The copper mines of Cabrières (France) and results of the experimental metallurgy

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Abstract: Since the works of E. Sangmeister (Junghans and al., 1960 to 1974), the spectrographic analyses by X-rays of Chalcolithic metal artefacts have shown the frequency, in the Southern part of France, of copper ores containing antimony and silver. The discovery of the prehistoric mines of Cabrières (Ambert and al., 1984) has provided the proof of this hypothesis by means of:

- the analysis of copper ores (carbonates, tetrahedrites) discovered in some mines (Pioch Farrus I and IV) dated to the Chalcolithic;
- the analysis of ores, globules and slags recovered from the centre of the ditches of Roquefenestre, and now by those of Pioch-Farrus 448, dated to 4310 ± 75 B.P., the more old site of Cabrières prehistoric exploitation (Espérou et al., 1994).

First metallurgical experiments carried out at Archéodrome de Beaune (Happ et al., 1994) with ores from Cabrières were made on two samples of ores (malachite from the mine La Vierge, polymetallic copper from Pioch-Farrus 448) discovered within prehistoric sites. A third sample (malachite from Pioch-Farrus 4) was taken from the richest vein (showing the presence of native deposit copper) discovered now in Cabrières. The metallurgical experiments using a knapping strategy are detailed here, and allowed us to obtain in all cases slags and beads of copper. The spectrographic analyses of these are identical to those chalcolithic yields from Cabrières. Achieved from ores collected in the prehistoric deposits, the comparative analysis of the products of the Chalcolithic metallurgy and of those from archaeometallurgy has enabled us to show that, 5000 years ago at Cabrières, the prehistoric peoples were using a veritable Copper - Antimony - Silver alloy, a bronze where the tin had been replaced by Antimony and Silver, and this more than five centuries (perhaps ten) before the appearance, in the South of France, of Bronze -sensu stricto-. That is an essential piece of knowledge, and until more copious information is obtained, Cabrières, would appear for this reason as an important landmark towards the West of the technological transfer which inaugurated the Atlantic Bronze Age.

I. PRESENT STATE OF KNOWLEDGE

This has been noted in several articles recently published (Ambert, 1990; Ambert, 1992; Ambert and Barge, 1991; Espérou, 1992; Espérou et al., 1992; Happ et al., 1994) or in course of publication (Ambert, in preparation, Espérou et al., 1994). These works have established precisely the importance, both quantitative and qualitative, of an exploitation (shown in figure 13) that is at present the most ancient in France.

1.1. Age of the first exploitations

The ditch of Pioch Farrus 448 (Espérou et al., 1994) has provided the oldest Carbon 14 date from the Cabrières district (Lv : 2039 - 4310 ± 75 BP), that is to say, from about the beginning of the 3rd Millenium B.C. A deposit of coarse pottery is associated with fragments of crucibles and of mining and metallurgical products. This date is about contemporary with those (4470 ± 150 BP to 4020 ± 100 BP) obtained from the nearby site of Roquemengarde, a Late Neolithic site which has provided what is at present the most ancient metallic evidence found in Languedoc (Guilaine, 1992). The spectrographic analysis of several of these objects shows a composition compatible

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with that of the ores from Cabrières (Guilaine, oral communication). In consequence, there exists a good probability that the metallurgy of Cabrières started from the absolute beginning of the 3rd Millenium B.C, forming the first phase of the regional Chalcolithic.

On the other hand, the Carbon 14 dates obtained from the site of Roque Fenestre - 3900 ± 130 BP (Ly-2689), 4010 ± 70 BP (Ly-1724), and 3870 ± 80 BP (Ly-1723) - testify to a second phase, to a more established metallurgy (Late Chalcolithic).

The Carbon 14 dates obtained from the mining spoil from Neuf Bouches in the Vallarade combine to confirm the age of this exploitation. Two Carbon 14 dates have been achieved at the Laboratory of Gif-sur-Yvette, from charcoal derived from the Context C2 - almost on the surface - and C9 - by contrast situated in the lower sequence of the spoil. These have given, respectively, the ages of : - Gif. 9571 : 4120 ± 40 BP (2872, 2506, Cal. B.C.) for the C2 context ; and Gif. 9570 : 2870 ± 50 BP (1199, 905 Cal. B.C.). The explanation for this chronostratigraphic inversion can be found without too much difficulty in the theory of secondary removal of Chalcolithic spoil in the course of an exploitation during the transition from the Bronze Age to the Iron Age.

1.2. Copper ores from Cabrières

The numerous analyses of ore show that the principal ore from Cabrières used by the Chalcolithic workers (at Roque Fenestre and Pioch-Farrus) were an argentiferous Tetrahedrite, - 'Grey Copper', of composition (Cu ± Ag, Zn, Fe)\textsubscript{12} S\textsubscript{2} \textsubscript{14} S\textsubscript{3} often associated with Chalcopyrite - Cu Fe S\textsubscript{2}. At Pioch-Farrus the numerous analyses of ore (Malachite, Grey Copper) make it possible to define the classical ore of Cabrières : Copper with a notably high proportion of Antimony (10 to 20 %) and of Silver (2 %), with proportions of Arsenic and Zinc less constant and weaker. This ore is above all characterised by the almost complete absence of Lead (often less than 0.005 %). The successive metallurgical treatments, recognised at Roque Fenestre (Ambert, 1990 ; Espérou, 1993), or achieved experimentally at the Archéodrome at Beaune (Happ et al, 1994), which allow the recovery of metal, eliminate the Zinc, diminish to an appreciable degree the proportions of Arsenic and Antimony, and tend to concentrate the Silver. Taking account of the initially high proportions of Antimony (up to 20 %) in the ore (presence of Pyrargite - Ag Sb S\textsubscript{3} - at Pioch-Farrus I), at the end of the cycle the metal produced from the ore of Cabrières could be defined as a copper with a low lead content and high Antimony-Silver (the presence of Arsenic being more unreliable). This is the copper of the Types E10, E11B, and above all E11A of the classification of Sangmeister (Junghans et al, 1960, 1968, 1974) widely represented in the Midi of France (Espérou, 1992).

The analyses of the ores and metallurgical products of Pioch Farrus 448 have disclosed some exceptions to this observation (Espérou et al, 1994), one part only of the slags and globules of metal from Pioch Farrus 448 corresponding to this definition. A fact unique at Cabrières, - several of the products from the Chalcolithic metallurgy, show proportions (considered as) abnormally high in Lead (between 0.3 and 0.6 %). This fact which is still incompletely explained, does not seem to be capable of resulting from a hypothetical importation (more difficult to conceive in this epoch). In fact, some ores of Chalcopyrite type of which we do not have the evidence of a Chalcolithic use, have a composition similar to these slags and have been discovered at Cabrières. Moreover, all the ores of Cabrières, in particular the Malachites, do not possess the
Antimony-Silver characteristic. In this case, they are the ores which are practically pure (cf. the mine of La Vierge, fig. 11 and 12).

1.3. The origin of the metallurgy of Cabrières

The discovery of a Palmella Point, near to the mines of Cabrières (Ambert et al., 1986) had led us to consider a hypothetical link between Portugal, the principal supplier of these points, and the development of the exploitation at Cabrières. The analytical spectrum of this object, which does not relate clearly to the composition of the ores of Cabrières, supported the hypothesis. The high technical level reached even in the epoch of the Roque Fenestre ditches (Ambert, 1990) undoubtedly suggests diffusionism, and necessitates research into the origin of this metallurgy. Henceforth the dates of Pioch Farrus 448, those of Roquemengarde, and the late position of the Palmella points in the Beaker culture, make - on the sole evidence of this Point - the hypothesis of a link with Portugal insecure.

The origin of the Languedoc metallurgy, that of Cabrières in particular, therefore still remains to be established precisely, although the relations with the rich metal-using culture of Remedello, and with Central Europe via Switzerland, indicate an Eastern origin.

1.4. Principal products of the ditches of Roque Fenestre

These ditches, integrated within the perimeter of the areas of metallurgical work, could have been - at a certain stage in their filling - used as basins; at other stages they served as receptacles for the peripheral activities.

The excavations (Espérou, 1993) have revealed four juxtaposed ditches, hollowed out of the schist, containing an original archaeological material (fig. 14). The list of the principal tools used include some pounders, smaller than those found on the edges of the mines, some grindstones, a shovel made from the scapula of an ovid, and most of all some cup-marked stones of which the two principal forms are represented (slabs, generally made of Basalt, which mostly possess one cup mark on each flat face; and 3-sided forms in hard rock (quartz) with double cup-marks) - tools which have never been encountered in the mines. We should also mention a 'pick' (?) with two points, some grindstones, and a fairly abundant supply of pottery.

The ditches of Roque Fenestre show several stages in the transformation into metal of the local copper ore:

a. the placing in water, at least temporarily, of basins used as washing areas, is certain. It is shown in particular by the bedding of sediments in the ditches, deposited there following a vertical gravitational gradient from the heavier to the lighter, sands, haematiferous muds, and then beds of charcoal.

b. the mass of debris derived from the workshop activity peripheral to the ditches contributes very largely to their filling. To the items mentioned previously should be added blocks of Dolomite and Quartz of about 20 cms. length which derive without doubt from the discarding of sterile rock, following a last sorting of the ore carried out nearby. In fact, the mineralised vein quartz was made the object of a careful crushing and a granulometric sorting in the presence of water. Several inclined beds, formed from gravel of a homogeneous size (2 to 3 cms.) which lean against the edge of Ditch n° 2, bear witness to that.

c. This sorting, previous to the roasting or reduction in the furnaces, must have had the object of isolating the most mineralised parts. The discovery of an indurated wall of quartz sands,
coloured green, very finely crushed, and heated, provides final evidence for the existence of reduction furnaces, situated on the top of the fill or near to the ditches, suggested equally by the slags and the metallic globules. The ditches of Roque Fenestre present an example unique in France, of the principal technical stages in the transformation from copper ore to metal. The spectrographic analyses of these products relate without exception to the classical spectrum of the ores of Cabrières.

1.5. The techniques used in the mining extraction

Our knowledge rests on the direct study of the mines (see below) and on that of the objects (pounders), - hammers of varied size and weight (from 250 to 14,500 gr. at the extreme limits) taken from the siliceous spoil of the exploited vein- (Espérou et al, 1992), from the Dolomites of the packing, but also from the veins of Quartz left in relief by erosion (Roussignol, Roques Blanches). This retrieval from the silicified Dolomite, splintered by impact, producing blocks that are more or less cubical or speroidal, elevated to the rank of hammers in replacement for an out-of-use pounder, is as practical as it is functional. The material being used up at the same time as the ore, this technique is more especially rational. it permits an increase in the removal of the sterile part of the rock which otherwise would rapidly obstruct these small excavations. On Vallarade, where in our present state of knowledge dykes of Quartz do not appear, the very numerous quartz pounders which bestrew the surface of the ground (see below) could have been collected, even prepared, in the less mineralised veins of Le Broum and Roques Blanches.

A typological study devoted to the pounders (Espérou et al, 1992) has allowed one to conclude that these tools - generally having a double or multiple working surface - may be grouped principally in two forms-functions. The sub-speroidal pounders (fig. 14, n°1) represent 43% of the objects, and the «wedges», much less numerous, bearing a cutting edge or a point. There exist some intermediary objects, of a pick type, and also some objects of small size, perhaps finishing tools. It is not easy to deduce exactly the techniques of rock extraction used in the prehistoric exploitations. In particular the heating of the mineralised walls with fire, designed to loosened the rock prior to battering with a pounder (related to the Mitterberg, Tylecote, 1987) leaves few traces, and its use, although locally likely, is not proven. The speroidal niches in the smooth walls, well exemplified at the front of the quarry at Pioch Farrus IV, allows one to suggest a recourse to pyrotechnic excavation.

II. ATTEMPT AT QUANTIFICATION OF THE CHALCOHITIC MINING / METALLURGICAL EXPLOITATION

The enumeration of sites recognised to date shows the density of the Chalcolithic exploitation of Cabrières. One can count as a minimum: - 5 or 6 mine entrances, one potential treatment area at La Roussignole where 265 pounders have been counted (Espérou et al, 1992).

The Pioch Farrus sector contains about 10 mine sites, with the exceptional sites of Pioch Farrus I and IV (Fig. 11) and the mine of La Vierge. The typological reckoning has enumerated 185 pounders there (Espérou et al, 1992). The sectors producing cup-marked stones, treatment areas, or habitations, are particularly well-represented. In fact, besides the 4 ditches of Roque Fenestre (Ambert et al, 1984, Espérou, 1993), more than half a dozen points have produced either cup-marked stones, such as Pioch Farrus 438, (Ambert, 1990) or Chalcolithic remains with-mining and metallurgical spoil (Espérou et al, 1994). The sector of Vallarade has produced very numerous pounders (369 have been counted there), and more than about fifteen mining sites, both Chalcolithic and proto-historic.

The preceding remarks underline the fact that the cup-marked stones (Fig. 14, n° 2, 4, 6), like the pounders are representative of the prehistoric metallurgical exploitation. Indeed, the cup-marked stones are clearly connected with metallurgy. In fact, the majority of sites which augment the list may produce traces of early metallurgy (cf. the region of Villefranche de Rouergue, unpublished oral communication, L. Carrozza). A recent reassessment (Ambert, 1992) associates the sites of Ouveillan and Roc d'en Gabi (Carcassonne) in a Verzian context, with those of the district of Cabrières (Pitrous, Les Figuières, Pioch Farrus 438, and the more peripheral site of Tribes à Bassan). The majority of the sites listed above in the district of Cabrières under the heading of metallurgical areas (about ten in all) have produced one or several examples, while in the ditches of Roque Fenestre (Espérou, 1993) twenty-eight of these objects have been found.

III. RESULTS OF THE EXPERIMENTAL METALLURGY

The first results have been recently presented (Happ et al, 1994). These researches, carried out
at the Archéodrome at Beaune, on the ores discovered in the prehistoric sites - ores which have been smelted following the techniques and with the equipment of which the archaeological excavations in Chalcolithic sites have revealed convincing traces. The ores used, and then, after smelting, the slags and the globules of copper obtained - have been subjected to spectrographic emission analysis at the laboratory of Rennes in order that they may be compared with the copper ores collected in the district of Cabrières and with the slags and globules brought to light at Roque Fenestre.

III.1. Chain of operations used in the experimental metallurgy

The metallurgy used for the reduction of the copper ores differs according to their nature. The processes of reduction of the Carbonates, such as Malachite (Cu₂(OH₂)₂CO₃) transforms the oxide of Black Copper (Cu O) and Red Copper (Cu O) relatively easily into metallic copper. The treatment of the Copper Sulphides (Grey Copper, Chalcopyrite, Bornite, etc.) is more complex. It demands a series of operations, successive or joined together, in order to separate the parts which are rich in copper from the other elements and impurities (S, Fe, Sb, As and elements) contained in the ore.

The experimental installations used for the results achieved are derived from the synthesis made between our present knowledge of metallurgy and the archaeological evidence. It consists of a metallurgy of copper by the dry method. At this level, the complexity presented by the melting of the Sulphides increases considerably, while at the same time our knowledge remains small and the archaeological indications insignificant. For the Sulphide ores it is preferable to practice roasting (Happ et al, 1994), previous to the slag-producing smelting developed after this. The minimum treatment needed for success:

a. Concentration

The raw ore according to its richness in copper is concentrated according to different methods:

- Sorting by eye, before or during the crushing.
- Decanting into ditches.

The product thus obtained, partly separated from its gangue, will have a mesh size between 0.5 and 1 cm maximum. The powders will be gathered together in the form of pellets (Happ et al, 1994).

b. Smelting producing a slag

General principles

The reaction of the Malachite is very simple, its aim is the reduction of the Carbonates and the elimination of the CO₂ and the H₂O under the effect of the temperature (500 to 600°C). There remains the Cu₂O which reacts in the flame with the CO (Cu₂O + CO → 2 Cu + CO₂ which is released). In the fire only the CO produces the reaction and not the Carbon. The carbon by itself with the ore would involve much too slow a reaction. The distribution of the temperature and the gases in the reducing furnace is an absolutely vital factor in the success of the experiment.

The separation of the copper from the slag is achieved by the difference in density. The nature of the smelting is linked to the composition of the ore. It is always antagonistic to its gangue. The metallurgist must essentially obtain a «good» slag in order to liberate the maximum amount of copper. However in the majority of the experiments certain nodules of copper will remain imprisoned in the slag. They will later be recovered by crushing and sieving.

Experimental fulfilment at the Archéodrome

The prototype of the furnace used for this experimentation is in every way similar to the smelting furnaces already known from the Bronze Age, at Mitterberg or in Northern Italy. It is built out of stones packed together with clay in its upper part, the structure being rough-cast with clay to ensure a minimum of heat loss. The bottom is dug into the soil. The tuyère emerges at this level. All the dimensions, diameters, heights, depths, and inclination of the tuyère are empirical, but related nevertheless to observation of the furnaces of Mitterberg and the Alpine domain.

The conduct of the furnace is essentially a delicate operation. The furnace must at first be pre-heated with flaming wood. Then the ore is introduced in successive layers, alternating with charcoal. The size of the charcoal is of extreme importance. It must be as near as possible to that of the ore; this is to ensure the progressive and concomitant descent towards the base of the furnace. A too rapid descent of the ore will tend to diminish the output of the operation. In a case where the slag is not totally purged of its copper, it will be possible to pass this product a second time through the furnace in the course of a second phase of reduction. This repetition of the operation will be the rule for the ores which are more difficult to reduce.

The temperature at the heart of the furnace above the tuyère is from 1200°C. In front of the tuyère the maximum point measured is from 1350°C. A chromatic scale of colours is nonetheless sufficient for appreciating these temperatures for a trained metallurgist...

This technique, used for the Carbonates,
proves powerless to eliminate the Sulphur contained in the Tetrahedrite of Pioch Farrus 448 (Cu, Fe) 12 Sb₄ S₁₃. Its presence makes the operation impossible, hence the need for roasting (that is to say an at least partial oxidation) the only treatment capable of reducing the Sulphur. Therefore, before the reduction, it is necessary to oxidise the ore, thus schematically achieving the operation Cu₃S + O₂ → (Cu, Cu₂O, CuO) + SO₂ (which is released).

After recoiling of the furnace some metallic nodules are formed, of which certain ones with a brilliant grey metallic aspect are very (too much) rich in Antimony. At the end of the reductions the globules of copper obtained have been resmelting in a crucible. The technique used is that brought to fruition by W. Fasnacht. A shallow ditch is dug in the soil, filled with charcoal, and this is brought to incandescence with an angled tuyère and two hand-bellows. A very porous crucible containing the globules of the first reduction has been placed at the top of the combustible matter since the start of the operation and has registered the rise of temperature in the same rhythm. The whole mass is then covered again with charcoal brought to incandescence in their turn, always with the aid of the angled tuyère, of which the snout is placed vertically above the centre of the crucible. This technique, known in Central Europe since the Bronze Age at least, has the advantage of punctually concentrating the heat on the ore or the globules to be smelted, and diminishing the risk of shattering the crucible.

In the conditions of the experiment, a heating time of a quarter of an hour has been enough to produce a little ovoid ingot of which we give the analysis.

III.2. The results of the experimental metallurgy

A very interesting result : the spectrographic analyses of these productions are identical to those from the Chalcolithic productions of Cabrières, demonstrating in particular the use of Tetrahedrite by the Chalcolithic workers.

The pursuit of the experiments aims to ensure the control of the proportions of Antimony and Silver between the ore and the metal. In fact, during our first experiments, the use of Tetrahedrite from Cabrières - (Cu, Fe) 12 Sb₄ S₁₃ complicated by the presence of Pyrrargite - Ag Sb S₃ - has given us some brittle coppers with proportions of Antimony near to 20 %. The composition of the Chalcolithic globules, on the other hand, formed of copper with 6 - 7 % of Antimony/Silver, show that this transfer, conveniently controlled, allowed the production of a veritable Copper-Antimony-Silver alloy which melted and flowed much better than would have occurred with a very pure copper. It is this that the experimental production of the copper ingot has allowed us to verify. All the secondary impurities, and in particular the proportions of Arsenic (which could reach up to 5 %) have been oxidised as well as a high proportion of the Antimony contained in the globules. At this stage the ingot is characterised by a binary association - Antimony (3.5 %) - Silver (1.35 %) - which, associated with the low proportion of Lead (0.02) allows us to identify the metallurgical productions of Cabrières. This copper, used since the beginning of the 3rd Millenium (PF 448 ; LV 4310 ± 75 BP) presents some essential, remarkable characteristics very likely put to profit by the chalcolithic workers. We underline that a metal such as this possesses the technological qualities of a Bronze. This is a Bronze where the Tin would have been replaced by the Antimony and the Silver, and that more than five centuries (perhaps ten) before the appearance in the South of France, of Bronze "sensu stricto". One understands better, now, the interest of the copper ore of Cabrières for our remote ancestors.

IV. CONCLUSION

This text presents the principal results obtained in the most ancient mining and metallurgical sector of France. Beyond that, it has seemed necessary to us to return to the results of the programme of experimental metallurgy rapidly described here. Achieved from ores collected in the prehistoric deposits, the comparative analysis of the products of the Chalcolithic metallurgy and of those from archaeometallurgy has enabled us to show that, 5000 years ago at Cabrières, the prehistoric peoples were using a veritable Cooper - Antimony - Silver alloy, a bronze where the tin had been replaced by Antimony and Silver, and this more than five centuries (perhaps ten) before the appearance, in the South of France, of Bronze "sensu stricto".

That is an essential piece of knowledge, and until more copious information is obtained, Cabrières, would appear for this reason as an important landmark towards the West of the technological transfer which inaugurated the Atlantic Bronze Age.

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