PAUL-LOUIS CYFFLÉ'S (1724–1806) *TERRE DE LORRAINE*: A TECHNOLOGICAL STUDY*

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Fragments of four Terre de Lorraine biscuit figurines were subjected to porosity analysis, X-ray fluorescence analysis, X-ray diffraction analysis, backscattered-electron image analysis—coupled with energy dispersive spectrometry—and electron backscatter diffraction analysis to determine the porosity, bulk, major, minor and trace element compositions, and the composition and the proportion of their constituent phases. Cyfflé's Terre de Lorraine wares embrace two distinct types of paste, a calcareous and an aluminous–siliceous one. Both are porous (9–25% water adsorption). The former consists of a mixture of different proportions of ground quartz or calcined flint, ground Pb-bearing glass and calcium carbonate with a refractory clay. The firing temperature was between 950 and 1050°C. For the latter, Cyfflé mixed ground pure amorphous SiO_2, ground quartz or calcined flint, ground porcelain, ground Na–Ca-glass and coarse-grained kaolinite with a fine-grained kaolinitic clay. The figurines were fired below 1000°C. The result was a porous, hard paste porcelain-like material. Cyfflé's recipes for both pastes can be calculated from the chemical and the modal analyses.

KEYWORDS: TERRE DE LORRAINE, PAUL-LOUIS CYFFLÉ, LUNÉVILLE, CHEMISTRY, MINERALOGY, TECHNOLOGY

INTRODUCTION

Biscuit and bisque

Ceramic objects with a whitish body (e.g., creamware, porcelain) are fired at least twice. The first firing is called the biscuit stage. The resulting ceramic body, the so-called *biscuit*, is brittle and porous. Dipping it in a glaze suspension leads to the absorption of the glazing elements at its surface. A second firing will cause the glazing elements to melt, and after cooling the result is a vitrified or glassy covering.

Whitish ceramic figurines can be produced with or without a glaze covering. If the body is porous, a glaze is necessary to protect the ceramic object against contamination. But such a glaze is not necessary in non-porous material such as the *bisque* porcelain, which is an unglazed, hard fired, non-porous and translucent material. It is often called *biscuit*.

French white figurines of the 18th century

In France, figurines with a white body were initially made from soft paste porcelain, before kaolin was discovered. This French soft paste or artificial porcelain made without kaolin was first

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Table 1 *Summary of the historical evolution of hard porcelain production in France*

Year	Action
c. 1740	Creation of the soft paste porcelain manufacture of Vincennes
1745	Charles Adam obtains a royal privilege with the exclusive right to produce figurines of humans and animals imitating those of Meissen in Saxe
1751	Production of the first French hard paste porcelain by Paul Hannong in Strasbourg
1752 (21 November)	Soft paste porcelain biscuits currently produced by Vincennes
1754	Paul Hannong's Strasbourg manufacture transferred to Frankenthal in Germany because of the privilege of Vincennes
1756	Vincennes manufacture transferred to Sèvres
1759	Sèvres becomes exclusive royal property. First attempts by Beyerlé in Niderviller to produce hard paste porcelain with kaolin from Passau in Austria with the help of craftsmen from Strasbourg and Saxe
1763	Paul Hannong's son Pierre Antoine sells the secret of the composition of the hard paste porcelain to Sèvres, which then produces hard paste porcelain based on the same principle as the Meissen porcelain (D'Albis 2003)
1764	Niderviller has a shop selling porcelain in Strasbourg
1766 (15 February)	Royal edict protecting the royal manufacture of Sèvres

produced in Rouen by Edme Poterat shortly after 1673 (Soudée Lacombe 2006), followed by Saint-Cloud in 1697, Chantilly in 1726 and later by Mennecy in 1734 (D'Albis 2003). The formula of this type of porcelain, also called *frit porcelain*, is much more complex than that of real, hard paste porcelain (D'Albis 1983). It is obtained from a frit made by melting sand with such flux as sea salt (NaCl), saltpetre (e.g., KNO_3), alum (e.g., $KAl[SO_4]_2.12H_2O$), Alicante soda (Na_2CO_3) and gypsum ($CaSO_4.2H_2O$). This frit was then crushed and finely ground together with white chalk and a marl which turns white after firing. From a technical point of view, the frit was the binding agent and the lime and the marl gave plasticity to the raw paste.

The early history of French hard paste porcelain can be summed up by a few significant dates (Table 1). According to Brongniart (1877), the early Sèvres hard paste porcelain was composed of a mixture of 70 wt% kaolinitic clay, 12 wt% coarse sandy kaolin (where the quartz and kaolin are visible with the naked eye), 9.2 wt% kaolinitic clay sand, 5.3 wt% Aumont sand and 3.5 wt% lime (equivalent to 6.3% limestone, $CaCO_3$). Aumont sand came from the village of Aumont-en-Halatte, situated north of Paris in the department of Oise. Both types of kaolin sand result from the washing of raw kaolin and contain quartz and feldspar. According to Brongniart (1877), hard porcelain paste used for sculpture was obtained by mixing 62 wt% coarse kaolin clay, 17 wt% feldspar, 17 wt% Aumont sand and 4 wt% chalk ($CaCO_3$). This kaolin clay is a nearly pure kaolinite obtained by a mechanical treatment of coarse kaolins. Sèvres manufacture's book of experiments for the period 1768–1780 mentions the testing of more than 200 different recipes (Treppoz and d'Albis 1987).

Paul-Louis Cyfflé (1724–1806) and his Terre de Lorraine

Paul-Louis Cyfflé was born in Bruges on 6 January 1724, and probably attended the Academy of Fine Arts of the town, where he was the pupil of the sculptor Jean Van Hecke (Morey 1871;

Noël 1961; Thomaes and Van den Abeele 2008). The death of his mother caused him to leave Bruges for Paris in 1741. He stayed there for a few years, most likely studying drawing and sculpture before going to Lorraine. In fact, between his departure from Bruges in April 1741 and his arrival in Lunéville at an uncertain date—1746 has been suggested—no one knows exactly where he went or what he did. In Lunéville, he soon became the pupil and then the friend of the sculptor Barthélémy Guibal (1699–1757). On 7 January 1751, he married Catherine Marchal (1728–1795) with whom he had eight children, born between 1751 and 1767. He must have become quickly famous and appreciated, as the former polish king Stanislas Leszczynski (1677–1766), Duke of Lorraine since 1737, agreed to be godfather of his firstborn son in 1751. From then until 1763, the archives refer to him as 'modeler, chiseler and sculptor to the King'. His main important known achievements are the standing statue of Louis XV on Place Royale which he made with Guibal, and the impressive *Fountain of Alliance* on the square of the same name in Nancy.

In Lunéville (Fig. 1), Jacques Chambrette was the first to use a local white clay that he called *Terre de Lorraine*, with which he made ceramics of a beautiful white colour comparable to porcelain, in a separate manufacture he was allowed to set up in Lunéville in 1749. He also created another manufacture in Saint-Clément, situated not far away in the territory of the 'Three Bishoprics' where taxes were lower. After Chambrette's death, the Saint-Clément manufacture was sold in 1763, and Cyfflé worked there, experimenting with different recipes. In 1766, King Stanislas died and Lorraine was annexed to the kingdom of France. At the end of the same year, Cyfflé asked Trudaine de Montigny—general inspector of the 'Ponts & chaussées' (roads and bridges) and in charge of mining administration, also a famous chemist and a member of the
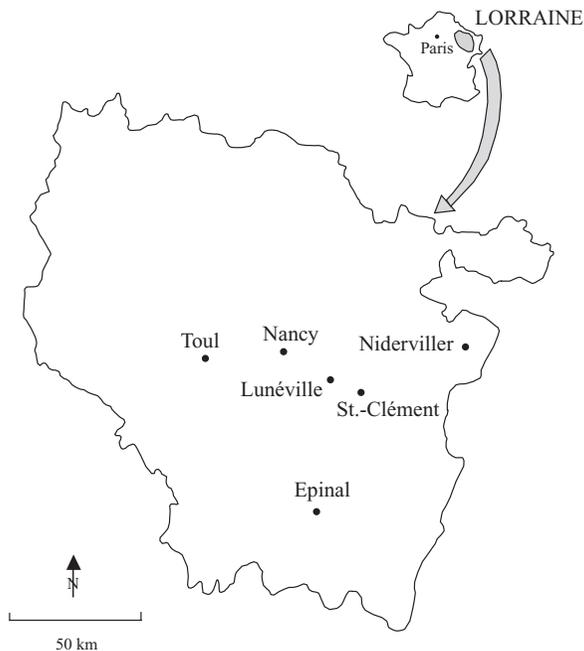
Figure 1   *Map of Lorraine showing the location of places mentioned in the text.*

Academy of Sciences—permission to found his own manufacture of *Terre de Lorraine*, saying that he intended 'to produce at home in Lunéville earthenware which, without actually being porcelain, would be more beautiful than pipe clay, and called *Terre de Lorraine*' (Houry 1954; Noël 1961, 87). On 3 May 1768, an edict of the Council of State authorized him to produce in his Lunéville manufacture rue de Viller 'earthenware named *Terre de Lorraine*, as well as common and ordinary faience using pipe clay', but Cyfflé nevertheless specialized in the production of small unglazed and undecorated white figurines which he had probably already been experimenting with there since the middle of 1765 (Noël 1961, 87–90, 92). According to the same author, in a letter of 1769 he wrote: 'After much care and many renewed experiments, I finally managed to find the solidity of my marble paste which, among other advantages, can withstand washing'. The same year, Trudaine wrote a letter to the minister of finance Bertin, saying that Cyfflé had given him as an experiment a head made of what he called 'marble paste', which Trudaine sent to Bertin, adding 'You will undoubtedly judge that this head could only be made from porcelain paste that the applicant has made harder' (Noël 1961, 90).

His first collaborator there was his young son Joseph, soon followed by many others: in 1775, 27 people were employed there, among whom were nine pupils, but no painter. During these years, Cyfflé acquired a lasting reputation. In 1778, these numbers went down to 11, then eight in 1779, and the manufacture finally closed down in 1780. After failing to set up a new manufacture in Bruges, Cyfflé created another one in 1783 in Hastière-Lavaux, in the county of Namur in Belgium, but it was destroyed during the French Revolution and Cyfflé took refuge in Bruxelles where his children lived. He then retired in the neighbouring suburb of Ixelles where he died on 24 August 1806.

Cyfflé's figurines immediately met with considerable success. The first were sometimes marked with his name *CYFFLÉ À LUNÉVILLE* (Nancy, Musée Lorrain, inv. ML 95.869), along with *TERRE DE LORRAINE* or *TDL*, stamped under the base of several groups. But it is very difficult today to decide whether the remaining items should be attributed to Cyfflé himself or to his numerous and various imitators. As early as 1776, in his admission speech to Nancy Science and Literature Academy, Lecreulx, speaking of Cyfflé's 'Bélisaire', 'Henry IV and Sully' and other groups, said: 'These pieces have been copied everywhere' (Noël 1961, 152). A number of moulds were initially sold when Cyfflé's Lunéville manufacture closed down: in a letter to Lanfrey of Niderviller written in April 1780, Cyfflé said that he had 'moulds to sell' (Noël 1961, 124). Concerning the selling of these same moulds, the *Affiches des Évêchés* of 13 November 1783 (n 46, p. 361), mentioned 'well-preserved moulds, groups as well as figurines and vases, made by famous artists'.

Among others, the manufactures of Saint-Clément, Niderviller, Toul (Morey 1871), but also Épinal are known to have used some of these moulds—sometimes marking their pieces—up to the middle of the 19th century. Maurice Noël even wrote: 'Taking into consideration the fact that Cyfflé's moulds can be found in most Lorraine manufactures, one can wonder in the end which Lorraine manufacture did *not* make so-called 'Cyfflé figurines' (Noël 1961, 166). As to the composition of these particular ceramics, he adds: 'One should not attempt to attach to the name *Terre de Lorraine* too specific a meaning, because of the variety in the composition of the raw material used, as well as because of the difficulty in identifying the manufactures where these little figurines were made. It is almost impossible to distinguish the production of Cyfflé's manufacture from those of Saint-Clément, Lunéville, Toul, Niderviller and even Ottweiler where he briefly stayed before acquiring Saint-Clément' (Noël 1977, 77).

Up to the present, no document has been found giving additional details on how these moulds were acquired, which ones and by whom. Assuredly, Cyfflé did not sell all of them in 1780, and

kept a certain number for himself in view of the transfer of his manufacture first to Bruges, then to Hastière. Some of them may also have found their way to neighbouring manufactures such as Saint-Servais, as shown by Dardenne (Noël 1961, 150).

The secret recipe for making *Terre de Lorraine* biscuit has never been published or handed down to succeeding generations. Cyfflé himself speaks of a marble-like paste (Noël 1961, 89). Whether he introduced pure, white marble ($CaCO_3$), pure limestone ($CaCO_3$), lime (CaO) or hydrated lime (portlandite $Ca(OH)_2$) as flux into the batch is not known. Some authors think that ground bone was one of the elements used to make the paste (Noël 1961, 95). Peiffer (2007, 146) mentions the recipe for soft paste porcelain given by Oppenheim (1807, 272) which could well be very similar to that of Cyfflé: 'It is a mixture of niter, a little sea salt, Alicante soda, alum, gypsum and a lot of siliceous sand fritted together, and then heated to a state of molten paste. After cooling down, finely ground white marl is added to the mixture'. Peiffer (2000, 60) adds that it is a perfectly vitrified paste with a fine grain and a light ivory colour. Peiffer (2007, 148) compares the *Terre de Lorraine* to Tournai soft paste porcelain made from a blend of a frit of Alicante soda (Na_2CO_3) and sand mixed with 40 or 50% marl and lime (Treppoz and d'Albis 1987). According to Peiffer (2003, 2007), *Terre de Lorraine* could be made of refractory mineral particles such as kaolin or any other suitable clay, bound together by a substance either vitreous or in flux added to the paste (frit, glass, calcium phosphate). One would then have to distinguish between: an amorphous body made of a finely ground grog of pipe clay or *derle*—a kaolinitic clay from Belgium (Hauregard 2007)—fired at a high temperature and whose paste contains a large part of calcined stone. This element also serves as a temper agent; a plastic element used in the shaping process, a clay which turns white after firing, whether calcareous or feldspathic; and a powerful vitrifying binding element consisting either of a white chalky clay, a calcium carbonate such as alabaster, a calcium sulphate (gypsum or selenite), or of an alkaline frit bringing about the cohesion of the body. This is not a homogeneous vitrification as in the case of porcelain, but a completely non-porous, heterogeneous bond of the 'fine stoneware' type.

Therefore, following Peiffer (2007, 144–5), several points remained to be cleared up. Is *Terre de Lorraine* a phosphatic soft paste porcelain? Or a frit porcelain with a *terre de pipe* flux? Or a *terre de pipe* body fired at temperatures of white stoneware? Or a Ca-bearing hard porcelain, comparable to early Meissen porcelain?

EXPERIMENTAL

Sampling strategy

Four unglazed figurines from the collections of the Castle of Lunéville, destroyed in the blaze of 2 January 2003, were sampled (Table 2, Fig. 2). These pieces were selected because they could reasonably be attributed to Cyfflé's workshop, either by iconographic evidence, the archives of the museum having also been destroyed by the fire, or thanks to the currently accepted stylistic analyses. Nearly all the pieces of a single figurine bear evident scars caused by the fire, either because they have been blackened to the core, or because they are covered with crusts of glass, molten ceramics or metal coming from the melted showcases. Only the fragments with the least possible damage were chosen. In order to evaluate and analyse the amount of a possible contamination caused by the fire or the water from the firehoses, five different samples were taken from the same figurine 'Hercules and Omphale'.

Table 2 *Description of the analysed specimens from the collections of the Castle Museum in Lunéville*

An. No.	Description
TBL 17	A fragment from the body of a famous group called 'Henri IV and Sully', which is the translation of the main scene of a play called 'Henry IV's Hunting Party' by Collé. Cyfflé took up a print by Gravelot illustrating the 1762 edition of Collé's work, which was first performed in Nancy in September 1766 (Noël 1961, 65). The first figurine was made in 1768, and several examples are known to have been kept in historical collections such as those of Catherine II of Russia or the Prince of Salm in Senones. The Musée Lorrain in Nancy keeps a specimen (ML 95-872) with the quotation from the play inscribed on the base (Nancy 1997, 85, no. 44). For a photograph see Peiffer (2000, 61).
TBL 25	A fragment of a body belonging to a figurine that has not been identified.
TBL 24, 26–29	Fragments of the same group representing 'Hercules and Omphale', which is also one of Cyfflé's most famous figurines. *c.* 1770. Wishing to expiate the murder of one of his friends, Hercules consulted the oracle of Apollo, who advised him to enter the service of Omphale, Queen of Lydia. Although Hercules was the son of Zeus and was famed for his invincible strength, he submitted to the tasks the queen devised for him to expiate his crime. Omphale fell in love with Hercules for his strength and physical beauty, and the two married. A soft paste porcelain figurine representing this scene was first issued by the manufacture of Vincennes in 1752, which probably was at the origin of Cyfflé's version. The Musée Lorrain in Nancy keeps a specimen marked *TERRE DE LORRAINE* with the initials *J G*—for Jean-Baptiste Grandel, who worked in Cyfflé's workshop—engraved in a seal, under the base (ML 60.21.1, ill. Céramique lorraine 1990, 184; Guillemé-Brulon 1995, 74) (Fig. 2). TBL 24: fragment of the head of Omphale; TBL 26: fragment of the banquette on which Omphale is sitting; TBL 27: fragment of the carpet on which Hercules is sitting; TBL 28, 29: fragments of the base.
TBL 34	Most probably a fragment from a mythological figurine representing 'The Sheperd Paris', identified by Mrs Maïté Horiot. It is supposed to bear the stamped mark *TERRE DE LORRAINE* under the base, but unfortunately we could not check it. Cyfflé derived his inspiration from a piece made by Louis Gillet, a sculptor from Lorraine, for his reception at the Academy (Nancy 1968, no. 83). The same subject was also made at Sèvres and Niderviller porcelain manufactures, and the latter still possesses the original mould of this sculpture (ref. F21). Two similar specimens are kept by the Musée Historique Lorrain in Nancy (Inv III 582 D, h. = 230 mm, l = 90 mm, gift of Mr R. Mougenot, and Inv TS 121). The first one bears the stamped mark *TERRE DE LORRAINE* under the base, with the inscription 'Jacque' made by hand with a sharp tool (Noël 1968, 251).

Powder preparation

From the ceramic objects, a small sample was obtained by cutting with a saw. This sample (4–16 g) was ground in a tungsten carbide mill after careful removal of the possibly contaminated surface.

Porosity

Water adsorption of whole fragments (1.2–3.6 g) of seven specimens was measured applying DIN 18132 (1995). Not enough material was available for TBL 25. The samples were dried for 24 h at a temperature of 100°C. After each measurement, the samples were dried again at 100°C and measured again. For every sample, both measurements only differed by 0.01%. The values adopted are the mean between both measures.

Figure 2 *Biscuit figurine in Cyfflé's* Terre de Lorraine *representing 'Hercules and Omphale', 1765–1775. Height: 315 mm; width 300 mm. Mark:* TERRE DE LORRAINE; *initials J. G. (Jean-Baptiste Grandel) engraved in a seal, under the base. © Musée Historique Lorrain, Nancy (inv. 60.21.1). Photo Martine Beck Coppola.*

Chemical analyses by X-ray fluorescence

Two grams of powdered sample were calcined at 1000°C for 1 h to obtain the loss on ignition (LOI); 0.700 g of calcined powder were carefully mixed with 6.650 g of Merck spectromelt A10 ($Li_2B_4O_7$) and 0.350 g of Merck lithium fluoride (LiF). This mixture was put into a platinum crucible and melted at 1150°C for 10 min (Philips PERL X-2) to obtain a glassy tablet. These tablets were analysed for major, minor and trace elements using a Philips PW 2400 wavelength-dispersive spectrometer (rhodium tube, 60 kV and 30 mA). Calibration was made on 40 international standards. Accuracy and precision were checked using laboratory reference samples. Error has been evaluated to be less than 5% for all elements analysed.

Modal compositions

Phase proportions of three representative *Terre de Lorraine* samples were determined by digitized backscattered-electron (BSE) image analysis using the program Adobe Illustrator (Patharea-Cs2-0-1.1b2.sit), integrating an area of $250 \times 192\ \mu m = 52{,}000$ points (TBL 17), $52.0 \times 29.6\ \mu m = 51{,}985$ points (TBL 28) and $150.0 \times 117.7\ \mu m = 30{,}322$ points (TBL 34).

Mineralogical analyses by X-ray diffractometry (XRD)

The mineralogical composition was determined through powder X-ray diffraction (Philips PW 1800 diffractometer, CuKα, 40 kV, 40 mA, 2ϑ 2-65°, measuring time 1 s/step).

Scanning electron microscopy

Backscattered-electron images (BSE) were collected with a scintillator type detector out of polished samples, using a Philips FEI XL30 Sirion FEG electron scanning microscope. TBL 25 was not analysed due to the small size of the sample. The samples were mounted in an epoxy block, flatly polished with a 0.5 µm diamond paste and then coated with a thin carbon layer. Chemical compositions were determined by energy-dispersive X-ray spectrometry, operated at a beam acceleration voltage at 20 kV and a beam current of 6.5 nA. Standardless quantification was performed using an EDAX-ZAF correction procedure of the intensities, using spot analyses (2 µm diameter) as well as larger area analyses (TBL 17: matrix 30×30 µm; Pb-bearing glass particles 8×10 µm; TBL 24, 27–29, 34: 3.6×4.7 µm) of homogeneous areas. In TBL 17, bulk compositions of the relict glass particles were measured in representative areas, integrating both glass and silica polymorphs. The detection limit for most elements was about 0.2 wt%. The reliability of the results was proven by measuring well-known glass standards (DLH2, Corning B, C, D and Obsidian). The relative mean deviation for major and minor oxide components was 2% for concentrations in the range of 20–100 wt%, 4% for 5–20 wt%, 10–20% for 1–5 wt% and >50% for >1 wt%.

Electron backscatter diffraction

The crystalline or amorphous nature of the major phases in TBL 28 and TBL 34 were determined by electron backscatter diffraction (EBSD) (Schwartz *et al.* 2000), following the procedure of Vonlanthen (2007). EBSD measurements require a pristine crystalline lattice extending to within a few nanometres of the specimen surface. If not, the quality of the Kikuchi pattern would be seriously affected. To maximize the EBSD data acquisition, a chemical–mechanical lapping (Fynn and Powell 1979; Lloyd 1987) with a basic colloidal solution (particle size 0.025 µm) was performed. After 4 h of lapping, the samples were rinsed in water. To reduce the charging effects under the scanning electon microscope (SEM) electron beam, silver painting was applied on the ridges of the sample blocks. Finally, each sample was coated with a 2 nm carbon layer using a BalTec MED 020 high-vacuum coating system equipped with a quartz film thickness monitor. The minute carbon thickness applied was sufficient to avoid charging and did not deteriorate the EBSD patterns. The University of Fribourg's EBSD system is mounted on a Philips FEI XL30 SFEG SEM, equipped with a two-sector solid-state forewardscattered electron detector. The diffraction patterns collected on the phosphor screen are recorded with a Firewire 1412 CCD camera. The video frames are fed online into the processing software EDAX (TSL) OIM Data Collection 5.2 on a computer. Typical operating conditions to collect EBSD patterns included an acceleration voltage of between 20 and 30 kV for a probe current of 20 nA (in spot 5). A working distance of 15 mm was used. The tilting of the sample was achieved without a pre-tilt sample holder through a whole stage rotation of 70°. To determine the crystallographic orientation of the source point, i.e., to identify the crystal species, the EBSD Kikuchi patterns were processed through an automated indexing procedure. The EDAX (TSL) OIM Data Collection software proposes three parameters to assess

the reliability of indexing: the confidence index (CI), the fit and the image quality. CI is the most important and may range from 0 to 1. A threshold of CI ≥ 0.2 was used in this study.

<div align="center">RESULTS</div>

Porosity

The material studied is very porous, as shown by the water adsorption (WA), varying between 9 and 25% (WA in %: TBL 17 = 15.9, TBL 24 = 9.1, TBL 26 = 17.5, TBL 27 = 17.7, TBL 28 = 18.8, TBL 29 = 17.9, TBL 34 = 25.2). Four out of the five fragments of the 'Hercules and Omphale' group show very similar WA results, contrary to TBL 24 which has much lower ones. This fragment—Omphale's head—was probably hit harder by the fire than the rest of the group, which caused a decrease of the porosity due to a harder sintering.

Bulk compositions

The analysed sherds comprise two compositional groupings: calcareous, i.e., CaO-rich bodies ($n = 2$, TBL 17, 25); and non-calcareous, aluminous–siliceous bodies ($n = 6$, TBL 24, 25–29, 34) (Table 3, Fig. 3). Both CaO-rich samples TBL 17 and 25 show great differences in their major and minor oxide concentrations. Five aluminous–siliceous bodies (TBL 24, 26–29) show little variation in their major and minor oxides, which is not surprising, as all of them are fragments of the same figurine. Comparatively, TBL 34 is richer in SiO_2 and TiO_2, but lower in Al_2O_3, K_2O and Na_2O. For all the samples, zinc values vary a lot, ranging from 13 ppm to more than 505 ppm (Fig. 3 (d)). It is surprising to see that in the five samples from the 'Hercules and Omphale' group (TBL 24, 26–29) zinc values are not comparable—as is the case for the other oxides and elements—ranging from 118 to 505 ppm. Such a variation is not compatible with a homogeneous paste coming from the same and sole piece. Brehler and Wedepohl (1969) have shown that the zinc values of 454 clays and shales from all over the world, low in bituminous and carbonaceous matter, were below 160 ppm and the one of 1106 granitic rocks, also from all over the world, below 120 ppm. We can therefore conclude that values beyond 200 ppm—as is the case for TBL 24, 26–28—correspond to abnormalities, very probably reflecting a chemical contamination by zinc vapours during the blaze of 2 January 2003 coming out of melting metallic objects. The 118 ppm Zn of sample TBL 29 most likely corresponds to the initial value, or at least to the least contaminated one.

Microstructures and phase compositions

Calcareous body The body of TBL 17 is composed of angular fragments of quartz whose diameters do not exceed 50–70 µm (Figs 4 (a), 5 (a)). Some fragments of sharp-edged K-feldspar can also be observed. Both minerals do not show any sign of reaction with the matrix. Larger circular pores correspond to primary particles rich in CaO, i.e., carbonate or portlandite $Ca(OH)_2$, having reacted with the clay paste during the firing. The diameters of the pores increase as they are closer to the surface of the biscuit, which is the sign of a localized fire blast during the firing of the object or, which is more likely, of the fire action during the January 2003 blaze of the museum. The body also contains sharp-edged fragments of a lead-bearing glass or frit (Table 4) with laths of a SiO_2 polymorph, probably cristobalite in view of their crystal habitus (Fig. 4 (b)). The idiomorphic character of these polymorphs indicates that they crystallized as a liquidus

Table 3   Bulk compositions of the analysed specimens by X-ray fluorescence. Oxides and LOI in wt%, elements in ppm. Fe_2O_3 = total Fe as Fe_2O_3. Other analyses are porcelain body compositions of different 17th and 18th century manufactures (Brongniart 1877), Chinese porcelain bodies (Tite et al. 1984) and inferred composition of the matrix clay. B = Berlin, M = Meissen, S = Sèvres (SF = biscuit figurine), V = Vienna. The number after the letter indicates the production year

An. No.	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Ba	Cr	Cu	Nb	Ni	Pb	Rb	Sr	Y	Zn	Zr	Total	LOI
Cyfflé's Terre de Lorraine bodies																							
TBL17	56.44	0.49	21.37	0.84	0.01	0.42	16.08	0.88	2.46	0.06	318	108	46	15	38	19737	127	342	37	146	146	101.16	0.38
TBL24	63.88	0.07	29.25	0.33	0.01	0.26	1.28	2.15	2.27	0.04	421	13	13	11	31	464	90	145	9	323	88	99.70	0.77
TBL25	81.55	0.32	9.33	0.34	0.01	0.23	6.35	0.94	1.12	0.04	131	36	5	11	10	85	61	116	13	13	87	100.27	0.25
TBL26	64.04	0.11	29.73	0.32	0.01	0.22	1.31	2.22	2.28	0.03	386	3	5	10	26	590	92	146	9	215	94	100.42	0.78
TBL27	63.58	0.08	29.86	0.33	0.01	0.29	1.22	2.11	2.32	0.04	371	23	14	9	32	546	92	146	7	505	87	100.02	0.65
TBL28	63.38	0.07	30.00	0.33	0.01	0.29	1.22	2.21	2.35	0.04	397	6	20	9	33	232	92	146	9	201	93	100.02	0.91
TBL29	63.54	0.08	29.37	0.33	0.01	0.27	1.26	2.20	2.34	0.04	416	9	15	10	33	364	90	145	8	118	93	99.56	0.64
TBL34	71.45	0.30	24.67	0.55	0.01	0.12	0.47	0.92	1.69	0.12	392	29	56	15	19	259	134	130	14	91	63	100.42	0.58
Average of the figurine 'Hercules and Omphale'																							
	63.97	0.11	29.97	0.32	0.01	0.22	1.25	2.23	2.31														
Porcelain bodies from other manufactures (Brongniart 1877)																							
B 1808	66.60		28.00	0.70		0.60	0.30		3.40														
M 1806	58.10		36.70	0.70		0.40	0.20		3.40														
S 1770–1836	58.00		34.50				4.50		3.00														
SF 1794–1834	64.23		30.05				2.89		2.79														
V 1806	61.50		31.60	0.80		1.40	1.80		2.20														
Chinese porcelain bodies (Tite et al. 1984)																							
Yan dynasty underglaze blue	71.1		21.3	1.5		0.1	0.1	1.9	3.9	0.2													
Sung/Yuan dynasty yingding porcelain	79.2		16.1	0.6		0.3	0.7	0.5	2.4	0.3													
Inferred composition of the matrix clay																							
TBL 17	58.9	0.75	33.09	1.3	0.01	0.65		1.36	3.81	0.09													
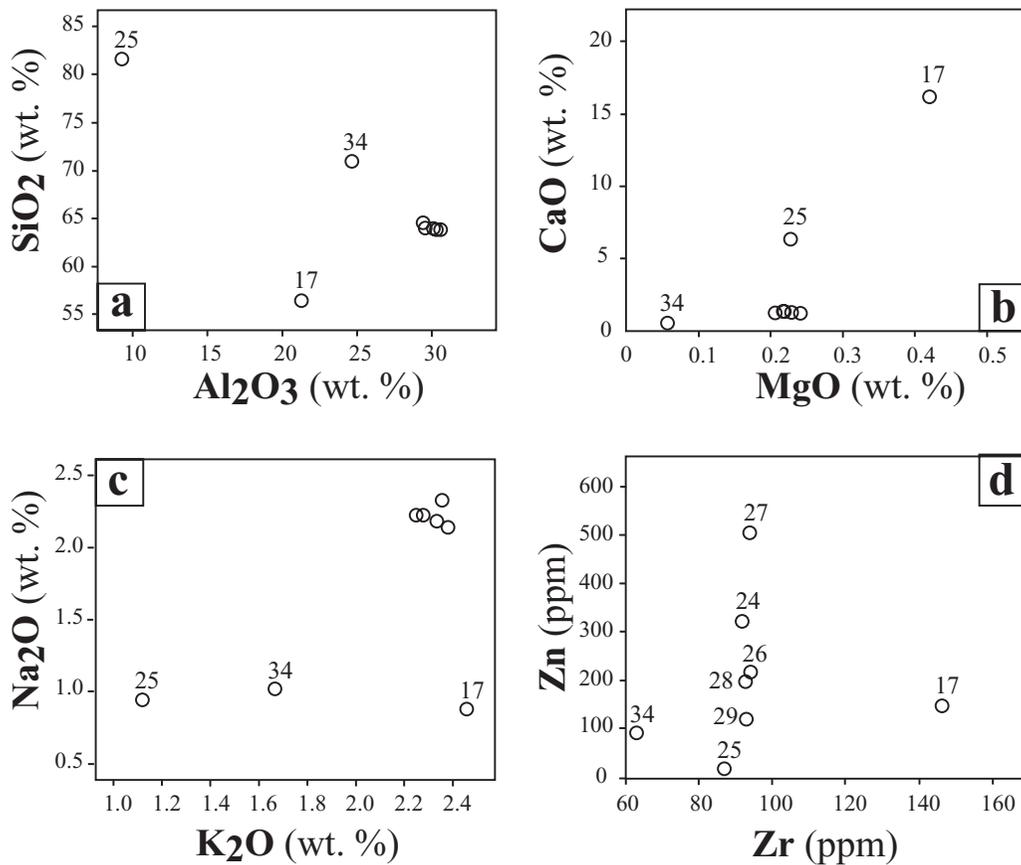
Figure 3   *Body (bulk) compositions for the analysed samples displayed on bivariate plots of selected oxide and elements.*
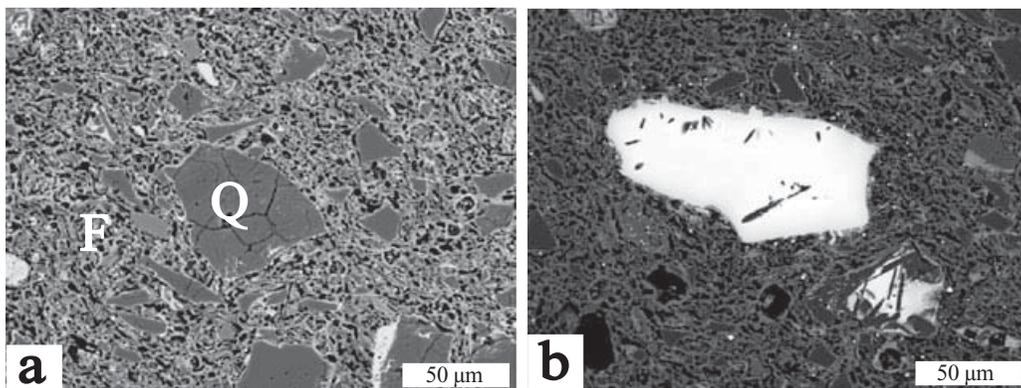
Figure 4   *Backscattered-electron images of calcareous sample TBL 17. (a) The body showing angular, unreacted quartz (Q) and K-feldspar (F) set in an extensively vitrified, porous matrix. (b) Two lead-bearing glass fragments (white) with precipitated SiO$_2$ polymorph needles and laths (grey), probably cristobalite.*
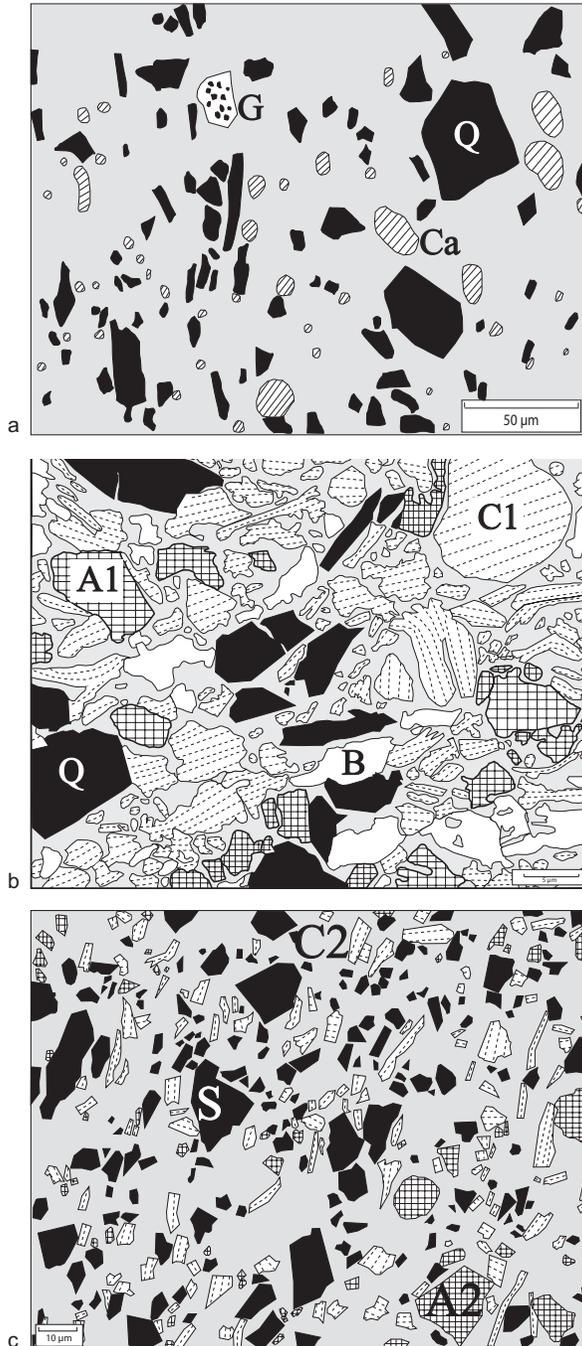
Figure 5 *Line drawings from backscattered electron images and element mappings. Grey = matrix and pores. (a) Calcareous figurine TBL 17. Ca = CaO-rich area; G = lead-bearing glass; Q = quartz. (b) Fine-grained, aluminous–siliceous figurine TBL 28 with phases A1, B, C1 (meta-kaolinite) and Q (quartz). (c) Coarse-grained, aluminous–siliceous figurine TBL 34 with phases A2, C2 (meta-kaolinite) and S (amorphous SiO$_2$ or quartz).*

Table 4 Energy-dispersive X-ray spectrometry analyses of Pb-bearing glass fragments (four representative analyses) and the matrix of the calcareous figurine TBL 17 as well as of the main phases in Cyfflés non-calcareous, aluminous–siliceous Terre de Lorraine

	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	P_2O_5	PbO	Al_2O_3/K_2O	Al_2O_3/SiO_2
Pb-bearing glasses (frits)												
TBL 17-h	60.9		4.9	0.8	0.5	2.7		5.3		24.9		
TBL 17-k	79.7		3.4	1.0	0.3	1.3		3.3		11.0		
TBL 1-7n	72.2	0.2	3.9	0.5	1.4	10.2		2.5	0.7	8.4		
TBL 1-7u	55.7	0.2	6.3	0.7	3.0	17.7		3.9	0.9	11.6		
Matrix												
TBL 17 (*n* = 10)	47.0	0.6	26.2	1.1	0.7	18.8	0.2	2.2		3.2		
Non-calcareous, fine-grained body (TBL 24, 27–29)												
Type A1 (*n* = 12)	73.5		19.1	0.3	0.6	0.7	2.1	3.9				
Type B (*n* = 11)	61.2		25.9	0.3	0.2	5.9	5.2	1.2				
Type C1 (*n* = 22)	51.8		42.4	0.5	0.9	0.5	1.6	2.7	0.2		15.70	0.81
Matrix (*n* = 2)	57.1		37.5		0.4	0.9	1.2	2.8			13.39	
Non-calcareous, coarse-grained body (TBL 34)												
Type A2 (*n* = 12)	68.1		20.8	0.3		0.6	1.7	9.0				
Type C2 (*n* = 10)	53.7		42.2	0.4		0.5	0.7	1.8	1.3		23.44	0.78
Amorphous SiO_2 (*n* = 10)	97.7		2.3									
Matrix (*n* = 10)	65.0	0.3	31.4	0.7	0.5		0.3	1.7			18.47	
Kaolinites (Grim 1968)												
Zettlitz, CS	54.5		43.4	0.8	0.3	0.3	0.5	1.0				0.79
Mexia, Texas	53.7		45.3	1.1	0.4	0.5						0.84
Macon, Georgia	53.7		44.0	0.4	0.6	0.6	0.4	0.6				0.82
St. Austell, GB	54.1		43.7	0.7	0.3	0.2	0.1	1.7				0.81
Anna, Illinois	53.5		44.2	1.4	0.5	1.2	0.2	0.4				0.83

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Table 5 *Modal compositions and calculated recipes*

An. no.	Flint (TBL 17, 28) or amorphous SiO₂ and few flint (TBL 34)	Coarse CaO-rich area	Frit	A1 (TBL 28), A2 (TBL 34)	B	C1 (TBL 28), C2 (TBL 34)	Matrix	Pores	Clay	CaCO₃	Ca(OH)₂
Modal analyses (vol. %)											
TBL 17	15.4	3.8	0.7				64.2	15.9			
TBL 28	13.4			10.3	10.8	33.9	12.8	18.8			
TBL 34	17.1			4.9		9.9	42.9	25.2			
Calculated recipe (TBL 17: wt%, TBL 28,34: vol.%)											
TBL 17a	18.4		2.0						50.9	28.7	
TBL 17b	18.4		2.0						58.4		21.2
TBL 28	17.5			13.5	14.1	44.2			10.7		
TBL 34	23.0			6.5		13.2			57.3		

phase during cooling and not during a subsolidus reaction, such as devitrification. The chemical composition of these glass particles is highly variable (Table 4). Matrix analyses revealed amounts of PbO around 3.2 wt% (Table 4). The proportions of the major phases (in vol.%) of TBL 17, based on quantitative BSE image analysis of Figure 5 (a), are given in Table 5.

Aluminous–siliceous bodies   The five samples of the group 'Hercules and Omphale', characterized by a homogeneous chemical composition, show a very fine-grained texture, with grain diameters or lengths below 5–10 μm (Fig. 6 (a)). These bodies contain four major phases (Fig. 6 (b) to 6 (d)), embedded in a small quantity of a K + Na + Ca-bearing, aluminous and siliceous matrix made up of small platy grains of former clay minerals: angular fragments of a silica polymorph, identified by EBSD as α-quartz (Fig. 6 (e) and 6 (f)); roundish grains of a K-rich, aluminous–siliceous phase A1; roundish grains of a Ca–Na-rich aluminous–siliceous phase B; and platy grains of a Al-rich siliceous phase C1 with a marked, phyllosilicate-type cleavage. Moreover, some sporadic rutiles can be observed.

TBL 34 shows a more heterogeneous body than the five fine-grained fragments (Fig. 7 (a)). The constituents are also coarser, some of them reaching 70 μm. One can recognize a great quantity of angular fragments of, as evidenced by EBSD measurements, amorphous SiO₂ of very variable sizes, together with very few angular fragments of α-quartz grains like those of the fine group, as well as A2 type grains and C2 fragments with a fold-like structure (Fig. 7 (b) to 7 (d)). The porous matrix is composed of a blend of small fold-like particles with quartz grains.

According to EBSD, phases A1 and A2 are both mostly amorphous. The grains A1 in the fine-grained bodies show two types of minute crystalline inclusions, set in a homogeneous glassy matrix: irregularly shaped grains, identified by EBSD (Fig. 8 (a), 8 (c), 8 (e)) analyses as α-quartz, are interpreted as corroded quartz relicts, set in a glassy matrix, due to a prograde firing (Fig. 6 (c)); elongated, acicular and lens-shaped crystallites with a high Al₂O₃ to SiO₂ ratio, typical for mullite (3Al₂O₃.2SiO₂). The euhedral outlines most probably resulted from crystallization from a melt during a retrograde reaction, i.e., the cooling of this liquid (Fig. 6 (d)). Their shape is compatible with the orthorhombic {110} prism, typical for mullite—the diamond pattern in Figure 6 (d) corresponding to (001) cross-sections, the needles to longitudinal sections of such a prism. The EBSD patterns of these crystallites correspond to mullite (Fig. 8 (b), 8 (d), 8 (f)). No
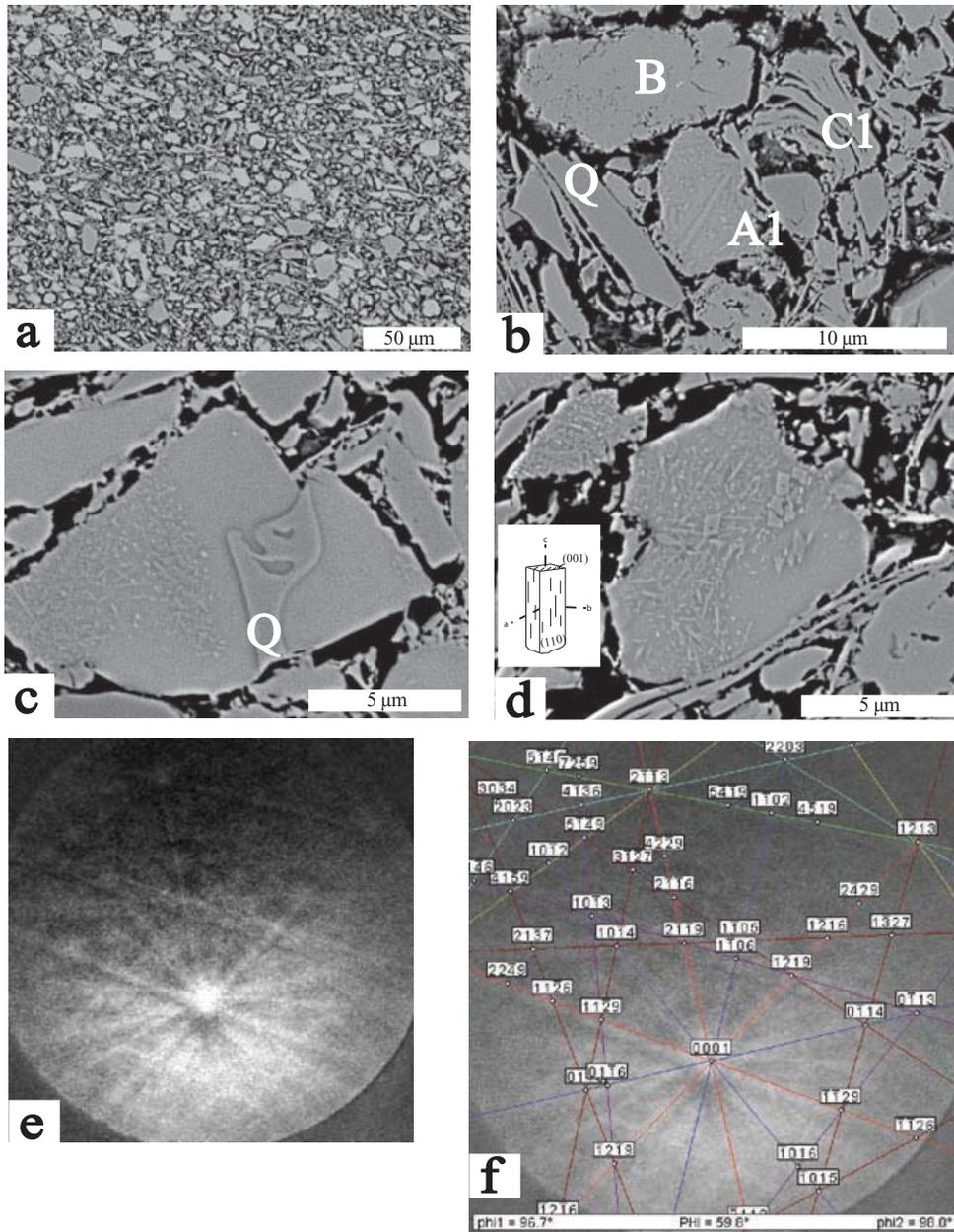
Figure 6 *Backscattered-electron images of the fine-grained, aluminous–siliceous* Terre de Lorraine. *(a) Sample TBL 27 showing a homogeneous distribution of angular, platy and fibrous grains. (b) Sample TBL 28 showing foliated and bent meta-kaolinite grains (C1) and angular quartz (Q) associated with rounded grains type A1 and B. In the A1 phase, acicular microcrystals of mullite are visible. (c) A glassy grain type A1 with tiny mullite needles (left) and an irregularly shaped, corroded α-quartz crystal (Q), implying resorption by the surrounding former melt. Sample TBL 28. (d) A glassy A1 grain showing diamond pattern (001) cross-sections and elongated (110) prismatic sections of mullite (white), embedded in a homogeneous glassy matrix. The idealized orthorhombic crystal habit is shown in the insert. Sample TBL 28. (e) EBSD Kikuchi pattern of a α-quartz (b). (f) EBSD pattern (e) indexed as α-quartz. Confidence index = 0.2.*
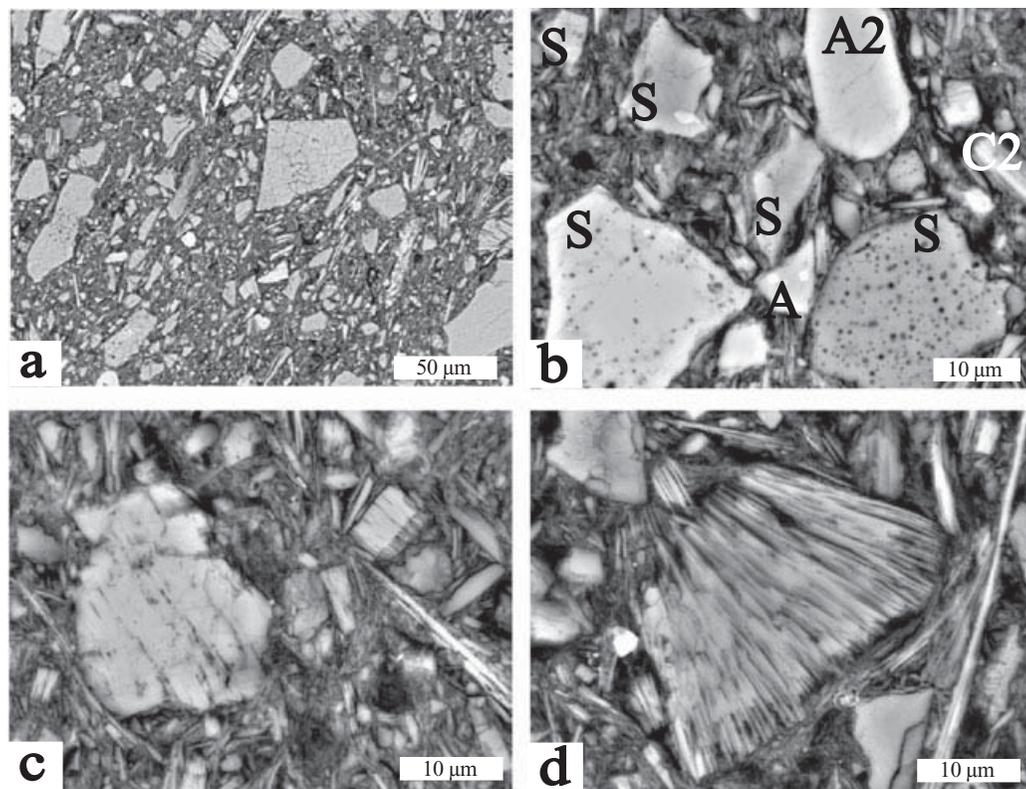
Figure 7 *Backscattered-electron images of the coarse, aluminous–siliceous* Terre de Lorraine *TBL 34. (a) Angular quartzes and amorphous SiO₂ and platy as well as fibrous meta-kaolinite grains in a fine-grained matrix. (b) The typical association of phase A2, amorphous SiO₂ with tiny hollows (S) and meta-kaolinite (C2). (c) Aspect of phase A2 with typical striation. (d) Coarse meta-kaolinite flake C2.*

discrete crystalline inclusions were detected in the A2 phases of the coarse-grained body TBL 34. They show a parallel alignment of very tiny bubbles, typical of a former melt (Fig. 7 (c)). Phase B, present in the fine-grained, but lacking in the coarse-grained body is, according to the EBSD measurements, also amorphous. The microgranular aspect evokes a glassy frit (Fig. 6 (b)). In phases C1 and C2, no microcrystalline inclusions could be detected, even at high magnifications. These phases are amorphous as well, as shown by EBSD.

Representative compositions of the phases A1, A2, B, C1, C2 and of the two matrices are listed in Table 4. The phases A1, A2 and B have compositions which cannot be related to a known mineral. Phases C1 and C2 are chemically not very different in both bodies. Their compositions are very similar to those of the mineral kaolinite (Table 4). The higher K_2O and Na_2O are interpreted to pertain to relict mica, converting to kaolinite (Tite *et al.* 1984). The Al_2O_3/K_2O ratio is different for C1 (15.70) and C2 (23.44), proof that different meta-kaolinites were used as ingredients for the two pastes.

The matrices of the two bodies differ chemically, as well as the coarse meta-kaolinites C1 and C2. Their higher SiO_2 and their lower Al_2O_3 contents can be explained by the presence of very small grains of α-quartz. Their Al_2O_3/K_2O ratio differs (fine-grained body: 13.39; coarse-grained

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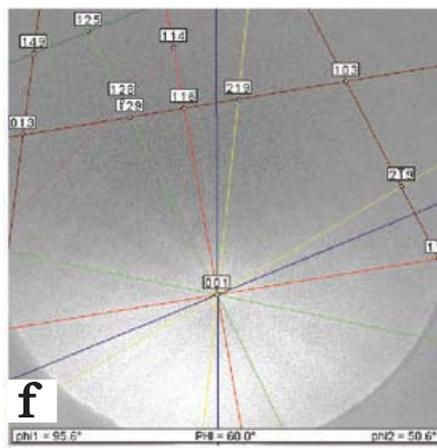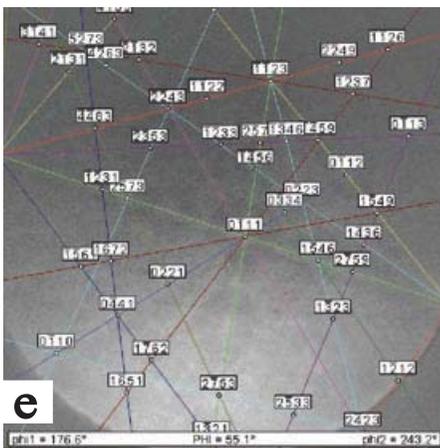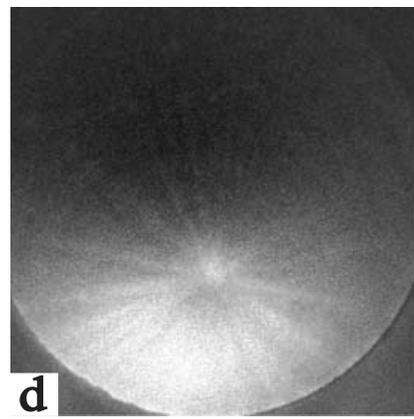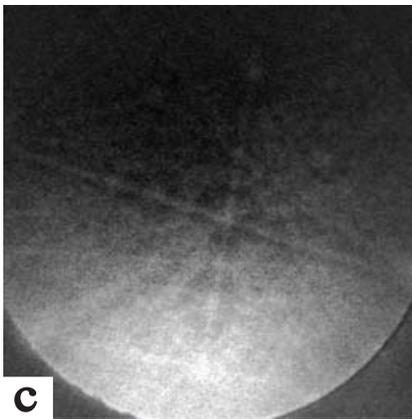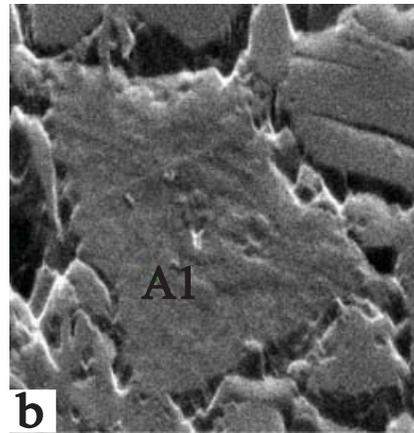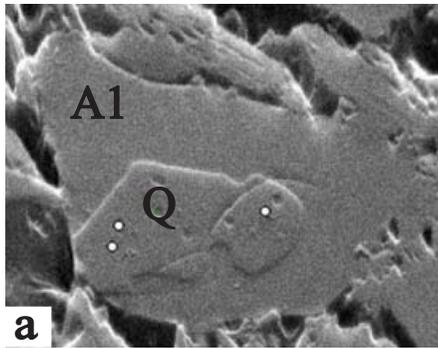
Figure 8   *Sample TBL 28. (a) Secondary electron image of a grain A1 with relictic quartz (Q). Stage in EBSD position, tilted by 70°. Circles = position of pattern centres. (b) Secondary electron image of a grain A1 filled with needles of mullite. Stage in EBSD position, tilted by 70°. (c) EBSD Kikuchi pattern of the quartz from (a). (d) EBSD Kikuchi pattern of a mullite needle from (b). (e) EBSD pattern (c) indexed as α-quartz. Confidence index = 0.35. (f) EBSD pattern (d) indexed as mullite. Confidence index = 0.30.*

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body: 18.47), as well as that of their coarse meta-kaolinites C1 and C2. The chemical composition resembles that of a quartzo-kaolinitic clay.

It is apparent from Figure 9 that the plotted points of the amorphous phases occupy different areas of the diagrams. Grains A1 and A2 are richer in SiO_2 and K_2O, but lower in CaO and Na_2O than the glassy phase B. Very high SiO_2 values, occurring in some A1 analyses, are due to the presence of relictic α-quartz. A2 is significantly richer in K_2O than A1. The SiO_2 versus Al_2O_3 plot shows a strong negative correlation for the A1, B and C1 phases, and a clustering of phases A2 and C2 (Fig. 9 (a)). A positive Na_2O/K_2O correlation is evidenced for phase B in Figure 9 (b).

Modal compositions of the fine-grained *Terre de Lorraine* TBL 28 (Fig. 5 (b)) and the coarse-grained TBL 34 (Fig. 5 (c)) are given in Table 5.

X ray diffraction

The studied samples can be assigned, as shown by their X-ray diffractograms, to three different mineral associations:
(a) α-Quartz + plagioclase + hematite + wollastonite + gehlenite + calcite (TBL 17)
(b) α-Quartz + cristobalite + plagioclase + calcite (TBL 25)
(c) α-Quartz + cristobalite + sanidine + mullite (TBL 24, 26–29, 34)
Associations (a) and (b) pertain to the calcareous, and association (c) to the aluminous–siliceous bodies. Calcite is a post-firing, secondary phase (Maggetti 1994) in the first two associations, as revealed by optical microscopy analysis. In association (c), both low- and high-temperature silica polymorphs are present. As shown by BSE imaging, two types of α-quartz occur in these bodies: coarse, crushed and unreacted grains; and relict, xenomorphic inclusions in the glassy phase A1. No cristobalite or sanidine was detected in the SEM analyses. Consequently, both are interpreted to occur as minute (sub-µm) crystallites in one or more of the phases A1, A2 and B. Mullite occurs as equilibrium crystallization produced during the cooling of the former melt of phase A1 and is probably also present in the meta-kaolinitic phases C1 and C2.

<div align="center">DISCUSSION</div>

The preceding text has shown the composition complexity of the *Terre de Lorraine* specimen likely to have been produced by Paul Louis Cyfflé in his Lunéville workshop during the years 1768–1780, indicating clearly that several recipes were used by the ceramic sculptor for his production.

Classification

Ancient classification   In 18th and 19th century France, the names and classification of white earthenware (named '*faïence fine*') were complex and often contradictory. The wide range of these names was discussed by Peiffer (2003) from a technical and historical point of view, and more recently by Maire (2008). If we want to have the opinion of a specialist from the beginning of the 19th century, we can for instance refer to Brongniart (1770–1847), director of the porcelain manufacture of Sèvres from 1800 to 1847, who distinguishes three types of white earthenware in the first edition (1844) of his fundamental treatise (Brongniart 1877): the so-called *terre de pipe* ('faïence fine marnée') with a calcareous body; the creamware or cream-coloured earthenware ('faïence fine caillootée', 'cailloutage', 'terre anglaise'); and the ironstone ('faïence fine dure', 'faïence fine feldspathique').
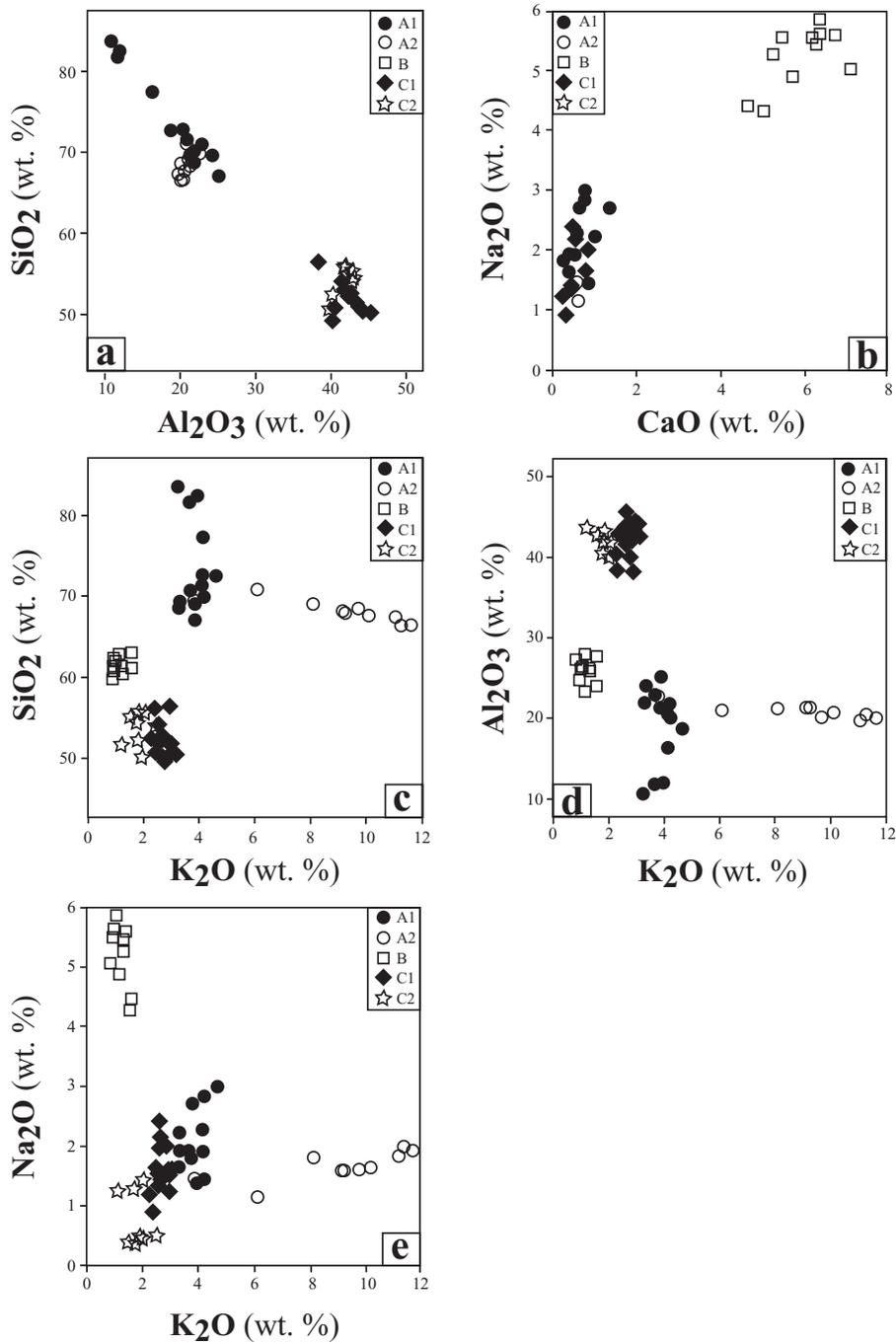
Figure 9   *Bivariate plots showing the compositions of the glasses A1 (fine-grained body), A2 (coarse-grained body), the frit B, and the meta-kaolinites C1 (fine-grained body) and C2 (coarse-grained body).*

The first body is a production from Lorraine, created before 1750 in the faience manufactures of Lunéville and Saint-Clément in northeastern France (Fig. 1). We have already seen that in 1749, Jacques Chambrette was authorized to add to the existing manufacture which produced tin glazed earthenware a second unit specializing in '*terre de pipe* imitating that which was produced in England'. He must have experimented with clays that turn white after firing at a much earlier date, for in 1748 he invited the Court of the Duke of Lorraine along with Voltaire and Madame du Châtelet to be present at his experiments with this new paste (Noël 1961, 38). But the production of objects made of *terre de pipe* must have occurred even earlier, because Chambrette could have used recipes that already existed. We must not forget all the previous experiments made and the results obtained in and around Paris in the early 1740s: in 1743, Claude Imbert Gérin had already obtained an exclusive privilege for the production of white-bodied earthenware 'as it is made in England' in his Paris manufacture, and the new 'Royal Manufacture of French Faience imitating that of England' was set up in Pont-aux-Choux in 1749.

The other two ceramic bodies were invented in England. Creamware bodies appeared in Staffordshire in the 1750s (Towner 1978; Lockett and Halfpenny, 1986). Josiah Wedgwood I (1730–1795), using refined ball clay and calcined flint, produced his famous Queen's ware. From 1775, the addition of Cornish china clay and china stone resulted in a body called 'ironstone'.

The three types of *white earthenware* are artificial pastes, obtained by mixing: one or more plastic materials such as kaolin or China clay, or highly plastic, kaolinitic and refractory clays (ball clays); one or more non-plastic materials such as calcined and finely milled flint, quartz and quartz-rich sands or ground fired clay (= grog); and fluxing materials such as lime, chalk, limestone, dolomite, giobertite, feldspar, Cornish stone, pegmatite, frit, glass, fusible sand and bone ash. The fluxing material of *terre de pipe* is lime, the analyses of which give CaO amounts of around 10–15 wt%, and feldspar for the third group.

Modern classification   Clay-based ceramic objects with a white body belong either to the family of the *white earthenware*, if they have a water adsorption >2%, or to the *porcelains*, if their water adsorption is <2% (Salmang and Scholze 1968, Table 45). The ceramic objects that have been studied here therefore all belong to the first category. Today, four types of *white earthenware* are distinguished: calcareous white earthenware; siliceous white earthenware; feldspathic white earthenware; and mixed white earthenware which contains lime and feldspar as fluxing elements. The first type is also called soft white earthenware, and the others hard white earthenware.

If the classification of TBL 17 and 25 in the calcareous white earthenware group is not a problem, that of the other two figurines made from an aluminous–siliceous paste (TBL 24, 26–29, 34) is much less evident, given the heterogeneity of the paste. If one considers the rather high values of alkalis and the small quantity of CaO in the bulk analyses, and if one takes into account the high porosity, these objects could be classified in the group of mixed white earthenware. But the bulk composition of the fine paste is very similar to that of Vienna hard paste porcelain (Table 3). It is also comparable to that of hard paste porcelain Sèvres figurines but, because of its low CaO value, it is different from early Sèvres hard paste porcelain, which is contemporary to *Terre de Lorraine*. On the contrary, from a chemical point of view, TBL 34's coarse paste is notably different from the hard paste porcelain of these manufactures, and could rather be compared to a Chinese hard paste porcelain (Table 3).

The porcelain phases A1 and A2

Collectively, SiO_2, Al_2O_3 and K_2O constitute 94–98 wt% of the A1 and A2 phases in the fine- or the coarse-grained aluminous–siliceous *Terre de Lorraine* bodies. Consequently, phase diagrams
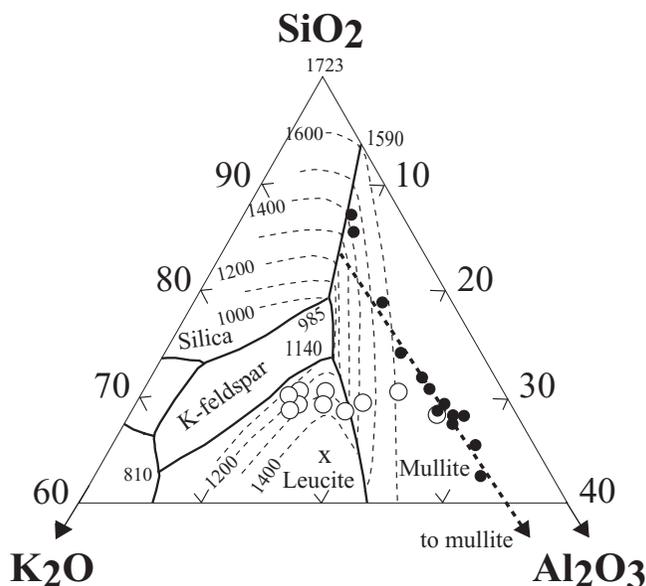
Figure 10 *Compositions (wt. %) of phases A1 (dots) and A2 (open circles) plotted on the high-silica part of the SiO₂–K₂O–Al₂O₃ phase diagram (simplified after Osborn and Muan 1960). Temperatures in °C. Isotherms are shown every hundred degrees in the silica, leucite and mullite primary fields. The heavy dashed line is a projection between mullite through the alignment of the A1 plots towards the mullite–silica cotectic line. X = K-feldspar.*

such as the ternary SiO_2–K_2O–Al_2O_3 system, can be used to constrain the origin and firing temperature of these phases (Fig. 10). CaO, Fe_2O_3, MgO and Na_2O contents were assumed to be negligible. It can be seen from this figure that the A1 compositions all fall within the mullite primary field of the phase diagram.

A clustering around 30 wt% SiO_2 can be observed in a part of the diagram where Chinese porcelain can also be found (Tite *et al.* 1984; papers in Shixiong 1986). The A1 fragments are therefore interpreted as finely crushed porcelain bodies, consisting of varying glass + crystallite mixtures.

The diagram also shows a visible positioning of the analyses on a line ranging from the representative point of the mullite to the silica + mullite cotectic at 1200–1250°C. This peculiarity can be explained by a lack of chemical homogeneity of these very fine particles, which, at micron scale, show variable proportions of quartz and of kaolinitic clay in the original paste, the K-feldspar flux remaining proportionally constant. Under equilibrium conditions and if other flux such as Na_2O is not taken into account, the original paste begins to melt at 985°C, i.e., the ternary eutectic point. After dissolution of all the K-feldspars, it evolves along the silica + mullite cotectic until the isotherms of 1200–1250°C. At these temperatures and for compositions plotting on the line mullite–silica + mullite cotectic, all quartz will have melted. The melt migrates in the primary field of mullite along a line directed to the mullite point on the binary system Al_2O_3–SiO_2. The presence of disequilibrium quartzes and equilibrium mullite crystallites in mostly all A1 grains suggests that the firing was below these cotectic temperatures of 1200–1250°C, which are close to the 1200–1300°C range of Chinese porcelain firing (Wood 1986). The A2 phases are clearly richer in K_2O than the A1 phases. This suggests a larger proportion of K-feldspar in the body made of kaolinitic clay + quartz + K-feldspar. Accordingly, the great majority of the A2

analyses cluster together in the field of leucite. As shown in Figure 10, the analyses gather along a horizontal line which goes from the major concentration of the A1 around 30% SiO_2 to values richer in potassium. This suggests that Cyfflé probably took paste A1 to begin with, keeping silica constant, but varying the quantities of added K-feldspar and kaolinitic clay. For the compositions situated right of the line SiO_2–K-feldspar, the fusion begins at the eutectic temperature of 985°C. For many of the A2 compositions, after the dissolution of all the quartz, the melt follows the cotectic K-feldspar + mullite towards the peritectic point of 1140°C and eventually the cotectic leucite + mullite. For analyses plotted on the left side of this line and under equilibrium conditions, the first melt is generated in the ternary peritectic of 810°C and follows the cotectic leucite + K-feldspar. No primary quartz can therefore survive in these grains, which is proved by SEM observations where this mineral could not be detected, contrary to A2 grains. One also realizes that most A2 grains must be completely melted at temperatures of 1150–1350°C. This is perfectly in keeping with their amorphous nature revealed by EBSD.

How did Cyfflé make these porcelains? In this context, it must be recalled that Claude Humbert Gérin made hard paste porcelain at the manufacture of Vincennes around the year 1747, using a kaolinitic clay called 'Terre de Chimay' from the Principauté of Liège (D'Albis 1984). Cyfflé could have succeeded in making porcelain by himself if he had used a high-temperature kiln capable of reaching 1200–1250°C or if he had placed the mixture of different phases fired to a biscuit state beforehand on the floor of a tin glaze faience kiln (Diderot and d'Alembert 2002), where at the time the enamel was usually prepared with a frit at temperatures going far above 1100°C (Rosen 1995; Maggetti 2007b). But Cyfflé could also have bought refuse sherds—these must have been biscuit fired bodies, because no glaze was found in the A1 and A2 fragments—from other porcelain manufactures, which would explain the different chemical composition. This hypothesis must not be retained, for porcelain biscuits are normally fired at temperatures much lower than 1200–1350°C. He may very well have purchased broken, unglazed porcelain kiln furniture from other manufactures. This ceramic material suffered many high-temperature firing cycles in the porcelain kilns and the recycling of these huge amounts of useless material would have been very welcome for the porcelain manufactures.

The amorphous SiO_2 phase

The nature and the origin of this phase is puzzling. It cannot be a siliceous glass, because in order to produce it, the pure quartz would have to be melted at a temperature of 1713°C. The little aluminium (2.2 wt%, Table 4) of the vesicular portions lowers the fusion point to 1590°C, corresponding to the temperature of the binary eutectic point situated in the siliceous part of the binary system SiO_2–Al_2O_3, which is the place where the first melt is generated (Fig. 10). All the same, this temperature is still too high for the kilns of the 18th century. This enigmatic phase is, rather, a naturally white amorphous substance that Cyfflé used without any previous thermic treatment. It could be either flint or opal (amorphous $SiO_2.nH_2O$).

Recipes

As stated in the introduction, none of Cyfflé's recipes have been handed down to posterity. One therefore has to rest on scientific analyses for indications about the process he used. The historical data clearly show that he produced his *Terre de Lorraine* along with more traditional pipe clay (*terre de pipe*). If all the pieces analysed here have indeed been made in his manufacture, these analyses clearly confirm that he used two major recipes, i.e., a calcareous and an aluminous–siliceous one.

Calcareous body (TBL 17)   Brongniart says little about *terre de pipe*, the bulk of which would mostly be composed of plastic clay extracted in the quarries on the right bank of the Rhine, around Köln and Koblenz, with added calcined flint, chalk or alkaline frit (Brongniart 1877, 112). SEM observations and chemical composition tend to agree with Brongniart. The use of Rhine clays rich in Al_2O_3 (Schüller 1974) results in a high content of this oxide in both pieces analysed. The angular edge of the quartz crystals, i.e., the calcined flint pebbles, is evidence that these grains were added to the paste after grinding. The addition of glass fragments was also observed (Fig. 4). It is not an alkaline frit, as Brongniart wrote, but a lead-bearing frit. The presence of such glassy fragments explains the high lead content of sample TBL 17. One can ask why these fragments, as shown by the SEM pictures, survived the firing as separate particles and hadn't seemingly much effect as a flux. But their overall lead content (Table 4) is, as evidenced by relevant phase diagrams, too low for melting at the inferred figurine's firing temperature of 900–1050°C (see below). On the other hand, the lead of the matrix could derive from originally very tiny glass particles, free of SiO_2 polymorphs and containing up to 45 wt% PbO, as measured in corresponding wholly glassy areas of the coarser, unmelted particles. These compositions would be completely molten at the above-mentioned firing temperatures. Such an addition and absence of any reaction with the matrix was also noted in the pipe clay products of the Bois d'Épense manufacture (Maggetti 2007a). If the porosity is excluded from the modal analysis, all the lead attributed to the glass and the CaO from the bulk analysis either transformed into Ca-carbonate (calcite $CaCO_3$) or Ca-hydroxide (portlandite $Ca(OH)_2$), two recipes can be calculated for TBL 17 (Table 5). Kalifeldspar was not included in the calculations, because this phase occurs only in insignificant amounts. Furthermore, the composition of the clay can be inferred by subtracting the 18.4 wt% of the quartz from the total SiO_2 of the bulk analysis and eliminating all the CaO from the bulk analysis (Table 3). This composition is perfectly compatible with the Rhine clays (Schüller 1974). Once again, we can only assume that Cyfflé used this recipe if we are certain that TBL 17 was really made in his workshop.

Aluminous–siliceous bodies (TBL 24, 26–29, 34)   From the modal analysis of sample TBL 28, one can calculate the proportions of the different ingredients used in the fine-grained aluminous–siliceous *Terre de Lorraine* (Table 5). It seems quite plausible that Cyfflé used calcined and ground flint pebbles as for the calcareous figurine, which were mixed with ground A1 porcelain and B glass, together with coarse kaolinite and with a kaolinitic clay used as a matrix bonding agent. There is a possibiliy that Cyfflé had already fritted coarse kaolinite in order to diminish as much as possible the shrinkage of the unfired figurine during the drying and firing steps.

The recipe for the coarse-grained body TBL 34 (Table 5) is different from the fine body because of: the absence of B-type glass; the small proportion of calcined flint (= quartz); the high value of amorphous SiO_2; the different chemical composition of the A2 porcelain fragments, of the C2 meta-kaolinite and of the matricial clay if compared to the equivalent phases of the fine-grained body. Therefore, to make this object, Cyfflé used other ingredients and a smaller proportion of coarse kaolin. To sum up, both recipes of siliceous *Terre de Lorraine* recall the manufacturing processes of two different ceramics: on the one hand, because of the use of kaolin, hard paste porcelain, and on the other hand, through the introduction of glass, French soft paste porcelain.

Firing conditions of the figurines

Calcareous body   The microstructure of sample TBL 17 can be classified as 'extensive vitrification', characterized by a network of glass filaments and spongy pores, which develop at firing

temperatures of about 1000°C (Maniatis and Tite 1981; Kilikoglou 1994; Wolf 2002). The ancient firing temperatures of TBL 17 and TBL 25 can also be estimated by comparison of their phase associations (a) and (b) with the phase evolution in experimentally fired raw materials with a similar calcareous composition (Maggetti 1982). These inferred temperatures were most probably in the range 900–1050°C, matching the BSE-derived conclusions. Such temperatures fall in the usual range of the French faience kilns of the 18th century (Rosen 1995; Maggetti 2007b).

Aluminous–siliceous bodies   Scanning electron micrographs of the matrices of both bodies show platy clay minerals with no or little signs of vitrification, that is, a partial fusion along the grain boundaries of the clay platelets. This suggests firing temperatures somewhere in the interval between 550°C (decomposition of the matrix's kaolinite) and 800°C (in analogy to calcareous clays, where vitrification starts only above this temperature; Maniatis and Tite 1981; Kilikoglou 1994; Wolf 2002).

   The contours of the fragments A1 and A2 are roundish and show very small reaction rims with the matrix. This indicates firing temperatures just exceeding the 985°C of the ternary eutectic point (for A1) or the 810°C of the ternary eutectic point (for part of the A2), as the originally angular fragments will begin to melt at these temperatures. The firing temperatures of both figurines were therefore below or close to 1000°C, which are the normal temperatures in a faience kiln. X-ray diffractometry phase association (c) is characterized by crystals of high temperature such as cristobalite and mullite. As shown by experimental firings of refractory, Fe-poor clays (Maggetti and Rossmanith 1981; Maggetti 1982), mullite forms at temperatures over 950°C and cristobalite at temperatures higher than 1050°C. Considering the heterogeneous nature of the body, these values are obviously not related to the firing of the figurine, but to that of the vitreous phases that compose it.

<div align="center">CONCLUSIONS</div>

Out of four figurines which have been examined, two were made in the local tradition with a *terre de pipe* body. This body is a mixture of different proportions of calcined flint with a refractory clay, ground lead-bearing glass and with calcium hydroxide or calcium carbonate. They were fired at temperatures of 950–1050°C, which are the normal temperatures of a faience kiln. As these two figurines have no mark, one can wonder whether they were really made in Cyfflé's workshop. But, as we have seen previously, this does not go against historical evidence. The chemical composition of the other two pieces is indeed very different. It is not a French soft paste frit porcelain body nor an English type bone China, but a hard paste porcelain of the Meissen type. It is not a classical homogeneous hard paste porcelain, but a heterogeneous, porous body. The texture analysis reveals a complex paste composed of different elements, diverging from one figurine to the other. For the fine-grained piece, Cyfflé used 17.5 vol.% calcined and milled flint, + 13.5 ground porcelain + 14.1 ground Na–Ca-glass + 44.2 coarse kaolinite, all mixed up with 10.7 of a fine kaolinitic clay. The recipe used for the coarser-grained piece is as follows: 23.0 vol.% calcined and milled amorphous SiO_2 with some flint + 6.5 ground porcelain + 13.2 coarse kaolinite, all mixed up with 57.3 of a fine kaolinitic clay. The ground ingredients, i.e., the porcelain and the Na–Ca-glass, were synthesized at a temperature of around 1200°C. There is also the possibility that the coarse kaolinites underwent a thermal treatment prior to being added to the kaolinitic clay. Both *Terre de Lorraine* bodies were then fired at a temperature lower than 1000°C, resulting in porous bodies (9–25% porosity). On the one hand, because he used vitreous temper, Cyfflé's particular technique recalls the one applied in the making of French soft paste

porcelain; on the other hand, the introduction of quartz, kaolinites and K-feldspar is very similar to the recipe of hard paste porcelain of the Meissen type. His clever choice of this hybrid technique may very well have been Cyfflé's answer to the French king's prohibition of the making of porcelain, in which case one better understands the sibylline message in his sentence ' . . . without actually being porcelain, would be more beautiful than *terre de pipe*'.

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