

SKETCHES IN THE HISTORY OF METEORITICS 2: THE EARLY CHEMICAL AND MINERALOGICAL WORK

D.W. Sears and H. Sears

*Department of Metallurgy, University of Manchester
Great Britain*

The chemical and mineralogical work on meteorites over the period 1800 to 1840 is reviewed. The number of elements known to be present in meteorites rose from six to 19. Chemical techniques advanced rapidly so that by 1815 the procedure was essentially that of modern wet chemical analysts: removal of the magnetic material, dissolution of the acid-soluble portion and fusion of the remainder with alkali. After Bournon's work in 1802 much mineralogical progress was made during the 1820's, notably by G. Rose. Berzelius made important contributions by his own analyses and synthesis of the work of others. By 1840 ordinary chondrites, carbonaceous chondrites, plagioclase-pyroxene achondrites, Chassigny, pallasites and octahedrites could all be distinguished.

INTRODUCTION

In the first paper in this series it was argued that an important factor in establishing the serious study of meteorites was the discovery that, wherever they fell, meteorites were physically and chemically alike (Sears, 1975). It was therefore natural that these aspects should be developed first. It is the purpose of this second paper to pursue this point, outlining the chemical and mineralogical work on meteorites from 1800 to 1840.

The first decades of the 19th century saw enormous developments in analytical chemistry and mineralogy (Szabadvary, 1966; Weeks, 1968). The reasons for this are to be found in the progress of chemistry as a whole. Lavoisier's work on combustion and Dalton's atomic theory and its corollaries meant that at the opening of the century chemistry was just becoming a quantitative science and there was a pressing need for comprehensive and reliable atomic weight tables. This need was to be met in large part by the giant of main-stream and analytical chemistry, Jons Jacob Berzelius. Equally impressive advances in mineralogy were being made, especially in crystallography and crystal chemistry. It was against this background that meteoritics was initially developed.

ANALYTICAL TECHNIQUE AND THE IDENTIFICATION OF THE ELEMENTS IN METEORITES

It was recognised from the beginning of the century that the study of meteorites presented particular problems for the analyst but they also promised special rewards (Faraday, 1826):

I have always been struck by the circumstance that notwithstanding the apparently independent sources of meteoric matter both stones and alloys with iron and also the minute chemical examination which had frequently been made of them that no new metal or element of any kind has been found in them nothing but what has previously been recognised in the materials of our globe. I think that if meteoric masses had been unexamined and I had been looking out for something in which to discover new bodies I should at once have gone to them.

The extraterrestrial origin of meteorites suggested that they might contain terrestrially unknown elements, certainly they seemed to contain in abundance elements rare on Earth, for example nickel and chromium. The main concern therefore was to establish what elements were present, Table 1. This led inevitably to refined techniques, since some elements exist only in phases in which an inappropriate procedure would be insensitive. Nickel, aluminium and chromium are the major examples of this. There were two major innovations in the analytical technique for meteorites which were associated with the discovery of new elements in them. The first was the need to separate the metal, which was apparent after Howard's (1802) discovery of nickel in stony meteorites. After commenting on its absence in the previous analyses Howard stated that, "with respect to the nickel, I am confident it would have been found in all, had the metallic particles been separately examined." His separation of the metal was prompted by two factors. The major one was that Howard worked with a mineralogist; large sections of his paper were authored by Bournon, and it was this which prompted the separation of the metal and this in turn ensured his discovery of nickel. The second factor Howard (1802) stated in a criticism of the academician's analysis of the Lucé meteorite (Fougeroux *et al.*, 1772; Lavoisier, 1772):

It was unfortunately made on an aggregate portion, and not of each distinct substance; irregularly disseminated through it. The proportions obtained were, consequently, as accidental as the arrangement of every substance in the mass.

The rareness of nickel terrestrially and its presence in all the stones and irons examined was essential in establishing the authenticity of meteorite falls in 1802/3 (Sears, 1975). In analytical technique its presence may also have been important as the reason for hydrogen sulphide becoming an analytical reagent in meteorite analysis long before its use became widespread. The only alternative to sulphide for the precipitation of nickel was cyanide. According to Szavabadvary (1966) H₂S was generally introduced to analysis by Pfaff

Table 1
Element identifications made before about 1840*

Element	Meteorite in which found	Method of discovery	Method most frequently used	Reference to discovery
Sulphur	Albareto/Lucé	see text	H ₂ S evolved with acid passed through Pb-acetate or CuSO ₄	see text
Magnesium	Ensisheim	— —	ppt carbonate	Barthold (1800)
Silicon	Ensisheim	— —	repeated evaporation with HCl	Barthold (1800)
Iron	Albareto	see text	ppt carbonate or hydroxide	Troili (1766)
Calcium	Ensisheim	ppt oxalate	as left	Barthold (1800)
Nickel	Campo del Cielo	Solution colour, no ppt in neutral solution, NiSO ₄ with H ₂ SO ₄	1) Difference (in metal). 2) ppt sulphide	Proust (1799)
Manganese	Sena	Blow-pipe on sulphide	1) ppt Mn(CN) ₂ 2) ppt Mn(CO ₃)	Proust (1805)
Chromium	Vago and four others	ppt HgCrO ₄	as left	Laugier (1806)
Carbon	Alais	lime water test	various eg as left	Thenard (1806)
Aluminium	L'Aigle, Salles	ppt hydroxide	as left	Sage (1808)
Sodium	Erxleben		Difference on removing K from soluble carbonates as K ₂ PtCl ₆	Stromeyer (1812)
Cobalt	Eichstädt/Cape of Good Hope	Colour change of ammonia complex on heating	Philips' method	Klaproth (1795-1815) Stromeyer (1817)
Potassium	Juvinas	ppt K ₂ PtCl ₆	as left	Vauquelin (1821)
Copper	Juvinas	ppt Cu(CN) ₂	as left	Vauquelin (1821) Laugier (1821)
Phosphorus	Bohumilitz	Blow-pipe on Mg ₃ (PO ₄) ₂		Berzelius (1832)
Chlorine	Stannern	ppt AgNO ₃	as left	Vauquelin (1809) Jackson (1838)
Tin	Blansko/Chantonnay	Blow-pipe		Berzelius (1834)

*Arsenic and zinc were also said by Humboldt (1849) to have been found in meteorites during this period but the specimens were either non-meteoritic or details have not yet been located.

(1821) although Boyle described its use in the analysis of mineral waters in 1685. In meteorites it appears to have been in use since the beginning of the century. Vauquelin (1802, Fig. 1) used hydrogen sulphide to precipitate nickel in his analysis of Benares, his second method, and Fourcroy (1803) actually utilised the hydrogen sulphide evolved in pouring acid on the powdered meteorite, reporting that the “water charged with sulphurated hydrogen was afterwards employed to separate the oxide of nickel . . .” The reference to oxide instead of sulphide reflects the inadequacy of atomic weight tables because all precipitates were heated to redness to convert them to oxides, the molecular weights of which were better known.

As a consequence of the discovery of nickel it became virtually universal practise to separate the metal at the beginning of the analysis and to give its composition separately. The result quoted was usually for the non-magnetic portion and the iron therefore quoted as the oxide. A noteworthy exception was one report of Klaproth's, Fig. 1. In connection with his analysis of Lissa, Klaproth (1812) wrote:

I have supposed, that all the iron in the aerolite was in the metallic state. Formerly such as could be extracted by the magnet was alone so reckoned, the rest being considered as oxide of iron. But there is no sign of oxidation in this aerolite recently fallen, it is evident, that shining points, which did not adhere to the magnet, were pyrites, in which the iron was originally in the metallic state.

He asserted therefore that the oxidised iron usually detected was the result of weathering and that all iron was originally metallic or in the sulphide. Most considered that some of the iron existed originally in the oxidised form, but it was uncertain which other elements it was combined with. For example, Stromeyer (1812, Fig. 1) calculated the distribution of the iron in some detail and was one of the first to report the metallic and oxidised iron separately in the same bulk analysis. Another error Klaproth made was his continued refusal to admit the presence of chromium, although by then every major chemist had confirmed its presence.

The second major innovation to meteorite analytical technique associated with the discovery of elements was the use of alkali fusion. Howard (1802) describes this procedure at the beginning of his analysis of the silicate portion, “I treated 100 grains with potash, in a silver crucible; and, after the usual application of red heat, separated as much silica as possible, by muriatic acid and evaporation.” Alkali fusion was also used by Klaproth (1803) and he was the first to use this technique for initial dissolution in routine analysis (Szavadbary, 1966). It was not used by the early French chemists, Fig. 2. Vauquelin (1802), Fourcroy (1803) and Proust (1805) used hydrochloric acid directly on the powdered meteorite, sometimes with a little nitric acid, and assumed that all material insoluble after boiling was silica. The difference in procedure is apparent in the analysis results. The French chemists missed

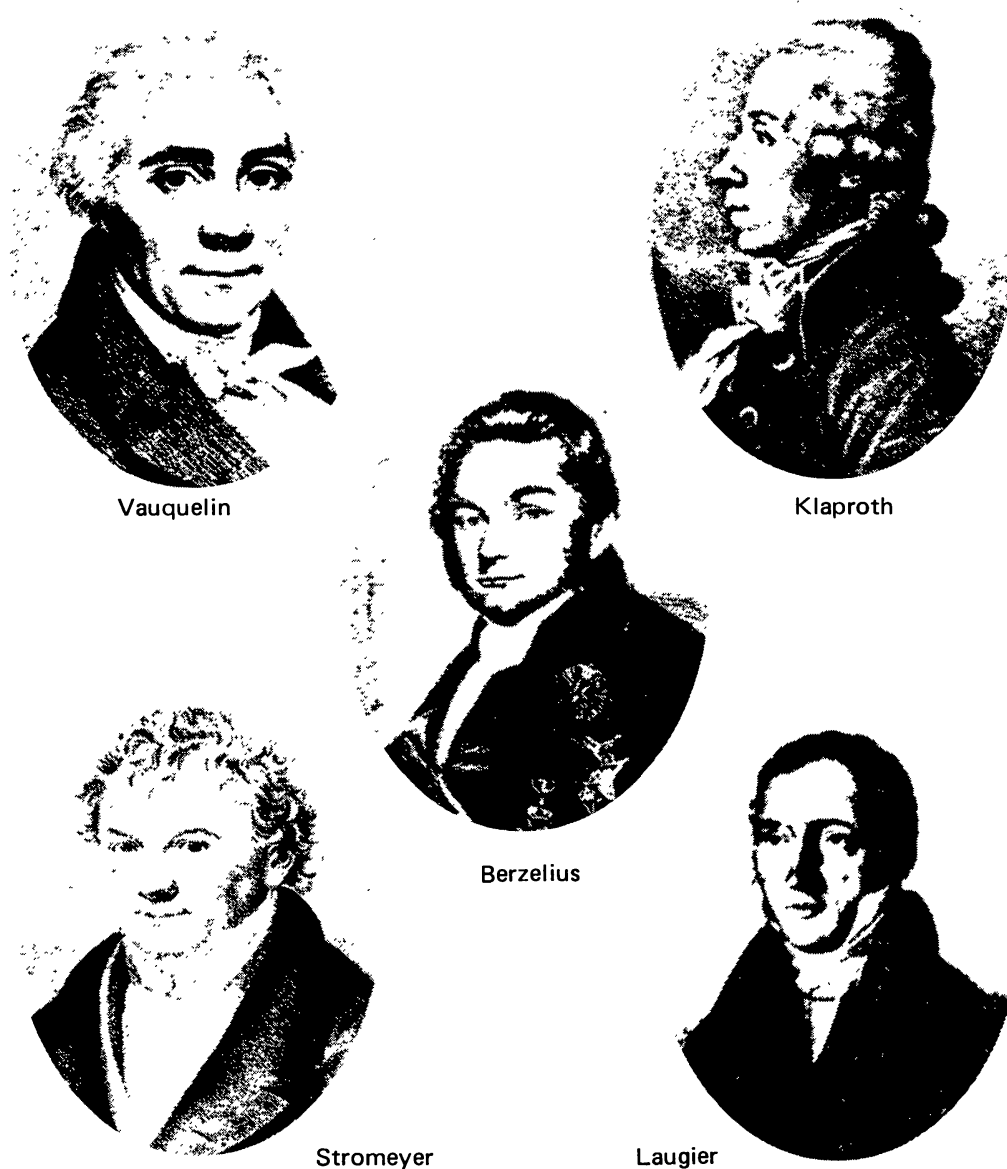


Fig. 1 Early 19th century meteorite analysts. Nicolas-Louis Vauquelin (1763-1829), Professor at the Ecole Polytechnique and School of Mines in Paris, and Martin Heinrich Klaproth (1743-1817), Professor of Chemistry at Berlin, were instrumental in the establishment of meteoritics. Both analysed a large number of meteorites, Vauquelin discovering potassium, copper and chlorine in meteorites. André Laugier (1770-1832), Fourcroy's cousin and Professor at the Museum of Natural History, Paris, discovered chromium and copper in meteorites and made major innovations in technique. Friedrich Stromeyer (1776-1835), student of Vauquelin who became Inspector-General of Hanoverian apothecary shops, discovered sodium and cobalt in meteorites and was among the first to report both reduced and oxidised metal in the same bulk analysis. Jöns Jacob Berzelius (1779-1848) discovered phosphorus and tin in meteorites and identified the major minerals. (All portraits from M.E. Weeks and H.M. Leicester "Discovery of the Elements," 7th edition published by the Journal of Chemical Education 1968, except Laugier which is from G. Kersaint, "Antoine de Fourcroy", Mémoires du Museum National d'Histoire Naturelle, Nouvelle Série D, Sciences physico-chimique, Tome 2, 1966.)

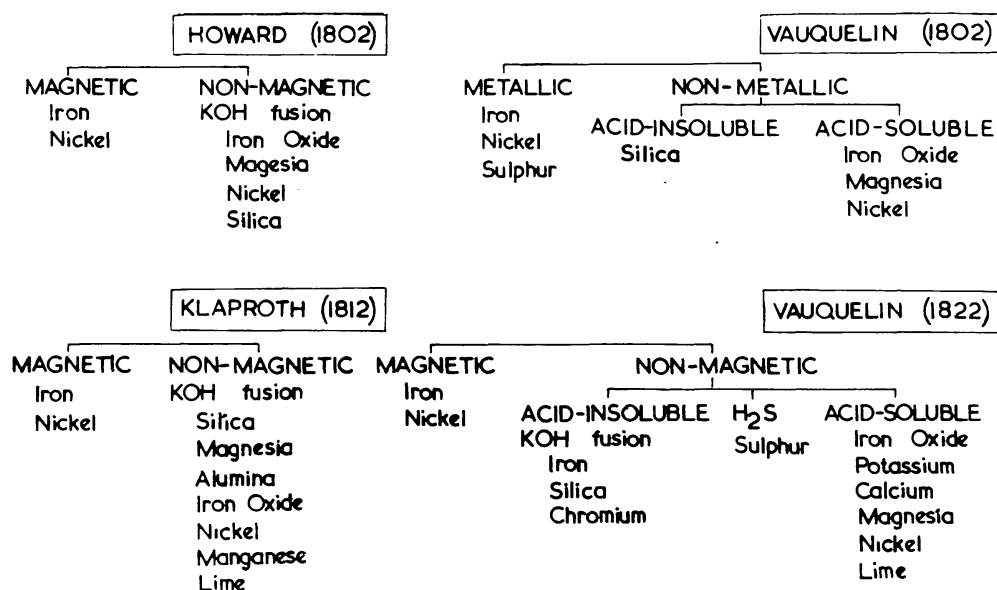


Fig. 2 The analysis procedures shown schematically used by Howard (1802) on Benares, Wold Cottage, Ensisheim and Tabor; by Vauquelin (1802) on Benares, Creon and Barbotan; by Klaproth (1812) on Lissa; and by Vauquelin (1822) on Epinal.

the magnesia in the acid-insoluble silicates and reported values of 13, 9 and 12% (Sears, 1975, table 2) whereas Howard and Klaproth gave five values averaging 22% magnesia. The silica values reported by the French analysts are consequently higher than those determined by alkali fusion. The difference seems not to have been appreciated at the time; indeed, the most noteworthy feature was their overall similarity (Sears, 1975). In 1806 André Laugier, Fourcroy's cousin, discovered chromium in five meteorites after he had used alkali fusion. His success induced him to conclude that:

... In many cases, in order to attain the requisite precision of chemical analysis, it may perhaps be expedient to treat the same substance with both acids and alkalies; as experience demonstrates that a principle may be overlooked in one case, which will be obvious in the other.

The need to use alkali fusion was also apparent in the discovery of aluminium in meteorites. Like chromium, aluminium exists primarily in the acid-insoluble material, feldspar, and consequently when using acid dissolution Vauquelin always missed the aluminium. Sage (1808) discovered the element in Salles after vitriolisation – boiling with concentrated sulphuric acid – and although it was confirmed by Klaproth (1809), Vauquelin (1808a) challenged the observation since he had examined the same meteorite (Dree, 1803). However, later in 1808 Vauquelin analysed Stannern. This meteorite not only contains much aluminium, being a eucrite, but is also free of olivine and therefore contains no acid soluble material. Vauquelin had to resort to alkali

Table 2
Results of the analyses (percentages) schematically shown in figure 2

	Howard (1802)*	Vauquelin (1802)*	Klaproth (1812)**	Vauquelin (1822)*
Silica	50	46	43	35
Iron Oxide	11	38	—	63
Magnesia	25	15	22	4.25
Chrome Oxide	—	—	—	0.25
Sulphur	present	present	3.5 (incl. loss)	2.25
Nickel Oxide	1.3	—	—	0.5
Lime	—	2	0.5	
Potash				12.5
Manganese	—	—	0.25	—
Alumina	—	—	1.25	—
Iron Metal	—	—	0.29	—
Nickel Metal	—	—	1.50	—

*Non-magnetic portion only.

**All iron reported as metal.

fusion in his analysis of Stannern (Vauquelin, 1808b) and he used the technique in subsequent analyses. The discovery of chromium and aluminium ensured that alkali fusion was used by most analysts, but these ideas were new at the beginning of the second decade of the 19th century and analytical techniques varied widely. Klaproth (Reuss, 1812) fused all the non-magnetic portion with alkali, Warden (1810) fused only the acid-insoluble portion while Higgins (1811) was still assuming all the acid-insoluble portion was silica.

Whereas nickel, chromium and aluminium were elements whose discovery prompted major changes in technique, sulphur, though readily detectable (Troili, 1766; Fougroux *et al.*, 1772; Lavoisier, 1772), was responsible for many variations in procedure because of the difficulty of its determination. In the 18th century the academicians had to resort to calculation by difference and arrived at a figure of eight percent. The early 19th century practise was to add hydrochloric acid to the powdered stone, which dissolved some of the silicates and produced a fine suspension of sulphur. This was usually decanted and filtered after the insoluble silicates had settled (Howard, 1802). The difficulty of this process was apparent in Webster's (1824) analysis of the Nobleborough eucrite where a value of 18.3% sulphur indicated incomplete separation of the silicates. In later years the standard procedure was to add HCl to the powdered stone dropwise with heating until the evolution of H₂S ceased. The gas was passed through lead

acetate or copper sulphate solution and the amount of sulphide precipitate determined. It was realised by Howard (1802) and Proust (1805) that the sulphur was located entirely in the sulphide and that meteoritic sulphide was distinct from terrestrial pyrites because of its solubility in acid and lower proportion of sulphur.

From around the middle of the second decade of the 19th century it became standard procedure when analysing meteorites to remove the magnetic material, to remove the acid-soluble constituents and finally to fuse the remainder with alkali, Fig. 2. By the end of the decade sulphur was usually determined by passing the H_2S evolved on adding acid through lead or copper solution. It does not seem to have been realised that the presence of acid-soluble and acid-insoluble portions suggests that two silicates are present until Berzelius pointed it out in 1828. This conclusion was based as much on the mineralogical work as the chemistry because from the middle of the second decade the two became even more inseparable.

EARLY MINERALOGICAL WORK

Some early mineralogical observations were made by Bournon, who identified the major phases in stony meteorites as chondrules, metal, sulphide and matrix and suggested that the “glass” in Krasnojarsk was olivine. Many casual observations were made during the chemical work. For example, Howard’s (1802) and Proust’s (1805) observations on the sulphide and Stromeyer’s (1812) and Vauquelin’s (1816) recognition of chromite may be said to be observations of this kind. Stromeyer (1812) suggested that green grains in the Erxleben meteorite (probably chondrules) were olivine and, since he could not detect sodium in the metal, concluded that it was present in the ground-mass, probably as feldspar.

The first major mineralogical work after Bournon’s was that of Rose (1825). Rose examined Juvinas and to a lesser extent Stannern. The two kinds of grain present were found to be augite (recognised from its crystallography) and a feldspar which the chemistry suggested was labradorite. In addition the sulphide was analysed to see if its non-magnetic property could be ascribed to some nickel content (unlike FeS , NiS is non-magnetic) but Ni could not be detected. These two meteorites closely resembled Jonzac and Loutalax and all resembled dolerite in appearance, chemistry and mineralogy.

Rose’s observations had a considerable influence although many did not appreciate the atypical character of these meteorites. Thus Shepard in his description of Richmond (L5), describes feldspar as “... one of the most common ingredients of meteorites, although in the present specimen it forms somewhat less than one quarter of the mass.” The reason Rose chose to examine these two in depth was no doubt their comparatively coarse texture

which was open to study by the chemical crystallographic techniques then under active development and not because they were typical.

The olivine of Krasnojarsk was the subject of much study because of the widespread specimens, its macroscopic nature and the fame of Pallas' travels. Biot (1820) conducted the first thorough optical study and analyses were made by Walmstedt (1825) and Stromeyer (1825). The outcome of this work was that it was realised that the olivine closely resembled terrestrial olivines whose composition could be represented $\begin{matrix} M \\ f \end{matrix} \} S$ where M, f and S represent the oxides of magnesium, iron and silicon respectively.

THE DIVERSITY OF METEORITE CLASSES

Ever since the English chemist, Mr. Howard, called the attention of philosophers and naturalists towards the stones called meteoric, all chemists who have repeated the experiments laid down in his interesting memoir, have obtained similar results. They all agree that whatever the time, or wherever the place in which these stones have fallen, their component principles have been the same, viz. silex, iron, magnesia, sulphur, nickel with a few accidental traces of lime and alumine.

The norm having been thus defined (Laugier, 1806), attention subsequently seemed to focus particularly on atypical meteorites. This led to one of the major achievements in the first few decades of meteoritics – recognition that meteorites existed with highly diverse properties. The first unusual meteorite to be examined was the Alais C1 chondrite. Thenard's (1806) treatment was the same as for other meteorites of the time being analysed in France, except that he assumed the acid-insoluble portion was a mixture of silex and carbon. He then determined the carbon by noting the loss in weight on heating to redness. Vauquelin's (1806) method was very different; he passed the carbon dioxide given off while fusing the stone with potassium nitrate through lime water. The amount of carbon was then calculated from the carbonate precipitate. The fused mixture was analysed in the usual way. However, although it was realised that Alais contained much carbon, the apparent similarity of its bulk chemistry to that of earlier stones meant that it was not considered to be significantly different from them at this time.

The Stannern meteorite, which fell in 1808, had a fundamentally different bulk chemistry. This was first reported by an obscure Vienna apothecary by the name of Moser and his results prompted a repetition of the analysis by Vauquelin (1808b) who wrote that:

This aerolite is of a different species from those that have hereto been analysed, since it contains neither magnesia, nor chrome, substances constantly found in the other aerolites; and in containing a considerable quantity of alumine, traces of which alone have been found in others.

With Chassigny, which Vauquelin analysed in 1816, the most notable features apparent were not just its slightly different bulk chemistry but also the lack of metal and nickel; and again he realised that this was of a different class. The iron was “totally oxidated” and combined with magnesium and iron but Vauquelin does not appear to have realised that it was olivine. The acid-insoluble material was taken by Vauquelin to be “silex as sand” (*i.e.* SiO_2) and gray grains containing chromium and possibly iron (chromite).

The “native irons” attracted considerable interest in their chemistry as well as their internal structure. The discovery of etch patterns in meteorites, Fig. 4, has been reviewed in some detail elsewhere (Paneth, 1960; Smith, 1962; Mehl, 1965). The major motivation for the chemist was to maintain a link between them and the stones through the discovery of common elements, especially after nickel-free stones were discovered. A significant development was the distinction between pallasites and octahedrites which appears first to have been made by Chladni in his catalogue (1819, 1826).

There were considerable developments in the 1820's as a consequence of the almost simultaneous fall of two more eucrites, Jonzac in 1819 and Juvinas in 1821, and because of the application of mineralogical studies. A third eucrite (Nobleborough) fell in the United States in 1823 but attracted little attention in Europe. The analyses of Jonzac and Juvinas (Laugier, 1821; Vauquelin, 1821) showed them to resemble Stannern and terrestrial basalts (Klaproth, 1815; Vauquelin, 1808b). Rose (1825) found further similarities in his mineralogical work. For the chemists they seemed particularly rich in new elements, and this may have acted as a spur to new work. In 1834, therefore, Berzelius suggested that Juvinas, Jonzac and Stannern had a common origin.

By the end of the second decade, therefore, plagioclase-pyroxene achondrites could be distinguished from ordinary chondrites, and Chassigny was recognised as a unique meteorite. Alais was known to contain carbon but this does not seem to have been considered significant. Pallasites were also recognised as being significantly distinct from octahedrites, whereas previously they were taken together as “native irons.” These results were largely dispersed in a number of papers on the individual meteorites and were made over a period of time when analytical techniques were undergoing considerable changes. Their full significance was only to be appreciated when they were repeated by a competent analyst using unquestionable procedures and presented in a single synthesis of progress so far. This was the contribution of Jons Jacob Berzelius and his paper of 1834.

BERZELIUS

Berzelius published analyses on Seres and Bohumiltz in individual papers and Blansko, Chantonay, Loutolox, Alais, Krasnojarsk and Elbogen

collectively (Berzelius, 1828, 1832 and 1834, respectively). He examined the magnetic portion, the acid-soluble portion and the acid-insoluble portion which he fused with sodium carbonate. In addition a portion of the original stones was fused with barium carbonate to determine the alkali metals, Fig. 3, Table 3. One can safely assume that the innumerable techniques developed by Berzelius described by Szavadbary (1966) were also applied to meteorites. Although it had been in common use for many years, Berzelius made great use of the blowpipe for which his book became the standard text (Berzelius, 1820). Every substance was initially examined this way.

Under the blow pipe Seres responded in a manner similar to olivine and this was clearly the acid-soluble constituent. In addition to the metal and sulphide, Berzelius identified the acid-insoluble portion with a fourth mineral which was a silicate of alkali, alumina, iron oxide, manganese oxide, magnesia and lime. He seems at the time of his analysis of Seres to be unaware of the discovery of chromite.

The Bohumiltz analysis was particularly successful, because Berzelius not only identified considerable quantities of phosphorous but located the phase they were in as “metallic phosphorets” (schreibersite) with the composition 65.977% iron, 15.008% nickel and 14.023% phosphorous (also 2.037% silica and 1.422% carbon). He later found these in Elbogen and Krasnojarsk. In addition he found a phase less soluble than the bulk of the metal which he described as “. . . a skeleton, of the form of the iron, black, very light and porous” which from its analysis appears to have been taenite (34.00% nickel).

The 1834 analyses confirmed the earlier results; but, more important, they were extended to include a wide variety of meteorite classes. Blansko and Chantonay could be analysed by the usual techniques, Berzelius now finding chrome-iron (chromite) in Blansko; but, as with its earlier analysts, Berzelius had to resort to different methods for Alais. Berzelius’ analysis of Alais was one of the most complicated performed on meteorites involving a magnetic portion (which he identified as Fe_3O_4), a gas evolved at 100 °C (water and organics), a water-soluble portion (organics and sulphates), a water-insoluble carbonaceous portion and finally the residue which was analysed by alkali fusion. A more comprehensive treatment of Berzelius’ method with Alais is given by Nagy (1975), who also presents a translation of his analysis procedure shown schematically in Fig. 3. Berzelius summarised his views as follows:

It is therefore evident that the Alais meteorite is not the same as the foregoing ones. Neither can it be considered as merely a lump of earth. The presence of metallic iron and its sulphuret, and of oxides of nickel, cobalt, tin, copper, and chromium, which occur in it, proves that this earth has been formed from the usual meteoric mass, which was in this place chiefly composed of

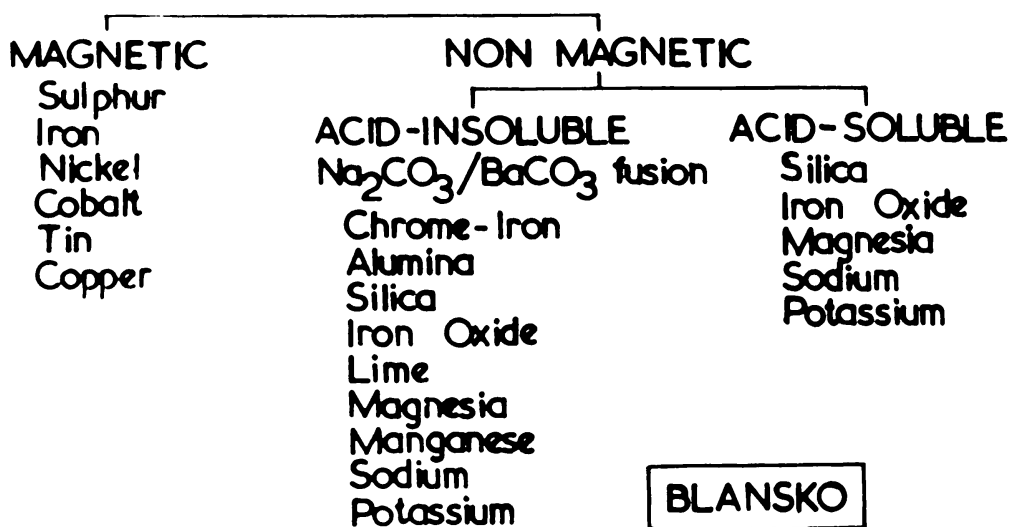
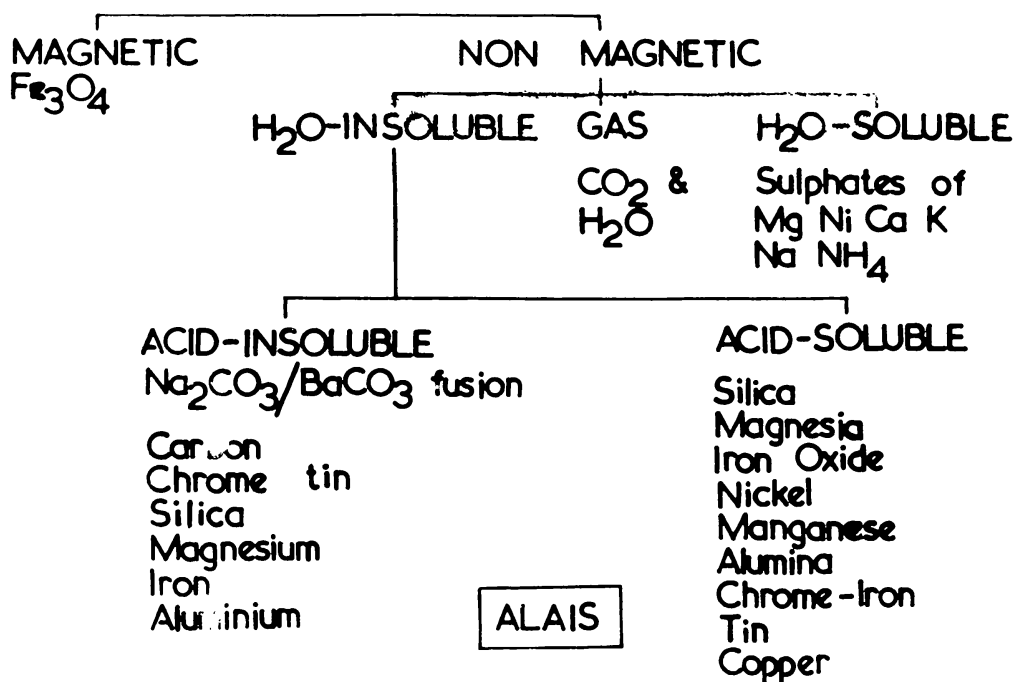


Fig. 3 Schematic illustrations of the analysis procedures used by Berzelius (1834) on Alais and Blansko.

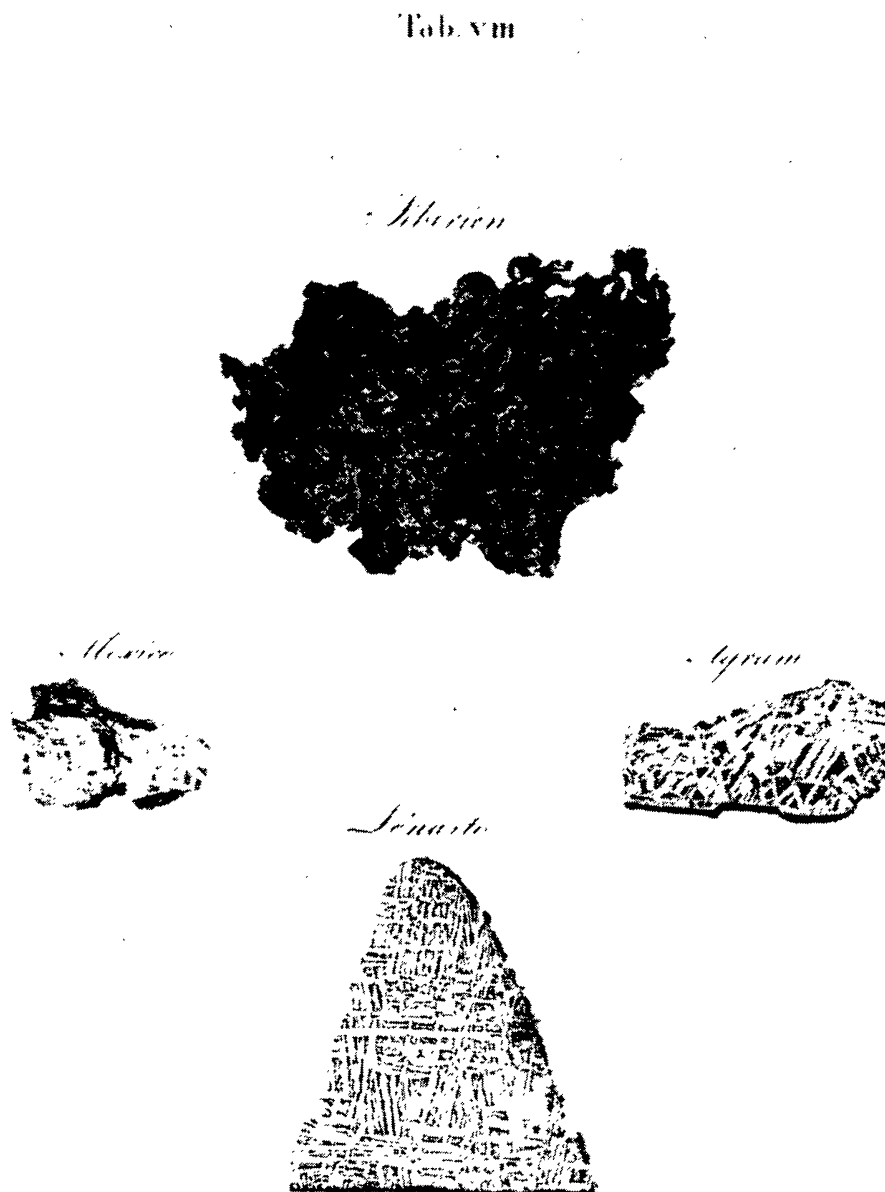


Fig. 4 Direct typographical imprints of the Krasnojarsk pallasite, which has been hand-painted, Toluca ("Mexico"), Hraschina (Agram) and the Lenarto iron. From C.v. Schreibers' "Beytrage Zur Geschichte und Kenntniss meteorischer Stein-und Metall-massen," Vienna, 1820.

meteoric olivine. So there can be no doubt that this stone, in spite of the difference of its external appearance, is but a meteorite which in all probability had its origin in the same situation as other meteoric stones.

In the case of Luotolax there appears to have been a confusion over specimens. Luotolax is a pyroxene-plagioclase achondrite and its similarity to

Table 3
Results of the analyses (percentages) by Berzelius
shown schematically in figure 3

	BLANSKO		ALAIS	
	Acid-soluble	Acid-insoluble	Acid-soluble*	Acid-insoluble**
<i>Non-magnetic</i>				
Silica	33.084	57.145	34.19	38.50
Magnesia	36.143	21.843	24.32	4.17
Lime	3.106	3.106	0.25	—
Protoxide of Iron	26.935	8.592	31.78	22.17
Protoxide of Mn	0.465	0.724	0.28	—
Oxide of Ni+Sn,Cu	0.465	0.021	1.50	4.58***
Alumina	0.329	5.590	2.57	2.08
Soda	0.857	0.931	—	—
Potash	0.429	0.010	—	—
Cr-Fe, + Sn	—	1.533	—	4.38
Loss	1.273	0.505	3.51	
<i>Magnetic</i>				
Iron	93.816		Magnetite	
Nickel	5.053			
Cobalt	0.347			
Tin & Copper	0.460			
Sulphur	0.324			
Phosphorus	trace			

*Also 0.69 chromium and iron and 0.87 oxide of tin mixed with copper.

**Also 21.55 carbon.

***Nickel oxide only. Oxide of tin given 1.67.

the other members of its class was pointed out by Rose (1825). Berzelius found the specimen to consist of olivine and magnetite and clearly therefore this was not authentic Luotolax material; possibly it was Alais without its carbonaceous matter, or Chassigny with its fusion crust. A confusion with Luotolax also occurred in the British Museum collection 30 years later (Maskelyne, 1863).

Berzelius was in an excellent position to reconcile the chemical and mineralogical results. This was because of his own expertise in both fields, and more especially because of the mineralogical work performed during the 1820's. He ended his 1834 paper by summarising the mineralogy of stony meteorites which we may briefly outline:

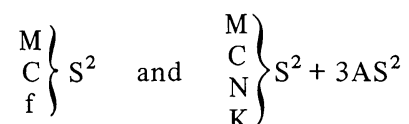
Native iron: usually as small grains in the stone but sometimes constituting the whole mass “generally it is full of cavities containing a gangue.” Unlike Chladni he did not realise the atypical character of Krasnojarsk. It contains nickel in variable amounts and small quantities of other elements.

Iron sulphide: distinct from terrestrial pyrites “containing an equivalent of each constituent” (*i.e.* FeS) which explains the feeble magnetism and acid-solubility.

Magnetic iron oxide: this had been found by Berzelius in Alais (and Luotolax).

Meteoritic olivine: constitutes half the non-magnetic portion having the same formula as common olivine. It contains “as isomorphous substitutes” nickel and manganese silicates and tin. In this respect it also resembled terrestrial olivine. “. . . it is worthy of remark that it scarcely ever contains lime.”

Acid-insoluble silicates: “. . . in which the oxygen of the silica is double that of the bases.” Berzelius suggested that there were two silicates, analogous to pyroxene and leucite and with the formulae,



respectively, where the oxides of calcium, sodium, potassium and aluminium are represented C, N, K and A respectively.

Alloy of iron and chrome: “It is very remarkable that this mineral so constantly accompanies meteoric stones.”

Oxide of tin: contains copper and is found dissolved in the metal and chrome-iron.

An analogous list for the irons would have included native iron, sulphide, “metallic phosphorets,” and the skeletal black iron (taenite).

Berzelius’ 1834 paper was extremely influential and was probably one of the most important written about meteorites since Howard’s in 1802. A point worth stressing is the variety of meteorite classes he examined, and this perhaps reflects a considerable literature survey before commencing work. In all, Berzelius examined what we would now recognise as two H chondrites, an L chondrite, a Cl chondrite, a howardite, IA and IID irons and a pallasite. It must have been apparent to him that there was an enormous range of classes of meteorite.

SUMMARY

The main aim of the early chemists working on meteorites was the determination of the elements present in meteorites and over the period 1800 to 1840 the number rose from six to 19. The discovery of many of them led

to major developments in analytical techniques. The separation of the magnetic phase was universal after the work of Howard (1802) but, although alkali fusion was used by Howard and Klaproth, it was not common in France until after about 1810. The modern procedure of fusing only the acid-insoluble portion did not become universal until about 1815.

The first major mineralogical paper after Bournon's observations in 1802 was that of Rose (1825), although numerous observations had been made during chemical analysis; for example, the unique character of meteoritic sulphide and the presence of "chrome-iron" (chromite). Rose described the mineralogy of Juvinas and Stannern as augite and labradorite from chemical crystallographic observations and stressed their similarity to dolerite.

Berzelius examined a wide variety of meteorite classes, and summarised the major minerals in stony meteorites as metal, sulphide, olivine, pyroxene, feldspar and "chrome-iron." He also found Fe_3O_4 in Alais. In irons he found schreibersite and taenite. Chladni distinguished between pallasites and octahedrites. By 1840 ordinary chondrites, carbonaceous chondrites, plagioclase-pyroxene achondrites, Chassigny, pallasites and octahedrites could all be distinguished.

ACKNOWLEDGMENTS

We are grateful to Dr. R. Hutchison and Mr. A. Bevin (British Museum, Natural History) for help and hospitality and for providing Fig. 4, and Mr. D.E. Secretan (French Department, University of Manchester), Professor A.J. Meadows (University of Leicester) and Mme. Hautecoeur (Librarian, French Academy) for help and information. Dr. R.S. Clarke and Dr. B. Mason provided constructive reviews for which we are also very grateful.

ERRATA AND APPENDIX

Two errors appeared in the first paper of this series (Sears, 1975) which are corrected here. It was Olbers and not LaPlace who previously advocated a terrestrial volcanic origin for meteorites (page 220) and Bakewell (1819) did not favour an extraterrestrial origin but rather a planetary degassing theory (page 223). The importance he attached to the analyses compared with Biot's report is nevertheless apparent. Neither of these errors in any way affects the conclusions of the paper.

Several analyses were consulted and found useful but are not specifically cited in the text above. These were Chilton (1826, Nanjemoy), Stromeyer (1819, Pohlitz), Klaproth (1795-1815, Stannern), Laugier (1827, Renazzo), Faraday (1839, Cold Bokkeveld) and Shepard (1829, Red River).

REFERENCES

- Barthold, C., 1800. Analyse de la pierre de Tonnerre. *J. de Physique*, Ventose, An 8, 169-176.
- Berzelius, J.J., 1820. *Abhandlung om Blasrörets anwendande i kemien och mineralogien*.
- Berzelius, J.J., 1828. Undersökning af en Meteorsten. *Handl. Svenska Vetenskaps-Akad.* 156-163. Also appeared in *Ann. Physik (Poggendorf's)* **16**, 611-618 (1829) in German, and an English abstract appeared in *Edin. J. Sci.* **2**, 373 (1829-1830).
- Berzelius, J.J., 1832. Undersökning af en vid Bohumiliz i Böhmen funnen jernmassa. *Handl. Svenska Vetenskaps-Akad.* 106-119. Also appeared in *Ann. Physik. (Poggendorf's)* **27**, 118-132 (1833).
- Berzelius, J.J., 1834. Om Meteorstenar. *Handl. Svenska Vetenskaps-Akad.* 115-183. Also appeared in *Ann. Physik (Poggendorf's)* **33**, 1-32, 113-148 (1834) and an English summary appeared in *Phil. Mag.* **9**, 429-441 (1836).
- Berzelius, J.J., 1836. *Lehrbuch der Chimie*. Dresden-Leipzig.
- Biot, J.B., 1820. Sur la structure de la substance verte, qui se trouvée dans les cavités de la masse de fer natif découverte en Sibérie par Pallas. *Bull. Soc. Philomath. Paris*, 89-90.
- Chilton, G., 1826. Analysis of the Maryland Aerolite. *Amer. J. Sci. (Silliman's)* **10**, 131-137.
- Chladni, E.F.F., 1819. *Über Feuer-Meteore, und über die mit denselben herabgefallen Massen*. Heuber, Wein.
- Chladni, E.F.F., 1826. Neue Beiträge zur Kenntniss der Feuermeteore und der herabgefallen Massen. *Ann. Physik (Poggendorf's)* **6**, 21-34, 161-182, **8**, 45-60. English abstract appeared in *Phil. Mag.* **67**, 3-20, 179-181 (1826).
- Dree, Et. de., 1803. Account and description of a stone which fell from the clouds in the Commune of Salles, near Ville-Franche, in the Department of Rhone. *Bibliothèque Britanique* **22**, 371-386. English translation appeared in *Phil. Mag. (Tilloch's)* **16**, 217-224 (1803).
- Faraday, M., 1826. In a letter to J. Herschel dated 16 August and published in *The selected correspondence of Michael Faraday* (ed. L. Pearce Williams) **1**, 166-167 (1971).
- Faraday, M., 1839. Chemical account of the Cold Bokkeveld meteoric stone. *Phil. Trans. Roy. Soc.* **129**, 86-87.
- Fourcroy, A.F. de, 1803. Memoir on the stones which have fallen from the atmosphere particularly near L'Aigle, in the Department of l'Orme, on the 26th April last. *Phil. Mag. (Tilloch's)* **16**, 299-305.
- Fougeroux, A.D., L.C. Cadet and A. Lavoisier, 1772. Rapport fait a l'Academie Royale des Science, d'une observation communique par M.

- L'Abbe Bachelay, sur une pierre qu'on prétend être tombée du Ciel pendant un orage. *Observations sur la physique, sur l'histoire naturelle, et sur les Arts (J. de Phys.)* 2, 251-255 (printed and dated 1777).
- Higgins, W., 1811. Description and analysis of a meteoric stone which fell in the County of Tipperary, in Ireland, in the month of August 1810. *Phil. Mag. (Tilloch's)* 38, 262-267.
- Higgins, W., 1819. Account of a shower of meteoric stones which fell in the County of Limerick. *Phil. Mag.* 51, 355-358.
- Howard, E., 1802. Experiments and observations on certain stony substances, which at different times are said to have fallen on the Earth; also on various kinds of native iron. *Phil. Trans.* 92, 168-212.
- Humboldt, A.v., 1849. *Cosmos: A Sketch of a Physical Description of the Universe*. E.C. Ottée's translation, H.G. Bohn, London. 1, 97-125.
- Jackson, C.T., 1838. Chemical analysis of meteoric iron, from Claiborne, Clarke Co., Alabama. *Amer. J. Sci.* 34, 332-337.
- Klaproth, M.H., 1795-1815. *Beiträge zur chemischen Kenntniss der Mineralkörper*. 6 volumes, Posen - Berlin.
- Klaproth, M.H., 1803. Bestandteile mehrerer meteorischer Stein und Metallmassen. *Ann. Physik. (Gilbert's)* 13, 337-342.
- Klaproth, M.H., 1809. Extrait d'une lettre sur les pierres météoriques. *Ann. Chim. et Phys.* 70, 181-184.
- Klaproth, M.H., 1810. Chemische Untersuchung einiger der jüngst gefallen Meteorstein. *Beiträge zur chemischen Kenntniss der Mineralkörper* 5, 245-264.
- Laugier, A., 1806. Memoire sur l'existence de chrome dans les pierres météoriques lu à l'institut de France, le 10 mars 1806. *Ann. Muséum Natl. Nat. Paris* 7, 392-397. Translation in *J. Nat. Phil. (Nicholson's)* 15, 147-149.
- Laugier, A., 1821. Analyse de la pierre météorique de Juvenas. *Ann. Chim. et Phys.* 19, 264-273.
- Laugier, A., 1827. Note sur la composition de la pierre météorique de Ferrare. *Ann. Chim. et Phys.* 34, 139-142. English abstract appeared in *Phil. Mag.* 2, 70 (1827).
- Lavoisier, A., 1772. Sur un effet singulier de tonnerre. *Observations sur la physique, sur la histoire naturelle, et sur les Arts (J. de Phys.)* 2, 310-312 (printed and dated 1777).
- Maskelyne, N.S., 1863. Notices on aërolites. *Phil. Mag.* 25, 437-453.
- Mehl, R.F., 1965. On the Widmanstätten structure. In C.S. Smith (ed.) *Sorby Centennial Symposium on History of Metallurgy*, 245-269. Gordon and Breach, New York.
- Nagy, B., 1975. *Carbonaceous Meteorites*. Elsevier.
- Paneth, F.A., 1960. The discovery and earliest reproductions of the Widmanstätten figures. *Geochim. Cosmochim. Acta* 18, 176-182.

- Pfaff, C.H., 1821. *Handbuch der analytischen Chemie*. Altona.
- Proust, J.L., 1800. Reported in *J. Nat. Phil. (Nicholson's)* **4**, 356-357 and mentioned by him in *J. de Phys.* **57**, 169-204 (1803).
- Proust, J.L., 1805. Sur une pierre météoriques tombée aux environs de Sigena, en Aragon, dans l'année 1773. *J. de Phys.* **60**, 185-204. Abstract appeared in English in *J. Nat. Phil. (Nicholson's)* **12**, 2-7 (1805).
- Reuss, F.A., 1812. Mémoire sur les aérolithes tombées près de Lissa en Bohème, le 3 Sept. 1808. *Ann. Chim. et Phys.* **74**, 84-87. English translation appeared in *J. Nat. Phil. (Nicholson's)* **31**, 224-229.
- Rose, G., 1825. Ueber die in den Meteorsteinen vorkommenden krystallisirten Mineralien. *Ann. Physik (Poggendorf's)* **4**, 173-204. Abstract in English appeared in *Edinburgh Phil. J.* **13**, 368 (1825).
- Sage, B.G., 1808. Du procédé que j'ai employé pour déterminer l'existence de l'alumine dans les pierres meteoriques. *J. Phys. Chim. D'hist. Nat.* **66**, 460-462. English translation appeared in *J. Nat. Phil. (Nicholson's)* **24**, 190-192 (1809).
- Sears, D.W., 1975. Sketches in the history of meteoritics 1: The birth of the science. *Meteoritics* **10**, 215-225.
- Sears, D.W., 1976. Edward Charles Howard and an early British contribution to meteoritics. *J. Brit. Astron. Assoc.* **86**, 133-139.
- Shepard, C.U., 1829. A mineralogical and chemical description of the Virginia aerolite. *Amer. J. Sci. (Silliman's)* **16**, 191-203.
- Shepard, C.U., 1829. Analysis of the meteoric iron of Louisiana (and discovery of Stanniferous columbite in Massachusetts). *Amer. J. Sci. (Silliman's)* **16**, 217-219.
- Smith, C.S., 1962. *A History of Metallography*. Univ. Chicago Press, London & Chicago.
- Stromeyer, F., 1812. Analyse du zu Erxleben im Elb-Department am 15 Apr. 1812 herabdefallenen Meteorsteins. *Ann. Physik (Gilbert's)* **42**, 105-110.
- Stromeyer, F., 1817. Auffindung von Kobalt in dem Meteor-Eisen. *Ann. Physik (Gilbert's)* **56**, 191-194.
- Stromeyer, F., 1819. Chemische Zerlegung des Kostritzer Meteorsteins. *Ann. Physik (Poggendorf's)* **63**, 451-455.
- Stromeyer, F., 1825. Abhandlung über Olivin, Chrysolith und das Fossil in der Pallasischen Meteoreisenmasse. *J. Chem. Physik* **44**, 265-277. Abstract in English in *Amer. J. Sci. (Silliman's)* **13**, 184-185 (1828).
- Szabadváry, F., 1966. *History of Analytical Chemistry*. Pergamon Press.
- Thenard, L.J., 1806. Analyse d'un aérolithe tombée dans l'arrondissement d'Alais, le 15 mars 1806. *Ann. Chim. et Phys.* **59**, 103-110.
- Troili, D., 1776. *Ragionamento Della Caduta di un Sasso Doll'Aria*. Modena.
- Vauquelin, L.N., 1802. Sur les pierres dites tombées du ciel. *Ann. Chim. et Phys.* **45**, 225-246.

- Vauquelin, L.N.**, 1809. Sur l'aérolithe tombée aux environs de Parme pour y découvrir la présence de l'aluminé, annoncé par M. Sage. *Ann. Chim. et Phys.* **69**, 280-284.
- Vauquelin, L.N.**, 1809b. De l'aérolithe tombée à Stannern en Moravie, le 22 Mai 1808. *Ann. Chim. et Phys.* **70**, 321-330. Abstract in English appeared in *J. Nat. Phil. (Nicholson's)* **25**, 54-59 (1810).
- Vauquelin, L.N.**, 1816. Analyse de l'aérolithe tombée aux environs de Langres, et envoyé a l'institut par M. Pistorlet. *Ann. Chim. et Phys.* **1**, 49-54. Translation in English appeared in *Phil. Mag.* **47**, 349-353 (1816).
- Vauquelin, L.N.**, 1821. Aérolite tombée a Juvinas, pres d'Aubenas, Departement de l'Ardeche. *Ann. Chim. et Phys.* **18**, 421-423.
- Vauquelin, L.N.**, 1822. Examen fait, d'après l'invitation de l'académie des sciences d'une aérolithe tombée aux environs d'Epinal, le 13 Septembre 1822, à l'entrée de la foret de Taunière, à trois quart de lieue de la Baffe (Vosges). *Ann. Chim. et Phys.* **21**, 324-328 (1822).
- Walmstedt, L.P.**, 1825. Olivin-ähnliches Fossil aus der Pallasischen Meteor-eisenmasse. *J. Chem. Physik* **44**, 261.
- Warden, D.B.**, 1810. Description et analyse d'une pierre météorique tombée a Weston dans l'Amérique septentrionale, le 4. decembre 1807. *Ann. Chim. et Phys.* **74**, 293-299. English translation appeared in *Phil. Mag. (Tilloch's)* **36**, 32 (1810).
- Webster, J.W.**, 1824. Chemical examination of a fragment of a meteor which fell in Maine, August, 1823. *Phil. Mag.* **63**, 16-19.
- Weeks, M.E.**, 1968. *Discovery of the Elements*. 7th Edition revised and updated by H.M. Leicester. *Jour. Chem. Ed.*

Manuscript received 1/14/77