Mediterranean Archaeology and Archaeometry, Vol. 17, No 1, (2017), pp. 217-231

DOI: 10.5281/zenodo.400779

ARCHAEOMETRIC CONTRIBUTION TO THE INTERPRETATION OF THE LATE BRONZE AGE "HOARD" FROM PORTO DO CONCELHO (MAÇÃO, CENTRAL PORTUGAL)

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Received: 20/10/2016
Accepted: 18/03/2017
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ABSTRACT

This paper presents and discusses the results of the multi-analytical study carried out on a group of 39 Late Bronze Age artefacts from the site of Porto do Concelho, Central Portugal. The chemical composition of the objects was determined by Energy Dispersive X-ray Fluorescence (EDXRF) and their microstructural features were identified by Optical Microscopy (OM) and Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM-EDS). The results show that metals are binary alloys (Cu+Sn) and leaded bronzes (Cu+Sn+>2.0 wt.% Pb), with a variable content of impurities, such as Pb (when lower than 2.0 wt.%), Fe, As, Ag, Sb and Ni. The microstructural characterization carried out on 21 artefacts allowed the identification of two main operational sequences: 10 objects show a dendritic microstructure suggesting that metals have not undergone any post-casting treatment; 11 artefacts display the presence of annealing twins with, in some cases, slip bands, resulting from the application of thermal and mechanical treatments.
The analytical results are compared to those of other metal collections from regional LBA, and together with the bibliographic information available become an opportunity to question and rediscuss the real nature of a collection of metals that most of archaeologists consider as a typical LBA founder's hoard.

KEYWORDS: Hoards, Late Bronze Age, Central Portugal, EDXRF, SEM-EDS, OM.

1. INTRODUCTION

The study of metal hoards belonging to the Late Bronze Age (LBA) from Western Iberia has been a subject of interest for the Portuguese archaeologists since the second half of the 19th century (Sarmento 1888; Veiga 1891; Azevedo 1895; Pereira 1898). Ever since, this topic has been irregularly regarded, generally in the occasion of new findings (Fortes 1905/ 1908a; Fortes 1905/ 1908b; Fortes 1905/1908c; Viana 1938; Cortez 1951; Costa 1963; Júnior 1968; Pereira 1971; Cardoso et al. 1992), while it was only in recent years that this phenomenon has been studied in a more comprehensive and systematic way, also through multidisciplinary approaches (Vilaça 2006; Gutiérrez Neira et al. 2011; Bottaini 2012; Bottaini et al. 2016).

The steady constraints to be faced in the study of this archaeological evidence are well known by the researchers and reside mainly in the antiquity of most of the findings and in the fortuitous nature of the discoveries published to date. In fact, most of the hoards from Western Iberia were found by chance between the late nineteenth century and the last quarter of the last century, usually by individuals engaged in rural activities.

In this scenario, after having been discovered, the objects were generally recovered in a precipitous way and then dispersed among the finders, sold to antiquarians or remelted in order to check the nature of the metal, with no major concerns about the record of the objects' contexts of provenance. Thus, information about the archaeological contexts, the circumstances of the findings and the exact content of the hoard are almost always incomplete and often divergent, if not irretrievably lost (Vilaça 2006: 29-37). Furthermore, it is not rare to have more or less arbitrary interpretations of contents and contexts perpetuating uncritically in the scientific community, acknowledging hoards that have never actually existed in the way they are described in literature. For all these reasons, the study of bronze deposits from Portuguese territory requires working with partial information whose authenticity is frequently hard to check.

Unlike this framework, the way the metals from Porto do Concelho were uncovered and the exact location of the finding place have been described in some detail in a paper published shortly after the discovery of the artefacts (Jalhay 1944). Despite this group of metals being considered one of the most significant evidences of the LBA metallurgy from Central Portugal, the artefacts have never been properly characterized from an analytical point of view, usually being valued for their typological features (Pereira 1970; Coffyn 1985) and for the relationship between the place of hoarding and other elements of the cultural landscape, i.e. nearby settlements and hoards (Delfino 2016).

In this paper, the application of chemical and micro-structural techniques has been used as a tool to better understand the nature of the metals from Porto do Concelho. For this purpose, a portable Energy Dispersive X-ray Fluorescence (EDXRF) spectrometer was adopted to determine the chemical composition of the objects; Optical Microscopy (OM) and Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM-EDS) was used to characterize the microstructure of the metals, in order to define the operational sequences that the ancient metalworker applied in the manufacture of the artefacts and to identify the presence of inclusions, casting defects and phases.

The aim of this paper is to understand the technological choices that the ancient metalworker made during the metal production process, allowing to answer to some specific questions: which alloys were employed for the production of these objects? How were they manufactured? How many operational sequences may we recognize within this group of metals? Is the metallurgy from Porto do Concelho coherent with that from LBA Central Portugal?

Moreover, through the discussion of the archaeometric data, preliminarily presented in Bottaini (2012) and Bottaini et al. (2015), and the critical reading of the information available in the bibliography, a new interpretation of Porto do Concelho is proposed.

2. PORTO DO CONCELHO METALS

The group of metals presented in this paper was discovered between March and July 1943 at the Porto do Concelho site, in Central Portugal (Fig. 1).

Figure 1: Location of Porto do Concelho and other sites mentioned in the text, namely: 1. Vila Cova de Perrinho; 2. Travasso: 3. Moura da Serra; 4. Coles de Samuel; 5. Castro de Argemela; 6. Quinta de Ervedal; 7. Gruta da Nascente do Algarinho; 8. Cabeço de Maria Candal (or Freixianda); 9. Senhora da Moita; 10. Castelo Velho do Caratão; 11. Casais de Fiéis de Deus; 12. Casais da Pedreira; 13. Moinho do Raposo.

The collection of metals, currently stored in the Museu de Arte Pré-Histórica e do Sagrado no Vale do Tejo, consists of tools, weapons, ornamental objects and artefacts with unknown function/functionality, being composed by 41 items, belonging to 40 artefacts, namely: two looped palstaves (PC127 and PC128); two Rocanes-type talon sickles (PC129 and PC130); three spearheads (PC131, PC132 and PC133); five swords (PC134, PC135, PC136, PC137 and PC138); four daggers (PC140, PC141, PC144 and PC150); two bracelets (PC143+PC 146 and PC145); one fibula (PC148); eleven entire ring-shaped objects (both closed and opened) (PC157; PC152; PC155; PC156; PC153; PC159; PC158; PC162; PC160; PC154 and PC163); two blades (PC139 and PC142); one conical-shape object (PC149); seven fragments (PC164; PC151; PC147; PC161; PC166; PC 168 and PC167). A sword is currently lost. The metal reaches a total weight of about 1.5 kg (Fig. 2).

Figure 2: Artefact collection from Porto do Concelho, including the lost sword (upper right box, according with Pereira 1970).

According to E. Jalhay, these objects were not all discovered at the same time, resulting instead from the assemblage of artefacts found in three different occasions: a first group composed by 35 objects was found under a stone slab by Joaquim Pires Caratão and a group of workers while building a country road, on March 6th 1943 (Fig. 3).

Figure 3: The founders of the first group of metals discovered on March 6th 1943 (Jalhay 1944) (upper). The site where the metals would be found, with a commemorative plaque of the finding (photograph of Museu de Arte Pré-Histórica e do Sagrado no Vale do Tejo) (lower).

Immediately after the discovery, the metals were dispersed among the finders, being recovered two days later by João Calado Rodrigues, the one who, during a survey carried out on March 9th 1943 in the site of Porto do Concelho, also proceeded to the discovery of four more artefacts. Finally, on July 8th 1943, Eugénio Jalhay, in the occasion of a new prospection, found the last group of metals, composed by two ring-shaped objects and one fibula (Jalhay 1944).

In a recent paper, the number of items assigned to this metal collection rose to 45 units (Delfino 2016: 94), although no further information has been provided about the typology and the circumstances in which these other artefacts would have been collected. Thus, in the absence of new and duly justified data, the artefacts to be attributed to the site of Porto do Concelho are the 42 described by E. Jalhay.

Within the traditional stigma of classification and categorization of hoards, the artefacts found in these three occasions have been interpreted as part of a single founder's hoard, i.e. as a group of materials temporarily stored to be recovered within the recycling and the consumption processes of metal (Jalhay 1944; Pereira 1970; Coffyn 1985; Ruiz-Gálvez Priego 1995; Melo 2000; Vilaça 2006: Cardoso 2007; Delfino 2016).

The seeming lack of logic in the composition of the collection, both at a typological and functional level, the physical state of the materials, with whole and fragmented objects, and the localization of the site where the artefacts were uncovered, i.e. in a strategic zone which would have apparently served as a crossing point, played a crucial role for this kind of interpretation.

From a typological point of view, the artefacts denote a strong regional character: in fact, although with a Mediterranean dissemination (Giardino 1995), the single-looped palstaves (Monteagudo 1977), the Rocanes-type sickles (Coffyn 1985), the Porto de Mós type of daggers (Fernández-García 1997), the socketed spearheads (Cardoso et al. 1992) and the swords (Meijide Cameselle 1988; Brandherm 2007) fall into the typical LBA productions documented in several settlements and hoards from Central Portugal (Coffyn 1985; Vilaça 2006; Bottaini 2012).

Only PC 148, corresponding to one fibula, and PC142 differ from that indigenous matrix. In the latter case, the function of the artefact, unique in the Portuguese LBA panorama, is unknown. Taking into account its elongated conical shape and its longitudinal drilling, this object has been interpreted as a metallic tuyères (Pereira 1970: 204), i.e. an object used to blow air in the forge, although the reduced size of the diameter of the hole makes this interpretation unlikely (Fig. 4).

Fig. 4: Two different views of the conical-shape object PC142.

Most of the artefacts are fragmented: palstaves, sickles, spearheads, swords, daggers, as well as some of the rings have fractures that clearly make them useless for the tasks for which they have been produced, although the antiquity of these fractures is, in some cases, questionable.

Indeed, and according to M. H. Pereira (1970: 184), one of the two sickles was broken at the time of the finding, which could also have happened with other artefacts, possibly damaged by the finders while verifying the nature of the alloy. Thus, it is hardly possible to distinguish the ancient fractures from the recent ones, also because in the meantime the objects were treated and preserved.

In any case, it must be noted that both tools and weapons are fractured. The main difference lies in the fact that the former, i.e. palstaves and sickles, despite fractured, are complete, while the latter, namely swords (except the lost specimen) and daggers (except PC144), appear to be limited to a single part of the artefact: the lower part of the blade for the swords, the handle for the daggers.

3. EXPERIMENTAL

According to the morphology and the state of conservation of the objects, different preparation methodologies were adopted: in order to avoid contamination from superficial corrosion patinas, the most fragile and deteriorated metals were mechanically cleaned in a small area to obtain the chemical composition of the bulk alloy. In the material studied by optical and electron microscopies, the analyses were performed on cross-sections of samples of about 3mm^2, manually removed from the artefacts with a jeweler's saw and mounted in resin.

Chemical analyses were performed by EDXRF with a portable spectrometer INNOV-X Alpha of the National Archaeological Museum of Madrid, equipped with a silver anode X-ray tube. Working conditions: 35kV, 2μA. The acquisition time was set at 40 s and the quantitative values were calculated from a calibration validated by certified standards. In the case of silver and antimony the detection limit is 0.15 wt.%, while for the remaining elements it is set to 0.02 wt.%.

Principal component analysis (PCA) has been used to correlate X-ray fluorescence data from Porto do Concelho with those from other Central Portugal LBA hoards. PCA was carried out on Sn, Pb and Fe, in order to understand compositional differences among the considered groups of metals.

The microstructural observation was carried out on samples mounted in epoxy resin and polished with silicon carbide papers (220-1200 μm) and diamond pastes (up to 1μm). The metallographic study was made with a Leica optical microscope, model DMLM, equipped with a DFC480 digital camera. In order to reveal the microstructure of the metal, the samples were etched with a ferric chloride acid solution ($FeCl_3+HCl+H_2O$) (Scott 1991).

Analyses by SEM-EDS were performed with an Hiachi S-3700N interfaced with a Quanta EDS microanalysis system with a Bruker AXS Xflash Silicon Drift Detector (129 eV Spectral Resolution at FWHM/Mn Kα). Data processing has been carried out with the Bruker ESPRIT 1.9 software.

4. RESULTS

4.1. *Alloy composition*

Some of the metals from Porto do Concelho were analysed in the past, although the results have never been published or discussed in detail. A group of objects, whose typology is not referred, was analysed by X-ray Diffraction (XRD) in the sixties of the last century. The results obtained are manifestly incompatible with the LBA metallurgy, since it was determined that metals were made with copper and zinc as main constituents (Pereira 1970: 24).

Some years later, A. Coffyn reports to have analysed four objects, without mentioning neither which objects were studied nor the analytical technique used. The results were summarily published, leading the author to state that the artefacts were made with alloys of copper and tin, with an average of 13.26 wt.% Sn, and low levels of lead (Coffyn 1998: 175).

In the present occasion, the whole collection from Porto do Concelho has been analysed, with the exception of three artefacts: PC161 and PC168 for being very small fragments and bearing a thick patina; and a sword for being lost. Chemical analysis by EDXRF was carried out on 40 items (corresponding to 39 objects since PC 143 and PC1466 belongs to the same bracelet).

The EDXRF elemental composition results are shown in the table 1. According to the data, copper is present in variable percentages between 64.6 wt.% (PC155) and 92.2 wt.% (PC132), with a major concentration in the range between 84.0 wt.% and 86.0 wt.% (fig. 5A). Tin also presents a very remarkable variability, ranging from 7.3 wt.% (PC132) to 29.1 wt.% (PC153), being the most recurrent range from 14.0 wt.% to 16.0 wt.% (fig. 5B).

The high tin content suggests that metals from Porto do Concelho were produced with fresh tin and not with scrap bronzes. In fact, experimental studies have shown how in bronze alloys produced from scraps without the addition of fresh tin, the tin content decreases significantly already from the first re-castings, resulting in alloys poorer in tin (Sarabia-Herrero 1992).

It is worth to be recalled that the control over the tin content is an essential condition in order to produce objects with good mechanical properties, since an excessive amount of tin (above 14.0 – 15.0 wt.% Sn) led to the formation of the α+δ eutectoid phase that makes the alloy increasingly brittle and difficult to deform without causing a crack.

Table 1: Results from EDXRF and OM. EDXRF elemental composition (wt.%); n.d.: not detected. Manufacture column: C: Casting; A: Annealing; F: Forging; FF: Final Forging. N.A.: not analyzed.

Object	Ref.	Cu	Sn	Pb	Fe	As	Ag	Sb	Ni	Manufacture
Palstave	PC127	85.4	14.3	0.18	n.d.	0.12	n.d.	n.d.	n.d.	C+(F+A)
Palstave	PC128	90.7	9.11	0.21	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)+FF
Sickle	PC129	81.7	15.1	0.18	n.d.	2.97	n.d.	n.d.	n.d.	C
Sickle	PC130	84.7	14.8	0.2	n.d.	0.26	n.d.	n.d.	0.34	C
Spearhead	PC131	86.5	12.6	0.57	n.d.	n.d.	n.d.	n.d.	0.16	C
Spearhead	PC132	92.2	7.3	0.46	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)
Spearhead	PC133	85.0	14.2	0.33	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)
Sword	PC134	88.3	10.8	0.91	n.d.	n.d.	n.d.	n.d.	0.21	C
Sword	PC135	88.5	11.2	0.29	n.d.	n.d.	n.d.	n.d.	n.d.	C
Sword	PC136	80.7	18.3	0.23	n.d.	n.d.	n.d.	n.d.	n.d.	C
Sword	PC137	87.7	11.9	0.39	n.d.	n.d.	n.d.	n.d.	n.d.	C
Sword	PC138	82.1	17.6	0.25	n.d.	n.d.	n.d.	n.d.	n.d.	C
Blade	PC139	80.5	18.6	0.35	n.d.	n.d.	n.d.	n.d.	0.54	C
Dagger	PC140	85.4	14.2	0.37	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)
Dagger	PC141	84.0	15.6	0.3	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)
Undefined object	PC142	90.0	8.72	1.09	0.13	n.d.	n.d.	n.d.	0.09	C+(F+A)
Bracelet shaped object	PC143	90.0	9.92	0.1	n.d.	n.d.	n.d.	n.d.	1.14	C+(F+A)
	PC146	88.6	11.2	0.15	n.d.	n.d.	n.d.	n.d.	1.06	N.A.
Dagger	PC144	86.7	12.9	0.4	n.d.	n.d.	n.d.	n.d.	n.d.	C
Bracelet shaped object	PC145	85.7	14.0	0.24	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)+FF
Ring shaped object	PC147	89.1	10.5	0.33	0.09	n.d.	n.d.	n.d.	n.d.	N.A.
Fibulae	PC148	91.8	7.89	0.24	n.d.	0.08	n.d.	n.d.	n.d.	N.A
Undefined object	PC149	75.6	22.5	0.08	n.d.	0.98	0.17	0.41	0.14	N.A.
Dagger	PC150	88.2	11.3	0.27	n.d.	n.d.	n.d.	n.d.	0.18	C+(F+A)+FF
Undefined object	PC151	88.9	10.9	0.16	n.d.	n.d.	n.d.	n.d.	n.d.	C+(F+A)
Ring shaped object	PC152	82.3	14.0	3.67	n.d.	n.d.	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC153	69.1	29.1	0.51	n.d.	1.31	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC154	84.8	14.7	0.25	n.d.	0.12	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC155	64.6	20.9	14.5	n.d.	n.d.	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC156	75.6	23.1	0.83	n.d.	0.43	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC157	74.3	23.7	1.62	n.d.	0.25	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC158	76.6	22.3	0.26	n.d.	0.84	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC159	72.2	26.1	0.61	0.48	0.57	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC160	80.4	19.3	0.28	n.d.	n.d.	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC162	85.74	9.16	5.0	0.11	n.d.	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC163	77.4	20.90	0.63	0.63	0.46	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC164	85.8	12.2	1.98	n.d.	n.d.	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC165	77.6	20.6	1.33	0.1	0.29	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC166	85.5	12.0	2.53	n.d.	n.d.	n.d.	n.d.	n.d.	N.A.
Ring shaped object	PC167	87.4	8.42	4.14	n.d.	n.d.	n.d.	n.d.	0.07	N.A.

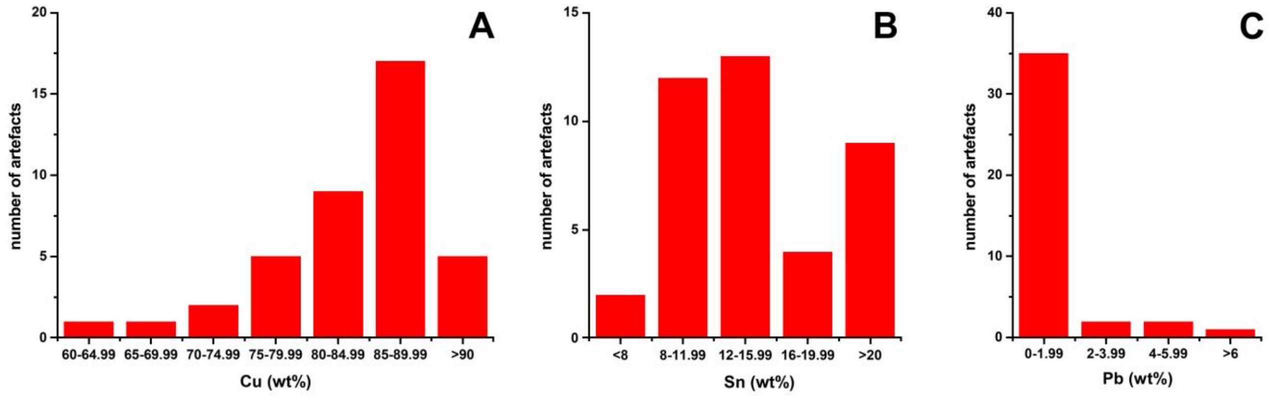

Figure 5: Histograms with the distribution of Cu (A), Sn (B) and Pb (C).

The results from Porto do Concelho show that in the objects that supposedly required higher mechanical strengths, such as palstaves, sickles and most of the weapons, tin is kept below the limit of 15.0 wt.%, whilst it is higher in most of the ring-shaped objects, wherein half of the specimens have a content above 20.0 wt.% Sn, with a global average of ~18.0 wt.% Sn.

Metals with high tin content are not an absolute novelty in regional terms, as already demonstrated by a ring from the LBA settlement of Castro de Argemela (27.6 wt. % Sn) (Vilaça et al. 2012b).

Since alloys with tin content between 20.0 and 30.0 wt.% produce a characteristic silver colour (Giardino 1998: 142), more suitable for ornamental objects, it may be admitted that high tin content could be an intentional technological choice in the production of certain type of artefacts.

Lead is systematically identified in all the artefacts, ranging from 0.1 wt.% (PC143) to 14.5 wt.% (PC155) (Fig. 5C). Most of the analysed objects (35 out 39) contain a low amount of lead (< 2.0 wt.%), also visible by SEM-EDS images (Fig. 6), suggesting that its occurrence is not the result of an intentional addition, instead resulting from presence of this element in the ores used within the fabrication of the alloys or providing a clue for the use of recycled metals.

Conversely, the higher lead content in four ring-shaped objects (from 2.53 up to 14.5 wt.% Pb) probably results by a clear technological option. Although the function of ring-shaped objects is rather ambiguous, the high lead content may confirm that these type of artefacts were used for tasks that did not require high mechanical strength. In fact, the low miscibility of Pb in Cu+Sn alloys leads to the formation of unalloyed segregations that increases brittleness and reduces the strength of the alloy.

Besides the major elements, impurities were identified as well, namely Ag, Sb, Fe, As, Ni and Pb (<2.0 wt.%). PC149 is the only artefact containing small amounts of both Ag and Sb, respectively, 0.17 wt.% and 0.41 wt.%, demonstrating the uniqueness of this artefact not only from the morphological point of view, as well as in terms of chemical composition.

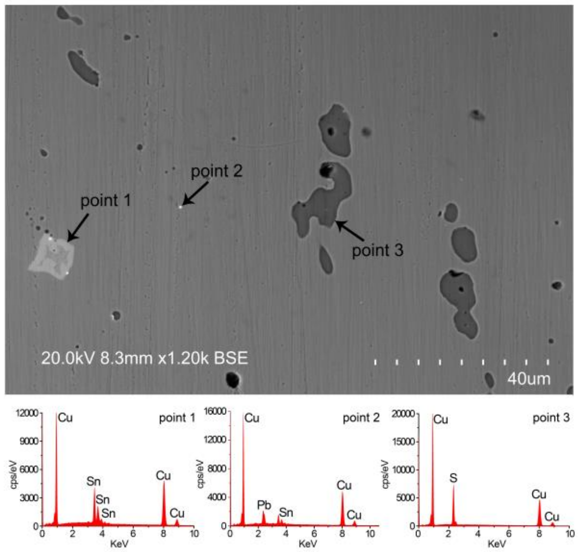

Figure 6: Backscattered electron image with EDS spectra of the bracelet PC145, showing the α+δ eutectoid phase (27.0 wt.%) (point 1); Pb-rich inclusion (point 2); Cu-S inclusion (point 3).

The occurrence of Ag and Sb is a common feature of the LBA metallurgy from Central Portugal, although with values generally below 0.1 wt.%, as the analyses by Particle-induced X-ray emission (PIXE) on the metals from Cabeço de Maria Candal (also known as Freixianda) hoard proved (Gutiérrez Neira et al. 2011). In the case of Porto do Concelho, the high detection limits for both Ag and Sb (0.15 wt.%) did not allow to properly quantify their presence.

To overcome this limitation, SEM-EDS analysis was performed which allowed the identification in several pieces of small inclusions enriched in Ag, such as in PC130, PC137, PC139, PC141 and PC143 (Fig. 7).

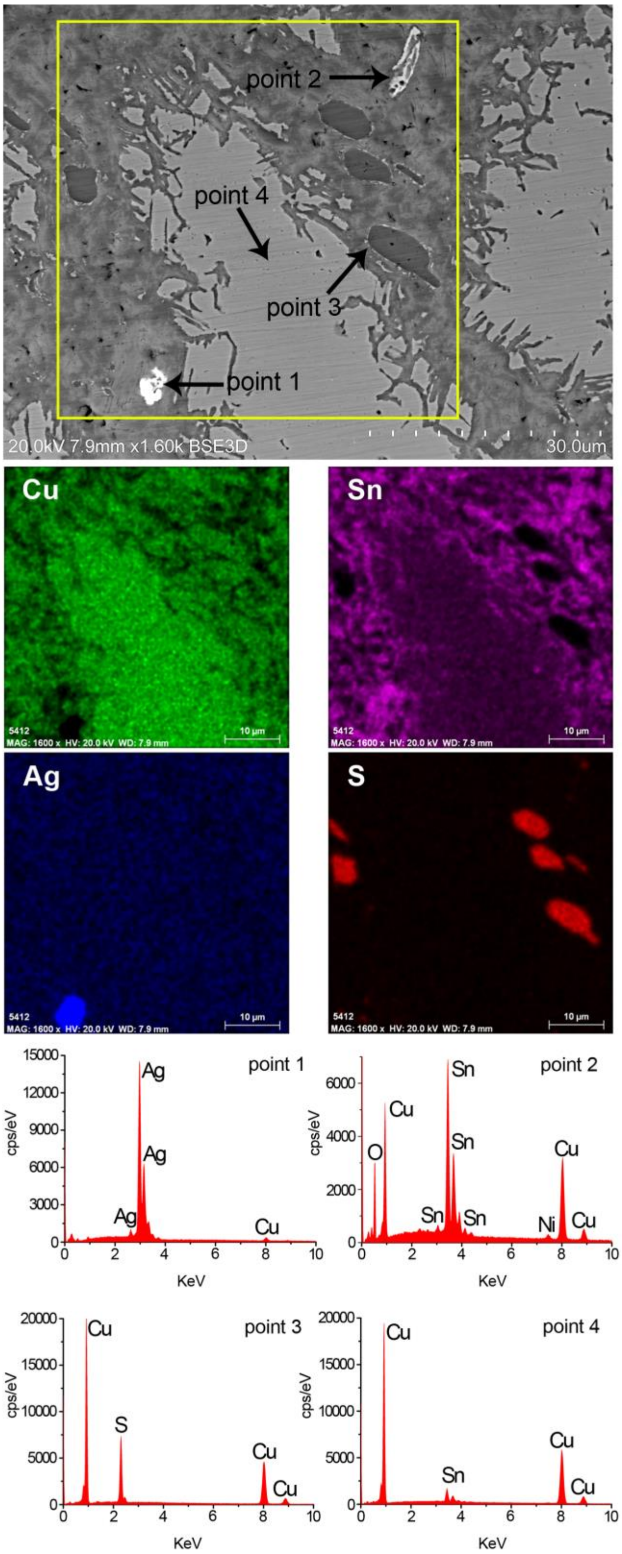

Figure 7: Backscattered electron image with EDS elemental distribution maps and EDS spectra of the blade PC139, showing the presence of the an Ag-rich inclusion (point 1); a+δ eutectoid phase (49.5 wt.% Sn) (point 2); S-rich inclusion (point 3); a-phase (10.4 wt.% Sn) (point 4).

One undefined object (PC142) and five rings (PC147, PC159, PC162, PC163, PC165) contain iron, with amounts ranging from 0.09 and 0.63 wt.%, suggesting the use of copper smelting furnaces able to achieve temperatures sufficiently high to reduce the iron impurities present in the ores, much more efficient than the traditional furnace vessels (Craddock and Meeks 1987; Rovira and Ambert 2002), probably used for the production of the rest of the objects. Arsenic is present in 13 objects with values between 0.08 wt.% and 2.97 wt.%., whilst 10 objects contain variable quantities of nickel (to 0.07 up 1.14 wt.%). The occurrence of both As and Ni is probably due to the presence of impurities contained in the ores used within the production process and carried out through to the finished artefact.

Thus, the artefacts from Porto do Concelho appear to have a more heterogeneous elemental composition when compared with binary bronzes with few impurities typical of the LBA metallurgy from Central Portugal.

Fig. 8 displays a statistical multivariate analysis (PCA) carried out on a matrix dataset in which XRF data from Porto do Concelho and from LBA hoards from Central Portugal are compared. The data confirms that within a group of metals from Porto do Concelho, tin, iron and lead achieves a slightly higher average content with respect to the metals found in the rest of the regional collections. Finally, it should be highlighted that the total impurities amount (Fe, As, Ag, Sb, Ni and <2.0 wt.% Pb) identified in 13 objects from Porto do Concelho reaches higher values (> 1.0 wt.% of the alloy composition) than those expected for the region.

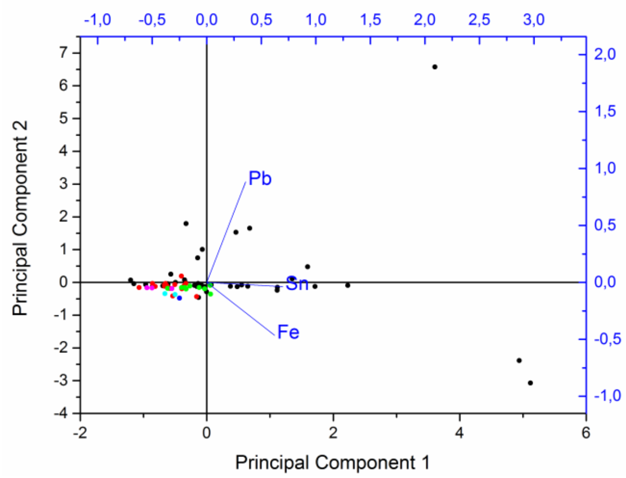

Figure 8: PCA discriminating among the Sn, Pb and Fe amount in Cu-based artefacts which shows the deviation of the elemental composition of the Porto do Concelho metals relative to other metal sets in the LBA of Central Portugal. Porto do Concelho (black dots); Coles de Samuel (red dots); Cabeço de Maria Candal (green dots); Moinho do Raposo (dark blue dots); Casais da Pedreira (light blue dots); Travasso (purple dots).

4.2. Microstructure

The metallographic study focused on 21 artefacts, namely: two one-looped palstaves (PC127 and PC 128), two sickles (PC129 and PC130), three spearheads (PC131, PC132 and PC133), five swords (PC 134, PC135, PC136, PC137 and PC138), two blades (PC139 and PC142), four daggers (PC140, PC141, PC 144 and PC150), two bracelets (PC145 and PC146+ PC143) and one undefined object (PC151).

Based on the data, two different manufacturing techniques were identified. Two sickles (PC129 and PC 130), one spearhead (PC131), five swords (PC134, PC 135, PC136, PC137 and PC138), one blade (PC139) and one dagger (PC144) show a dendritic micro-structure (Fig. 9), which suggests that after being removed from the mould, the ancient metal-worker did not apply any thermomechanical treatment to the artefacts.

As for the rest of the objects, two palstaves (PC127 and PC128), two spearheads (PC132 and PC133), three daggers (PC140, PC141 and PC150), two bracelets (PC145 and PC143) and two undefined objects (PC142 and PC151) show the presence of recrystallized twinned grains with, in some cases, slip bands (PC128, PC145 and PC150). This kind of microstructure suggests that after being removed from the moulds, artefacts were heated under the melting point to make it more ductile and more suitable to be plastically deformed through forging. This way, cycles of hot/cold working were repeated until the objects had the desired shape and hardness. The presence of slip bands indicates a finishing treatment through the application of a more intense forging cycle (Fig. 10).

The observation by SEM allowed to identify several inclusions dispersed in the microstructures of the alloys: in addition to those mentioned above (α+δ eutectoidc and Ag), segregations of Cu-S, nickel-rich inclusions (Fig. 11A) and unalloyed copper inclusions (Fig. 11B) were also detected. The presence of the latter could be explained as a remnant of incomplete mixing or as the result of a copper re-deposition process resulting from corrosion phenomena (Bosi et al. 2002; Emami et al. 2013).

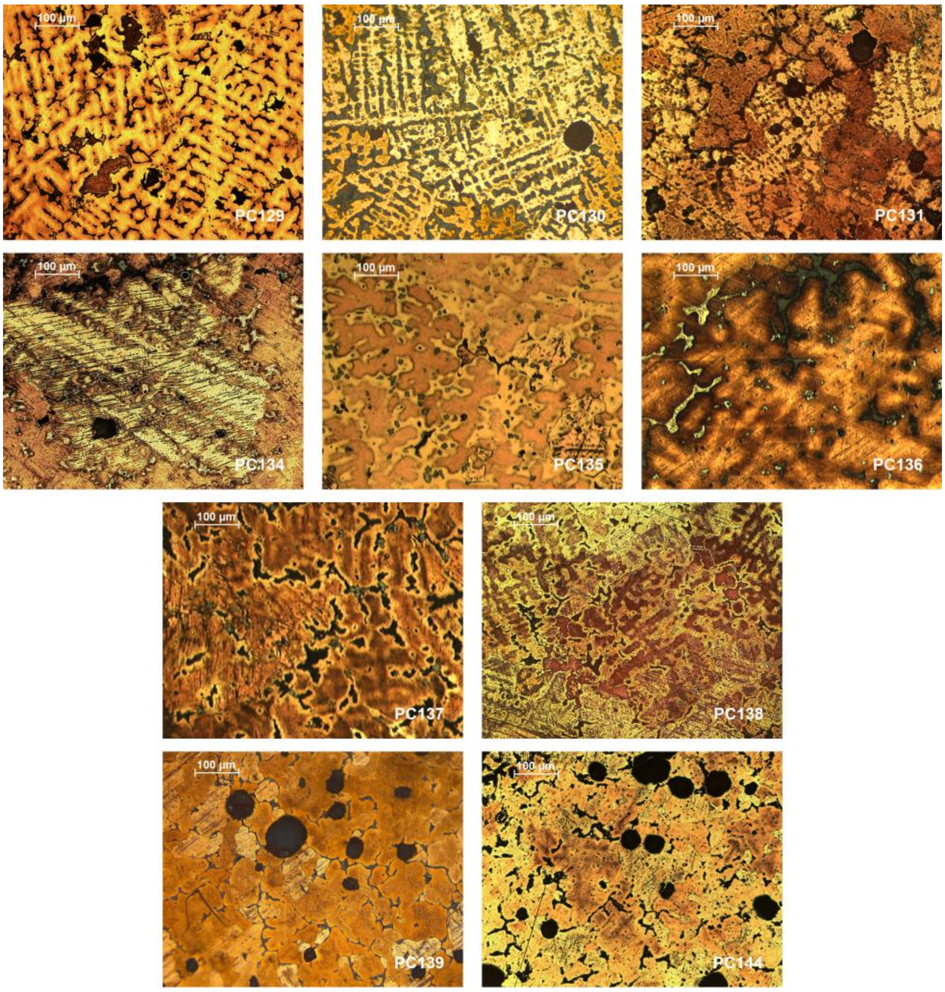

Figure 9: Microstructures of artefacts showing the dendritic microstructure typical of metals that did not undergone to any post casting treatment.

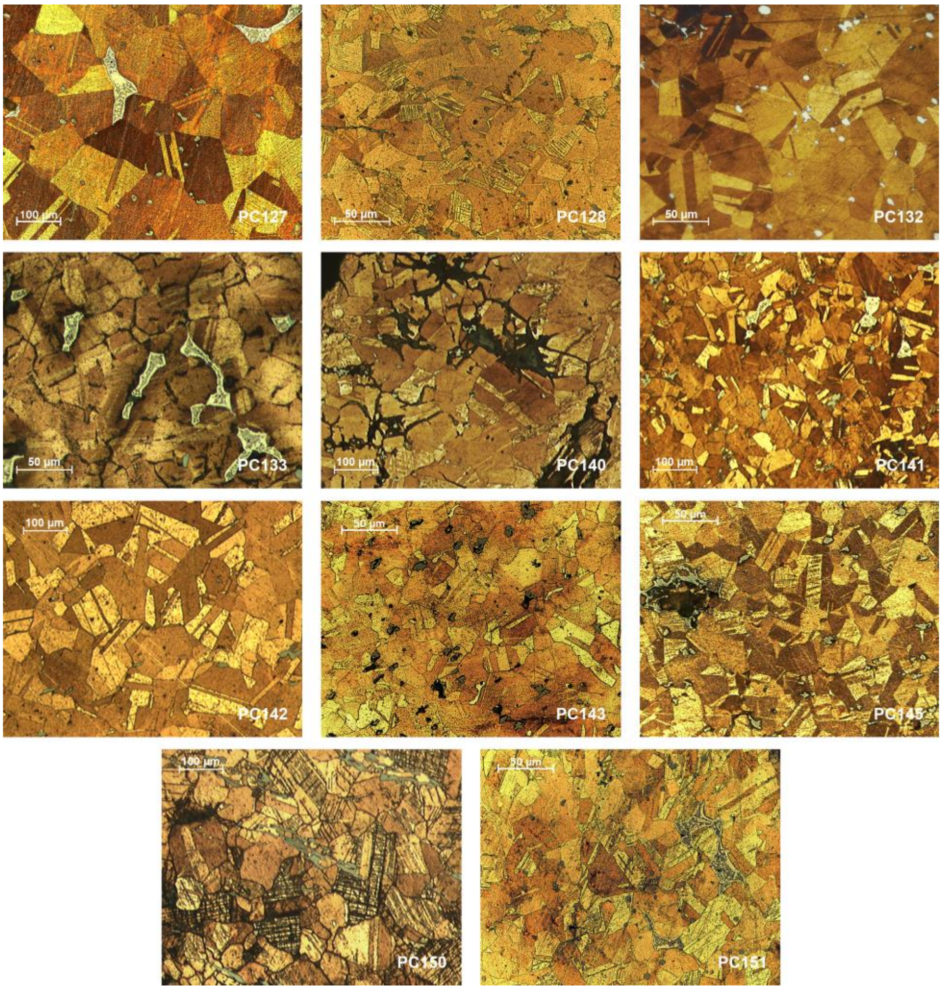

Figure 10: Microstructures of artefacts showing recrystallized grains and sliding bands (PC128, PC145 and PC150), which suggests the applying of post-casting treatment (forging+annealing).

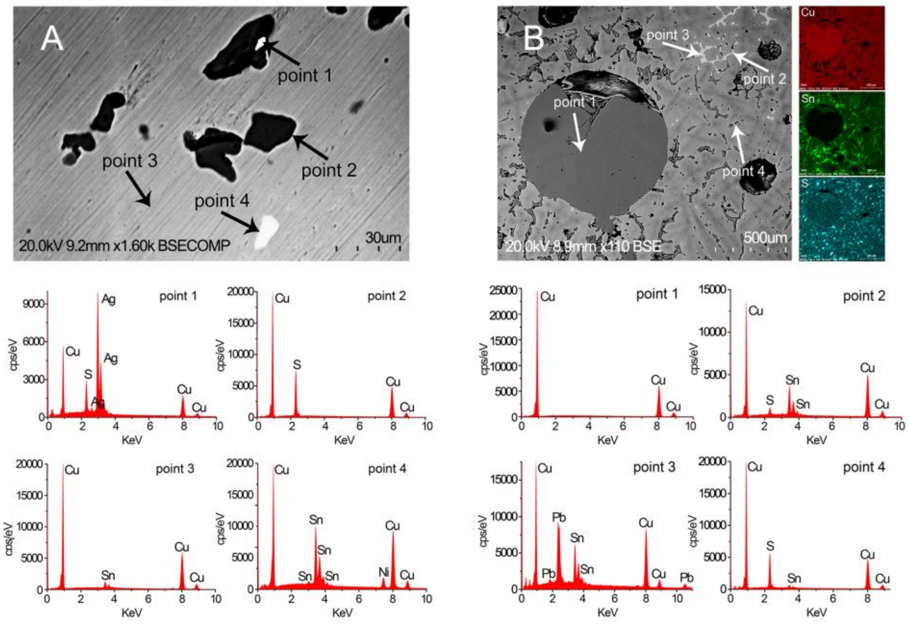

Figure 11: A) Backscattered electron image with EDS spectra of the bracelet PC143 showing an Ag-rich inclusion (point 1); S-Cu inclusion (point 2); a-phase (7.8 wt.% Sn) (point 3); Ni-rich inclusion (point 4). B) Backscattered electron image with EDS elemental distribution maps and EDS spectra of the sickle PC130 with a globular unalloyed Cu-inclusion (point 1); a+δ eutectoid phase (24.15 wt.% Sn) (point 2); Pb rich-inclusion (point 3); Cu-S inclusion (point 4).

5. DISCUSSION

The description of the circumstances in which the metals from Porto do Concelho were found has been used to support the interpretation of this collection as a founder's hoard. However, the paper published in 1944 does not provide suitable information on the real composition of the collection and leads one to wonder about its real archaeological nature.

In fact, the artefacts were found in at least three circumstances and by different people in an area generically designated with the toponym of Porto do Concelho. The spatial relationship between the exact location where each single group of metals stood is not referred and there is not any effective control over the type and the number of artefacts assigned to the first finding. In respect of the latter, the 1944 paper informs that it was composed by 35 artefacts, among which rings, daggers, bracelets and axes. But are these artefacts those that were discovered in that occasion or are just those that J.C. Rodrigues was able to recover two days later? It stands to reason that the artefacts recovered might not correspond to those actually found under the stone slab.

In fact, it is worth mentioning that other sites with LBA metallurgy are known close to Porto do Concelho, namely Senhora da Moita and Castelo Velho do Caratão, and that metals from these sites were found casually at about the same time of those from Porto do Concelho, being dispersed among local people as well (Pereira 1970). Given the lack of more detailed information on the objects found at Porto do Concelho on March 6th 1943 and those recovered from the hands of the finders two days later, it is admissible that other artefacts coming from nearby sites could also have been mixed and arbitrarily attributed to the collection from Porto do Concelho.

Thus, the strict reading of the information presented by E. Jalhay would eventually open up the possibility of being faced with a different reality than the one that has been perpetrated until now in the bibliography. In fact, the widespread of different groups of metals within an area where neither settlement nor grave have been apparently found, suggests that we are not dealing with a single hoard but rather with different hoards that ancient local communities buried in an evocative space possibly devoted to the deposition of artefacts with culturally shared values (Fontijn 2002-2003; Vilaça 2006: 39). This is a well-known reality in other European regions, such as in the Armorican region, in the Northwest of France (Verron 1983; Gabillot 2003), where single or multiple depositions, not necessarily with chronological relation, occurred in areas with clearly defined borders, possibly chosen for meanings reflecting the perception that ancient societies had of such a place.

In fact, the group of metals from Porto do Concelho shows internal elements that are distinctive of the hoarding practices from Central Portugal and that seem to point out the existence of selective deposition processes, especially at the level of combinations of certain metal types. The association between palstaves, sickles, bracelets, daggers, swords and spearheads, for example, appears to be documented in several LBA regional hoards, such as Cabeço de Maria Candal (palstave + dagger) (Gutiérrez Neira et al. 2011; Vilaça et al. 2012a), Coles de Samuel (palstave + bracelet + sickle) (Bottaini et al. 2016), Travasso (palstave + sickle) (Leitão e Lopes 1984), Moura da Serra (palstave + sickle) (Nunes 1957) and Casal de Fiéis de Deus (bracelet + dagger + sword + spearhead) (Vasconcelos 1919-1920; Melo 2000).

At the same time, certain other types of metals present at Porto do Concelho, such as the fibula (PC 148), the object interpreted as a "tuyères" (PC149) and the ring-shaped objects, seem to be deliberately avoided within the hoarding practices known for Central Portugal.

From an analytical point of view, the comparison between the elemental composition of the metals from Porto do Concelho and others from LBA Central Portugal hoards also highlights a contrasting situation. Unlike the Late Bronze Age / Early Iron Age metallurgy from Northwestern Iberia, where the addition of lead to copper-based artefacts was a common practice (Montero et al. 2003), LBA metals from Central Portugal are generally composed of binary Cu-Sn alloys (with 8.0-15.0 wt.% Sn in most of cases), with a reduced amount of impurities (Vilaça 1997; Figueiredo et al. 2010). The analysis carried out on Cabeço de Maria Candal hoard show that artefacts were made of bronze with a slightly higher average of tin contents (~14.0 wt.% Sn) and reduced amount of impurities (<1.0 wt%) (Gutiérrez Neira et al. 2011); similarly, the 18 artefacts from the Coles de Samuel hoard are composed by binary bronzes (~11.0 wt% Sn) with impurities not exceeding 1.1 wt.% (Bottaini et al. 2016).

These data are not entirely consistent with the results obtained at Porto do Concelho, where different groups of copper-based alloys may be identified. Most of the artefacts are composed by bronze alloys, with tin not exceeding 15.0 wt.% and generally reduced impurities (<1.0 wt.%). Fifteen artefacts can be classified as high tin bronzes (>15.0 wt.%), while five rings are leaded bronzes (Cu + Sn + >2.0 wt.% Pb). Ring-shaped objects show a clearly different chemical composition when compared with the rest of the material. Five rings contain more than 2.0 wt.% of lead, whilst five have more than 0.05 wt.% of iron, not being possible to establish any relationship between the occurrence of these two elements.

The observation of the artefacts' microstructure provides information very useful from an interpretative point of view. At a first glance, it can be observed that both ascast metals and artefacts which suffered post casting treatments coexist in the same context, being these results coherent with metallographic studies carried out on bronze artefacts from Cabeço de Maria Candal (Vilaça et al. 2012a), Coles de Samuel (Bottaini et al. 2016), Gruta da Nascente do Algarinho (Figueiredo et al. 2011), Vila Cova de Perrinho, Casais da Pedreira and Moinho do Raposo (Bottaini 2012). In a broader perspective, the data from these hoards suggest that metals had distinct biographical trajectories, some retrieved from circulation without being adequately prepared for practical purposes, others presumably thrown out after being used.

According to a visual and a microscopic observation of the artefacts, five swords (PC134, PC135, PC136, PC137 and PC138) and three daggers (PC140, PC141 and PC150) seem to stand out from the rest of the artefacts from Porto do Concelho, revealing a situation probably more complex than that accepted for their interpretation as scrap metals. As already high-lighted, the five swords are reduced to the lower part of the blade and the tip, with the only exception of PC134; they have a dendritic microstructure, whilst the blades show irregularities (i.e. asymmetrical blades and presence of nicks) that suggest some kind of impact. The three daggers are limited to the handle and, in the cases of PC140 and PC141, to the upper part of the blade; the microstructure suggest that these artefacts were subjected to a plastic deformation by annealing and forging.

The deposition of fragmented weapons, often with signs of deliberate destruction, damage and breaking, is recurrent in the LBA hoards from the European Atlantic façade (Bradley 1990; York 2002; Quilliec 2008), being also documented in Central Portugal, namely in the deposits from Quinta do Ervedal (Villas-Bôas 1947), Casais de Fiéis de Deus (Vasconcelos 1919-1920; Melo 2000) and Cabeço de Maria Candal (Vilaça et al. 2012a).

The repetitiveness of this depositional pattern over a wide geographical area, as well as the physical conditions and the microstructural features observed in the swords and in the daggers from Porto do Concelho, would allow one to consider these two groups of artefacts not as a simply accumulation of metal arbitrarily stored to be later recycled (i.e. as part of a founder's hoard), rather as the materialization of codified and structured depositional practices in which weapons (as well as others types of artefacts) could be ritually broken before being deposited (Nebelsick 2000; Fontijn 2002-2003). In this way, and according to an understanding of the hoarding phenomenon that goes beyond the dichotomy between profane and ritual hoards, the act of fragmentation itself would be part of the biography of object identities, being a crucial event able to legitimate the voluntary deposition of a certain object (Chapman and Gaydarska 2007).

Thus, the analyses carried out on the Porto do Concelho collection show a quite heterogeneous situation in terms of both chemical composition and microstructural characterization, partially in agreement with the characteristics of the LBA metallurgy from Central Portugal. According to these data, metals from Porto do Concelho could have been made by craftsman with different levels of technological know-how and technical skills, able to produce binary bronzes, leaded alloys as well as metals with higher iron amount (which could indicate a different smelting technology) and to give these objects the desired shape through casting or forging and annealing cycles. Hence, it is possible that the artefacts from Porto do Concelho were produced at different times, being hoarded at the same time or, in a vision of "open hoard", over several times. But none of the analytical data confirms that the objects have been imported from other regions, as has been recently suggested (Delfino 2014).

6. CONCLUSIONS

The study of the metal hoards from the Portuguese territory requires working with very partial and sometimes equivocal information, which makes the interpretation of each single hoard, as well as of the hoarding phenomenon in general, even more difficult. In the historiography on this topic, the collection from Porto do Concelho represents a very paradigmatic case: resulting of fortuitous findings discovered in three different circumstances, this group of artefacts was immediately regarded as an example of a typical LBA founder's hoard.

The lack of control over the findings and over the recovery of the artefacts dispersed among the finders after the unearthing of the first group of objects, raise questions on the authentic nature of the metals from Porto do Concelho. The analytical results seem to strengthen these doubts: while the LBA hoards from Central Portugal show a fairly homogeneous chemical composition, i.e. pure bronze alloys with reduced impurities, the metals from Porto do Concelho are characterized by a more diverse composition, coexisting binary Cu-Sn alloys and leaded bronzes (Cu+Sn+>2.0 wt.% Pb), containing amounts of impurities, namely Fe, that in some cases appear to be very significant. Furthermore, the high Sn content is a relevant issue, since if we consider the metals of Porto do Concelho as a founder's hoard whose existence is justified within the metal recycling practices, a lower Sn content might be expected.

Metallographic observations enable to describe the microstructure of the alloys, thus providing additional information about technological aspects. In particular, this technique made possible the identification of two main different microstructures: a group of artefacts presents a dendritic microstructure typical of objects that have not been subjected to any post-casting treatment; a second one shows the presence of recrystallized grains, sometimes with slip bands, which suggests the use of alternate cycles of annealing and forging applied in order to obtain metals with higher resistance.

In conclusion, based on archaeological information and on the analytical data, and unlike the interpretation generally accepted in the bibliography, the idea that metals from Porto do Concelho were part of a single hoard temporarily stored by an ancient founder does not seem plausible. Anyway, at the same time, the presence of recurring elements within the phenomenon of selective deposition from Central Portugal, i.e. the occurrence of characteristic typological combinations of certain objects, the physical features of the swords and daggers, etc., does not exclude the existence, within the whole collection from Porto do Concelho, of smaller groups of artefacts that may have been voluntarily removed from circulation and separately hoarded in an area specially designed for this purpose.

ACKNOWLEDGEMENTS

This work has been financed by national funds by FCT – Foundation for Science and Technology under the UID project UID/HIS/00057/2013 (POCI-01-0145-FEDER-007702) and UID/Multi/04449/2013 (POCI-01-0145-FEDER-007649), COMPETE, FEDER, Portugal2020. The contribution of IMR and RV is part of the I+D+i project HAR2014-52981-R funded by the Spanish Ministerio de Economía y Competitividad. The authors would like to acknowledge Dr. Luiz Oosterbeek (Instituto Politécnico de Tomar and Director of the Prehistoric Art Museum of Mação) for allowing the study of the metal artefacts and the Museu Dom Diogo de Sousa (Braga) for the restoration of the objects. The first author also thanks the FCT for the SFRH/BPD/111039/2015 grant and Neil Young for the support during the preparation of this paper.

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