



## Pierre Oscar Figuiet

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### Resumen

Pierre Oscar Figuiet (1759-1817), farmacéutico francés cuya principal contribución fue el descubrimiento del uso de carbón vegetal para blanquear sustancias inorgánicas y orgánicas. Figuiet también realizó un trabajo sustancial sobre la química del oro, la síntesis de muchos de sus compuestos, particularmente una sal triple de cloruro de oro y cloruro de sodio. Determinó la composición de los garbanzos y del cardo estrellado.

### Palabras clave

Blanqueo; cardo estrellado; compuestos de oro; garbanzos.

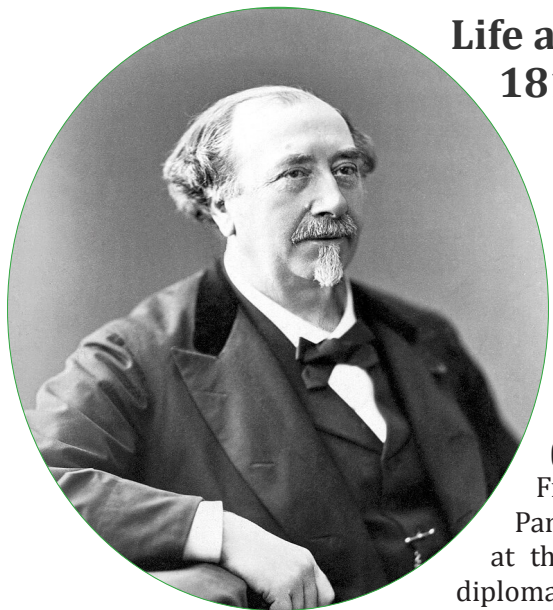
### Abstract

Pierre Oscar Figuiet (1759-1817), a French pharmacist whose main contribution was the discovery of using animal charcoal for bleaching inorganic and organic substances. Figuiet also carried substantial work on the chemistry of gold, the synthesis of many of its compounds, particularly a triple salt of gold chloride and sodium chloride. He determined the composition of purple thistle and chickpeas.

### Keywords

Chickpeas; discoloring; gold compounds; purple star thistle.

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## Life and career (Charlot, 2005; Boullay, 1817)

**P**ierre Oscar Figuiet was born in 1759, in Sommières, department of Gard, to a well-to-do family. After finishing his basic education, he was sent to Montpellier to begin a three-year apprenticeship in the pharmacy of Chonney. After finishing it, one of his aunts invited him to Geneva to continue his training in the pharmacy of Royer and Tingri. This he did for five years. In 1790, he moved to Paris and studied for two years under Antoine-François Fourcroy (1750-1809) at the College of the Jardin du Roi, and Jacques-François Demachy (1728-1803) at the Collège de Pharmacie de Paris. He then returned to Montpellier and completed his studies at the École de Pharmacie de Montpellier. After receiving his diploma of pharmacien (1792), he opened a pharmacy in Montpellier and shortly thereafter he was admitted to the guild of pharmacists. After a successful professional career, he was appointed professor of general chemistry in the École Spéciale de Pharmacie of Montpellier (1803 to 1817).

Pierre Figuiet passed away in Montpellier on March 28, 1817. His brother took over the management of the pharmacy, and his children continued the profession.

### Scientific contribution

Figuiet wrote about 20 papers and books (i.e., Figuiet, 1812a, 1813) about inorganic chemistry, chemical synthesis, plants composition, mineral waters, etc. In addition to the subjects described below, he studied sodium hydrosulfide and its use in the preparation of sodium hydroxide (Figuiet, 1807, 1812c); he analyzed mineral waters (Figuiet, 1809c, 1810, 1812), studied the preparation of the Seignette salt (potassium sodium tartrate tetrahydrate) and sodium phosphate (Figuiet, 1812b); and of potassium acetate (Figuiet, 1813); etc.

### *Gold compounds*

In 1811, the Montpellier physician André-Jean Chrestien (1758-1840) published the third edition of his book about a new method for treating venereal and lymphatic diseases (Chrestien, 1811), based on the iatraliptic method for administering remedies promoted by Peter Clare (1738-1786). This method was founded on the application of the desired medicine by rubbing it on the gums (Clare, 1779). Chrestien tried first a very fine gold prepared by triturating leaf gold with seven times its weight of mercury and then expelling the mercury by heat or exposition to the sun or dissolving it out by pure nitric acid. The beneficial results obtained by rubbing this preparation led Chrestien to speculate that perhaps they were due to the small quantities of mercury, which the gold had retained. To test this possibility, he repeated the rubbing process using this time gold oxide (prepared by the action of KOH on a dissolution of gold in regal water) instead of the gold amalgam. Again, the resultant results were very good, which showed that mercury had no role on the therapy. The gold oxide prepared by tin was then substituted for that prepared by

KOH, but Chrestien, fearing now that the effects obtained could be attributed to the small quantities of tin retained by gold oxide, resolved to try only gold chloride. Unfortunately, this substance was found to be too caustic, Hence, he definitely substituted it by a mixture of gold and sodium chlorides, which was justified by the ease of administration. He also understood that different compounds of gold, having different activity, could provide a great homogeneity of therapeutic action. Chrestien's book reported the results of several experiments, which illustrated the positive action of his treatment (Chrestien, 1811).

According to Figuier, Chrestien's publication spoke of the various gold prescriptions he had made with the metal, but did not provide details how they were obtained. As a result, some physicians had prepared them as they understood them, unfortunately in the wrong manner. This had caused erroneous results and unfavorable recommendations. For these reasons, he requested from Figuier to describe these preparations in the proper manner and publish them in a scientific journal (Figuier, 1811a).

Figuier's paper contained the preparation of gold oxide precipitated by tin or potassium, the preparation of finely divided gold, and of the triple salt of gold and sodium chlorides. For example, the gold oxide was prepared by precipitation with tin, as follows: One part of pure gold reduced to granules was mixed with eight to ten parts of aqua regia made with equal parts of nitric acid and HCl. After the effervescence had abated, the liquid was heated to boiling over a sand bath until the dissolution of all the gold. The resulting neutral solution was evaporated until the consistency of clear syrup and then mixed with twenty parts of its weight of distilled water, and with pure tin reduced in films. The mixture was then left alone to let the gold precipitate. At the end of this process, the tin not dissolved was removed and the solution filtered. The solid deposit was washed with distilled water until the liquor came out tasteless. It was then dried in the shade, pulverized, sieved through a silk sieve, and kept it in a crystal bottle (Figuier, 1811a).

Chrestien's report induced other several scientists to study the chemistry of gold and the synthesis of several of his derivatives. Thus, Louis Nicolas Vauquelin (1763-1829) wrote that gold had been employed, so far, in four different states, (1) finely divided, (2) as chloride, (3) as oxide, precipitated from a solution by KOH, and (4) a precipitate obtained by treating a solution of gold chloride with tin (Vauquelin, 1811). According to Vauquelin, there was some difficulty in obtaining these preparations constantly in the same state; for this reason, he described in detail the processes and improvements he had developed to solve the problem. For example, it was common practice to prepare aqua regia by mixing two parts of nitric acid with one of HCl. Vauquelin reasoned that gold required only a very small amount of oxygen for its solution, hence, it was possible to obtain the same result using aqua regia composed of the two acids in the *opposite* ratio. Vauquelin's results indicated that three parts of aqua regia composed of one part of nitric acid and two of HCl were enough to dissolve one part of fine gold, instead of the four parts of the traditional mixture. KOH, NaOH, baryte, and lime had no action on the resulting solution; they only changed its color from yellow to deep red. The solution of gold, when properly evaporated, appeared as yellow prismatic crystals; this process had to be conducted carefully to avoid the partial decomposition of the salt into metallic gold. Vauquelin gave a detailed description about the action on the solution of gold chloride of various reagents in different concentration, particularly, the precipitation of gold oxide by means of fixed alkalis. An important result was finding the existence of a triple salt composed of gold chloride and potassium chloride (Vauquelin, 1811).

In the same year, A. S. Duportal and Pierre-Joseph Pelletier (1788-1842) recommended that instead of using the fine gold in the form prepared by Chrestien, it was better to precipitate a solution of gold chloride with the minimum amount of iron sulfate, followed by filtering, washing the precipitate with water acidulated with HCl to eliminate the retained iron oxide, more washing, and drying. The resulting dry gold appeared as a deep brown powder, a consequence of its minute size. They also described an improved method for preparing gold oxide by precipitation with KOH, and also the precautions to take to avoid the product being caustic. The precipitation with a solution of tin required that this metal be in a fixed state of oxidation, otherwise the product would vary both in nature and quantity (Duportal and Pelletier, 1811).

Oberkampf provided additional information about the dissolution of gold in aqua regia and HCl, the precipitation of gold by streaming hydrogen, hydrogen sulfide, or phosphine, through a solution of gold chloride, or by mixing the latter with alkalis. (Oberkampf, 1811). The latter result contradicted Vauquelin's claim that the alkalis did not act upon a water solution of gold chloride. Oberkampf thought he could account for this difference by the excess of acid of the solution, which gave rise, with the precipitating alkali, to the formation of a great quantity of triple salt, indecomposable, at least totally, by an excess of base. His results indicated that the cold method produced no precipitate in the space of several minutes, even with ammonia, when the excess of acid was sufficient. Oberkampf analyzed the gold oxide provided by different methods and concluded that it contained, by weight, 90.9% gold and 9.10% oxygen. He also found that the triple salts of gold and sodium chlorides were very soluble and little deliquescent, and that they crystallized with difficulty. The precipitates of gold by tin chloride differed much according to the circumstances. For example, mixing very concentrated solutions of gold chloride and tin chloride at the minimum, in whatever proportion, resulted in the precipitation of metallic gold. In the presence of an excess of chloride, the precipitate was blackish (Oberkampf, 1814).

The contradictory results about the action of alkalis led Figuier to conduct additional experiences of the subject and to prove that the excess of acid of the solution of gold did not prevent the precipitation of the oxide by an alkali. Nearly the same amount of gold oxide was obtained in both the acid and neutral cases (Figuier, 1816ab).

Figuier dissolved 6 g of dry gold chloride in 150 g of distilled water and separated the yellow filtrate into two equal parts. Into one, he added 4 g of HCl and then neutralized both solutions with aqueous KOH. This turned the yellow color into deep red (as reported by Vauquelin). A short time afterwards, the liquors became turbid, and a flaky gray precipitate was developed; these precipitates increased after some time and became deeper. After forty-eight hours, they appeared to be at their maximum of augmentation and color. Figuier observed no difference between the two samples, except that the need to employ a greater portion of KOH in the acid sample of gold chloride (Figuier, 1816ab).

In a following paper, Figuier described the different experiments he had conducted to prepare the crystalline triple salt of gold and sodium chloride and to demonstrate that it was really a salt and not a mixture of the two chlorides. In this manner, he could use gold chloride as a medicine, while avoiding its caustic taste when used alone (Figuier, 1820, 1821, 1822).

### *Chemical analysis of purple thistle*

Figuier wrote that the scarcity of cinchona, its excessive price, and the fears of seeing this precious bark completely lacking, had urged physicians and chemists to search for native plants to be used for the same purposes. One of the promising plants was *Cardus stellatus* (*Centaurea calcitrapa*, L.), known for a long time, as a melting, aperitive, diuretic and particularly, febrifuge. These properties led Figuier to carry experiments to determine the principles contained in this plant (Figuier, 1809a).

The plant, collected at the time of its flowering, was separated from the woody stems, dried, and pulverized. The resulting green material had a herbaceous nauseating odor; 60 g of this powder were macerated with agitation, with 0.5 kg of distilled water for twenty-four hours. The filtrate was bitter and reddened tournesol paper, indicating the presence of a free acid. Mixed with various reagents, it produced colored precipitates, as follows: (1) barium nitrate, silver nitrate, and ammonium oxalate; white; (2) lead acetate; yellowish white (3) gallnut tincture; a fluffy gray deposit; (4) tannin; dark fawn; and (5) ferric sulfate; bluish green. In addition, it did not react with the antimony potassium tartrate solution, and the gelatin solution slightly altered its transparency only after several hours. These results indicated the presence of a sulfate, a chloride, calcium, animal matter, a resin-like substance, and the absence of tannin (Figuier, 1809a).

In another experiment, 120 g of *Calcitrapa centaurea* were digested for 48 hours in 700 g of distilled alcohol. The deep red filtrate had a very bitter flavor; mixed with water, it became milky. It was evaporated in a sand bath until the consistency of a thick extract, weighing 4.4 g, and being deliquescent. It was redissolved in alcohol and evaporated to the consistency of clear syrup. Mixed with distilled water, it became milky white. After standing for several hours, the floating liquid was separated from the resinous deposit formed and found to redden tournesol tincture. The liquid reacted with crystallized citric acid and oxalic acid producing a crystalline white precipitate and with mercuric nitrate, a deposit composed of flaky crystals. All these properties indicated the presence of the potassium acetate, which was probably the reason for deliquescence. The residue of the plant, which had been treated with alcohol, was digested in distilled water. The filtrate was mucilaginous, little colored, and slightly flavored. Upon evaporation, it left an extractive brown material, not very tasty, soluble in water, and slightly soluble in alcohol. Treated with twice its weight of nitric acid, it supplied mucous acid. The ashes of the plant were found to contain calcium sulfate, chlorides, and silica (Figuier, 1809a).

From all these results. Figuier determined that *Calcitrapa centaurea* contained (1) A woody material; (2) a gummy substance; (3) a resin-like material; (4) an animalized substance; (5) potassium acetate, (6) potassium and calcium sulfates; (7) calcium and potassium chlorides; (8) a green coloring matter; (9) a small amount of acid, probably acetic acid; and (10) a little silica (Figuier, 1809a).

### *Chemical analysis of chickpeas*

According to Figuier, the plant chickpea (*Cicer arietinum*) presented an interesting phenomenon: When in full vegetation, its stem, its leaves, and the envelope of its seeds were covered with a large quantity of hairs, which exuded a transparent, colorless and odorless liquor (Figuier, 1809b). Several scientists had analyzed this fluid and agreed it



was acidic but offered conflicting opinions regarding its composition, for example, Nicolas Deyeux (1744-1837) claimed it was liquid oxalic acid (Deyeux, 1798); Pierre Dispan believed it was a new acid, different from all known vegetable acids, which he proposed naming *ciceric acid* (Dispan, 1799), and Louis Nicolas Vauquelin (1763-1829) postulated that this acid was actually a mixture of 10% of oxalic acid and 90% of malic acid (Vauquelin, 1800). No other chemical examinations had been done, although chickpea was used as food, its fruit recommended as a medicine for treating renal colic, jaundice, bilious and atrabiliar diseases, and roasted, recommended as a replacement of coffee. Figuier added that Chrestien had suggested that he carry a more detailed examination of the seeds of the plant (Figuier, 1809b).

Figuier mixed 400 g of chickpeas flour with enough water to form a paste, which yielded 140 g white starch, quite soft to the touch, and tasting very similar to ordinary starch. The accompanying liquid was viscous, tasteless, had a nauseating smell and reddened tournesol tincture. Different reagents produced the following-colored precipitates, and their meaning: (1) lead acetate, white, and soluble in acetic acid; malic and oxalic acids (2) limewater, white; phosphoric or oxalic acid; (3) alcohol, fluffy; mucous (4) mercuric chloride, white; albumin; (5) acids, white coagulum; mucous (6) gallic acid, slightly dark brown; vegeto-animal substance (7) ferric sulfate, brown; (8) silver nitrate, dirty white; chloride; and (10) tannin infusion, slight turbidity; albumin. Additional tests signaled the presence of a gummy matter, a material analogous to gluten, an oily body, and a resin. The infusion produced from roasted chickpeas offered similar phenomena as the infused unroasted peas, with the difference of the flavor, which was bitter. Calcination of the peas in a platinum crucible produced an ash that reddened turmeric paper and did not react with ammonia or with ammonium oxalate, troubled limewater, and precipitated a solution of silver chloride.

Figuier concluded that the seed of *Cicer arietinum* contained: (1) starch; (2) albumin; (3) vegeto-animal material; (4) mucous membrane; (5) a resin-like substance; (6) a fixed oil; (7) potassium and calcium malate; (8) potassium chloride; (9) calcium and magnesium phosphate; and (10) iron (Figuier, 1809b).

### *Discoloration with animal carbon*

The discoloration of substances with animal carbon is probably the most important scientific contribution of Figuier.

In 1790, Lowitz read a memoir to the Société Libre d'Économie of Saint-Petersburg, where he reported that vegetable carbon had the property of eliminating the odor of water and of putrid animal substances (Lowitz, 1793). Everyone knew that water was very liable to spoil itself and to contract by putrefaction, qualities, which rendered its use dangerous, a circumstance especially significant in sea voyages. It was undoubtedly of a major interest to know how one could prevent the corruption of water and to make very pure as that which had entered putrefaction. Lowitz added, during his study of the depurative property of coals, he had noticed that they also had the property of almost suddenly removing from the most decomposed water its bad odor. This led him to conduct additional experiments to find a method of preserving water against decomposition using vegetable charcoal (Lowitz, 1793).

Properly filtered pure water was not subject to corruption, but it was very difficult to keep it for a long time in this state, because of its dissolving property. Large quantities

of water were stored in wood casks, which had the great disadvantage of dissolving in water many mucilaginous and extractive molecules, which accelerated their corruption. As written by Lowitz, "these molecules, in a divided state, present an innumerable quantity; to be alive, whose almost continual and uninterrupted destruction and regeneration impart to water that degree of corruption and putrefaction, which makes its use so dangerous. It is therefore not the water itself, but the substances which it holds in solution, and which, by their continual decomposition, dispose it in a state of corruption or putrefaction." He recommended that the barrels be maintained in the greatest cleanliness possible, since the least residue of the parts previously corrupted acted like a real leaven. For this purpose, it was necessary to wash them with hot water and sand or any other substance capable of removing the mucilaginous parts, and lastly using a certain quantity of charcoal powder, preferable mixed with concentrated sulfuric acid, which would effectively remove any putrid or moldy odor with which the interior of such barrels may be infected. The best mixture was 42.52 g of powdered charcoal and 24 drops of concentrated sulfuric acid for each kilo or rotten water, which did not impart water a perceptible taste of acidity. Charcoal powder and sulfuric acid were two anti-putrid substances, the first prevented the yellow color, which water ordinarily acquired with time, and the acid helped to clarify the water, which the charcoal powder employed by itself often made cloudy (Lowitz, 1793).

Lowitz's publication led many chemists to repeat his experiences, with mixed results. The most successful were related to the purification of potable water, using the information that the process was improved by previously humidifying the charcoal and exposing it to the action of solar light.

One of these scientists was Pierre Figuiet. Repeated experiments showed him that vegetable coal had a very weak and slow bleaching power. This led him to investigate the possibility of repeating Lowitz's experiments using other varieties of charcoal. Among them, he utilized animal bones to prepare what he called *carbon animal* (animal charcoal) *noir d'os* (bone black), or *noir animal* (animal black). These were prepared by calcining the bones in a closed crucible, followed by pounding the product in a mortar. Figuiet was surprised at the results: the new material bleached hot red wine almost instantly. This led him to study the bleaching of red *vinegar* in more detail because vinegar was the oldest and most common vegetable acid (Figuiet, 1811b). The easy of its preparation in large scale and modest price, multiplied its use in domestic economy and industry, appearing as one of the constituent principles of many chemical and pharmaceutical preparations. The sale of this acid was one of the mayor branches of French commerce and had encouraged chemists to search for ways of perfecting its manufacture, purification, and applications (P. Figuiet, 1811b, L. Figuiet, 1869, 1895).

Vinegar prepared with wine was preferable to that obtained by the fermentation of other vegetable substances; of this, two varieties (red and white) were available commercially. Red vinegar came from the acidification of red wine, while white vinegar came from white wine. The latter was more appreciated because it generally fulfilled better the indications that the one proposed of its use, it contained less extractive coloring matter than red vinegar. This explained efforts to deprive the latter of part of this coloring matter, to bring it closer to the qualities of white vinegar; one discolored, even this one which, in fact, it had a yellowish red color (Figuiet, 1811b).

The various processes for rendering vinegars less colored than in their natural state were based on the use of an entrainer for dragging the coloring matter away. This could be done by adding one or two egg whites to a liter of vinegar and boiling the mixture; the coagulated albumen carried with it part of the coloring matter. The cooled liquor was then filtered through Joseph paper (a highly absorbing variety), and the filtrate was vinegar less colored than it was before having undergone the operation. Another possibility was mixing a glass of milk with five or six liters of heated vinegar; the solidifying casein entrained a significant part of the coloring matter, and the filtrate was less colored vinegar less colored. A third process used the fact that the pomace of white grapes also had the property of discoloring the vinegar; this method was considerably used in large establishments, notably in Sète (Occitanie, in southern France), where there was a considerable trade in white vinegar which was sent to the North. In this process, the white grape pomace was put in large vats, which were then filled with vinegar and left alone for a few days. Afterwards, the slightly discolored vinegar was drained through an opening in the lower part of the vats and added to another vat, which contained marc, which has not been used. The process was repeated as many times as necessary until the desired degree of bleaching was achieved (Figuiet, 1811b).

As mentioned above, Figuiet process was based on the use of animal charcoal as bleaching material. It was very easy to use, it was very economical, and it could be practiced in both small and large batches. The resulting vinegar was colorless as pure water. Lowitz's vegetable charcoal could also be used, but was less effective and much slower than animal charcoal. Basically, the procedure consisted in adding 45 grams of bone charcoal to one liter of red vinegar, at room temperature and under agitation. The bleaching process was completed in two or three days. The filtered liquid was clear as water and had not lost its flavor, odor, or acidity. The same procedure could be applied in large scale by throwing the animal charcoal into a barrel containing the vinegar. The resulting color could be regulated by using different amounts of animal carbon. Figuiet remarked that the resulting light color vinegar could be kept for months without showing signs of alteration. It could be flavored by addition of appropriate plants or parts of them, before operating its discoloration, or by adding to the bleached vinegar a small quantity of alcohol loaded with the adequate aromatic principle (Figuiet, 1811b).

Figuiet found that the same procedure could be applied to red wine, distilled brandy, and the black residue of the preparation of sulfuric ether. The last case was particularly interesting because this residue contained the greater part of the acid, which has been employed in its preparation. Several chemists had proposed methods of purifying it in order to reuse it in subsequent operations. Figuiet tried them all and found that none resulted in full recovery, but that animal charcoal allowed achieving the goal. Basically, his proceeded consisted of the following steps: The residue was first mixed with a quantity of water equal to its own weight to make it less viscous and allow elimination by filtering of the carbonaceous and oily matter formed by the action of the acid on the alcohol. A liter of the filtrate was mixed with 50 g of bone black and stirred occasionally for two days. After this time, the new filtrate was completely discolored; evaporated in a glass vessel eliminated the excess water and left a residue, which could be used to prepare a new batch of ether (Figuiet, 1811b).



The publication described in detail the preparation of animal charcoal. The compact parts of the bones of oxen or of sheep were introduced in a crucible and the apparatus sealed except for a small hole left to allow release of the volatile material. The crucible was placed in a forge furnace, gradually heated until red-hot, and left in this state until the flame produced by the combustion of the oily and gelatinous parts of the bones had ceased. The small hole was reduced in size and the temperature increased substantially. This step eliminated the remaining volatiles. The crucible was left to cool, and the charcoal crushed in a mortar. Ivory black was produced by the same procedure. Figuiet found that although both charcoals lost their activity after being used once, the property could be recovered by heating them strongly in a closed vessel (Figuiet, 1811b).

Figuiet also applied his method to produced bleached Seignette salt, sodium phosphate, and potassium acetate (Figuiet, 1812b, 1813).

An additional paper described the discolorizing of a variety of substances using animal carbon and agents such as, magnesium oxide calcined, alumina calcined, and calcium carbonate. The substances tested were madder, Campeche, cochineal, wine, saffron, toasted chicken peas, and indigo (Figuiet, 1818).

Louis Figuiet, the nephew of Pierre Figuiet, described in detail the use of animal black in the bleaching of cane and beet sugar, and the benefits it brought to this industry (L. Figuiet, 1860, 1895).

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