into elaidic acid, while it had no analogous action on the other (Pelouze & Boudet, 1838).

Boudet and Pelouze also found that treating with hyponitric acid the combination of olein and margarine extracted from olive oil, converted the first into elaidin, and left the margarine intact, so that the product obtained presented a melting point intermediate between that of elaidin and of margarine. In addition, the saponified product could be easily partitioned by means of alcohol into margaric acid fusible at 60°C , which crystallized first, and elaidic acid melting at 45°C (Pelouze & Boudet, 1838).

Fermentation of fatty substances

In 1856, Boudet presented to the Académie des Sciences his findings about some physiological consequences of the fermentable property of fatty materials (Boudet, 1856). In the first part, he described the historical development of the discovery that fatty materials were also fermentable. He mentioned that until 1838 fats and oils were known to be decomposable only by the action of temperature or chemical reactions; there was no hint about the possibility that this process could also be done with ferment (enzymes). In that year Pelouze and Boudet reported that they had found glycerin, palmitic acid, and oleic acid in an old sample of palm oil, a finding that suggested the possible existence of a ferment able to split fatty substances in the same manner that sugar was split unto alcohol and CO_2 (Pelouze & Boudet, 1838). This curious fact led Edmund Frémy (1814-1894) to look further into old palm oil and to discover an additional fatty acid present in the oil, which he named *acide palmitique* (palmitic acid) (Frémy, 1840b). Frémy separated the solid fatty acid from the oil, purified it by ordinary procedures, and found that it had a strong analogy with margaric acid (palmitic acid). The acid melted at 60°C and contained

75.4% carbon, 12.5% hydrogen, and 12.1% oxygen (the correct composition is 74.94% carbon, 12.58 hydrogen, and 12.48% oxygen). Frémy prepared a large number of palmitates and determined their composition. Frémy also analyzed the composition of the brain in different states and of different ages and found that it contained a variable quantity of free fatty acids, which sometimes increased when the fatty matters were left in a closed bottle. He explained this phenomenon by quoting Michel Eugène Chevreul's (1786-1889) observations on the fat of cadavers (Chevreul, 1815) and quoting again Pelouze and Boudet remark about the spontaneous saponification of palm oil. Frémy showed that under the influence of the albuminous matter of the brain, the oleophosphoric acid was decomposed into olein and phosphoric acid, and that the proportions of free oleic and margaric acids, which he had recognized to exist in the fresh brain beside margarine and olein, increased rapidly as brain matter deteriorated further (Frémy, 1840c).

In 1844, Claude Bernard (1813-1878) and Charles-Louis Barreswil (1817-1870) found that the pancreatic juice not only had the property of emulsifying fatty substances, but that it could in a few hours split them into fatty acids and glycerin (Bernard & Barreswil, 1844). Finally, Pelouze provided additional examples of fatty matter fermentation and that oils, which were perfectly neutral in oilseeds, split quickly into fatty acids and glycerin as soon as, by breaking the cells, which contained them, were brought into contact with the substances, with which they were accompanied in these same seeds, and which acted as real ferments (i.e., Pelouze, 1831).

Boudet conducted his first experiments using palm oil and brewer's yeast; palm oil because it deteriorated with no trouble and seemed to contain a ferment, and the yeast, because it was easily accessible (Boudet, 1856). In one open flask he left a certain amount of palm oil to the action of its ferment, and in another, the same amount of oil mixed with ten parts of fresh brewer's yeast. Both flasks were loosely covered with paper (Boudet, 1856).

The palm oil was seen to discolor slowly while becoming acid. Concentration of the liquid by distillation precipitated some white acid crystals, sparingly soluble in cold water and very soluble in boiling water. The purified crystals were found to be sebacic acid, melting at 127 °C (today, 131 to 134.5 °C). The liquid was also found to contain a significant amount of glycerin. Boudet assumed that the sebacic acid (a dicarboxylic acid) was the result of the oxidation of the palmitic acid generated by the fermentation process together with oleic acid (Boudet, 1856). Boudet added that sebacic acid had been discovered by Louis-Jacques Thenard (1777-1857) during the distillation of tallow (Thénard, 1801), afterwards obtained by Jules Bouis (1822-1886) during the distillation of castor oil (Bouis, 1855), and also by Joseph Johann Pohl (1825-1900) by distillation of palm oil (Pohl, 1854). Boudet speculated that olein was the main product of the action of the natural ferment produced. The palmitin slowly acidified, while the olein was transformed first into oleic acid and glycerin and then into sebacic acid, accompanied by a certain proportion of an as yet undetermined acidic matter (Boudet, 1856).

The product of the fermentation with brewer's yeast was significantly different: it was a spongy solid material, which could be easily powdered. Extraction with ether separated palmitin, melting at 50 °C, and another acid, melting at 48 °C and partly soluble in a diluted solution of sodium carbonate. Surprisingly, no olein, oleic acid, and sebacic acid were found, pointing that the transformation by means of yeast was deeper than the one caused by the natural ferment. (Boudet, 1856).

Boudet mentioned that C. G. Lehman had written that fat, once introduced in the blood, went through a variety of transformations, which depended on the functions which it was destined to fulfil. These transformations usually involved the formation of glycerin and the slow oxidation of the fatty acids released (Lehman, 1855). The pertinent chemical equations were not known, but the final result was recognized: the body economy transformed the ingested fatty materials into water and CO_2 or released them by perspiration in the form of formic, acetic, or butyric acids (Boudet, 1856).

Human blood serum

According to Boudet, the information available about the extractive substances present in blood was scarce and incomplete (Boudet, 1833b). Some of them were osmazome, impure sodium lactate, muco-extractive matter, etc. Chemical analysis had been reported only for albumin, the fatty matter of the brain, urea, and an oily matter recently found by Louis René Lecanu (Lecanu, 1831). This situation led Boudet to carry additional experiments on the subject, restricting himself, in the beginning, to the examination of the possible products extracted by alcohol from serum previously macerated with boiling water. This residue was first dried at low heat, followed by a second extraction with boiling water; dried again, powdered, and finally extracted with boiling alcohol. On cooling, the extract deposited white flakes of a greasy and pearly aspect, which were separated by filtration and found not to be crystalline. Boudet believed that this material was an immediate principle, which he named *seroline* (Boudet, 1833b).

The extract filtrate was evaporated to dryness and left a yellowish-brown, slightly dark residue, of acrid flavor, smelling like the fatty matter of the brain, and emulsifying with cold-water. Treatment with alcohol of 36 °C separated it into two fractions, one soluble in alcohol, which Boudet believed was the oily matter of Lecanu, and the other, the fatty matter of the brain described by Louis Nicolas Vauquelin (1763-1829) and Chevreul (i.e., Vauquelin, 1811). The latter was insoluble in cold alcohol and soluble in boiling alcohol and in ether. It crystallized in brilliant sheets, did not react with colored reagents and alkalis, and emulsified with cold water. The alcoholic solution, left to itself, deposited small crystalline plates looking like cholesterol and melted at 137 °C. The remaining liquid, evaporated to dryness, and treated with NaOH, generated a material, which had all the properties of soaps of oleic and margaric acids. Boudet compared the properties of his cholesterol with those of pure cholesterol, extracted from gallstones, and found them to be remarkably similar (Boudet, 1833b).

Under the microscope, serolin appeared to be formed of filaments swollen from distance to distance by small white and opaque globules, which gave them the appearance of strings. It melted at 36 °C, did not act on test papers, and reddened on contact with concentrated sulfuric acid. It did not emulsify with water; was soluble in ether and sparingly soluble in alcohol, cold or boiling, and did not react with KOH, HCl, nitric acid, and acetic acid.

Boudet's work on the constitution of blood culminated in his doctoral thesis (Boudet, 1833b).

Boudet's brother, Ernest Boudet, a physician, asked him to investigate the action of different reagents on pulmonary parenchyma. A literature search showed him that in spite of the importance of the subject, nothing had been published about it (Boudet, 1844). Anatomists knew that this parenchyma was formed of cellular tissue, and chemists, that

hot water transformed it into gelatin. Boudet found, on the one hand, that heating a certain quantity of pulmonary substance with distilled water, yielded a gelatinous solution, and, on the other hand, kneading a piece of man's lung in a knot of linen, under a stream of water, yielded a liquid, which cold acetic acid converted into a coagulum of hematosine and albumin, while leaving a residue composed of a fibrous, elastic substance behaving with HCl, sulfuric, and acetic acids, like fibrin, and exhibiting its properties (Boudet, 1844). This simple experiment proved that the lung contained fibrin, gelatin, and a substance, which was casein. Further treatment with solvents like alcohol and ether allowed detecting the presence of non-saponified fat, sodium oleate and palmitate, free lactic acid, cholesterol, and some extractive substances, including onesoluble in alcohol and boiling ether, completely insoluble in cold alcohol, and presenting the all the properties attributed by Frémy to his *cérébrique acide* (Frémy, 1840a). In addition, Boudet found that the ashes of pulmonary tissue contained about 78% sodium chloride, sulfate, phosphate, and carbonate, and about 22% silica, iron oxide, calcium carbonate and phosphate. This result completed a general picture of the composition of lung parenchyma,(Boudet, 1844).

The next stage was a comparison of the composition of lung parenchyma and lung tubercles. On the one hand, Boudet found that treating the lung tubercle with cold water yielded albumin, a matter precipitable by acetic acid, analogous to that which had been detected out in the normal lung under the name of casein, and reducible to a substance, which offered the characteristics of fibrin. On the other hand, alcohol extracted oleic and palmitic acid, neutral fat, free lactic acid, and sodium lactate, cerebriic acid, cholesterol, etc. The pertinent ashes contained sodium chloride, sulfate, carbonate, calcium phosphate and carbonate, silica, and iron oxide. These results were quite similar to those of lung parenchyma, indicating that the difference between the two parts was not a chemical one (Boudet, 1844).

Natural and artificial mineral waters

According to Bouquet, this way of looking at mineral waters had resulted in some successful applications such as the use of Vichy's salt or sodium bicarbonate, which Jean Darcet (1724-1801) had introduced into materia medical (Darcet, 1826), and the use of Sedlitz waters (a yellowish, somewhat oily-looking fluid, with a nauseous, intensely bitter taste)containing sulfides (Bouquet, 1831). He himself used it to suggest modifications to the Barèges and Bonnes waters for drinking. For example, the French Codex stipulated that 20.5 ounces of Barèges water should contain sixteen grains of sodium carbonate, 0.5 grain of sodium chloride, four ounces of water saturated with its own volume of hydrogen sulfide, mixed with sixteen ounces of distilled water. According to Bouquet, a more appropriate formulation should be: twenty-five grains of crystallized neutral sodium sulfide, sixteen grains of sodium carbonate, 0.5 g of sodium chloride, and twenty ounces of distilled water (Bouquet, 1831). For a sulfurized water bath, he suggested using a solution containing ten ounces of crystallized neutral sodium sulfide completely dissolved in enough water, mixed with ten ounces of Codex salt-gelatinous solution (Bouquet, 1831).

In the following paper, Bouquet surveyed the information available about sulfur and its presence in mineral waters (Bouquet, 1832a). Since old times, sulfur was well-known to have therapeutic properties and had been administered by physicians in a variety of forms, with livers of sulfur (a loosely defined mixture of potassium sulfide, potassium

polysulfide, potassium thiosulfate, and potassium bisulfide) being the most common. These combinations were known and used with no relation to the sulfurous waters available in many places. Eventually, chemists noticed the strong analogy that existed between the alkaline sulfides and the characteristic elements of sulfurous waters, although initially they had assumed that hepatic waters contained only hydrogen sulfide in the free gas state. Later on, Anglada and Étienne Ossian Henry (1798-1873) had postulated that this sulfur was present mostly in combination with various salifiable bases, an assumption that turned these sulfides to be an essential ingredient of sulfurous waters (Anglada, 1833). This discovery led promptly to include sulfides in the preparation of artificial mineral waters, although natural sulfurous waters had a very variable and complicated composition. At the time, not much information was available about sulfides. Vauquelin had determined that they had a special flavor and that they were soluble in water and in alcohol (Vauquelin, 1801). Louis-Jacques Thénard (1777-1857) had shown that Vauquelin's hydrosulfide lost one half of its acid when heated in aqueous dissolution (Thénard, 1812). Anglada had confirmed this claim and rigorously demonstrated that in most of sulfurous waters this hydrogen sulfide was combined with sodium in the proportions of a neutral salt, making the neutral sodium bisulfide the one to be used for their artificial reproduction. In particular, Anglada found that bubbling hydrogen sulfide through a concentrated solution of NaOH resulted in the spontaneous crystallization of a sodium sulfide identical with that present in sulfurous waters, and containing, by weight, 27.8% of sodium, 14.4% of hydrogen sulfide, and 47.8% of water (Anglada, 1833).

Boudet's interest in the fabrication of artificial mineral waters similar to the ones found in Barèges, led him to conduct some experiments and determine that Anglada claims were correct. His results indicated that the best way to manufacture sodium hydrosulfide was to bubble hydrogen sulfide, obtained by the action of sulfuric acid on ferric sulfide, through a solution of NaOH of specific gravity 1.330. After some time, the solution crystallized spontaneously, as stated by Anglada (Boudet, 1832b). This result indicated that in principle, it was quite easy to imitate the Barèges water, known to contain only three salts, namely, sodium bisulfate, carbonate, and chloride, although no one had yet established the relative proportions of water and these salts. To fill this gap, Bouquet conducted a careful analysis of the natural waters by evaporating 758 g of water in a porcelain capsule, followed by treating the residue with a few drops of sulfuric acid, and calcining the product in a platinum ware. The resulting sodium sulfate was found to be accompanied by some traces of earthy matter and silica and suggested that one liter of water contained 0.091 g of crystallized sodium carbonate, 0.015 g of sodium chloride, and 0.212 of crystallized sodium bisulfide (Bouquet, 1832b).

Potassium cyanide as a medicine

In 1832, Mathieu Joseph Bonaventure Orfila (1787-1853) reported that he had received a letter from a Dr. Trouvé informing him of the tragic result of the treatment given to a patient suffering from neuralgia of the trunk (Orfila, 1834). The patient had been administered five enemas, each consisting of six ounces of water and six grains of potassium cyanide in different conditions: moistened but still in a mass; cold; heated in a water bath; as a slurry; and finally, very dry. Some of these treatments were accompanied by temporary side effects (strong convulsions, violent contractions of the limbs, fixing of the eyes became, and dilated pupils), which eventually disappeared. Unfortunately, the last enema, the one prepared

with the same dose of very dry cyanide taken from a jar, which had not yet been uncorked, and which scarcely exhaled any odor, resulted in death, after the patient experienced general convulsions, beating of the heart, slow and difficult breathing, coldness of the limbs, dilatation of the pupil, and fixed eyes (Orfila, 1834).

Orfila commented that it was obvious that dry potassium cyanide was more dangerous in the dry state than in the humid one, and that this effect could be explained by potassium cyanide being decomposed by water, particularly when hot (Orfila, 1834). Théophile-Jules Pelouze (1807-1867) had recently proven that a concentrated solution of potassium cyanide boiled without contact with air, was easily transformed into ammonia and potassium formate (which were inert), with the corresponding dilution of the potassium cyanide. Calcination of dry potassium cyanide, in the absence of air, had no effect (Pelouze, 1831).

This publication caught the attention of Boudet, who added some additional comments. Potassium cyanide was known as a sedative for neuralgia, as long as it was used in low and constant doses; the slightest variations could lead to the gravest consequences for the patient, as shown in the paper by Orfila (Boudet, 1834). Pelouze and Philip Lorenz Geiger (1785-1836) had shown that a concentrated solution of potassium cyanide, boiled in the absence of air, decomposed into ammonia and potassium formate (Pelouze, 1831; Geiger, 1832). The same solution, boiled in contact with air, generated hydrogen cyanide, potassium and ammonia carbonates, potassium formate, and lesser amounts of ammonia cyanide. In addition, solid potassium cyanide, kept in an open or badly stoppered bottle, was transformed into hydrogen cyanide gas and potassium carbonate produced at the expense of the CO_2 of the air. The usual procedure for preparing potassium cyanide for medical use was relatively simple: A mixture of yellow potassium cyanide and iron was calcined, the residue extracted with water, and the solution evaporated to dryness (Boudet, 1834).

According to Boudet, it was easy to see that the final product was not pure and identical from batch to batch. The variability in concentration implied that the pertinent doses did not have the same potency and could lead to serious accidents. Boudet had found an easy manner of overcoming this problem. He had noted that when the retort in which the mixture of potassium cyanide and iron had been calcined, was broken, it contained a mass formed of chunks of potassium cyanide and chunks of iron quadracarbure. Breaking this mass carefully allowed separating a certain quantity of molten potassium cyanide into white, compact fragments of perfect purity, which could be immediately used in the medical treatment (Bouquet, 1832).

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