La química en la historia, para la enseñanza

Pierre Joseph Macquer

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Resumen
Pierre Joseph Macquer (1718-1784) es bien conocido a través de sus investigaciones acerca del platino, el arsénico y sobre el teñido textil, pero en particular, por su monumental *Diccionario de la Química*. Fue el primero que investigó en forma detallada las propiedades del platino y trató de fundirlo (sin éxito) usando todas las técnicas disponibles en su tiempo. Su *Diccionario de la Química* fue el primer intento en la historia de esta ciencia para organizar en forma sistemática toda la información disponible entonces acerca de los elementos y sus compuestos, así como acerca de las teorías químicas y físicas acerca de la constitución de la materia. Macquer fue durante toda su vida un firme partidario de la teoría del flogisto y trató arduamente de reconciliarla con los descubrimientos de Lavoisier.

Abstract
Pierre Joseph Macquer (1718-1784) is well known for his work on platinum, arsenic, and dyeing, but particularly for his monumental *Dictionary of Chemistry*, the first of his kind. He was the first to thoroughly investigate the properties of platinum and try to melt it (unsuccessfully) by the procedures available at his time. His *Dictionary of Chemistry* was the first attempt in the history of chemistry to organize in a systematic form all the information available in the eighteenth chemistry about the elements and their compounds, as well as and chemical and physical theories about the constitution of matter. Macquer remained all his life a strong supporter of the phlogiston theory, and tried hard to conciliate it with Lavoisier's findings.

Life and career

Pierre Joseph Macquer was born in Paris, on October 9,1718, the eldest son of Joseph Macquer and Marie-Anne Caillet.

There have been some arguments about the origin of the Macquer family in France, some claiming that they descended from a noble Scots family who had lost all for the Stuart cause and the Catholic faith (Partington, 1962); others that the name Macquer is not a corruption of a Scots name. McKie (MacKie, 1949) did a thorough search of the parish registers of St. Germain-en-Laye and found the name of Maguire, the family name of Lord Enniskillen, and also various spellings names such as Magoire, Mackower, Magower, Makaire, Magaire, etc., as well Macquer.To him, the curious variety of spelling in the registers of St. Germain leaves no doubt of the possibility of the corruption of Maquire to Makaire. French pronunciation would make no difficulty in corrupting Maguire to Macquer. The established alteration in spelling and the descent from a noble family suggest, therefore, that Macquer may have been a descendant of the Maguires, Barons of Enniskillen, and of Irish, rather than Scots, origins. From this information McKie concluded that the Macquer family came to France, not after the rising of 1715, but with James II in 1689, and were thus, of Irish origin.

Both Pierre and his younger brother, Philippe (1720-1770), received their early education from Charles Le Beau (1771-1778), whose literary training laid the foundation of the clarity and elegance of style, which characterized the writings of both brothers. Philippe went on to become a lawyer, a profession that abandoned because of health reasons, to become a historian (Vicq d'Azyr, 1782).

Pierre showed an early interest in science and decided to devote himself to research. However, his parents insisted on his qualifying in a profession, and he chose medicine. He studied at the medical faculty of the University of Paris, graduated in 1742 and practiced medicine for only a few years, being at one time doctor for the poor of the parishes of St.-Nicholas and St.-Sauveur. At the same time he studied chemistry under Guillaume François Rouelle (1703-1770) and soon began to do research.

Rouelle had been appointed *demonstrator* at the *Jardin du Roi* (today *Muséum National d'Histore Naturelle*) in 1742 by George-Louis Leclerc Buffon (1707-1788) to serve under Louis Claude Bourdelin, the chair professor of chemistry. Eventually he also started giving classes on theory, independently of Bourdelin. Both Bourdelin and Rouelle's courses followed essentially the outline given by Nicolas Lemery's (1645-1715) in his book *Cours de Chymie* (first edition, 1675), a little more than a practical handbook of pharmaceutical preparations. Rouelle was considered to be an exceptional teacher and eventually exerted a critical influence on French chemistry. His central tenets were the phlogiston theory and that air can be considered a chemical constituent of matter. His most distinguished students included Antoine Laurent de Lavoisier (1743-1794), Macquer, Jean D'Arcet (1725-1801), Denis Diderot (1713-1784), Jean-

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Jacques-Rousseau (1712-1778), Anne Robert Jacques Turgot (1727-1781), and Pierre Bayen (1725-1798); this roster led to Rouelle being considered the founder of French chemistry.

Bourdelin passed away in1777 and Buffon requested and obtained that the chair in chemistry at the Jardin du Roi be assigned to Macquer. He remained in the chair until his death 1784. It is said (Viel, 1986-1990) that he was a remarkable teacher and ardent follower of the phlogiston theory.

Since there was few experienced and able chemists competing for places in the *Académie Royale des Sciences*, Macquer was elected *adjoint chimiste* in the chemistry section in 1745, and in the same year he read his first experimental memoir on the solubility in alcohol of different oils (Macquer, 1745). He was promoted to *associé chimiste* in 1776, and *pensionnaire* in 1772, replacing Charles Marie de la Condamine (1701-1774). Eventually Macquer became *Directeur-Suppléant* (1773) and *Directeur* of the Académie (1774).

Macquer's medical background and interest in chemistry led to his becoming a founding member of the *Société Royale de Médecine* (1776). From 1750 on he was *Censeur Royale* (Royal Censor) of the government Bureau de Commerce, which controlled French trade and industry, and prepared many reports on memoirs and books on new discoveries and remedies offered them, chemistry, medicine, and natural history, submitted to the two societies he was a member. It was in this advisory capacity that in 1783 he approved Claude-Louis Berthollet's (1748-1822) proposal to build the first factory in France for the manufacture of sodium carbonate.

Together with Joseph Marie François Lassone (1717-1788) and others he published a report on *Traitment Contre le Ténia* (Treatment against taenia) (De Lassone, 1775). He enlarged his literary activities in 1768, when he succeeded Louis Charles Henry Macquart (1718-1784) as a scientific and medical member of the editorial board of the *Journal des Sçavans*, a state-owned monthly journal that published book reviews and occasional original memoirs dealing with physics, medicine, pharmacy, surgery, chemistry, astronomy, and natural history. He remained in that position until 1776. The broad scope of Macquer's activities and the key positions he occupied explains the significant influence he exerted in the scientific life of Paris, in spite of the relatively small number of papers he published.

Macquer began academic life in 1752 when he was elected professor of pharmacy in the Faculty of Medicine in Paris. A few years later, in1757, he initiated a long and fruitful collaboration with Antoine Baumé (1728-1804) when they began teaching a joint course in chemistry in Baumé's private laboratory located in Paris (Julien, 1992). This course was repeated during the following sixteen years and gave place to a joint publication, *Plan d'Un Cours de Chymie Expérimentale et Raisonnée, avec un Discours Historique Sur la Chymie* (Macquer, 1757), which followed very closely Macquer's book *Élemens de Chymie Theorique*, published in 1749 (Macquer, 1749). Each of these courses included about two thousand experimental demonstrations. At that time Baumé was *maître-apothicaire* and professor at the *École de Pharmacie* and the author of a large number of memoirs and books, particularly *Chymie Expérimentale et Raisonnée* (Paris, 1773, 3 vols) (Wisniak, 2001). The fee for the course was 86 livres per year for new students, and 80 livres for the ones taking it for the second stage. The syllabus started with a general discussion of the development of chemistry since the time of Paracelsus (1493-1541) and followed with a discussion of the four elements and the principal known compounds: acids, salts, earths, mineral waters, the products of the action of heat on animals and plants, and finally, the analysis of biological products resulting from fermentation and putrefaction. Some of these subjects were given with extensive detail. For the example, the analysis of animal matter included milk, blood, lymph, fats, bones, marrow, flesh, products helpful for digestion and those resultant from digestion (saliva, pancreatic sugar, bile, chyle); compounds that served for generation (eggs), and depositions such urine, etc.

By 1773, when the joint courses with Baumé ended, Macquer started teaching at the Jardin du Roi. Claude Bourdelin (1621-1699) had been professor of chemistry since 1743, but because of poor health, he was usually replaced by Paul Jacques Malouin (1701-1778). Buffon, the intendant, then granted Macquer the chair.

Like the majority of the chemists of his time Macquer did most of his work in the area of mineral chemistry; organic chemistry was little developed because the experimental techniques were ill adapted to the isolation and analysis of delicate organic materials. Macquer's first memoir was related to the possible relation between the changes that occurred in oils during distillation and solubility of various products in it; this experimental and theoretical memoir contributed little to subject (Macquer,1745). Even as late as 1778 he thought that fatty acids liberated by mineral acids from soaps, though freely soluble in alcohol, were practically identical with the original oils. In an investigation of the solubility of carefully dehydrated salts in alcohol he found that chlorides and nitrates are more soluble than sulfates; salts in which the acid is less strongly united with the base are more soluble (Macquer, 1776, 1778). Anyhow, his quantitative results were of value to chemists who used alcohol extraction in the analysis of residues from evaporated mineral waters.

Several of Macquer published papers are the outcome of his many official appointments, e.g., those on the dyeing of silk (Macquer, 1768), on the chemical examination of Prussian blue (Macquer,1752), on the mercurial tincture of the

Comte de la Garaye (Macquer, 1755); on the fusibility of clays (Macquer, 1758); and on the effect of strong heat on various substances (Macquer, 1767).

Macquer collaborated with Jean Hellot (1685-1766) and Antoine Baumé (1728-1804) in developing the famou French porcelain fabricated at the Royal Porcelain Works at Sèvres. The original Chinese porcelain was initially imitated at Meissen, Germany, in 1715, and eventually after much searching and testing by Macquer and Baumé, in 1765 the necessary kaolin was found in France, at St. Yrieix, near Limoges. Four years later, the production of the first true French porcelain was begun.

Macquer did most of his work while the phlogiston theory was the accepted explanation for many chemical and physical phenomena. Macquer was the first Frenchman who gave it full recognition and taught it as the central subject in chemistry. He lived to see Lavosier develop the alternative oxygen theory of combustion and work hard to reconcile the new experimental evidence to the old theory

In 1776 Macquer aided by Joseph Aignan Sigaud de Lafond (1730-1810), demonstrated for the first time the formation of water during the combustion of hydrogen.

Macquer was a member of the Madrid Academy of Medicine and also of the Academies of Stockholm, Turin, and Philadelphia. Between 1745 and 1780 he published in all twenty-three memoirs on his own name and twelve in collaboration with other workers.

Macquer married in 1748 a lady of good family but poor means. In the last months of his life he suffered from headaches, fainting, and palpitation. All attempts at treatment failed. He died on February 15,1784, in Paris, at the age of 65, leaving a widow and two daughters. In accordance with his wishes a post-mortem examination was held, and his death was found to have been due to "l'ossification de l'aorte et des concretions perreuses dans les cavités du coeur" (arteriosclerosis and arterial hypertension). Antoine-François Fourcroy (1750-1809) succeeded him at the Jardin du Roi and Claude-Louis Berthollet (1748-1822) as Director of the Dyeing Industries.

Scientific activities

Macquer's contributions can be divided in two large groups, one corresponding to his books and the other to his pure and applied chemistry works. These will be discussed separately.

A. Books

1. *Elemens de Chymie* (Macquer, 1749)

The book *Élemens de Chymie Théorique* appeared for the first time in Paris in 1749. It was in one volume containing 350 pages, illustrated with plates representing chemical apparatus, and a reproduction of Étienne François Geoffroy's (1672-1731) table of chemical affinities. Lemery's book was still in vogue in France, having been translated into English, German, Latin, Italian, and Spanish. Macquer's book did not make much impression; but it contained the principles of chemistry, the different categories of chemical bodies and their properties, but more important, it did not describe their pharmaceutical properties (in spite of Macquer being a physician). It was the first time that the subject of chemistry was presented to beginning chemists, as a science independent of medicine and pharmacy, and in a simple language (Ahlers, 1969).

This book was designed for general reading by people who had insufficient time to begin to study more detailed treatises, and began by assuming no knowledge of chemistry on the part of the reader. Macquer begun his book with a definition of chemistry, because in the preface he insisted in the difference between chemistry and alchemy: "The subject and purpose of chemistry is the separation of the different substances that compose a body, the particular examination of each of them, their properties and analogies, their further decomposition if possible, their combination with other substances; their reaction to recover the original body with all its properties…" (Ahlers, 1969).

The description of matter begun by considering the four elements, earth, fire, air, and water, and went on to deal with the next simplest class, namely, saline substances, mineral acids, fixed and volatile alkalis, and their different combinations, and metals. According to Macquer metals were not more complicated that saline substances. The following chapters were dedicated to the study of oils (vegetable, animal, and mineral), and fermentation (spirituous, acid, and putrid). The final chapters dealt with analysis, with an explanation of Geoffroy's Tables of Affinity: "Pour rassembler sous un seul point de vue les verités les plus essentielles & fondamentales dispersées dans tout l'Ouvrage" (to assembly under one point of view all the most essential and fundamental truths dispersed in the book).

Air, Macquer said, had weight, and entered into the composition of many substances, especially animal and vegetable, and was given off in analysis. Earth was the non-volatile constituent of earthy residues. Fire was probably the principle of fluidity; possible air would be solid if it lost sufficient heat. The particles of fire were small atoms, which could penetrate matter. Phlogiston was fire combined with some other substance to give a secondary element. The flammability of substance was a proof that it contained phlogiston, but the converse was not true, since certain metals abounded in phlogiston, although they were not inflammable (Coleby, 1938).

2. *Élemens de Chymie Pratique*

Two years later, Macquer published a new book, *Les Élemens de Chymie Pratique* (Macquer, 1751), in two volumes and containing more than one thousand pages. It constituted the continuation of the first book. In it the theoretical principles developed before, were used for completing the knowledge of chemistry. This book was divided into three parts dealing with the mineral, vegetable, and animal kingdoms respectively. The first part was further divided into three: operations on naturally occurring saline substances, on metals, and on semi-metals.

In 1756 the two books were joined in a three-volume edition, one volume for theoretical chemistry and two for applied chemistry, containing about 1500 pages (Macquer, 1775).

3. *Dictionnaire de Chymie* (Macquer, 1766)

Unquestionable, Macquer's greatest work is the *Dictionnaire de Chymie*, published in two editions in Paris, in 1766 and 1778, and should be considered the first attempt to embrace in a systematic and precise synthesis all the chemical knowledge of his time, both pure and applied (Figure 1). This dictionary constitutes a solid source of information about the chemical knowledge of that time, the different theories in vogue, and the discoveries of many scientists. Although Macquer's was a well-known figure, for some unknown reason the first edition was published anonymously.

The dictionary contributed significantly to the unification of the scattered chemical and technological information known before its appearance. It immediately became very popular; it passed through several editions in French and was translated into German (1767), English (1771), Danish (1771-1772), and Italian (1783-1784). It became the model for those that followed. William Nicholson's (1753-1815) famous *A Dictionary of Chemistry* (London, 1795) was patterned after Macquer's (McKie, 1957; Neville, 1966; Smeaton, 1954).

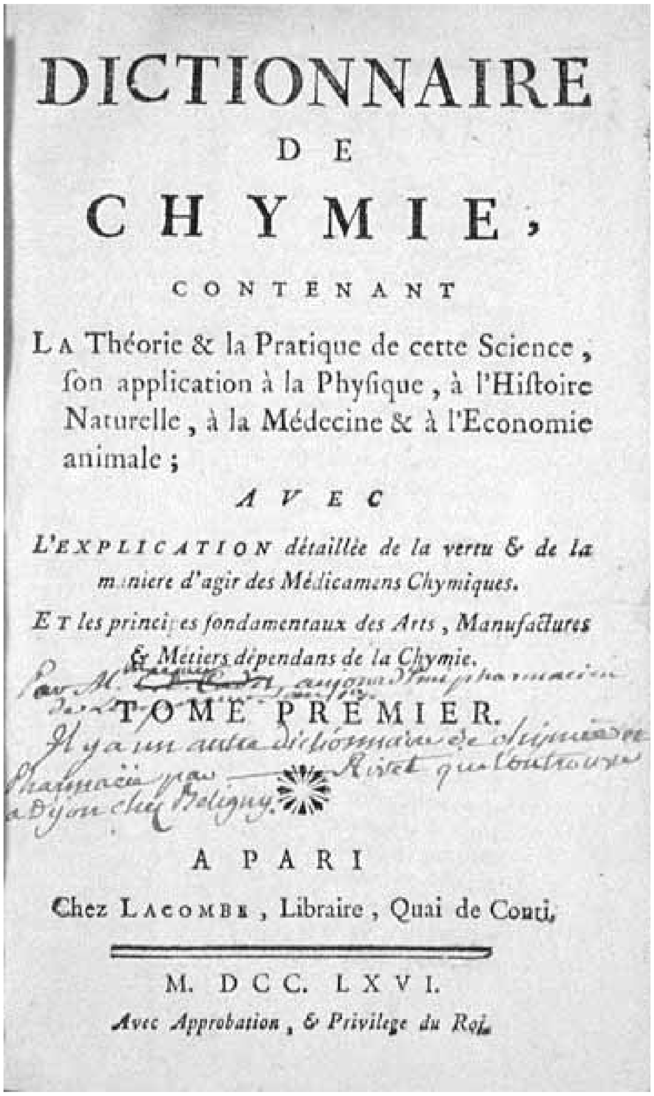
DICTIONNAIRE
DE
CHYMIE,
CONTENANT
La Théorie & la Pratique de cette Science, son application à la Physique, à l'Histoire Naturelle, à la Médecine & à l'Economie animale;
AVEC
L'EXPLICATION détaillée de la vertu & de la maniere d'agir des Médicamens Chymiques.
ET les principes fondamentaux des Arts, Manufactures & Métiers dépendans de la Chymie.
Par M. ~~[illegible]~~, aujourd'hui pharmacien [illegible]
TOME PREMIER.
Il y a un autre dictionnaire de chymie et pharmacie par — Rivet qu'on trouve a Dijon chez Boligny.
A PARI
Chez LACOMBE, Libraire, Quai de Conti,
M. DCC. LXVI.
Avec Approbation, & Privilege du Roi.

Figure 1. Macquer's *Dictionnaire de Chymie,* (1766).

The first French edition contained about 1300 pages, but it was more a compendium of chemistry than a dictionary. It carried 500 distinct articles alphabetically arranged, and adequately cross-indexed. It started with the entry *Acescent* (a word describing a substance that is turning sour) and ends with *Zinc* (Neville and Smeaton, 1981). The dictionary was preceded by a 22-page historical introduction, which briefly traced the progress of chemistry from its origins to the time of George Ernst Stahl (1660-1734) and Herman Boerhaave (1668-1738). Macquer's high regard for the phlogiston theory was evident, and he ended the historical introduction on a note of encouragement: "Nous avons l'avantage de voir enfin les plus beaux jours de la Chymie. Le goût de notre siecle pour les matieres philosophiques, la glorieuse protection des Princes, le zele d'une multitude d'amateurs illustres & éclairés, le profond favoir & l'ardeur de nos Chymistes modernes, que nous n'entreprenons pas de louer, parcequ'ils font au-dessus de nos éloges, tout semble nous promettre les plus grandes & les plus brillans succès. Nous avons vu la Chymie naître de la nécesssité, recevoir de la cupidité un accroissement lent & obscur; ce n'est qu'à la vraie Philosophie qu'il etoit réservé de la perfectioner" (We now have the advantage of seeing the best days of chemistry. The taste of our age for philosophical matters… and… the profound skill and ardour of modern chemistsseem altogether to promote the greatest and most brilliant success).

In his Dictionary Macquer proposed a general reform of chemical nomenclature that would put order in the chaos that existed on the subject. For example, he proposed to systematize the nomenclature of metallic salts by giving to all those of the same acid a common name, with an indication of the metal combined with the acid: "It would be proper to give the same denomination *vitriol* to all vitriolic salts with a metallic base, ant to name, for example, *vitriol d'or*, the vitriolic salt composed o vitriolic acid and gold; vitriol *d'argent, ou de lune*, the salt resulting from the union of the same acid with silver, and so on. Perhaps it would even be correct to include under the general name *vitriol*, all vitriolic salts whatsoever" (Macquer, 1776, vol 2, page 673).

Similarly he suggested the name *nitre* or *sel nitreux*, for all salts of nitrous acid (now called nitric acid); *sel marin*, or simply *sel*, for all salts of marine acid (hydrogen chloride; *sel tartareux*, or *tartre soluble*, *sel acéteux,* and *sel phosphorique*, for those of tartaric, acetic, and phosphoric acid respectively. (Macquer, 1776 vol 2, p. 430-436).

According to Macquer: "Metallic substances form a class of bodies not very numerous, of great importance in chemistry, medicine, arts, and the ordinary affairs of life." The physical and chemical properties of metals are discussed in detail; density, opacity, lustre, hardness, ductibility, malleability, fusibility, solubility in acids and bases, reactivity with salts, formation of alloys and amalgams, etc.

His definition for acids was: "simple saline substances" that could sometimes be obtained solid, but were usually found in the liquid state. According to Macquer, acids were "obtained from certain animal matters; as butter, fat, blood, ants, and most flies".

Macquer introduced the concept of *intermediate* to describe a reagent that could either generate a combination otherwise impossible (e.g., alkali by combining with oil mediated the combination of oil and water) or decompose a union otherwise resistant to disunion (e.g., sulfuric acid separates nitric or muriatic acid from combination with alkali) (Melhado, 1983).

A second edition of the Dictionnaire was published in Paris in 1778, by the imprimerie "*de Monsieur*", because it was under the patronage of the Comte de Provence, a young brother of Louis XVI. The two volumes contained about now 1,600 pages, including a 200-page index. It included the material included in the previous edition, enlarged, as well as additions reflecting the new information acquired in between.

The contents of the Dictionnaire are important today because they reflect the state of the art at that time and the prevailing scientific ideas. According to Macquer:

(a) Elements

"On donne en Chymie, le nom d'élemens aux corps qui sont d'une telle simplicité, que tous les efforts de l'art sont insuffisans pour le décomposer, & même pour les causer aucune espèce d'altération; & qui d'une autre part, entrent comme principles, ou parties constituantes, dans la combinaison des autres corps, qu'on nomme pour cette raison corps composes" (In Chemistry, we call elements those bodies having such simplicity that all the efforts of the art are not enough for decomposing them or for causing them any alteration, and on the other hand, they enter as principles or constituent parts, in the combination of other bodies, that for this reason are called composed bodies).

(b) Constitution of matter

"Les corps auxquels on a reconnu cette simplicité sont le feu, l'air, l'eau, & la terre la plus pure; parcequ'en effet les analyses les plus complettes & les plus exactes, qu'on ait pu faire jusqu'à présent, n'ont jamais produit autre chose en dernier ressort que les unes ou les autres de ces quatre substances, ou toutes les quatre suivant la nature des corps qui ont eté décomposés. Il est très possible que ces substances, quoique réputées simples, ne le soient pas, qu'elles résultent de l''union de plusieurs autres substances plus simples. Mais comme l'expérience n'apprend absolument rien sur cela, on peut sans aucun inconvénient, on doit même regarder en Chymie le feu, l'air, l'eau, & la terre, comme des crops simples; parcequ'en effet ils agissent comme tels dans toutes les opérations de cet art" (the bodies for which this simplicity has been recognized are fire, air, water and the most pure earth, for by the most complete analyses, which have been made, nothing has ever ultimately produced but some one, or more, of these four substances, according to the nature of the decomposed bodies. It is very possible that these substances although reputed as simple, they are not, that they are the result of the union of many other more simple substances. But since does not teach nothing about it, we can without inconvenient to regard in chemistry fire, air, water, and earth, as simple bodies, because they acted as such in all the operations of this art).

(c) Alchemy

The entry about alchemy is very short and is quoted here in its entirety: "Ce terme a été employé par les prétendus *Adeptes*, & par les chercheurs de *Pierre Philosophale*, pour désigner la Chymie par excellence, don't ils se flatten que la connoissance est réservée a eux seuls. Les Adeptes regardent la Chymie comme une science vulgaire, qui contient à peine les premiers Élemens de la science mystérieuse de l'Alchymie; mais jusque'à present ils n'ont rien produit, qui, au jugement des personnnes sensées, puisse donner le moindre fondement à une pareille prétention. Le vrais Chymistes regardent l'Alchymie comme une science imaginaire, & ceux qui s'y adonnent comme des gens qui, faute d'être suffisámment instruits, quittent la réalité pour courir après l'ombre."

According to Macquer *Adeptes* "le nom que se donnent les Alchymistes, qui prétendent avoir trouvé le secret de la pierre philosophale" and pierre philosophale was "le nom que les Alchymistes ont donné à la preparation par le moyen de laquelle on peut transmuter les métaux, faire de l'or & de l'argent, opérer en un mot toutes les merveilles du grand Oeuvr".

(d) Phlogiston

The dictionary also shows Macquer's strong position regarding the controversy between those advocating the phlogiston theory and those advocating the chemistry developed by Lavosier. He was a strong supporter of the original phlogiston hypothesis, but the increased experimental evidence provided by Lavoisier led him to modify his position. Macquer explained the increase in weight postulating that the metal first lost phlogiston (as in the classical theory), and then combined with a quantity of air that exceeded the weight of the phlogiston lost. Macquer continued to affirm that combustion and calcinations released phlogiston and that it was the constitution matter of light. Exchange of phlogiston between two bodies gave place to substantial changes and it was this property that allowed distinguishing it from pure fire and to consider it as the fire element combined with another substance.

Macquer described phlogiston as "le principle inflammable le plus pur & le plus simple" (the most pure and simple inflammable principle) …"Le phlogistique doit être regardé comme le fue elementaire, combine & devenu un des principes des corps combustibles" (phlogiston must be considered as elementary fire, combined and transformed into one of the principles of combustible bodies). "Ce principle ne pouvoir être separé d'avec les autres principes des corps, & obtenu seul & pur, & que par conséquent it a été impossible de reconnoître toutes celles de ses propriétés qui lui font particulieres, & qui le distinguent de toutes les autres substances" (It cannot be separated from the other principles of bodies, nor can be obtained pure, hence it is impossible to discover all the properties, which are peculiar to it and that distinguish it from all other substances).

Macquer's strong support of the phlogiston theory may explain why the great prestige with which he was held while he was alive decreased fast after his death. The successors of the chemical revolution judged a contemporary of Lavoisier according to his acceptance of the latter ideas. Scientists that did not convert to the new chemistry were considered pariahs.

B. Pure and applied Chemistry

1. Arsenic

Macquer's first work in mineral chemistry was investigating the properties of white arsenic (arsenious oxide) (Macquer, 1746, 1748). This non-metal and certain of its salts like the sulfides and arsenious oxide, were well known by the alchemists and chemists, who between the twelfth and sixteenth century have paid special attention to it because of its property of acting on metals like mercury, forming with pure metals or their alloys, other alloys which were fusible and colored like gold or silver (*teinture des métaux*). These results stimulated their search for a possible metallic transmutation. In addition, the reaction of white arsenic with nitre (potassium nitrate) was well known. Macquer was more interested in the nature of the reaction than its chimerical hopes. He showed that distillation of the reaction product left a neutral salt resulting from the "combination de la base du nitre et de l'acide particulier qui tire son origine de l'arsenic" (the combination of the base of nitre and the particular acid…originating from arsenic). He also demonstrated that similar salts were obtained if nitre was substituted by potash or chalk. He also discovered a new group of arsenic salts, the arsenates, fact that explains why neutral potassium arsenate is sometimes called Macquer's salt.

Macquer classified arsenic as a semi-metal, since white arsenic could be converted into a semi-metallic substance by combing it with phlogiston. The calx of arsenic (white arsenic), unlike calces was volatile, was soluble in water, had a violent corrosive action if taken internally, would combine easily with metals, rendering them brittle, and moreover, it would decompose nitre, not by combining with its acid, but by displacing the acid and combining with the alkali in the same way as sulfuric and phosphoric acid and the sedative salt (boric acid) did (Coleby, 1938).

Chemists such as George Ernst Stahl (1660-1734) and Johann Kunckel (1630-1702) had reacted arsenic with nitre and other compounds and obtained dense fumes of acids and colored materials. Macquer repeated and confirmed these experiments but paid particular attention to the solid residue (*caput mortuum*) that was left in the retort after distillation (Macquer, 1748). Solution of the residue in water followed by evaporation produced fine crystals of very regular shape, quite different from those of nitre, each crystal being a rectangular prism, terminated at each end by a rectangular pyramid.

Macquer also prepared another compound of alkali and arsenic by dissolving white arsenic in a "hot solution of nitre fixed by charcoal" (i.e., potassium carbonate). Solidification of the fused compound yielded a hard and brittle solid, which

Macquer called *liver of arsenic*. This salt was readily decomposed by acids and on calcinations gave ordinary fixed arsenic. It was soluble in water, but would not crystallize out; it gave precipitates with metallic salts different from those given by other compounds of arsenic with alkalis.

The two salts described by Macquer in this first memory were potassium dihydrogen arsenate (KH_2AsO_4) and impure potassium arsenate respectively

In his second memoir on arsenic Macquer described his many attempts to obtain neutral arsenical salts with other bases (Macquer, 1748). First, he tried unsuccessfully to react sodium chloride with arsenic hoping that it would chase out the acid (*chaser l'acide*) and combine with the base. He then tested (also unsuccessfully) the possibility of achieving the reaction by adding mercury, expecting that the latter would react with the acid part and form *sublimé corrosif* (corrosive sublimate, mercuric chloride) while sodium and arsenic would yield a neutral salt. Eventually his efforts fructified and he was able to obtain a liver of arsenic (*foie d'arsenic*) by boiling the compound with a solution of marine alkali (NaOH) or by heating it together with dry vegetable alkali. Macquer wrote that he was surprised to find that crystallization yielded a sodium salt very similar to the one obtained with nitre. According to him, this similarity on crystallization indicated an analogy between arsenic and the acid of the salt (chloride). This new salt, which he named sel *neutre* (neutral salt), did not decompose when heated strongly in closed vessels or under the action of strong acids. When added to incandescent coal in an open vessel, it melted rapidly and decomposed producing large amounts of arsenic vapors. Addition of the salt to a water solution of metallic nitrates yielded colored precipitates, white for lead and bismuth, yellow for mercury, turquoise blue for copper, and strong violet for silver. Macquer explained the formation of these precipitates as an example of double affinity: the acids were unable to remove the arsenic from his salts, the additional affinity of the base of the salt for the arsenic enabled the acid of the salt to chase it out from its combination with the alkali and thus enabled the acid to combine with the latter.

Macquer also tried to react arsenic with ammonia salts. He found that ammonium chloride behaved as common salt, no reaction took place. On the other hand, ammonium nitrate decomposed as other metal nitrates did yielding what Macquer considered an unusual neutral ammonium salt (ammonium arsenate). This arsenate decomposed on strong heating and treated with a fixed alkali (including calcium) gave ammoniacal vapors and left a neutral arsenical salt of calcium. Addition of the ammonium salt to a water solution metallic nitrates yielded colored precipitates as did the sodium salt.

Carl Wilhelm Scheele(1742-1786) first prepared arsenic acid and then obtained Macquer's neutral salts by neutralization with alkalis: "When acid of arsenic is saturated with alkali of tartar… The solution affords fine crystal, which wholly resemble Macquer's arsenical neutral salt in form as well as in other properties" (Scheele, 1775). He also showed that Macquer had not been able to obtain a neutral salt by fixation of nitre in a crucible because of the high temperature used.

2. Examination of platinum

Word of the existence in Mexico of an infusible metal reached Europe as early as the middle of the sixteenth century (the fusion point of platinum is 1755°C, a temperature that was unattainable during Macquer's time). About 1741, CharlesWood, and assayer of Jamaica, obtained some platinum from Cartagena and brought it to London. In 1749 William Watson(1715-1787) communicated to the Royal Society his results on platinum and those of William Brownrigg (1711-1800) (Watson,1750). According to Watson the Spaniards called the metal *Platina di Pinto* or *Platina* because it color resembled that of silver. The metal was collected in a very pure form although it contained small amounts of black iron ore. Watson was unable to melt the metal even in the presence of borax or other fluxes. Anyhow, he reported that the Spaniards were able to do so by addition of some unknown flux and thus cast it into sword hilts, buckles, snuffboxes, and other utensils. Four years later, William Lewis (1708-1781) published an extensive memoir describing the chemical properties of the element (Lewis, 1754). He indicated that pure platina was a white metallic substance, malleable in a small degree, *permanent* to fire, indestructible by nitre, unaffected by sulfur, and soluble in *hepar sulfuris* (a mixture of various compounds of potassium and sulfur made by fusing potassium carbonate and sulfur). Platina was similar to gold, the simple acid did not attack it but aqua regia did. Hence, the common methods of gold purification with *aqua fortis* (nitric acid), *aqua regis*, or *regal cement* were no longer reliable in the case that this metal was adulterated with platinum. Platina melted with an equal weight of metals (tin, lead, silver, gold, copper, and iron) making them stiffer and harder. It also alloyed with mercury, bismuth, zinc, and antimony (Condorcet, 1784: Vicq d'Azyr,1782).

In 1757 Antoine Baumé (1728-1804), received about a pound (*une livre*) of platinum from José Ortega y Hernández (1703-1761), Apothecary to the King Fernando VI of Spain, secretary to the Royal Academy of Medicine of Madrid since its foundation in 1734, and a corresponding member of the Paris Academy. Baumé and Macquer joined forces to make a physical and chemical study of the metal, particularly since Macquer owned some of the best furnaces constructed at that time.

Macquer and Baumé observed that calcination of the raw mineral led to an increase of its weight and the appearance of a friable gray material, facts that suggested that the platinum was contaminated with a base metal, probably iron. Although they were able to remove most of the impurities by cupellation with lead in a high-temperature porcelain oven at Sèvres, they failed again in their efforts to melt the metal. After many fruitless attempts to melting it using a variety of techniques, they came upon the idea of heating a small portion in the focus of a burning mirror (concave spherical mirror, *miroir ardent*), a method that no one had tried before. The mirror they used belonged to the Abbe Bouriot, it was made of silver glass, 22 inches (*pouce*) in diameter and with a focal length of 28 inches. With this mirror they were able to melt the metal in five or six places, the liquid globules produced remained attached to the mass and solidified as soon as they were moved out of the focus of the mirror. Purified platinum could only be dissolved with boiling aqua regia, the best composition being equal weights of nitric and hydrochloric acids. Macquer and Baumé considered that the mechanical properties of platinum coupled to its resistance to air, water, and most reagents made it an ideal material for making burning mirrors, telescope mirrors, and vessels for the kitchen and the chemical laboratory. They mistakenly thought that it had the same density as gold and might therefore be used to adulterate gold (Macquer and Baumé,1758). Eventually Baumé showed this result to be erroneous and that platinum was more dense than gold and hence it would not be used as an adulterant (Smeaton, 1984).

Macquer and Baumé obtained for the first time platinum black by calcining it alone or in the presence of borax (sodium borate decahydrate) or potassium tartrate, or the red platinum ammonium precipitate obtained by the action of ammonia on a solution of platinum dissolved by aqua regia.

According to Smeaton (Smeaton, 1984) Baumé evidently carried out further research alone because in his book *Chymie Experimentale et Raisonée* (1773) reported that cupelled platinum could be forged by hammering, without the addition of another metal.

3. Solubility of oils in spirits of wine

At the end of the eighteenth century the exact nature of fatty materials was unknown, in spite of what OttoTacken (?-1670) had written almost one century before: "les sels prennent naissance par l'action des acides sur les alcalis. Si dans le savon, la force alcaline de la potasse a disparu, c'est sans doute qu'il existe dans la graisse un acide cache" (salts originate by the action of acids on alkalis. If in a soap the alkaline force of potash has disappeared, there is no doubt that the fat contains a hidden acid) (Viel, 1986-1990). Fatty oils could be considered as a unique species. Since there was such a diversity of animal and vegetable fats and oils, this species contained many varieties. Chemists believed that the fat obtained by the acidification of soap was identical with the fat used in the preparation of the soap, that is, the fat was unchanged by the treatment with alkali in forming the soap. In other words, soap was binary combination of the *entire* fat with an alkali. It was also believed that heating a fat with an alkali removed oxygen from the atmosphere so that saponification involved a fixation of oxygen.

Macquer's first scientific paper (Macquer, 1745) dealt with the solubility of oils in spirits of wine. Although it did not contribute much to the problem of the composition of oils, it reported some curious conclusions: "toute huile est composée de phlogistique (!), d'acide, de eau, et de terre, car toutes ces principles se manifestent dans la décomposition des huile quelconques; mais peut être que l'eau et la terre qui font partie des huiles, y sont unies ensemble et n'existent dans le huile que sous la forme d'acide. La graisse n'est qu'une huile douce, concrete, non volatile quis ne doit sa consistence qu'à un acide qui lui est intimement uni, qu'on ne peut l'en separer que successivement et par des distillation réitérées" (every oil is composed of phlogiston, acid, water, and earth because all these principles manifest themselves during the decomposition of any oil; although it is possible that the water and earth that are part of the oil, are joined together and exist in the form of an acid. The fat is a sweet, solid, and non volatile oil that owes its consistence to an acid to which its it is intimately united, and that can only be separated by successive distillations).

According to Macquer, the flammability of oils demonstrated the presence of phlogiston. The proof that oils contain an acid were the facts that they combine with alkalis to form a neutral salt, that some metals are corroded by oils, and that acid crystals are found in some oils that have been stored for long periods. This acid served to unite phlogiston and the water, which have no affinity for each other, whereas acid has an affinity for both.

Oils were also a proximate principle of all animals and vegetables. Oil constituted the essential difference between the animal and vegetable kingdoms and the mineral kingdom. In principle, an oil in an absolutely pure state could be called the *oily principle*, the same in all animals and vegetables, and the difference between oils was due merely to extraneous matter. They were known to be composite bodies, but it was not known if certain components were essential or not to their properties.

Solubility in solvents was a common tool to investigate the nature of a body, simple or compounded, and accordingly Macquer selected spirit of wine as the solvent to study the properties of vegetable oils.

Macquer observed that when alcohol was added to olive oil, the supernatant alcoholic phase contained an acid substance, which increased in amount the more rancid the oil was. Fatty oils were quite insoluble in spirits of wine, but on rectification became lighter and soluble (Macquer, 1745). Repeated distillation of oils of different nature changed their solubility a different manner: the solubility of fatty oils increased while that of essential oils decreased. Macquer argued that substances of a similar composition had an affinity for each other, and since spirits of wine contained an oil we should expect it to dissolve all oils. On the other hand, spirit of wine also contained water, which did not dissolve oils. This two opposing effects could be explained only by the presence of a *third substance* showing affinity to both components. This third substance had to have an acid nature because essential oils could be rendered thick and heavy by treatment with acid and could be restored to their former condition by removing the acid by distillation. Fatty oils, unlike other oils, were neutral but became acid distillation, fact that gave weight to the hypothesis that an acid was the factor explaining solubility.

According to Macquer, the quantity of acid was less important that the form in which it was combined.Treatment of fatty oil with sulphuric acid yielded a product that was very soluble in spirit of wine. Repeated distillation resulted in a decrease in solubility, as in the case of essential oils.

Macquer determined the solubility of many salts in spirit of wine, among them: the salts of sodium, potassium, ammonium, calcium, silver, copper, iron, and mercury, with the three mineral acids (sulphuric, nitric and hydrogen chloride); the carbonates of sodium, potassium, ammonium; tartar emetic, and the tartrates of potassium, ammonium, and calcium. He understood that the solubility was dependent on the degree of dilution of the solvent, and for this reason he carefully distilled the spirit until he thought it was dry enough. In the same manner, salts that carried water of crystallization were made anhydrous before the measurements.

Analysis of the results led Macquer to conclude that neutral salts were more soluble in spirits of wine when their acid was less strongly combined with their base. The fact that all sulfates were insoluble proved the great affinity of the acid for the metallic base. Nitrates were much more soluble that the sulfates, and the solubility of the chlorides was similar to the nitrates. An exception to this rule was corrosive sublimate (mercuric chloride, $HgCl_2$) it was more soluble in spirit of wine than in water.

Antoine-Laurent Lavoisier (1743-1794) used Macquer results in his work on the use of spirits of wine in the analysis of mineral waters. Lavoisier wrote: “M. Macquer is the first who has undertaken a series of experiments on the solubility of salts in spirits of wine, and who has determined the extent of the solubility. The information known previously on the subject was not very extensive and was, moreover, scattered among different authors. The work of M. Macquer has assembled this, increased it tremendously and has completed to a certain extent this part of chemistry.”

4. Dyeing

Macquer’s first contribution to dyeing was to add to the fast blue dyes known at that time (pastel and indigo) a third more brilliant than either of the other. Macquer believed that he could contribute to the art; in order to do so he looked into the pigments used by painters and decided on Prussian blue as the best candidate. He based his selection on the basis of Hellot’s theories: The solutions of alum and green vitriol (ferrous sulfate heptahydrate, $FeSO_4 \cdot 7H_2O$) used in its preparation would serve as an excellent mordant[1] while the precipitate (*fécule*) would be very suitable for applying to fabrics because of its fine size. In his initial experiments he submerged a skein of flax, cotton, wool, and silk in an alkaline solution saturated with prussic acid, and then washed the cloth with acid water to eliminate the residual iron hydroxide. The material was tinted beautiful blue but irregularly. In the next set of experiments he boiled the cloth in a solution of alum and *tartre* (potassium tartrate), followed by a bath of Prussian blue. With this modification he obtained a uniform tint, although pale, and a cloth that was agreeable to the touch. Having shown that Prussian blue was a fast dye for wool and silk he went on to determine the conditions to perfect the process, looking for a way to render the fabric soft to the touch after dyeing, to reduce the cost as much as possible, to find a way of obtaining light and dark shades, and to perform operations on a large scale (Macquer, 1749, 1752).

Macquer believed that the color of Prussian blue was due to phlogiston: “cette matière colorante du bleu de Prusse est une matière inflammable dans un état singulier, & que je crois très-peu connu” (this coloring substance of Prussian blue is a flammable one in a particular state, which I believe unknown). Treatment of the dye with a solution of potassium hydroxide yielded a yellow neutral solution of a salt (potassium ferrocyanide), which Macquer believed to be compound of alkali and phlogiston (*alkali phlogistiqué*). This assumption was in agreement with Étienne François Geoffroy’s (1672-1731) theory that blood and other animal matter communicated to caustic potassium the necessary phlogiston to revitalize the green vitriol.

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[1] Mordants are earthy or metallic salts; they are decomposed by soluble dyes, which precipitate their base; this combines with the colored parts of the dye, and the color precipitates adhere to the textile. The mordant often changes the color of the dye.

Macquer made another important contribution to dyeing practice when he discovered a way of dyeing silk with cochineal (Macquer, 1768). Wool was dyed scarlet easily by dipping it in a bath containing a suspension of the active dye obtained by reacting cochineal with a solution of tin in a mixture of nitric acid and hydrogen chloride (aqua regia) as mordant. Unfortunately the same procedure was unable to dye silk. In a brilliant stroke of inspiration Macquer achieved silk dyeing by simply changing the order of the operations. He first impregnated silk with the mordant and then dipped it into a bath of cochineal to which no mordant had been added. The precipitate of tin and cochineal was thus formed on the silk itself, which was dyed a beautiful fast red. An interesting side angle of this research is that in the beginning the inhabitants of the near village suspected Macquer of practicing vivisection because of the red blood color of the water residues released by his laboratory.

After his success with Prussian blue, Macquer was appointed to assist Hellot as government inspector of the dyeing industries. Hellot passed away in1766 and Macquer succeeded him as inspector of dyeing.

The results of his experiences on dyeing were published as a book (Macquer, 1763) and as an entry in the Académie series, *Description des Arts et Métiers*.

5. The composition of Prussian blue

The first modern, artificially manufactured color, was Prussian blue. The color maker of Berlin, Diesbach, made the blue pigment about 1704 accidentally when experimenting with the oxidation of iron. During Macquer's time the manufacturing procedure consisted in adding a mixture of alum and green vitriol (iron sulfate) in solution to alkaline lye, which had been calcined with animal matter, followed by addition of an acid.

After successfully demonstrating the value of Prussian blue as a dyestuff (Macquer, 1749), Macquer initiated a research program to inquire into the nature and composition of the color. There were many conflicting opinions on the subject, some believed that the color, separated by the action of the alkali and precipitated on the white earth of alum; others believed that the color belonged to iron, which the organic lye precipitated and deposited on the earth of alum (Coleby, 1938).

Macquer found that dry washed Prussian blue was nonmagnetic but when calcined it released alkaline vapors and left a rust-brown residue that was attracted by a magnet. Addition of Prussian blue on fused nitre (KNO_3) resulted in a slight detonation. According to Macquer these findings indicated that the dye contained some fatty or flammable material and that the magnetic nature of the residue showed that the base of the color was the earth of iron. The color was not due to iron alone; it disappeared immediately in the presence of alkali leaving a dull grey powder, which on heating turned acquired a yellowish-brown. Macquer repeated the alkali treatment several times and then divided the residue into two parts. He treated one part with nitric acid and noticed total solution without return of the color. The second portion was calcined and the residue found to be totally attracted by a magnet. From these findings he concluded that the alkali treatment had removed the substance, which combined with the iron to form the dye. In addition, the alkali also dissolved some of the iron, and this matter liberated by further addition of the acid, combined with some of the coloring principle to give the precipitate observed (Coleby, 1938).

Macquer was now able to able to explain the need of alum (double sulfate of aluminium and potassium hydrated) for preparing the dye. Alum reduced the excess alkalinity of the lye and the amount of earth iron precipitated. This precipitate was then dissolved in the acid added in the following step. It had a white color and hence made the color of Prussian blue less intense. According to Macquer the blue color was not the natural color of iron; it resulted from the heterogeneous matter with which it was combined.

It was left to Scheelein 1782-1783 to push the analysis a stage further by isolating prussic acid by distilling the yellow crystals obtained from a lye saturated with Prussian blue, with dilute sulphuric acid. Berthollet showed a few years later that the yellow crystals contained iron (Berthollet,1791), a point sustained by Macquer.

6. Assay of silver and gold

At Macquer's time the standard procedure for assaying silver was cupellation with lead although Mathieu Tillet(1714-1791) had shown that the assay was not reliable enough because some silver was always absorbed by the cuple, thus yielding too low silver content. Certain earlier chemists believed that the difference was actually due by transmutation of lead. By royal command dated November 26, 1762, Hellot Tillet and Macquer were asked to find the best method of assaying gold and silver, and to determine the conditions under which the test should be carried out.

The standard cupellation assay procedure consisted in adding eight parts of lead to one of silver for alloys containing more than 50 per cent of silver, and sixteen parts of lead for alloys having less than 50 per cent of silver. Hellot, Tillet, and Macquer checked this recommendation and found that the assay could be made more accurate by varying the amount of lead according to the fineness of the alloy. They prepared a table indicating the appropriate proportions and reported it to the Council of State. They also concluded that it was impossible to recover all the silver by one cupellation and that the silver had not been produced from the lead, thus disproving the old belief that silver was formed during the process by transmutation of lead (Hellot *et al.*, 1763)

As a result of Hellot, Tillet, and Macquer's report to the Council, the French government issued regulations for silver assaying prescribing (a) the nature of the cupels, (b) their thickness, (c) the quality and history of the lead, (d) the proportions of lead to be used, and (e) the uniformity of the silver sheets, or special weights used in assaying.

7. *Rubber (Macquer, 1768)*

La Condamine visited Quito in 1736, with a French expedition sent by the Académie to explore tropical South America and determine the length of the arc of one degree in Quito. While at Quito he sent to the Académie several rolls of a new discovered resin called caoutchouc, with remarkable elastic properties. Pierre Bouguer (1698-1758) La Condamine sent to Paris some observations on the substance, but the Académie did not publish them. François Charles Fresneau (1703-1770) (discoverer of *Hevea Brasiliensis*), the resident engineer at Cayenne, investigated the resin and an account of his work reported to the Académie by de la Condamine (La Condamine, 1751). Fresneau and de la Condamine had observed that the resin was used in South American for waterproofing garments and for making flasks, and for these reasons they did experiments on the possibility of dissolving the material and recovering it from the solution without the loss of its peculiar and valuable properties. De la Condamine's experiments were unsuccessful, but Fresneau was able to dissolve the with nut oil on a sand bath.

Herissant and Macquer found that rubber treated with oil of harshorn (ammonium carbonate) turpentine, or ether softened it so that it could be moulded, and regained its former firmness and elasticity on exposure to smoke. Macquer found that common ether did not dissolve rubber, but it did do if distilled carefully. Evaporation of the clear solution led to formation of a layer of rubber as elastic as before solution. If water was added to the solution, it would not mix, but a solid membrane formed at the surface of separation, and this membrane could be stretched considerably without tearing, and would regain its former dimensions (Coleby, 1938)

8. Seawater

This is another interesting entry in Macquer's dictionary. He wrote that all seawaters had a salty taste, and slightly acrid bitter. Although the acridity and bitterness was usually attributed to the presence of bituminous matter, Macquer had been unable to detect its presence in seawater originating from many locations. He believed that the off taste was actually due to the presence of Glauber's salt and other salts. Chemists had tried to remove the supposed bituminous matter by distillation in the presence of several substances such as oil of tartar, seaweed, soap, wood ashes, etc. Macquer suggested that since all the potable water that fell from heavens was distilled water originating from the sea and made potable without addition of foreign substances, it should be possible to use the same procedure in a controlled fashion. In his dictionary Macquer reported that twenty-five years before the Académie des Sciences had appointed a commission composed of Roland Michel Barrin de la Galissonière (1693-1756), Bourdelin, and himself to examine on behalf of the Ministry of Marine a secret process offered to the Marine to make potable water from seawater. The test was performed in Macquer's laboratory on water provided by the naval authorities. The inventor added to the water a white powder and distilled the mixture, obtaining perfectly pure water. The examiners were very much impressed and were prepared to report favourably, but it occurred to Macquer to distil some more seawater without addition, and, to his surprise, by the simple precaution of not distilling too rapidly or too far, he obtained a distillate undistinguishable from the other. Macquer presented bottles of the two distillates to the inventor, who was unable to distinguish between them. According to Macquer "l'Auteur fit après cela sa reverence, sans dire un mot, & depuis on n'a plus entendu parler de lui" (after which the author of the secret made a reverence and left without saying a word, never to be heard again).

Macquer, Lavoisier, and Sage (Macquer *et al.* 1758) also analyzed water from the Dead Sea and found that it contained very large amounts of common salt and magnesium salts, but no bituminous matter. ◩

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