Contribution of the analytical work to the knowledge of the early metallurgy in the Iberian Peninsula

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Abstract

In this chapter, the state of the art on prehistoric metallurgy of the Iberian Peninsula is revised in the light of the results obtained by the Spanish Archaeometallurgy Project, with special reference to the method of copper obtaining using smelting crucibles and its related slags. Alternatives to the contentious issue of arsenical copper and solutions to the hitherto obscure problem of copper-tin bronze obtaining are also proposed. Early iron technology poses many problems still unresolved.

1. Introduction

It is a commonplace in the 20th century Spanish literature on Prehistory that the early metallurgical knowledge reached the Iberian Peninsula coming from the Eastern Mediterranean regions. The Chalcolithic and even the Argaric metallurgy was firstly explained as a consequence of the arrival of prospectors who should settle the Southeast territory where copper resources were abundant, giving rise to supposedly metallurgical societies such as those of Los Millares and El Argar cultures, through the third and second millennia cal BC. Louis Siret was the first who wrote on this topic in his very important book on early Spanish metallurgy (Siret, Siret 1890: 321). Over time, "the colonialist hypothesis" lost strength under the weight of new archaeological evidence but the notion *ex oriente lux* has strongly taken root in many scholars. In this sense, it is thought that Sicily and Sardinia, as islands in the route of the eastern prehistoric sailors, could have played an important role in the diffusion processes.

2. Archaeometallurgy in Spain

It is a fact that Spain enters very early in the archaeometallurgical world by the hand of Louis Siret, a Belgian mining engineer who worked at the large mining district of Almería province. He performed the excavation of dozens of archaeological sites in the region and the materials he published are today an essential source of information about the Millarian and Argaric cultures (Siret, Siret 1890). He also included many chemical analyses of metal objects, some ores and a few slags, as he was fully convinced of the importance of chemical analysis for understanding the archaeometallurgical problems.

After Siret, a long silence occurred until the well-known analytical program carried out in the sixty's by S. Junghans, E. Sangmeister and M. Schröder, the *Studien zu den Anfängen der Metallurgie*, which included more than one hundred analyses of prehistoric metals from the Iberian Peninsula. The definition of the metal group E 01 (arsenical copper), which also is present in Anatolia, Crete and Cyprus, was thought to be a strong argument in favour of the colonialist hypothesis.

A little latter, in the seventy's B. Rothenberg and A. Blanco Freijeiro developed the *Huelva Archaeometallurgical Survey* (Rothenberg, Blanco 1981). Despite not too much light on early metallurgy was obtained by this survey, it was, no doubt, interesting and fruitful for latter periods of copper and silver mining and metallurgy (Iron Age and Roman time).

At this point, a group of at that time young Spanish researchers (G. Delibes, M. Ruiz-Gálvez, C. Martín, M.D. Fernández-Posse, S. Rovira) leaded by M. Fernández-Miranda started in 1982 the *Arqueometalurgia de la Península Ibérica* project. Other

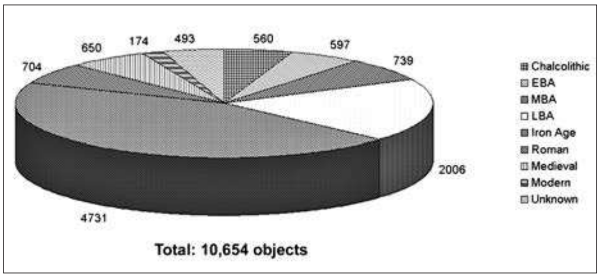

Fig. 1. - Analysis of metal objects performed by the "Arqueometalurgia de la Peninsula Ibérica" Project.

researcher joined few latter the initial group (I. Montero, J. Fernández Manzano). The project continues working up today. The figure 1 summarises the content of the database of elemental analysis of metal objects, by periods, reaching the amount of 10,654 all of them carried out by non-destructive XRF-ED spectrometry. To the before-mentioned number we must add 5,221 coins (mainly Punic, Iberian and Roman coins) and 1,350 ore samples (mainly copper ores) from many mines. More than one thousand metallographic studies complete the picture.

In 1996 we started the systematic analysis of slags, minerals and relevant metal samples using the scanning electron microscope (SEM). More than 300 samples have been analysed using SEM facilities.

Not all the results obtained by the project have yet been published, but those regarding the early metallurgical periods can be found in the books by Delibes and Fernández-Miranda (1988), Montero (1994), Rovira *et alii* (1997), Montero and Delibes (1999), Delibes *et alii* (1999) and Rovira and Gomez (2003), besides more than 100 articles.

Another research groups on archaeometallurgy have been created the last years in some universities (Cadiz, Madrid, Seville, Zaragoza, Valencia, Huelva and Barcelona). So, I expect that the knowledge on Spanish archaeometallurgy may be dramatically increased in future.

3. The inception of metallurgy in the Iberian Peninsula

"Colonialism and megalithismus", a well-know article by Renfrew (1967), was the first sound reaction against both the colonial hypothesis and the Aegean influence as statements to explain the Chalcolithic metallurgy in Iberia. This author, after reviewing the three basic arguments fenced by diffusion believers (fortified sites, copper metallurgy and collective burials into large monuments), concluded that the Iberian Chalcolithic developed locally, including the local invention of metallurgy, with a minimum of external influence. Short time latter, he proposed the date of 3500 BC as the starting point for metallurgy (Renfrew 1970: 308). However, available radiocarbon dates from "classical" sites of the Millarian culture (Full Chalcolithic) are not earlier than 3000 cal BC (Delibes, Montero 1997: 20).

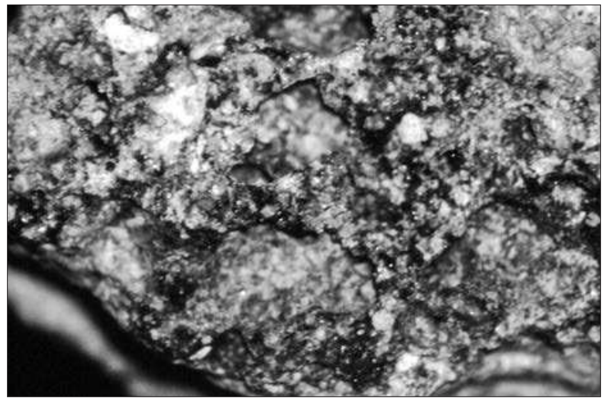

Fig. 2. - Cerro Virtud: crucible fragment; detail of the slagged surface, 20x.

The situation has changed since 1994, after the excavation by I. Montero of the Neolithic site of Cerro Virtud (Herrerias, Cuevas del Almanzora, Almeria). There, in an undisturbed stratum associated to the radiometric date of 5660±80 BP (Beta-90884), 4700-4350 cal BC (Ruiz Taboada, Montero 1999: 899), a crucible fragment was found (fig. 2). Despite the slagged ceramic show signs of severe leaching, XRF-ED analysis of a large area allowed to detect the presence of copper, antimony and lead (Ruiz Taboada, Montero 1999: 901). These elements are in a close connection with the copper ores from the Guadalupe mine at the foot of the hill where the site is located (Ruiz Taboada, Montero 1999: 901, table 3). New and more accurate analyses will be performed to the sherd in the immediate future.

Following these authors, the features of the slagged sherd lead to interpret it as a fragment of a crucible used for smelting copper ores in Neolithic time, as early as the first half of the 5th millennium cal BC.

A long hiatus of more than 1,500 years exists between the metallurgical find of Cerro Virtud and the well-established copper metallurgy at Almizaraque (Cuevas del Almanzora, Almería), a Chalcolithic site distant no more than 200 m from the Neolithic settlement. Obviously, metallurgy at Cerro Virtud cannot have been an isolated phenomenon and a review must be done of the metal objects recovered in earlier excavations of, for instance, collective megalithic burials associated with both Neolithic and Chalcolithic materials, which metal objects has been systematically ascribed to Chalcolithic owners. A clear case of forced situation may be mentioned in the cave La Cocina (Valencia): in a secure Neolithic stratigraphic context was found a copper awl (BERNABEU 1989: 136). The metal find was interpreted not as a proof of earlier metallurgy but as the persistence of Neolithic geometric microlithe industry through early Chalcolithic.

New evidences must be found before we may define more sharply the characteristics of the earliest metallurgy in Spain. Meanwhile, the hypothesis of the independent inception of copper metallurgy in the western Mediterranean is in the air as a possibility. The figure 3 shows the earliest dates of metallurgical finds in the Mediterranean.

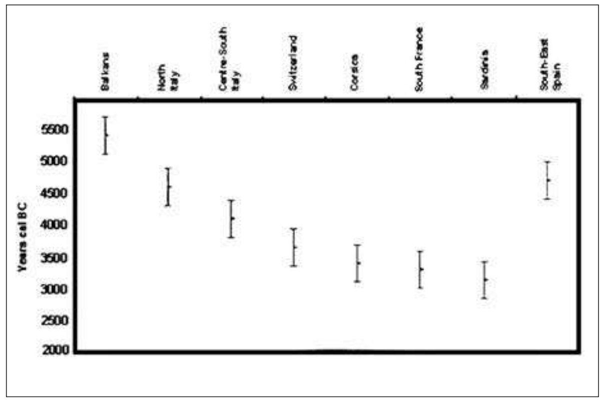

Fig. 3. - Chronological framework of the earliest metal finds at regions in or near the Mediterranean (after DELIBES, MONTERO 1997: 24, fig. 4).

4. Copper obtaining

The processes used by early smelters to get copper have been the central stress of our investigation during the last ten years, being focused on ores, slags and slagged material analysis. Some years before, in the eighty's, we could explain the absence of the remaining structures of smelting furnaces in Chalcolithic sites such as Almizaraque (Cuevas del Almanzora, Almeria) or Los Millares (Santa Fe de Mondujar, Almeria), which had given notable amounts of archaeometallurgical debris, by describing the smelting crucible technique. This subject will be treated latter with more details but can be said in advance (after the results of some smelting experiments recently done) that copper can be obtained very easily by this method, and only a common fireplace not much different than the one in the hut's kitchen is needed.

Due to its geological characteristics, the Iberian Peninsula beard huge mineral reserves in the past. Practically all the mountain ranges in Spain had abundant and easily accessible metal ores that Man worked when he had attained the necessary knowledge.

Copper minerals in the form of copper carbonates (malachite and azurite) and oxides (cuprite) were without doubt the most abundant outcrops in the past, as shown by present day remains. They have been synthesised in Rovira (2002a: 6-7).

4.1. *Ores worked by early metallurgists*

Many analyses of copper ores found at Chalcolithic sites have been published (ROVIRA *ET AL*. 1997). Those analyses were carried out using an XRF-ED spectrometer configured to measure elements with atomic number higher than 20. Then, information about gangue and real copper richness was not available in that way. In spite of this limitation, the aspect and density of minerals indicate they are low gangue oxidized ores, usually complex in composition, not only containing copper but also frequently iron, arsenic and other elements. X-ray diffraction analyses of some of these complex ores are shown in table 1.

The conclusion seems quite clear: the most analysed copper ores found in archaeological contexts or picked up from mines located near the sites are those of oxidized type. However, some recent analyses by SEM-EDX of new samples from Almizaraque and other Chalcolithic sites show some sulphur content. Isolated crystals of chalcopyrite and chalcocite have been identified by microscopical observation as remaining sulphides of the not completely weathered primary ore. Subsequently, smelting of fahl ores

Site	Mineral composition
Almizaraque (Cuevas del Almanzora, Almería) (1)	Malachite, azurite, cuprite (no gangue)
Almizaraque (Cuevas del Almanzora, Almería) (1)	Malachite, olivenite, chenevixite, alstonite?, quartz
Almizaraque (Cuevas del Almanzora, Almería) (1)	Azurite, malachite, olivenite, chenevixite, digenite?, alstonite?, quartz
Almizaraque (Cuevas del Almanzora, Almería) (1)	Olivenite (no gangue)
Cerro Minado mine (Huércal Overa, Almería) (2)	Azurite, dolomite
Sierra Cabrera 1 mine (Turre, Almería) (1)	Malachite, caolinite, mica moscovite, quartz
Cabezo de los Hilos mine (Mojácar, Almería) (1)	Malachite, mica moscovite, quartz
Los Pinares 4 mine (Los Gallardos, Almería) (1)	Malachite, azurite, hematite, mica moscovite, quartz
Los Pinares 4 mine (Los Gallardos, Almería) (1)	Malachite, azurite, hematite, mica moscov., feldspar
Loma de la Tejería mine (Albarracín, Teruel) (2)	Tenorite, cervantite, senarmonite, paxite, sandstone
La Ferrera mine (Rocabruna, Girona) (3)	Tenorite, paramelaconite, spertinite, hydroromarchite, valentinite, senarmonite, clinoclase, lammerite, discrasite
Can Manera mine (Manera, Girona) (3)	Cuprite, cassiterite, bronze (Cu6Sn%), enargite, eschafarzikite, hematite, magnetite
Amarguillo (Los Molares, Seville) (4)	Malachite (no gangue)
Amarguillo (Los Molares, Seville) (4)	Pseudomalachite, goethite
Amarguillo (Los Molares, Seville) (4)	Pseudomalachite, moscovite, quartz

Tab. 1. XRD analyses of copper ore samples from Chalcolithic sites and mines near Chalcolithic sites. (1) MONTERO 1994, (2) Montero, unpublished, (3) ALCALDE *ET AL*. 1998, (4) HUNT 2003.

played a short role (if any) in the Spanish early metallurgy (MÜLLER, REHREN, ROVIRA 1994), what is a distinctive feature that marks differences with other European regions as for instance the South East of France, where natural admixtures of sulphides and oxides were worked at Cabrières since the beginnings of the 3rd millennium cal BC (AMBERT 1999). Local mineralogy is determinative in all cases.

4.2. *Early slags of copper metallurgy*

No copper slag heaps are known in the Iberian Peninsula that could be dated before the Late Iron Age or Roman period. However, many Chalcolithic and Bronze Age dwelling sites dug in Spain use to provide evidences of metallurgical activities including few amounts of slags besides copper prills, ore pieces, crucible fragments and other by-products. Concerning Chalcolithic slags, full analytical results can be found in Sáez *et alii* (2003) and Rovira (2003).

Chalcolithic slags use to be found as small pieces, supposedly fragments of intentionally broken bigger pieces, of more or less vitrified, dense material of varied colours, from black to orange or reddish, showing green copper oxidation. The phase composition is very complex, usually far from the typical fayalite type slags. They use to entrap metal prills measuring from microns to more than one millimetre in diameter (fig. 5).

Not all the components in the slag have reacted (not in equilibrium system). Unreduced original ore and cuprite (in globular, dendritic and as prills structures) are frequently seen. Among the melted components, pyroxene, anorthite, melilite, akermanite, monticellite and other silicates use to be predominant. In iron-rich slags, plates of fayalite can occasionally be formed.

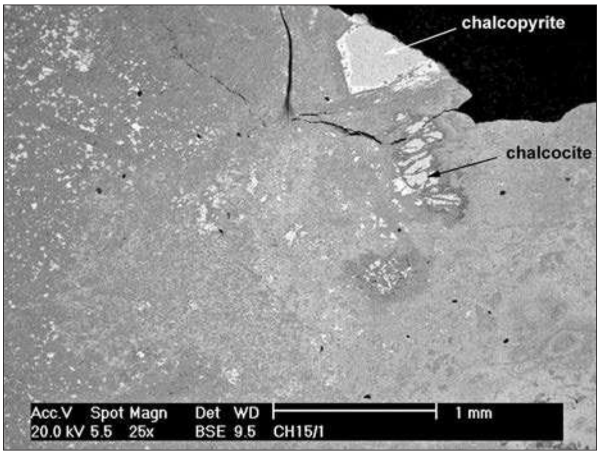

Fig. 4. - San Blas (Cheles, Badajoz): copper sulphide relicts in a malachite-cuprite matrix (SEM atomic number contrast image); copper ore from the Chalcolithic site.

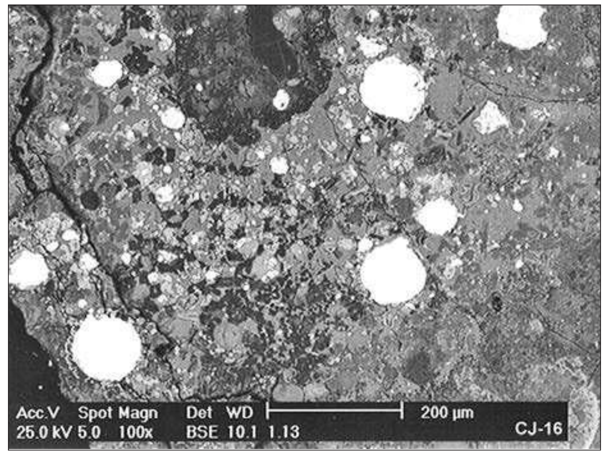

Fig. 5. - Cabezo Juré (Huelva): copper prills (white) entrapped in a slag from the Chalcolithic site (SEM atomic number contrast image).

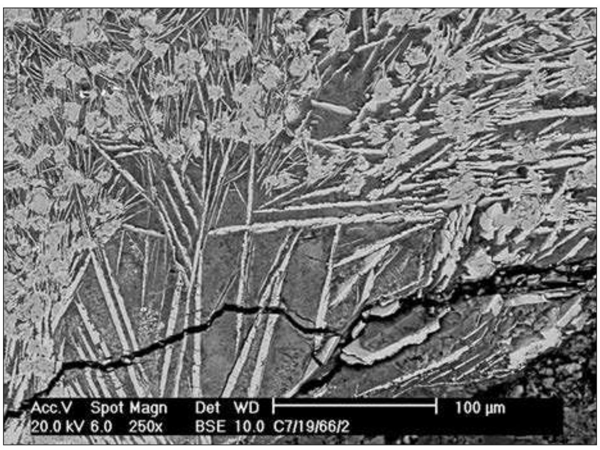

Fig. 6. - San Blas (Cheles, Badajoz): delafossite lathes in a copper slag from the Chalcolithic site (SEM atomic number contrast image).

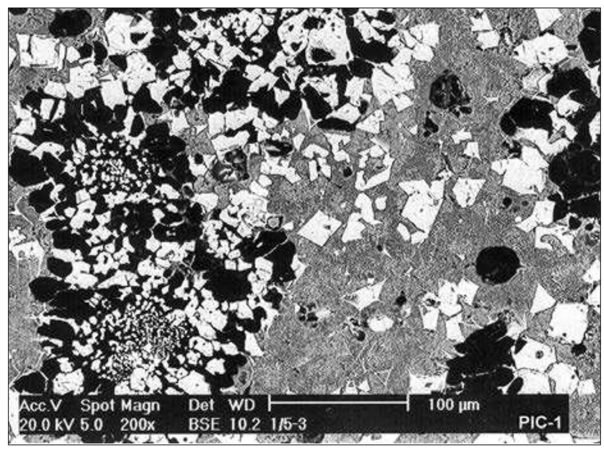

Fig. 7. - El Picacho (Carmona, Seville): Magnetite (white plates) in a slag from the Chalcolithic site (SEM atomic number contrast image).

Both microstructure and composition of these early slags suggest that direct reduction of the ore without fluxing was practised: all these compounds may be explained as self-reaction of the gangue in the smelted ore plus the contribution of charcoal ash, at high temperature.

Two compounds frequently found in the slags are of great interest for understanding the chemical environment the reactions took place. One is delafossite, a trivalent iron-copper oxide that is formed in oxidising atmosphere, at a temperature of more than 1.100º C (fig. 6). The other is magnetite or, more generically, trivalent iron oxide, formed by oxidation of divalent iron oxide (fig. 7).

These two compounds give a sound indication that smelting was performed in a fairly reducing-oxidising atmosphere, a condition that match quite well the crucible smelting process in an open fire.

This method for copper obtaining does not seem to change through the Early and Middle Bronze Ages, as shows the slag in figure 8. What occurred in the Late Bronze Age is not known, by the moment, as no slag analysis has been performed up to date.

4.3. *The crucible smelting process*

After many years of research, no furnace structure for copper smelting has been unearthed in either Chalcolithic or Full Bronze Age sites. What excavation depicts is the existence of fireplaces with abundant ash

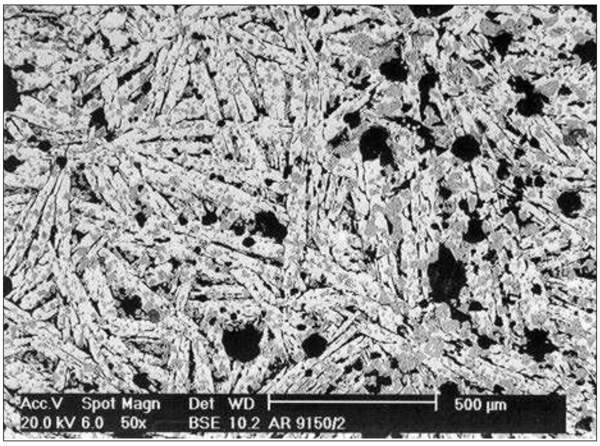

Fig. 8. - Cueva de Arangas (Cabrales, Asturias): large delafossite lathes (white) and magnetite (grey) in a slag from the Middle Bronze Age site (SEM atomic number contrast image).

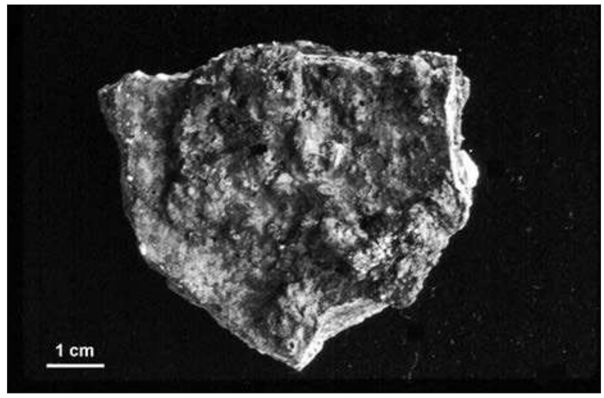

Fig. 9. - Almizaraque (Almería): slagged side of a smelting crucible fragment from the Chalcolithic site.

Fig. 10. - La Ceñuela (Murcia): reconstructed smelting crucible from the Chalcolithic site; maximum diameter about 45cm (Museo Arqueologico Nacional, Madrid).

layers including smelting debris. Particular kinds of debris are fragments of large clay trays and bowls, usually similar to cooking bowls or dishes, with their inner surface covered by slaggy and vitrified material (figg. 9-10).

After laboratory investigation they have been defined as remains of the containers used for copper smelting operations (Rovira 1989; Delibes *et al.* 1991; Craddock 1995; Rovira, Ambert 2002). The slaggy layer is formed by reaction among copper ore, gangue, ash and clay components of the crucible wall. Pyroxene, anorthite, akermanite, monticellite, melilite and other silicates are frequently identified in the thick layer of the vitrified surface, also containing copper compounds, delafossite, magnetite and macro and microscopical copper prills.

Smelting crucibles have been found in many other places outside the Iberian Peninsula (Zwicker *et al.* 1985; Hauptmann *et al.* 1996; Rovira, Ambert 2002), but what is singular and surprising is the long surviving of this technique in Spain, lasting up to the Iron Age.

After some experiments (Rovira 1999; Rovira, Gutierrez 2005) can be deduced that at the end of the smelting process within the reducing crucible some lumps and many prills of copper could be formed, together with or entrapped in a residual slaggy lumps also containing smaller prills, unreduced ore and slag. Then copper recovery should involve crushing the slag using stone hammers and, consequently, the residual amount of slag would be small and almost always as broken pieces (see Rovira, Ambert 2002 for a more precise description of the process).

The possibility of a two-step procedure for smelting has been recently proposed after the excavation of the Chalcolithic site of Cabezo Jure (Alosno, Huelva). More or less circular structures made of clay and small stones (fig. 11) measuring about 60 cm of interior diameter and about 20 cm high, containing porous, light slags, crucible fragments, ore pieces and some metal prills would be used in the first step for obtaining cuprite and a few of copper. These fire structures are built at the upper part of the hill were the settlement is located. The second step involving cuprite reduction and copper refining would be done into crucibles without any special fire structure, in the huts of the lower part of the site. The slags produced in the sec-

Fig. 11. - Cabezo Juré (Huelva): circular structure related to copper smelting (after Nocete 2001).

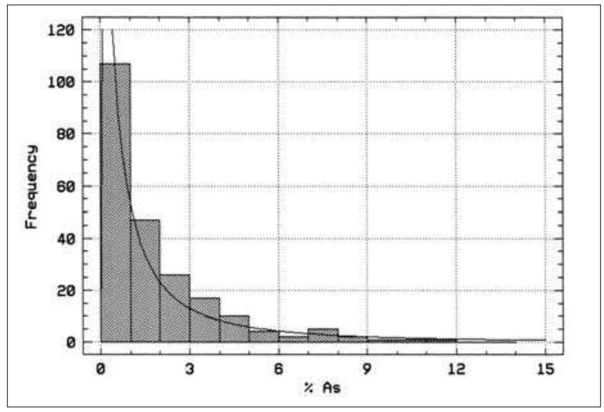

Fig. 12. - Arsenic in Chalcolithic copper objects from the SE of the Iberian Peninsula (analyses from Rovira, Montero, Consuergra 1997 and unpubished).

ond step are denser and slightly different in composition than the ones in the first step (Saez *et al.* 2003). An experiment made by Shalev *et alii* (2003) succeeded in obtaining about 200 g of molten copper in a crucible after direct smelting of cuprite recovered from a Chalcolithic site in Israel. No slag was produced. Hauptmann *et alii* (1996) suggested as well a two-step smelting process in the early Chalcolithic of Wadi Fidan (Jordan).

In conclusion, the scarceness of slags in the very early stages of copper metallurgy must not be surprising. It is a natural consequence both of the smelting of very pure oxidised copper ores, always available and easily distinguishable in the upper weathered zone of the copper outcrops, or were crushed for copper recovering and re-smelted.

5. The question of the arsenical copper

During many years has been thought that arsenical copper was an important step forward in alloying techniques to obtain better and better metals. So, the chain

copper → arsenical copper → tin bronze

has been considered the spinal column of the development of early metallurgy. Hundreds pages have been written on the topic and many scholars agree with this idea. However, we know no work based upon real archaeological evidences of smelting or melting practices that demonstrate how the alloying could be made in prehistoric times. The existence of arsenical copper objects as a real fact has lead to admit that the metallurgists would intentionally make them and many theories on arsenic alloying (including experimental way) have been built but no one, as far as we know, is wholly supported by the archaeological record. We believe that is a wrong way to investigate this subject.

What the evidences talk, at least for Spain is:

1. Copper and arsenical copper objects are found together, in the same archaeological levels, since the foundation moment, in all Chalcolithic sites having thick stratigraphic deposits such as Almizaraque (Almería) (Delibes *et al.* 1991) and Cabezo Jure (Huelva) (Bayona *et al.* 2003), both giving radiocarbon dates of the beginning of the 3rd millennium cal BC. The list of sites is longer.

2. Copper ores recovered in the sites use to be of polymetallic nature (tab. 1) (Rovira *et al.* 1997; Saez et al. 2003; Müller, Rehren, Rovira 1994).

3. There is no way to distinguish by colour differences the malachite also containing olivenite and/or conicalcite from pure malachite.

4. Slags, smelting crucible fragments and other debris indicate with no doubt that copper ores naturally bearing arsenic were smelted in situ (Rovira 2002b; Rovira, Ambert 2002; Saez et al. 2003; Müller, Rehren, Rovira 1994).

5. Arsenic content in Chalcolithic objects shows a distribution that match quite well the evolution of natural processes, having into account the arsenic lost in smelting and annealing operations, among other causes not so easy to evaluate (fig. 12). As it is well known, colour change and better properties of arseni-

cal copper start to be noticed when arsenic amount is higher than 3-4 %. If Chalcolithic metallurgists were concerned with intentional production, the histogram would reflect the effect as occurs with tin bronze (see below).

Therefore, in our opinion, these alloys were not deliberately made. Once again mineralogy is determinative. In fact we have detected regional differences in arsenic content related to the mineralogy and distance to the disposable resources, recycling, etc. (Rovira 1998).

Another matter is whether early metallurgist distinguished these arsenical bronzes or not. Probably the new colour was one of the more appreciated features in that time, after being detected by the current investigations that the improvement of mechanical properties not ever was properly applied, as demonstrate metallographic microstructure, arsenic content and function of the objects (Budd 1992; Rovira 1998; Rovira, Gomez 2003). But this is a matter that needs a long discussion in another scenario.

6. The first tin bronzes

Earliest tin bronzes of the Iberian Peninsula have been found in the site Bauma del Serrat del Pont (Tortella, Girona). It is a settlement inhabited for long time. An arrowhead and a metal waste made of low tin bronze were unearthed in Chalcolithic strata with Bell Beaker ceramics dated of 4200±70 BP (Beta-90622) and 4020±100 BP (Beta-64939), it is to say 2800-2450 cal BC. Evidences of tin bronze manufacture (smelting crucibles, metal waste, objects) are more numerous in this site in the following phase, Early Bronze (Alcalde *et al.* 1998).

As far as we know, tin bronze technology moved southward very slowly, reaching El Argar culture in the Southwest not early than 19th century cal BC. It is thought that this new technology arrived to Spain coming from outside, probably the South of France (Fernandez-Miranda, Montero, Rovira 1995).

An interesting aspect to detach, apart from the slow rhythm of territorial spreading, is that in no case tin bronze substituted copper technology. In Bauma del Serrat, for instance, copper and bronze still coexisted through the first half of the 2nd millennium cal BC. Moreover, taking into account the analyses of

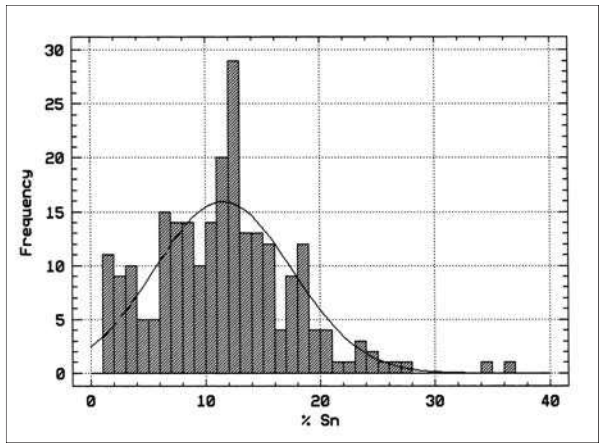

Fig. 13. - Distribution of tin percentages in the Middle Bronze Age objects of the Iberian Peninsula (data from Rovira, Montero, Consuergra 1997) and unpublished.

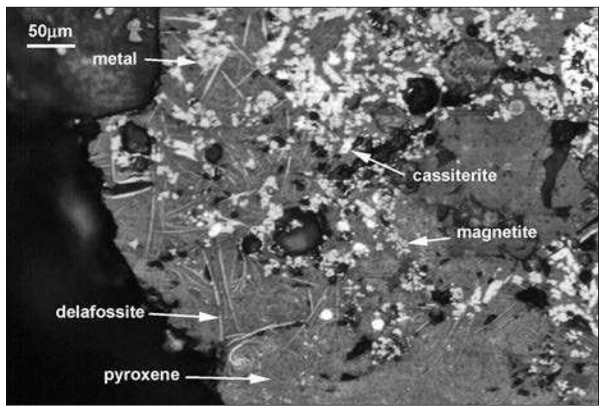

Fig. 14. - Monte Aguilar I (Bárdenas Reales, Navarra): optical microscope image of a bronze slag from the late EBA-early MBA site (not etched).

Middle Bronze Age objects performed by our project, about 80% of them were made of copper and 20% of tin bronze. It is striking that the most of Argaric objects made of bronze are ornamental ones (rings, bracelets, necklaces).

Tin percentages in the alloys are very irregular (fig. 13), what can be explained if we accept co-smelting of copper and tin minerals as the method used for alloying. Despite few slags from this period have been properly investigated, XRF-ED analyses of some slagged smelting crucibles show the presence of copper and tin compounds (Rovira, Montero, Consuergra 1997). These slags are quite similar to the Chalcolithic ones described before: a pyroxene matrix together with non-melted material, delafossite,

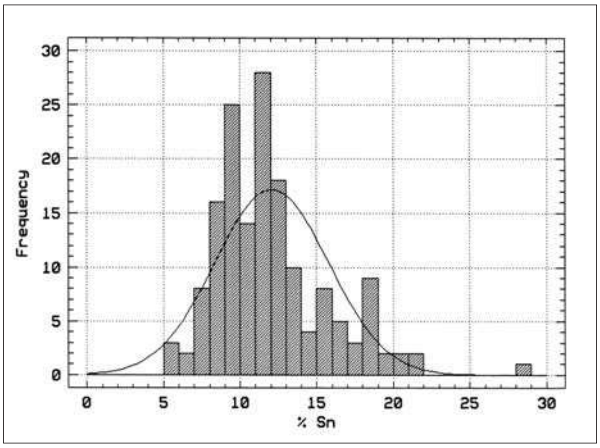

Fig. 15. - Tin content of swords and daggers Ría de Huelva type (data from ROVIRA 1995 and unpublished); compare with fig. 13.

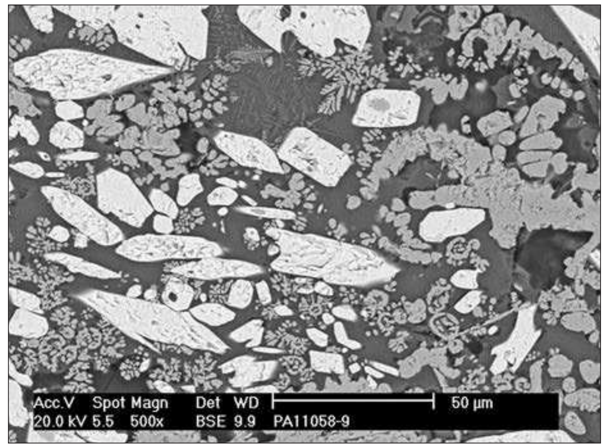

Fig. 16. - PUIG DE SANT ANDREU (GIRONA): copper-tin slag from the Iron Age site (SEM atomic number contrast image).

magnetite and metal prills. What is new is the presence of cassiterite both as globules and as rhombic crystals. The figure 14 shows an example.

Co-smelting to obtain copper-tin alloys has been verified by the experimental via by Rostoker and Dvorak (1991) among others. It is a process that makes very difficult to control the final proportion of components in the alloy because, on the one hand, the metallurgist did not know the exact composition of the minerals smelted together in the crucible. On the other hand, an unpredictable part of cassiterite is lost in the slag because it is not reduced to enter the alloy (fig. 14). Thermal and chemical environment in the smelting crucible, not easy to control, is of great importance. The presence of magnetite and rhombic cassiterite indicates a too much oxidising ambience, which negative effect on proper alloying has been reported by Dungworth (2000).

Tin content in bronzes seems to be more standardised in the Late Bronze Age. However, the range of compositions is still wide, as shows the histogram in figure 15 representing the alloys of Spanish swords and daggers of Ría de Huelva type. It is due, in our opinion, to the fact that co-smelting and/or copper cementation techniques continued in use as late as the whole Iron Age (ROVIRA 2004), as suggests a nodular slag from Puig de Sant Andreu (Ullastret, Girona) dated of the 5th century BC. As can be seen in the figure 16, dendritic cuprite (medium grey) coexists with globular and rhombic cassiterite (white), immersed in a matrix formed by complex silicates (dark grey). Metal prills having a wide composition range (from pure copper to more than 12% tin) are abundant in this slag, but no metallic tin prill has been found. There are more evidences from other Spanish sites in the database of our project.

Regarding the ternary alloy copper-tin-lead, it appears late in the Late Bronze Age, but heavily leaded bronzes are practically confined to the production of palstaves in Galicia and Portugal.

7. The first iron of the Iron Age

Some few iron objects have been reported in Late Bronze Age assemblages, being the two small bowl shaped pieces and the bracelet in the Villena treasure the most spectacular, circumstance that has lead to think that iron was considered an exotic and precious metal in that time.

Very recently we have had the opportunity to carry out metallographical sections to three knives and a saw still keeping up metallic nucleus, found in the Portuguese late LBA sites of Catchouca, Moreirinha and Monte do Trigo. Full results remain still unpublished but this is a good scenario to mention some notable features. One of the knives contains a large amount of slag inclusions, what could be interpreted as consequence of a rudimentary technology, but the other cutting objects were made of iron of a good quality. Carbon amount is very low except in one blade, made by piling two sheets, one of very mild steel and the other of carburised iron with about 0.4% C (fig. 17). Therefore, the skilfulness of the smiths

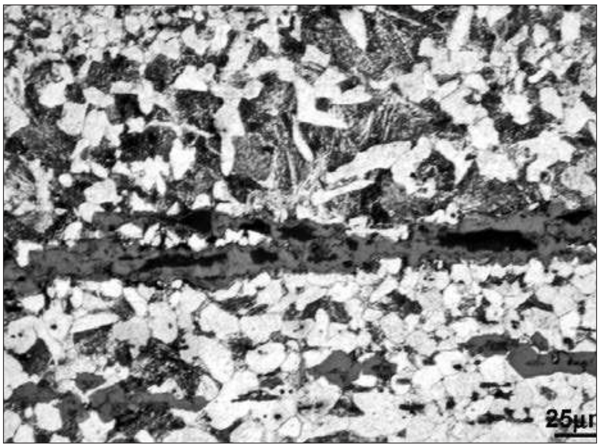

Fig. 17. - MOREIRINHA (PORTUGAL): metallography of the knife; detail of the union of the two piled sheets; some diffusion of carbon from the upper sheet to the lower one has occurred (etched with Nital).

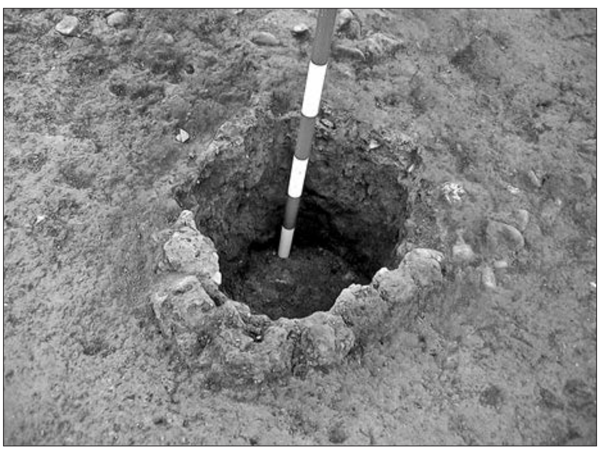

Fig. 18. - SEGEDA (ZARAGOZA): slag pit of an iron smelting furnace.

that made these objects was more or less the same that can be reckoned in the Iron Age metalworking. As iron slags have not been found in the sites, one can believe they are exotic goods acquired by trade, in a period that merchandising seems to be very active.

The spread of iron metallurgy in the Iberian Peninsula is linked to the proliferation of Phoenician settlements in the Mediterranean seaboard. The slags from Toscanos (Malaga) (KEESMANN *ET AL*. 1998) dated of the 7th-6th centuries BC seem to be related to a not too much developed smelting process, despite the authors said the contrary and reproduced a model of furnace purely imaginary (KEESMANN *ET AL*. 1998: 105, 107).

On the other hand, at an early Iberian culture site in Murcia province called El Castellar de Librilla, the platform where two probable "iron smelting furnaces" were sited was unearthed. The earliest one has been dated to the second half of the 8th century BC (ROS 1993). Its remaining structure is more likely a large forge than a furnace. The analyses of three slags (not correctly understood by the analysts who published them) containing lots of wüstite and low silica match well the composition of smithing slags (ARANA, PÉREZ 1993).

In any case, these sites are attesting the presence of iron workshops in a date as early as the late 8th century BC, what probably is in accord with the Portuguese finds above mentioned.

Despite archaeometallurgical study of early iron in Spain is in the beginning, one thing can be said: iron slags are frequently found, but not in large amount, in most Iron Age sites, no matter how large they are, what is an indication of the spread of iron technology in Iberia. The abundance in Spain of small rich iron oxidic ore deposits scattered on large territories favoured such dispersion. However, no significant slagheap has been detected by the moment that can be dated before Roman time. At the sight of these facts it is thought that smelting practices would be performed close to the mines, but this is a hypothesis that has not been proved, and systematic surveys must be done in future.

What do we know about early iron technology? Not too much, in few words. Notwithstanding this, it seems quite clear that smelting process was different in pre-Roman time. The study of slags from the early Iron Age of Aliseda (Cáceres), also inhabited in late Republican time, shows some slight differences in composition and microstructure between the two periods (ROVIRA, GOMEZ 1999). But the most amazing is that pre-Roman slags uses to be not tapped slags, what sometimes makes difficult to distinguish between smelting slags and smithing slags.

The only (probably) smelting furnace investigated up to the moment is one small slag-pit furnace unearthed in the Celtiberian site of Segeda (Zaragoza), dated to the middle of the 2nd century BC (ROVIRA, BURILLO 2003). The remains of the furnace consist of a small pit about 50 cm in diameter and 35 cm deep. The probably cylindrical body is lost, only remaining a clay wall few centimetres high surrounding the pit (fig. 18). Several plano-convex cakes and many nodular pieces of slag were found near this structure, but probably they are smithing slags.

Slag-pit furnaces were in use in France (CABBOÏ, Dunikowski 2004), England (Hodgkinson, Tebbut 1985) and Italy (Voss 1988) in pre-Roman time. The furnace from Segeda is much smaller than the French ones, but similar in size to the furnaces found at Baratti (Populonia).

Though not too much metallographic analysis of objects has been done, piling seems to be the standard method used to make the blades of the swords of the Iron Age (Nieto y Escalera 1970, Lorrio, Rovira, Gago 1998-1999). Carburising was normally applied but, surprisingly, some knives and cutting tools were made of ferritic iron or very mild steel. As many item have been found in graves where cremation ritual was practised, the thermal effect of the funeral pyre on the dead person's goods must be taken into account.

8. Some final thoughts

Archaeometallurgical research carried out in Spain by our project has been oriented to the evolution of metallurgical processes in Prehistory and its incidence in social changes than in other aspects. As far as I know, the Iberian Peninsula never yielded metal or ore or objects for trading outside its natural borders up to probably late Bronze Age and early Iron, centred in silver production and probably cassiterite mining, this last still remaining in the more extreme darkness. Before, during about two millennia, metallurgy, despite its general spreading on the Peninsula, seems to evolve under low social and economical pressure. Little amount of smelting and melting debris in the sites, non-existence of valuable hoards of metal objects up to the LBA, non existence of copper ingots up to late in the MBA (only a few are known) and the "primitive" features of copper obtaining leaded us to think that metallurgy was practised as a more or less domestic, not specialised activity (Rovira 2002a). It does not mean that the Iberian Peninsula as a whole remained out of the Mediterranean and Continental and Atlantic circuits of ideas. Not at all. A clear example of technological transference is the introduction of tin bronze by the end of Chalcolithic or EBA. But the slow spreading of this new technology, taking more than 500 years to reach the South, is a good proof of how a well-adapted metal technology was resistant to change. Another example of transference of ideas (but not of technology) is the adoption in LBA of some Atlantic types such as the swords. However, the alloys used to make that swords were different in Spain than in England or in France (Rovira 1995; Rovira, Gomez 1998).

Lead isotope analysis is being applied from more than ten years ago mainly to characterise the SW ore deposits in or near the Pyrite Belt and the possible provenance of metals produced in the area (Stos-Gale 2001; Hunt 2003). However the results are not too much consistent because the reference database is still small. If our hypothesis of little metal production and short distance circulation in early times is correct, lead isotope analysis may help to prove or discard it. By the moment, the suggestion by Stos-Gale (2001: 453-454) that four objects (one Chalcolithic and three Argaric) out of 23 may come from Sardinia and Liguria based upon the fact that their lead isotope signatures match quite well those of Sa Duchesa, Calabona and Ligurian ores respectively must be taken with reservations because the territory where these objects were found holds tens of copper mines but only three have been sampled, showing two different signatures. The Spanish geology is complex and the registered copper mines amount to some hundreds. So, the survey to get an acceptable lead isotope database will take much time and money.

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