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Arsenical copper and bronze in Middle Bronze Age burial sites of southern Portugal: the first bronzes in Southwestern Iberia

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ABSTRACT

Middle Bronze Age was a transition period in Iberia, characterised by the emergence of bronzes after more than a millennium of a conservative metallurgy of copper with arsenic. Despite its importance there are no relevant studies on MBA metallurgy in Southwestern Iberia due to the absence, until recently, of known settlements and the scarcity of metals. However, recent archaeological excavations have brought to light important finds dated to the SW Iberian Bronze Age such as new burial monuments and open settlements. About 50 artefacts from hypogea, cists and domestic contexts (pits) from Torre Velha 3 (Serpa) and Monte da Cabida 3 (Évora) were analysed by micro-EDXRF, reflected light microscopy, SEM–EDS and Vickers microhardness testing. Radiocarbon dating of their archaeological contexts established a chronology of ~1900–1300 cal BC. Despite presenting different burial practices both sites share the almost exclusive use of arsenical coppers (4.1 ± 1.0 and 4.2 ± 1.5 wt.% As, respectively). However, few awls and a dagger from Torre Velha 3 are among the earliest evidence of bronze in SW Iberia, being dated to the second quarter of the 2nd Millennium BC. These bronzes are similar (9.6 ± 1.2 wt.% Sn) to LBA alloys suggesting trade with a region with a developed bronze metallurgy. The emergence of bronze in SW Iberia during the first half of the 2nd Millennium BC points to an earlier introduction or a more rapid expansion than initially assumed. Nevertheless, these arsenical coppers and bronzes display a similar manufacture involving hammering and annealing cycles. A final hammering increased the hardness, which could be higher for bronzes. Arsenical coppers display variable operational conditions often with poorer thermomechanical work as expected from a prehistoric technology. A bronze dagger with silver rivets evidences the prestige value of early bronzes to MBA communities. Similarly, an arsenical copper dagger with silver coloured rivets shows the ability of MBA metallurgists to replicate prestige objects with indigenous knowledge.

1. Introduction

The development of prehistoric metallurgy was often a very slow process, being heavily dependent on technological knowledge and availability of metallic ores (Craddock, 1995). Early metals from Iberia were composed of copper with varying amounts of arsenic, a feature that persisted for more than a millennium, ~3000–1200 BC (Rovira, 2002). The true meaning of arsenical copper alloys (copper with more than 2 wt.% As) among ancient communities is still unclear. Arsenical copper does not seem to be an evolution in copper metallurgy since in some regions it is present among the first contexts with evidences of metals (Ruíz-Taboada and Montero-Ruíz, 1999). However, the possibility of attaining higher hardness or different colours was certainly among the first characteristics recognized by metallurgists. For instance, certain typologies at the Chalcolithic settlement of Zambujal (Torres Vedras), namely Palmela points, saws, long awls and tanged daggers, have higher arsenic content (Müller et al., 2007). In another example, a relation between arsenical coppers and tools/weapons has been identified at the Chalcolithic settlement of Castro de Vila Nova de São Pedro (Azambuja) pointing to a deliberate selection of alloys (Pereira

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et al., 2013). This work also suggested that colour, and not hardness, was the objective behind the production of those arsenical coppers. Later on, Middle Bronze Age (MBA) artefacts from Southwestern Iberia still have highly variable contents of arsenic. However, the limited number of studied examples inhibits a clear trend, with the exception of swords that show higher amounts of arsenic (Hunt Ortiz, 2003).

The conservative metallurgy of copper was slowly but progressively replaced by bronze (copper with more than 2 wt.% Sn). In Europe, very few bronzes can be securely dated to the 1st half of the 3rd Millennium BC. A double-edged knife from a 3080 to 2880 cal BC burial at Velika Gruda (Montenegro) is one of them, being considered to be an importation from the Near Eastern region (Primas, 2003). The earliest bronzes in the Atlantic region already belong to the 2nd half of the 2nd Millenium BC, such as a few burial offerings from 2580 to 1890 cal BC at Saint Lude 2, Bretagne (Briard, 1984). Likewise, the first true metalworking tradition in Britain belongs to 2300–2200 cal BC (Needham, 1996). Curiously, the only bronzes among the metallic collection from the 2300 to 2000 cal BC burial of Singen (Germany) have Atlantic typologies (Fernandez-Miranda et al., 1995). This evidences the complex pathways of the spread of bronze in prehistoric Europe. In the Western Mediterranean region bronze arises during almost the same period. In Italy, for instance, the bronze alloy becomes regular during the Early Bronze Age, 2200–1800 BC (Eaton, 1980), period after which the first bronzes appear in Sardinia, namely during 1800–1600 BC (Lo Schiavo, 1997).

The first bronzes in Iberia made their appearance in the northeastern region during the Early Bronze Age, perhaps due to the existence of exchange networks with other regions of Western Europe (Fernandez-Miranda et al., 1995). Archaeological works at Bauma del Serrat del Pont (Girona) recovered a bronze awl/needle and a bronze rod dated to 2560–1975 cal BC (Alcalde et al., 1998). According to this paper, however, the premature appearance of bronze in this settlement could be related with copper ores with high amounts of tin. These ores were already smelted during the Chalcolithic occupation of the site, but often failed to produce bronze due to unsuitable reducing conditions. Also in the northeastern region, namely at Monte Aguilar (Navarra) two bronze awls belonging to contexts dated to 1890–1750 BC were recorded (Sesma and García, 1994). During the second quarter of the 2nd Millennium BC bronzes arrived at the central and southeastern regions, namely at La Mancha, Levant and the Argaric region (Fernandez-Miranda et al., 1995). From northern Portugal, two metallic prills and a bronze bar suggesting the bronze production at the Bronze Age settlement of Sola (Braga) were also dated to the second quarter of the 2nd Millennium BC (Bettencourt, 2000). Until recently, the single securely dated context with early bronzes in Southwestern Iberia was the funerary hypogeum from Belmeque (Serpa), located in the southern Portuguese territory and dated to 1670–1390 cal BC (Soares, 1994; Alves et al., 2010). Nevertheless, the first evidence of bronze production in the region is attested by a mould for flat axes from Casarão da Mesquita 3 (Évora) exhibiting a later date of 1120–900 cal BC (Soares et al., 2007; Mataloto et al., 2013). Additionally, metallurgical production contexts from Entre Águas 5 (Serpa), dated to the first quarter of the 1st Millennium BC, point to the co-smelting of oxide copper ores with cassiterite (Valério et al., 2013).

The early bronzes in most of Western Europe exhibit variable concentrations of tin, but there was an alloy regularisation during the MBA, suggesting a regular trade of tin to areas without tin ores (Fernandez-Miranda et al., 1995). The diffusion of bronze artefacts was probably much faster than the spread of the technology itself, but in spite of arsenical copper and bronze being certainly distinguishable by colour, some technological expertise would be essential to locally produce the new alloy. Furthermore, the recently arrived bronze artefacts should reveal a clear mechanical or aesthetical benefit in order to instigate the replacement of the long established technology of arsenical coppers. Consequently, the MBA was an important period of slow but crucial metallurgical developments.

Despite the obvious importance of the MBA, there are no relevant studies on the coeval metallurgy in Southwestern Iberia. However, recently discovered MBA burial sites exhibiting numerous metallic artefacts have been providing a significant number of materials for study. Two of these archaeological sites are Torre Velha 3, located in the Portuguese left bank of the Guadiana River, and Monte da Cabida 3, situated not far from the city of Évora. Most of the artefacts are tools/weapons and ornaments from burial monuments, such as hypogea at Torre Velha 3 and cists at Monte da Cabida 3. Other metals belong to non-funerary contexts located in both sites. These distinct archaeological contexts were radiocarbon dated to get a precise and reliable chronology for the artefacts. Afterwards, it was possible to make an elemental and microstructural characterisation of the artefacts, thus providing for the first time an accurate comprehension on the MBA metallurgy in the region. Different alloys could be identified, while the manufacture of several typologies and alloys was compared, thus establishing the use and value of metal to those ancient communities inhabiting the Southwestern Iberia.

2. Archaeological background

2.1. Torre Velha 3 – metals from hypogea and pits

During 2008 the area directly affected by the upcoming construction of the Laje Dam (Serpa), included in an irrigation project connected with the Alqueva Dam, was subject to an archaeological survey (14 ha), followed by archaeological excavations of the structures and contexts that had been identified. Archaeological works exposed tens of negative structures containing artefacts and other materials that cover a large diachrony ranging from the Chalcolithic to the Late Antiquity (Alves et al., 2012). Nevertheless, the Bronze Age is the chronological period better represented at Torre Velha 3 comprising negative structures of different typologies such as hypogea and pits (Fig. 1).

Undoubtedly, a group of 25 hypogea are the most relevant archaeological structures recorded at Torre Velha 3 (Alves et al., 2010). These hypogea are MBA funerary structures (see Fig. 1) composed of an atrium and an underground burial chamber dug in the soil (Alves et al., 2010). At Torre Velha 3 these funerary monuments seem to have been grouped in two clusters plus an isolated hypogeum located approximately 40 m south of the main group. The set of burial offerings varies from one hypogeum to another but is mainly composed of ceramic vessels, metallic artefacts and meat offerings. About half of the hypogea of this necropolis include metals totalling about 20 artefacts. Most of the tombs exhibited an awl, while hypogeum [2215]–[2231] contained a dagger and a ring. Hypogea [1267]–[1792], [1298]–[1695] and [2417]–[2418] exhibited each one an awl and a dagger. These awls are thin bars with a rounded section opposite to a quadrangular section both ending with a sharp edge (Fig. 1). These offerings are typically associated to the gender of the buried individual (Castro-Martínez et al., 2006). Awls and small daggers (knives?) arise in female burials on both Southwestern Iberia (Pavón Soldevila, 2008) and Argaric region (Lull et al., 2011). The dagger from hypogeum [1267]–[1792] is an exceptional specimen since it has two rivets of silver attached to a small blade. Silver was also present among the grave goods from hypogeum [2550]–[2551] comprising a necklace with two silver beads, two copper beads plus a pair of beads made

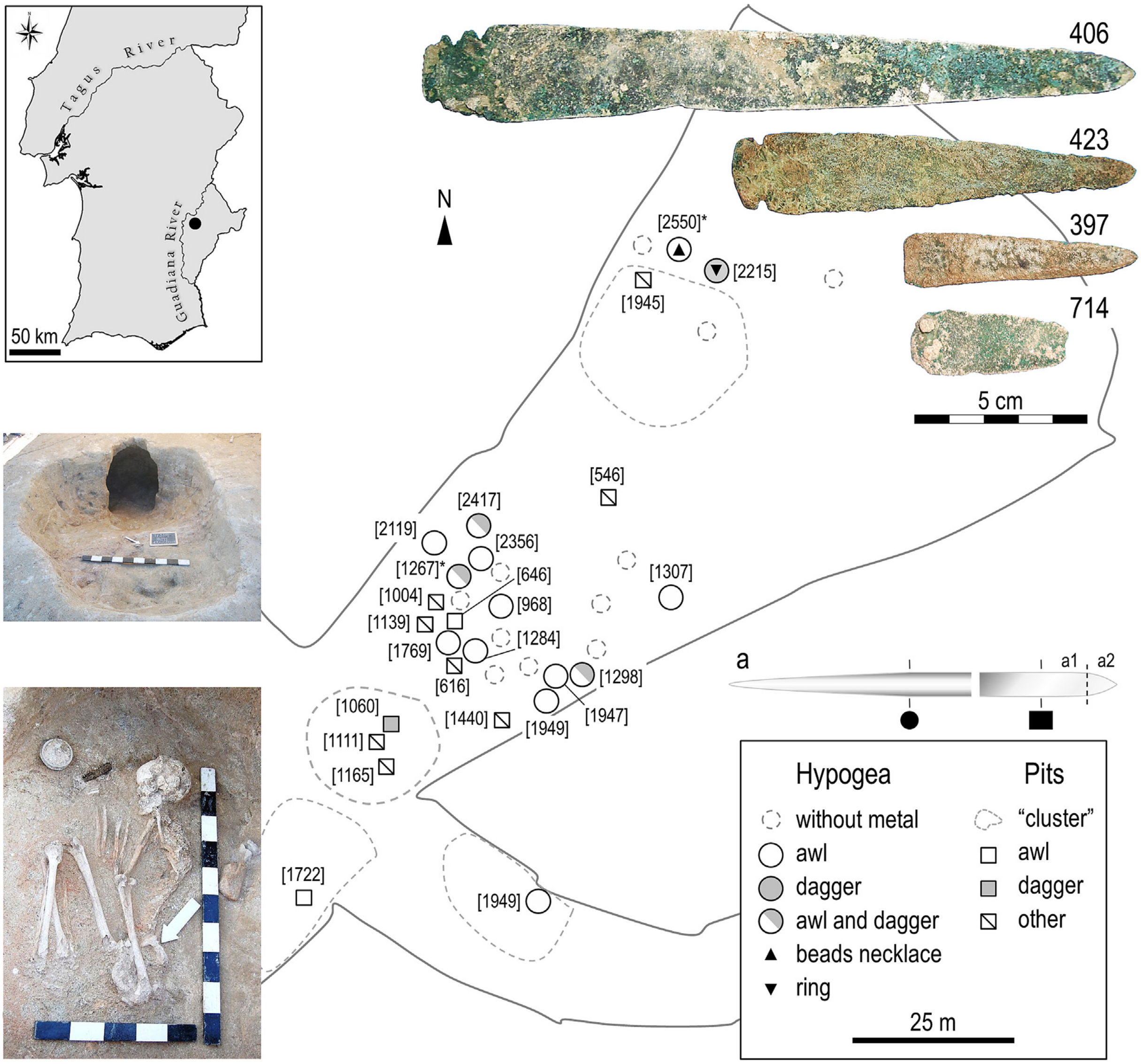

Fig. 1. Metallic artefacts, hypogea (referenced by the atrium number) and pits from Torre Velha 3 (left, top to bottom: site location in southern Portuguese territory, hypogeum [1489]–[1490] and burial chamber [2418] with human skeleton in foetal position plus metallic and ceramic offerings; top right: metallic daggers of variable size; a: awl scheme evidencing a rounded section opposite to the quadrangular section, a1, and ending with a sharp edge, a2).

of bone and a pair made of shells. The wealth of some burial offerings indicates the high social status of those buried individuals.

Hypogea have been considered typical of the Argaric Culture (2250–1450 BC) from Southeastern Iberia (Lull, 2000; Aranda Jiménez et al., 2009) and were practically unknown in the southwestern region of the peninsula. Until recently, only one funerary monument of this type had been identified in the southwestern region, the already mentioned hypogeum from Belmeque (Serpa). This hypogeum contained two inhumations with an unusually rich set of grave goods, including a finely decorated ceramic vessel, a bronze knife with gold rivets, a pair of copper and bronze daggers with silver rivets and several silver nails (Schubart, 1974; Soares, 1994; Soares et al., 2009). More recently, a group of 9 hypogea was discovered at Outeiro Alto 2 (Serpa) presenting a similar material culture composed by ceramic vessels and metallic awls and daggers (Valera and Filipe, 2010). Another set of 3 hypogea was excavated at Horta do Folgão (Serpa) comprising an awl and an exceptional sword (Valério et al., 2012a). Southwestern hypogea have been showing an important difference compared to the Argaric funerary monuments, which is the lower amount and lesser wealth of funerary offerings. Moreover, the Argaric graves are located underneath the houses, while in the Southwest they are located outside the houses and in the vicinity of the domestic space.

Pits are negative structures with a conical or cylindrical shape, usually with a domestic character and probably dug out for the storage of supplies or other materials. Some of the storage pits were later reused for burials although without any grave goods: of about 80 pits excavated in this archaeological site, 7 contained one or more human skeletons. Similar examples were found for instance at Casarão da Mesquita 3 (Évora) and Horta do Albardão 3 (Évora), whose inhumations have been dated to the 3rd quarter of the 2nd Millennium BC (MBA) or to the last quarter of the 2nd Millennium/1st quarter of the 1st Millennium BC (LBA) (Santos et al., 2008, 2009; Soares et al., 2009; Mataloto et al., 2013). Some of the pits without inhumations at Torre Velha 3 had indeed metallic artefacts. A total of 11 metals such as awls, chisels, a dagger and a saw were recovered in these archaeological contexts. Other pits also contained metals but clearly belonging to other chronological periods, such as a double-spring fibula ascribed to the Iron Age.

2.2. Monte da Cabida 3 – metals from cists and pits

The archaeological survey to minimize the impact from the construction of a road access to the irrigation block of Monte Novo, also connected with the Alqueva Dam project, led to the identification of the archaeological site of Monte da Cabida 3 (Évora). During 2007 archaeological works covered an area of about 0.2 ha and 64 negative structures were excavated including funerary monuments and pits (Antunes et al., 2012).

The more noteworthy funerary monuments of this site are cists, typical structures of the MBA in Southwestern Iberia. These funerary monuments consist of a rectangular burial chamber, usually bounded by 4 slabs forming the walls and a large one as the lid (Fig. 2). Four of the cists at Monte da Cabida 3 had metallic grave goods – awls (cists 9, 10 and 12) and a dagger (cist 8) – sometimes together with ceramic vessels. In the southern Portuguese region there are many parallels for this sort of funerary monuments, namely at Herdade do Montinho (Serpa), Carapetal (Vila Nova de São Bento), Santa Justa (Serpa), Barranco do Salto (Vila Verde de Ficalho), Bugalhos (Serpa), Talho do Chaparrinho (Vila Verde de Ficalho) and Carapinhais (Sobral da Adiça) (Soares, 1994; Soares et al., 2007).

Some of the pits from Monte da Cabida 3 were also used for Bronze Age inhumations, although, similarly to Torre Velha 3, without any grave goods. This distinction could point to the burial of individuals of low social status. The prehistoric metallic artefacts recovered outside the cists at Monte da Cabida 3 were two small shapeless fragments found inside two pits without inhumations.

2.3. Chronology of hypogea, cists and pits

Several radiocarbon dates were obtained with bone samples collected from significant archaeological contexts at Torre Velha 3 and Monte da Cabida 3 (Table 1). Bone samples belong to human inhumations, meat offerings or kitchen refuse recovered inside hypogea, cists or pits. The collection of dates indicates an occupation period between *c.* 1900 and 1300 cal BC. Additionally, the results show that all these different types of funerary monuments and domestic pits are broadly contemporary. There is also no chronological distinction among burials with and without metallic offerings, whose difference is probably mostly related with the social status of the buried individual. Finally, it should be emphasized the apparently identical chronology of the two burials with silver offerings, namely hypogeum [1267]–[1792] (1680–1450 cal BC) and hypogeum [2550]–[2551] (1670–1410 cal BC) despite their location in different areas of the necropolis.

3. Methodology

Preparation of artefacts for microanalyses consisted on removing the corrosion layer. A small area (∅ ~ 3–5 mm) on the surface of the artefact was polished with diamond pastes of progressively finer grit size (15 μm–1 μm). The area was observed with an optical microscope to confirm the achievement of a clean metal surface. In specific cases it was possible to cut a small section of the artefact, which was mounted in epoxide resin and polished with silicon carbide papers (1000, 2500 and 4000 grit size) and diamond pastes (3 μm and 1 μm).

Elemental characterization involved a micro-EDXRF ArtTAX Pro spectrometer with a 30 W Mo X-ray tube and an electrothermally cooled silicon drift detector (FWHM of 160 eV at 5.9 keV). Focussing polycapillary lens enables the analysis of a very small area (∅ < 100 μm). Three areas were analysed in each artefact using a tube voltage of 40 kV and a current intensity of 600 μA. Quantifications were made with the WinAxil software involving calibration factors calculated with reference materials. Phosphor Bronze 551 (British Chemical Standards) and IDLF5 (Industries de la Fonderie) were utilised for copper and bronze. Calibration for brass employed the SRM1103 (National Bureau of Standards), while for silver included silver-copper and silver-gold standard alloys (Araújo et al., 1993). Accuracy was assessed with the analysis of reference materials, namely Phosphor Bronze 552 and Leaded Gunmetal 183/3 (British Chemical Standards). Relative errors are less than 10% for alloying elements and less than 15% for minor elements. Quantification limits are 0.60 wt.% Ag, 0.50 wt.% Sn, 0.50 wt.% Sb, 0.10 wt.% Zn, 0.10 wt.% As, 0.10 wt.% Pb and 0.05 wt.% Fe.

Microstructural characterisation was made with a Leica DMI 5000M optical microscope using bright field, dark field and polarised light. An aqueous ferric chloride solution was used for etching. Representative samples were characterised by SEM–EDS using a conventional W filament scanning electron microscope (Zeiss DSM 962) with secondary electron and backscattered electron imaging modes. An Oxford Instruments INCAx-sight EDS spectrometer with an ultra-thin window allows the detection of low atomic number elements. Carbon conductive bridge was used to prevent charge accumulation. Analyses were made with a working distance of 25 mm, 20 kV of accelerating voltage, ~3 A of filament current and 70 μA of emission current.

Factor analysis was made with Statistica v.11 software package involving significant microstructural and compositional variables, namely the size of inclusions, deformation of inclusions, segregation bands, grain size, slip bands density and arsenic content.

Vickers microhardness testing involved a Zwick–Roell Indentec apparatus with 0.20 kgf load for 10 s. The average hardness comprises at least 3 indentation measures having a relative standard deviation less than 10%.

After analysis, the exposed artefact area was protected with a corrosion inhibitor – benzotriazol dissolved in ethanol – and coated with an acrylic layer of Paraloid B-72 dissolved in acetone. The coloration of surrounding patina was replicated with a mixture of pigments dissolved in an acrylic solution.

Fig. 2. Monte da Cabida 3: site location in southern Portuguese territory; cist 1 with human remains and ceramic offerings; dagger M4 with four rivets and vestiges of wooden handle (mineralized remains) and awl M1 with fitting to attach a wooden or bone handle.

Table 1
Radiocarbon dates of bone samples from Torre Velha 3 (TV3) and Monte da Cabida 3 (MCab3) (C: copper; B: bronze; S: silver; calendar ages using IntCal09, Reimer et al., 2009; OxCal 4.1.7, Bronk Ramsey, 2009).

Site	Reference	Structure	Metallic offering	Sample type	δ^{13}C (‰)	^{14}C Age (BP)	Calendar age (cal BC)	
							1σ	2σ
TV3	Sac-2825	Hyp. [1267]–[1792]	C + B + S	Human bone	−20.3	3280 ± 50	1610–1500	1680–1450
	Sac-2489	Hyp. [1489]–[1490]	None	*Bos* (radius)	−22.3	3300 ± 45	1630–1510	1690–1450
	Beta-262199	Hyp. [1662]–[1664]	None	*Ovis* (radius)	−20.5	3300 ± 40	1630–1520	1690–1490
	Sac-2490	Hyp. [1949]–[1950]	C	*Bos* (radius + ulna)	−21.5	3410 ± 60	1870–1620	1890–1530
	Sac-2465	Hyp. [2119]–[2120]	None	*Bos* (radius + ulna)	−22.0	3300 ± 50	1640–1510	1730–1450
	Sac-2827	Hyp. [2356]–[2357]	C + B	Human bone	−20.3	3340 ± 80	1700–1520	1780–1440
	Sac-2826	Hyp. [2417]–[2418]	C + B	Human bone	−20.5	3170 ± 90	1530–1320	1670–1250
	Sac-2480	Hyp. [2498]–[2497]	None	*Bos* (radius)	−21.5	3410 ± 60	1870–1620	1890–1530
	Sac-2466	Hyp. [2550]–[2551]	C + S	*Bos* (radius)	−21.8	3250 ± 60	1610–1450	1670–1410
	Sac-2882	Pit [969]	–	Human bone	−21.0	3330 ± 50	1680–1530	1740–1500
	Sac-2883	Pit [1991]	–	Human bone	−20.6	3290 ± 50	1620–1510	1690–1450
MCab3	Sac-2631	Cist 9/904	C	Human bone	−18.9	3290 ± 60	1630–1500	1690–1440
	Sac-2368	Pit 4/888	–	Bones (*Sus* sp.)	−16.1	3220 ± 90	1610–1420	1690–1300
	Sac-2888	Pit 41/931	–	Human bone	−19.4	3490 ± 50	1880–1750	1940–1690
	Sac-2437	Pit 64/960	–	Human bone	−20.5	3330 ± 45	1670–1530	1690–1510
	Sac-2326	Pit 51/883	–	Animal bones	−19.6	3410 ± 45	1760–1640	1830–1610
	Sac-2321	Pit 51/966	–	Animal bones	−21.0	3440 ± 50	1870–1690	1890–1630

4. Results

4.1. Elemental composition

4.1.1. MBA artefacts

The elemental composition of metals from Torre Velha 3 was determined by micro-EDXRF (Table 2). The collection of 33 metals (31 artefacts and 2 rivets from dagger 406) is mostly composed by arsenical coppers (82%: $n = 27$), whereas bronze and copper artefacts show a low incidence (15%: $n = 5$ and 3%: $n = 1$, respectively). However, one of the most outstanding results of this research was the identification of bronzes in this MBA set. The four bronze alloys recovered inside hypogea have an average value of tin close to 10 wt.% (9.6 ± 1.2 wt.% Sn) and very low iron contents (<0.05 wt.%). The "awl" 393 recovered inside a small pit is composed of a bronze alloy comparable to other bronzes.

Table 2
– Composition of MBA copper-based artefacts of hypogea and pits from Torre Velha 3 (length of awls and daggers in cm; content in wt.% as average ± standard deviation; n.d. = not detected).

Hypogeum [atrium]–[chamber]	Artefact (length)	Reference	Cu	As	Sn	Ag	Fe
[968]	Awl	395	88.5 ± 0.6	<0.10	11.4 ± 0.6	n.d.	<0.05
[1267]–[1792]	Awl (14.0)	410	95.2 ± 0.2	4.74 ± 0.18	n.d.	n.d.	<0.05
	Dagger	714	90.8 ± 0.4	0.12 ± 0.01	9.03 ± 0.32	n.d.	<0.05
[1284]–[1415]	Awl (4.8)	401	95.9 ± 0.1	4.03 ± 0.19	n.d.	n.d.	<0.05
[1298]–[1695]	Awl (8.9)	405	96.7 ± 0.7	3.24 ± 0.70	n.d.	n.d.	<0.05
	Dagger (20.7)	406	95.1 ± 0.4	4.82 ± 0.42	n.d.	n.d.	<0.05
	Rivet	406a	94.5 ± 0.3	5.45 ± 0.24	n.d.	n.d.	<0.05
	Rivet	406b	95.1 ± 0.2	4.81 ± 0.15	n.d.	n.d.	<0.05
[1307]–[1370]	Awl	403	97.5 ± 0.2	2.49 ± 0.21	n.d.	n.d.	<0.05
[1769]–[1770]	Awl (5.5)	408	94.3 ± 0.5	5.70 ± 0.51	n.d.	n.d.	<0.05
[1947]–[1948]	Awl (9.3)	411	95.5 ± 0.2	4.45 ± 0.22	n.d.	n.d.	<0.05
[1949]–[1950]	Awl (7.0)	412	91.4 ± 0.5	4.09 ± 0.35	n.d.	4.65 ± 0.21	<0.05
[2119]–[2120]	Awl (6.4)	419	95.5 ± 0.2	4.46 ± 0.21	n.d.	n.d.	<0.05
[2215]–[2231]	Dagger (11.2)	428	97.7 ± 0.1	2.33 ± 0.08	n.d.	n.d.	<0.05
	Ring	415	86.7 ± 0.4	13.3 ± 0.4	n.d.	n.d.	<0.05
[2236]–[2237]	Awl (5.89)	421	95.6 ± 0.1	4.38 ± 0.15	n.d.	n.d.	<0.05
[2356]–[2357]	Awl	418	90.1 ± 0.6	0.64 ± 0.03	9.23 ± 0.51	n.d.	<0.05
[2417]–[2418]	Awl	422	89.1 ± 0.1	2.17 ± 0.03	8.73 ± 0.06	n.d.	<0.05
	Dagger (11.7)	423	96.3 ± 0.6	3.87 ± 0.21	n.d.	n.d.	<0.05
[2550]–[2551]	Awl (4.4)	416	95.0 ± 0.1	4.91 ± 0.21	n.d.	n.d.	<0.05
	Bead	911	97.3 ± 0.1	2.60 ± 0.11	n.d.	n.d.	<0.05
	Bead	912	97.4 ± 0.2	2.57 ± 0.16	n.d.	n.d.	<0.05
Pit	**Artefact (length)**	**Reference**	**Cu**	**As**	**Sn**	**Ag**	**Fe**
[546]	Palmela point	392	95.7 ± 0.2	4.27 ± 0.18	n.d.	n.d.	<0.05
[616]	Rivet	391	94.6 ± 0.8	5.37 ± 0.74	n.d.	n.d.	<0.05
[646]	Awl (5.8)	390	95.6 ± 0.2	4.34 ± 0.21	n.d.	n.d.	<0.05
[1004]	Chisel	398	95.8 ± 0.1	4.15 ± 0.09	n.d.	n.d.	<0.05
[1060]	Dagger (6.7)	397	94.6 ± 0.1	5.31 ± 0.11	n.d.	n.d.	<0.05
[1111]	Fragment	429	97.4 ± 0.8	2.57 ± 0.76	n.d.	n.d.	<0.05
[1139]	"Awl"	393	90.0 ± 0.2	n.d.	10.0 ± 0.2	n.d.	<0.05
[1165]	Fragment	400	98.5 ± 0.1	1.46 ± 0.04	n.d.	n.d.	<0.05
[1440]	Chisel	615	95.3 ± 0.2	4.63 ± 0.24	n.d.	n.d.	<0.05
[1722]	Awl	407	95.3 ± 0.1	4.67 ± 0.16	n.d.	n.d.	<0.05
[1945]	Saw	388	97.4 ± 0.3	2.58 ± 0.27	n.d.	n.d.	<0.05

Artefacts from cists and pits at Monte da Cabida 3 were also analysed by micro-EDXRF to establish their elemental composition (Table 3). The set of 7 artefacts is mainly composed of arsenical copper alloys, plus a copper fragment with low arsenic content (0.94 wt.%).

Another significant finding from Torre Velha 3 was the existence of a few burial offerings and ornaments made of silver. Micro-EDXRF analysis of the bead 913 identified a silver with a high copper content, 4.78 wt.% (Table 4). Silver rivets of dagger 714 present a high copper content (4.3–5.1 wt.% Cu), but also significant amounts of gold, namely 4.9–5.4 wt.% Au (Table 4).

4.1.2. Non-prehistoric artefacts

A small group of other copper-based artefacts recovered from pits at Torre Velha 3, with chronologies between the Iron Age and the Late Antiquity, display quite different elemental compositions (Table 5). This set of 9 artefacts comprise copper (396 and 414), bronze (402, 404, 409 and 425), tin-antimony bronze (233), brass (424) and leaded brass (413).

4.2. Manufacture

4.2.1. MBA metalworking

Optical microscopy observations enabled the identification of microstructural characteristics of arsenical coppers from Torre Velha 3 and Monte da Cabida 3. Almost all artefacts have similar features such as deformed grains, annealing twins and slip bands (Fig. 3). The microstructure of awl 405 deviates from the other microstructures since it exhibits a residual dendritic segregation enveloping very small sized grains.

Moreover, some artefacts such as the saw 388 have a significant density of oxide inclusions due to oxidising conditions during melting. SEM–EDS analyses show that these inclusions are mixed Cu–As oxides of variable composition (Fig. 4). The arsenic retained in these inclusions reduces the arsenic content of the α-phase solid solution, thus decreasing its contribution to the mechanical properties of the alloy. Additionally, the arsenic-rich phase (Cu_3As) is present in several artefacts, being identified by SEM–EDS analyses on awls M1 and M2 (Fig. 4). Initially, the Cu_3As results of inverse segregation during cooling of copper alloys with high amounts of arsenic. Afterwards, the long-term segregation over archaeological time would deposit the As-rich phase on the grain boundaries, as proposed by Budd and Ottaway (1995).

Factor analysis involving the size and deformation of Cu–As–O inclusions, incidence of segregation bands, grain size and density of slip bands plus the content of arsenic was made to provide an overall assessment about the manufacture of the set of artefacts (Table 6). The arsenic content was later discarded since it does not statistically correlate with the remaining variables. The two factors extracted account for 64% of total variance. Factor loadings show that factor 1 is related with higher deformation of inclusions, decreasing grain size and increasing density of slip bands, while factor 2 can be associated with the decreasing size of oxidic inclusions and homogenisation of segregation bands.

Optical microscopy observations of bronzes mostly show microstructures with deformed grains with annealing twins and slip bands (Fig. 5). The dagger 714 exhibits a cored dendritic structure with minute grains and slip bands, suggesting limited thermomechanical work. Some alloys exhibit Cu–S inclusions among the α-phase solid solution, which were identified as Cu_2S by SEM–EDS analyses (Fig. 4). Cu_2S inclusions are often elongated evidencing the main direction of deformation of the artefact (e.g. awl 395, Fig. 5).

Table 3
– Composition of MBA copper-based artefacts of cists and pits from Monte da Cabida 3 (length of awls in cm; content in wt.% as average ± standard deviation).

Cist	Artefact (lenght)	Reference	Cu	As	Fe
10	Awl (5.2)	M1	97.1 ± 0.5	2.81 ± 0.45	<0.05
12	Awl (4.2)	M2	94.1 ± 0.4	5.88 ± 0.48	<0.05
9	Awl (4.6)	M3	94.6 ± 0.4	5.37 ± 0.38	<0.05
8	Dagger	M4	95.4 ± 0.6	4.59 ± 0.63	<0.05
	Rivet	M4a	73.5 ± 1.3	26.5 ± 1.3	<0.05
Pit	**Artefact**	**Reference**	**Cu**	**As**	**Fe**
16	Fragment	M8	97.4 ± 0.3	2.58 ± 0.27	<0.05
17	Fragment	M9	99.0 ± 0.2	0.94 ± 0.21	<0.05

Table 4
– Composition of MBA silver artefacts of hypogea from Torre Velha 3 (content in wt.% as average ± standard deviation; n.d. = not detected).

Hypogeum [atrium]–[chamber]	Artefact	Reference	Ag	Au	Cu
[1267]–[1792]	Rivet	714a	90.3 ± 0.3	5.40 ± 0.06	4.26 ± 0.34
	Rivet	714b	89.9 ± 0.2	4.94 ± 0.02	5.09 ± 0.16
[2550]–[2551]	Bead	913	95.2 ± 0.8	n.d.	4.78 ± 0.78

4.2.2. Non-prehistoric bronze metalworking

Among the non-prehistoric metals recovered at Torre Velha 3 there are some bronzes belonging to the Iron Age, namely a double-spring fibula (404) and two "blade" fragments (402 and 425). These bronze alloys were subjected to microstructural characterisation to identify possible differences with MBA bronzes. Iron Age bronzes present microstructures composed of heavily deformed grains with annealing twins and a high density of slip bands (Fig. 5).

4.3. Hardness

Mounted sections of arsenical copper and bronze artefacts were subjected to Vickers microhardness testing to determine the hardness (Table 7). Arsenical coppers belonging to the MBA have a wide range of hardnesses (73–144 HV0.2). Similarly, MBA bronzes show variable values (110–221 HV0.2), while bronzes belonging to the Iron Age have a somewhat higher hardness (171–235 HV0.2).

Microhardness profiles were obtained along the cross sections of a saw and a blade (Fig. 6). The saw 388 shows a central zone with uniform thickness (550 ± 30 μm) and low hardness (107 ± 2 HV0.2), while only the tooth was highly narrowed and hardened (143 ± 6 HV0.2). On the contrary, the blade from dagger 397 is much tougher (205 ± 4 HV0.2) and was gradually deformed thus resulting on an increasingly higher hardness from the middle to the cutting edge of the blade (up to 231 HV0.2).

5. Discussion

5.1. MBA common metallurgy

The large majority of MBA artefacts from Torre Velha 3 are arsenical coppers, thus showing a strong influence of the metallurgy inherited from the Chalcolithic Period. Metals from hypogea and domestic pits have an analogous and strong incidence of this alloy (about 80%). Moreover, excluding the unusually arsenic-rich ring 415, arsenical coppers from hypogea and pits also have similar arsenic contents (4.1 ± 1.1 wt.% As, $n = 17$ and 4.2 ± 1.0 wt.% As, $n = 9$, respectively). The technological resemblance suggests that burial offerings should have been everyday life tools or personal ornaments that were deposited in the graves. This implies that the manufacture of common artefacts and burial offerings should be the same. Nevertheless, it must be noted that some typologies, such as the saws, only appear among the world of the

Table 5
– Composition of non-prehistoric copper-based artefacts from Torre Velha 3 (content in wt.% as average ± standard deviation; n.d. = not detected).

Structure	Artefact	Reference	Cu	Sn	Zn	Pb	Sb	As	Fe
[56]	Fibula	233	88.9 ± 0.1	5.00 ± 0.20	n.d.	1.55 ± 0.23	2.66 ± 0.05	1.91 ± 0.04	<0.05
[799]	Needle	396	97.0 ± 0.1	1.64 ± 0.16	n.d.	n.d.	n.d.	0.50 ± 0.02	0.49 ± 0.03
[1344]	"Blade"	402	90.5 ± 0.2	8.77 ± 0.15	n.d.	0.52 ± 0.02	n.d.	n.d.	0.17 ± 0.01
[1377]	Fibula	404	88.3 ± 0.4	11.2 ± 0.3	n.d.	n.d.	n.d.	0.23 ± 0.01	0.32 ± 0.02
[1638]	Pin	409	95.8 ± 0.5	2.40 ± 0.39	1.3 ± 0.1	0.44 ± 0.09	n.d.	<0.10	0.12 ± 0.01
[1851]	Earring	413	79.6 ± 0.4	1.85 ± 0.10	15.2 ± 0.1	2.83 ± 0.34	n.d.	n.d.	0.27 ± 0.01
[1905]	Nail	414	99.8 ± 0.1	n.d.	n.d.	n.d.	n.d.	0.11 ± 0.01	<0.05
[2263]	Fragment	424	80.8 ± 0.1	n.d.	18.3 ± 0.1	0.37 ± 0.05	n.d.	n.d.	0.18 ± 0.01
[2277]	"Blade"	425	94.1 ± 0.2	5.51 ± 0.10	n.d.	0.11 ± 0.02	n.d.	<0.10	0.10 ± 0.01

living. Furthermore, there is neither a noticeable distinction between different typologies nor between different sizes. For instance, awls and daggers present similar alloy content (4.3 ± 0.8 wt.% As, $n = 12$ and 4.1 ± 1.3 wt.% As, $n = 4$, respectively) and there is no correlation between length and content of arsenic concerning awls or daggers.

The arsenical copper metallurgy is also preponderant at Monte da Cabida 3. The average arsenic content of these arsenical coppers (4.2 ± 1.5 wt.%, $n = 5$, excluding the arsenic-rich rivet M4a) is comparable to the value on Torre Velha 3 (4.1 ± 1.0 wt.% As, $n = 26$, excluding the arsenic-rich ring 415). The similarity indicates that those coeval MBA communities with distinct burial practices had the same metallurgy of arsenical coppers. Moreover, the high levels of arsenic of MBA artefacts from Torre Velha 3 and Monte da Cabida 3 show a type of metallurgy where the use of scrap was not entirely significant.

Chalcolithic collections from the Portuguese territory show a somewhat different trend. The study of metals from the Chalcolithic settlement of Leceia (Oeiras) indicated that only a minor fraction of the artefacts (38% of 32 artefacts) shows higher arsenical copper, which was reserved for long awls, saws, Palmela points and tanged daggers (Müller and Cardoso, 2008). The occurrence of arsenical copper is also small (37% of 53 artefacts) among the metals from the Chalcolithic settlement of Castro de Vila Nova de São Pedro (Azambuja), but a statistically significant association was found between this alloy and tools/weapons, namely arrowheads, daggers and knives (Pereira et al., 2013).

Generally, there seems to be a chronological trend towards the use of higher contents of arsenic on MBA metals from this region. Certain artefacts can provide some clues about the possible reasons for this chronological evolution. The ring 415 from Torre Velha 3 has a considerable higher content of arsenic (c. 13 wt.% As), i.e. an increased presence of the arsenic-rich phase γ (Cu_3As) producing a silver colour that would be highly valued on prestigious ornaments. Similarly, a layer of Cu_3As covers the rivet M4a (c. 26 wt.% As) of dagger M4 from Monte da Cabida 3 giving it a silvery sheen. Dagger M4 should be an attempt to produce an artefact with a high-status using an arsenical copper blade with four silvery colour rivets. Furthermore, this dagger is possibly a local reproduction of other highly prestigious artefacts such as dagger 714 from Torre Velha 3 displaying a handle with two silver rivets and a bronze blade of similar dimension.

5.2. MBA innovative metallurgy

The new technology of bronze is already present among metals from Torre Velha 3 despite the still more common metallurgy of arsenical coppers. The absence of bronze artefacts at Monte da Cabida 3 was predictable considering the low number of metals and

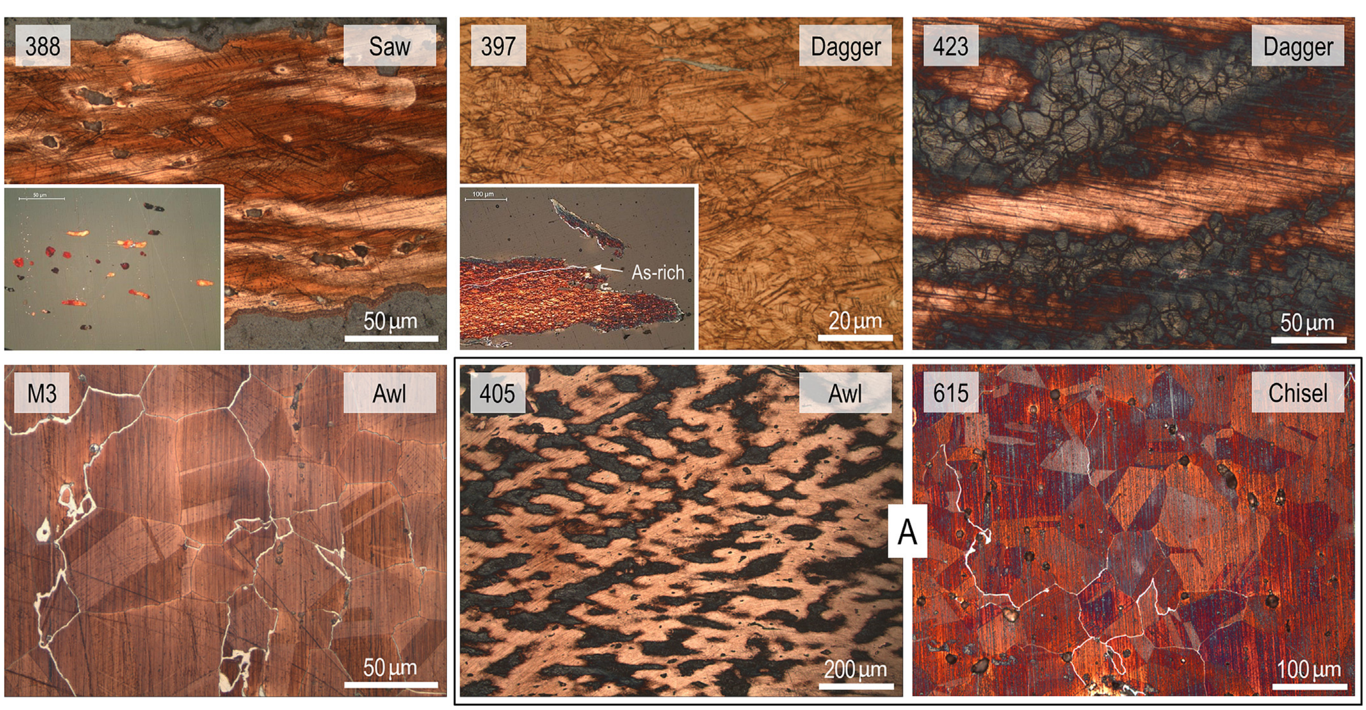

Fig. 3. Microstructures of MBA arsenical copper artefacts from Torre Velha 3 and Monte da Cabida 3 (A: unusual microstructures).

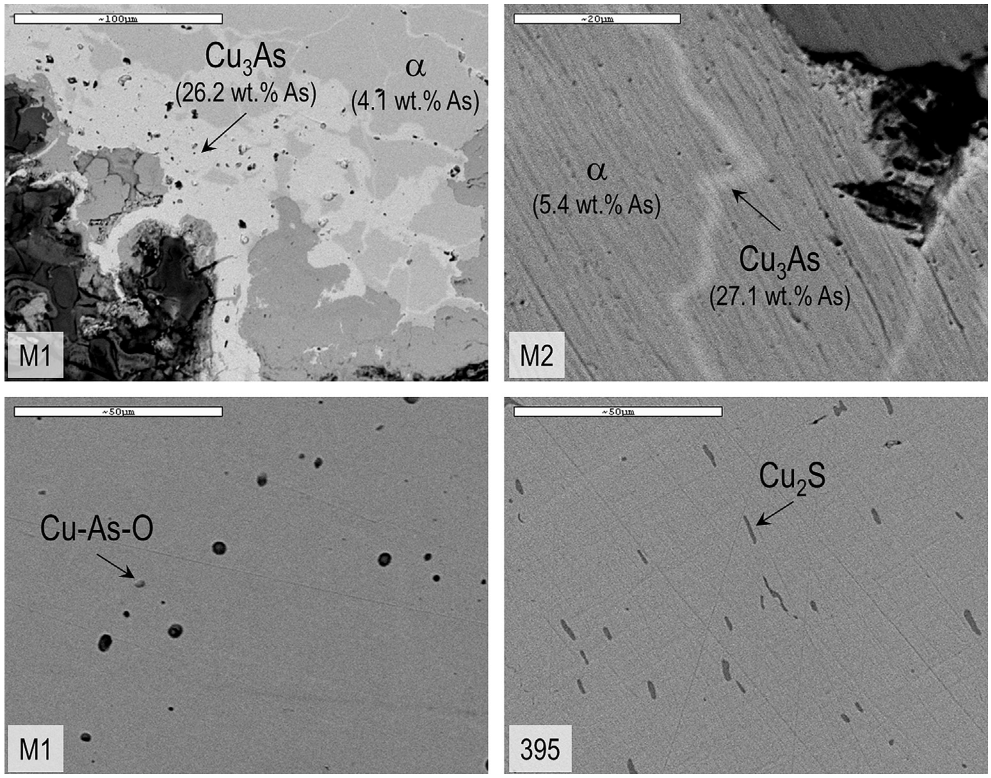

Fig. 4. SEM–BSE images and SEM–EDS spot quantifications of arsenical copper awls M1 and M2 from Monte da Cabida 3 and bronze awl 395 from Torre Velha 3.

Table 6
Factor loadings of principal components from microstructural data of MBA artefacts from Torre Velha 3 and Monte da Cabida 3 (5 variables; 30 samples; varimax normalised; bold: factor loadings with absolute value higher than 0.7).

Variable	Factor 1	Factor 2
Size of Cu–As–O inclusions	0.118	**−0.792**
Deformation of Cu–As–O inclusions	**0.734**	−0.069
Segregation bands	−0.023	**−0.770**
Grain size	**−0.763**	0.377
Density of slip bands	**0.719**	0.411
Explained variance	33.0%	30.7%

low incidence of bronzes during this chronological period, as inferred from coeval collections from Torre Velha 3 and other Iberian regions.

Most of the bronze artefacts from Torre Velha 3 belong to clearly closed contexts, the exception being the awl 395 that probably belongs to a hypogeum that was later disturbed by the construction of a pit. Funerary contexts of dagger 714 and awls 418 and 422 were radiocarbon dated: hypogeum [1267]–[1792] 1680–1450 cal BC, hypogeum [2356]–[2357] 1780–1440 cal BC and hypogeum [2417]–[2418] 1670–1250 cal BC, respectively (Table 1 and Fig. 7). As mentioned before the rich hypogeum of Belmeque was also radiocarbon dated: ICEN-142 3230 ± 60 BP (1670–1390 cal BC). These four archaeological contexts with bronze artefacts are the oldest in Southwestern Iberia that have been dated. Results point out that during the second quarter of the 2nd Millennium BC this new alloy made its appearance in this western end of the Iberian Peninsula. It seems, therefore, that the chronology concerning the introduction of bronze in Southwestern Iberia cannot be differentiated from the same event in the central, northwestern and southeastern regions, although being slightly later than its arrival at Northeastern Iberia.

However, unlike what happens in the aforementioned regions where early bronzes present highly variable tin contents, alloys from Torre Velha 3 present a suitable and almost constant content of tin (c. 10 wt.%). This suggests that bronzes from Torre Velha 3 might be importations of a region with an already well-established bronze technology. A good example of bronzes obtained during early metallurgical trials is a collection of Argaric metals exhibiting irregular amounts of tin, namely 15 poor tin alloys ($2 < Sn < 8$ wt.%), 7 regular bronzes ($8 < Sn < 12$ wt.%) and 4 high tin alloys ($Sn > 12$ wt.%) (Rovira, 2004). On the contrary, a collection of Bujões/Barcelos plain axes from central and northern Portugal and typologically attributed to the MBA (although without any absolute chronology) shows bronze alloys with tin contents (10.0 ± 1.6 wt.% Sn, $n = 10$, Figueiredo et al., 2012) very similar to the bronzes from our necropolis. These MBA bronze alloys have tin contents of typical bronzes from the LBA (~1200–800 BC) in the Portuguese territory (Figueiredo et al., 2010; Valério et al., 2013).

The bronze indigenous tradition was so well adapted to local characteristics and demands that it extended to the Iron Age, i.e. until the Post-Orientalizing Period, at least inland of Southwestern Iberia (Valério et al., 2010). Another indication of this indigenous technology is the very low iron content of copper-based artefacts (<0.05 wt.% Fe). The low iron amount of pre-Phoenician metals has been used as an indicator of smelting with a slightly reducing atmosphere, which inhibits the incorporation of iron in the metallic bath (Craddock and Meeks, 1987). The raw copper obtained in true smelting furnaces can be easily purified to iron contents of about 0.5 wt.% but higher reduction is difficult, and needless, since it does not conduct to noticeable improvements to mechanical properties (Northover, 2004).

Despite the advanced composition of those bronze burial offerings, one of the awls from the necropolis of Torre Velha 3 has also a high content of arsenic (422: 2.17 wt.%). Similarly, one of the

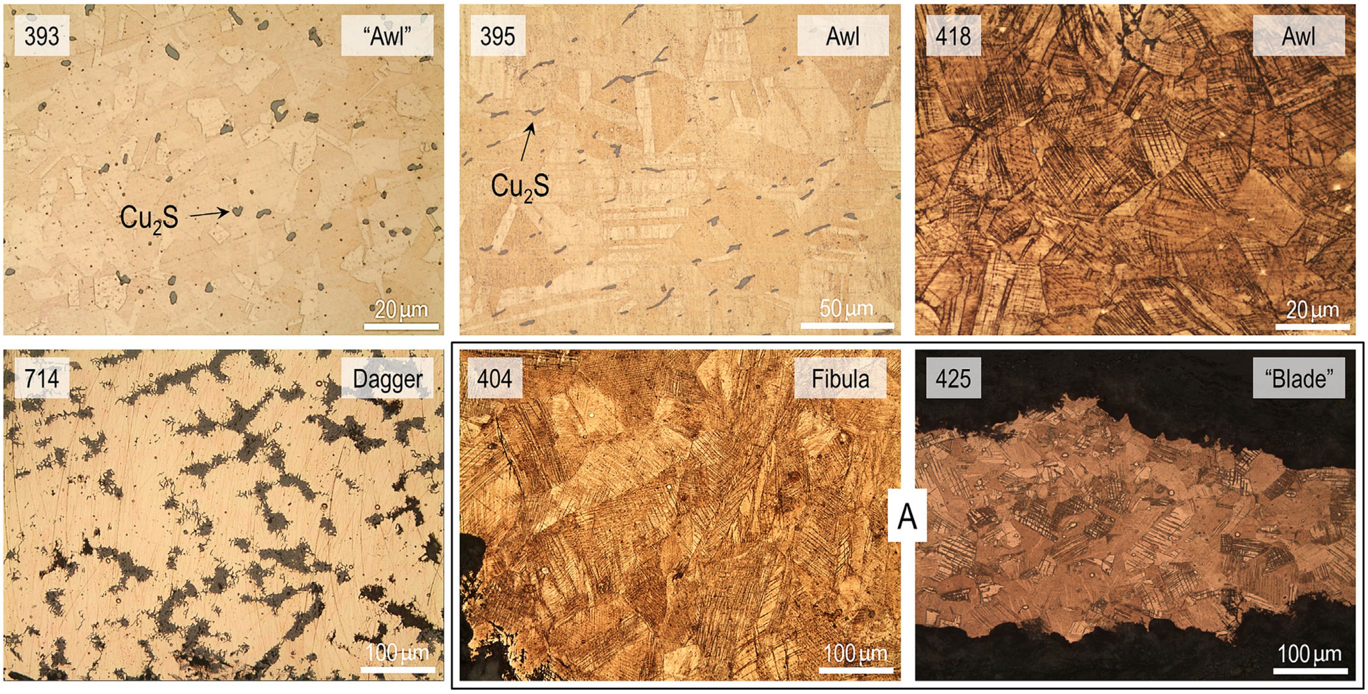

Fig. 5. Microstructures of MBA bronze artefacts from Torre Velha 3 (A: microstructures of Iron Age artefacts).

Bujões/Barcelos axes has also a high content of this element (2.2 wt.% As, Figueiredo et al., 2012) and these alloys are not entirely uncommon among the immense collection of bronzes of the Portuguese territory analysed by the SAM project (Junghans et al., 1968, 1974). Most of these arsenic-rich bronzes are plain axes that typologically belong to the Bronze Age, but the absence of secure archaeological contexts and radiocarbon dates hinders a finer chronology. Arsenic-rich bronzes are also present in the Argaric metallurgy, being attributed to the smelting of arsenical copper ores with cassiterite (Rovira, 2002). Ores recovered in Chalcolithic and MBA mines and settlements of Southwestern Iberia show the use of secondary copper minerals (e.g. cuprite, malachite and azurite) with varying contents of arsenic (Hunt Ortiz, 2003). In some instances, arsenic was found to be present as copper arsenides (algodonite, Cu_6As or domeykite, Cu_3As), such as the 3rd Millennium settlements from the neighbouring regions of Valencina de la Concepción, Sevilla (Nocete et al., 2008) and Almizaraque, Almería (Müller et al., 2004). The smelting of arsenic-rich copper ores was also established in Chalcolithic settlements in the Portuguese Estremadura, namely, Leceia (Müller and Cardoso, 2008) and Zambujal (Müller et al., 2007). The Cu–Sn–As "alloy" is not exclusive of Iberia as it appears, for instance, in the MBA metallurgy of the Levant and Mesopotamia (De Ryck et al., 2005), even though copper arsenide ores are uncommon in the region (Rosenfeld et al., 1997). The use of arsenical copper scrap as a source of copper also results in arsenic-rich bronzes. The long-term depletion of copper-arsenide deposits and/or an adaptation to the preferential use of copper oxides and carbonates, together with the arsenic losses by oxidation and evaporation of As_2O_3 fumes during recycling, would cause the overall reduction of the content of this element in bronze alloys from later periods.

Table 7
– Hardness of copper-based artefacts from Torre Velha 3 (average ± standard deviation).

Alloy	Artefact	Reference	HV0.2
Arsenical copper (MBA)	Awl	407	125 ± 5
	Chisel	615	73 ± 5
	Rivet	391	144 ± 11
	Fragment	400	97 ± 5
	Fragment	429	115 ± 8
	Fragment	M8	109 ± 4
	Fragment	M9	88 ± 5
Bronze (MBA)	"Awl"	393	117 ± 2
	Awl	395	110 ± 2
	Awl	418	221 ± 2
	Awl	422	154 ± 3
Bronze (Iron Age)	"Blade"	402	181 ± 4
	"Blade"	425	171 ± 4
	Fibula	404	235 ± 3

Silver artefacts constitute another metallurgical innovation present at Torre Velha 3. Actually, silver was one of the MBA metallurgical novelties in Southwestern Iberia, being recovered exclusively in burial contexts (Hunt Ortiz, 2003). The high copper content of bead 913 (4.78 wt.%) could point to a deliberate alloy. However, a recent study on metals of Southwestern Iberia suggested that MBA silver-copper alloys have resulted of the use of mixed silver-copper ores or silver and copper mined from the same ore deposit (Bartelheim et al., 2012). The same geologic provenance was established by the similar Pb isotopic ratios of pure silver and silver–copper artefacts. Copper oxide and carbonate ores with significant amounts of silver are documented in the Iberian pyrite belt (Rovira and Renzi, 2013). The use of mixed ores might explain the arsenical copper awl 412 of Torre Velha 3 showing a high content of silver (4.65 wt.% Ag). Copper with high silver content is rather uncommon, but other regional examples from this chronological period are known, namely an awl (10.9 wt.% Ag and 1.74 wt.% Sn) from the necropolis of Las Minitas, Badajoz (Gómez Ramos et al., 1998) and an earring (2.57 wt.% Ag) from the necropolis of Las Cruces, Sevilla (Hunt Ortiz, 2012).

The silver rivets of the dagger 714 also present high copper content (4.3–5.1 wt.%), but more significant is the unusually high amount of gold (4.9–5.4 wt.%). These silver alloys indicate the use

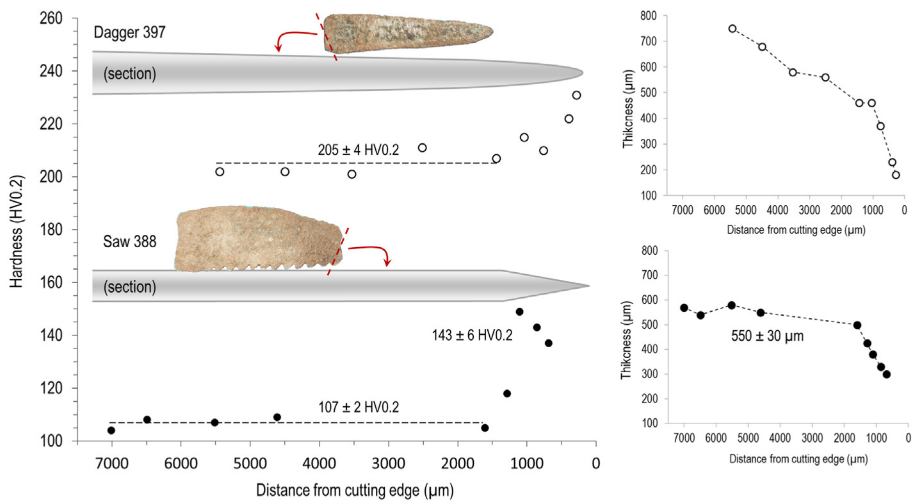

Fig. 6. Hardness and thickness profiles of MBA dagger 397 and saw 388 from Torre Velha 3.

of distinctive raw materials and silver with a high amount of gold is highly uncommon among prehistoric Iberian contexts. The known parallels are the rivets from a dagger recovered at Villacarrillo, Jaén (2.5 wt.% Cu and 3.0 wt.% Au, Montero Ruiz et al., 1995). The possibility of a foreign origin for this dagger is enhanced by its bronze blade, since as mentioned before this alloy was still very uncommon in the region during this period. Perhaps even more significant is the absence of Cu_2S inclusions in this blade. Copper sulphidic inclusions are rather common among prehistoric bronzes in the Portuguese territory, probably resulting from sulphur impurities in the smelted oxidic copper ores (Valério et al., 2013).

Finally, it must be noted that bronze and silver offerings from Torre Velha 3 are related with hypogea with a higher number of metallic goods. Hypogea [1267]–[1792] and [2417]–[2418] both present an awl and a dagger, while the hypogeum [2550]–[2551] had an awl and a necklace with silver and arsenical copper beads. These hypogea with higher wealth should point to the high social position of the individuals buried in the graves.

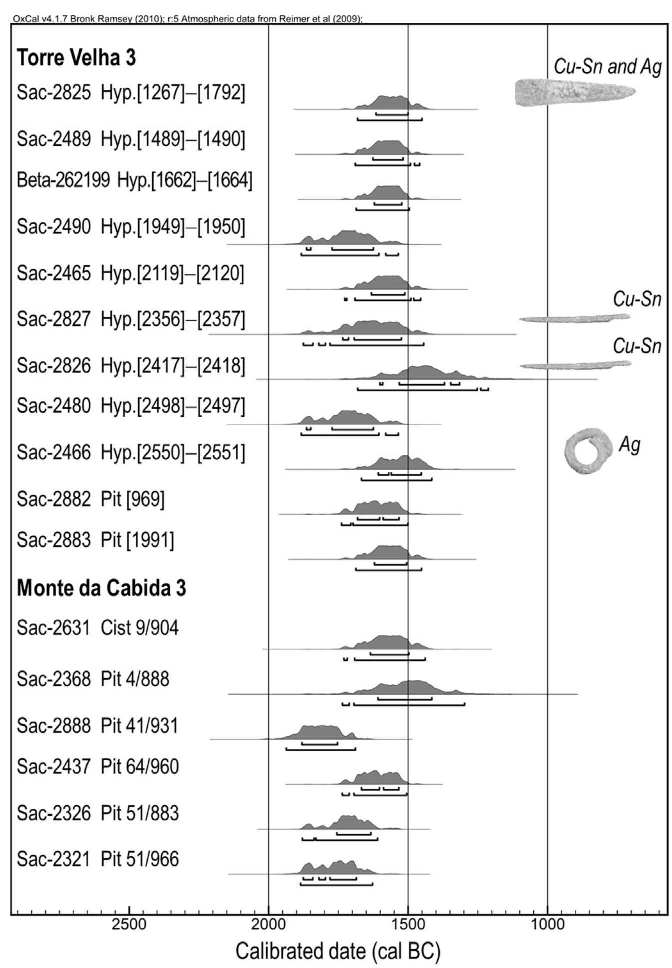

Fig. 7. Calibrated radiocarbon dates using the IntCal09 calibration curve (Reimer et al., 2009) and OxCal program (V4.1) (Bronk Ramsey, 2001, 2009) with indication of burials with bronze (Cu–Sn) and/or silver (Ag) artefacts.

5.3. Non-prehistoric metallurgy

One of the distinctive features of the non-prehistoric metallurgy is the use of a wide diversity of metals and alloys. In the set of 9 artefacts studied we found 5 different types, namely copper, bronze, tin-antimony bronze, brass and leaded brass. The study of the use of these alloys is beyond the scope of this work, but the compositional data endorses the late chronology of those artefacts, established by the associated pottery recovered from the infilling of the pits. A few of those metallic artefacts have typologies from the Iron Age, such as a double-spring fibula (404). Bronze and brass alloys have undoubtedly a posterior chronology and the higher iron content of artefacts can also be used as an indicator of non-prehistoric origin. The presence of these metals also confirms the long time span of use of some of those "storage" pits at Torre Velha 3.

5.4. Manufacture of arsenical copper and bronze artefacts

Microstructures of MBA arsenical coppers indicate that after casting an artefact would have been hammered, annealed and finished with a hammering operation. Since annealing softens the metal and allows an additional deformation, multiple cycles of hammering plus annealing could be used to obtain the desired shape. The final hammering would be used to increase the hardness or as a final smoothing of the surface. The manufacture of the chisel 615 differs slightly from the customary procedure, since the absence of slip bands suggests that there was no finishing hammering, at least on the middle section of the chisel. Mechanical work was also used to sharpen specific areas of the artefact, such as the edge of a dagger or the tip of an awl. A previous study on a sword of an MBA hypogeum at Horta do Folgão (Serpa) has identified the high skills of those metallurgists (Valério et al., 2012a). The blade was much more deformed at the tip and, especially at the cutting-edge, certainly to obtain a higher hardness. Therefore, considering the high number of awls available at Torre Velha 3, an attempt was made to attest if edges were subjected to specific work. It was not possible to sample multiple areas on each awl, but several awls were observed on the tip, while others were observed on the central section (Fig. 1: awl scheme – areas a1 and a2, respectively). No significant differences were observed in the grain size or the density of slip bands between tips and central sections. Therefore, if those edges have been sharpened, this has been done probably by abrasion and not by mechanical deformation.

Some microstructures clearly show that the prehistoric metallurgists often encountered problems during the manufacture of the artefact. For instance, the awl 405 exhibits a residual dendritic segregation. The recrystallization of an arsenical copper takes place between 300 °C and 400 °C, but higher temperatures are needed (600–700 °C) to homogenise the as-cast dendritic segregation in a reasonable time (Northover, 1989). Heavily segregated microstructures require higher temperatures to become fully homogenized since the segregation intensity is determinant regarding the homogenisation of arsenical coppers (Budd, 1991). Additionally, a low degree of deformation (inferred from the undeformed dendrites in the awl) limits the homogenization of the alloy. The dagger 423 and saw 388 also exhibit noticeable segregation bands, being other examples of incomplete homogenisation.

The As-rich phase initially formed by inverse segregation during solidification is also present at the grain boundaries of recrystallized grains. A high temperature annealing (e.g. indicated by the large grain size of the microstructure of chisel 615) could homogenize the alloy, but the long-term segregation of arsenic precipitates the As-rich phase at the new grain boundaries. The presence of this segregated arsenide weakens the metal, as evidenced by the accidental capture of a crack in the tip of dagger 397.

Despite sharing a similar overall manufacture these artefacts evidence quite different operational conditions. Differences are patent on the size and deformation of Cu–As–O inclusions, incidence of segregation bands, grain size and density of slip bands. A plot of factor scores gives a general idea about the variability of those features (Fig. 8). Results exhibit a large dispersion, as expected of prehistoric metallurgical works. About a third of the artefacts show larger grain size and lower density of slip bands evidencing an inferior thermomechanical work. It will not be surprising that the only ornament studied (ring 415) is found in this group. The remaining two-thirds of the artefacts show improved thermomechanical operations and will have a higher hardness. The manufacture often comprises a stronger hammering work, such as with dagger 397 or with awl 410 (the longest one). Among these are some non-homogenised alloys, but this would not have a visible effect in the alloy properties thus being ignored by those ancient metallurgists. Particularly significant is the fact that the arsenic content does not relates with manufacture features. This suggests that MBA metallurgists were not taking advantage of arsenic to improve the properties of the alloy, particularly to increase the hardness of tools and weapons.

Microstructures of MBA bronzes do not show a significant improvement in the manufacture compared to arsenical coppers. The usual procedure includes cycles of hammering and annealing plus a hammering finishing. However, certain artefacts such as the dagger 714 continue to exhibit limited thermomechanical work. Apparently, in this case the metallurgist gave priority to the

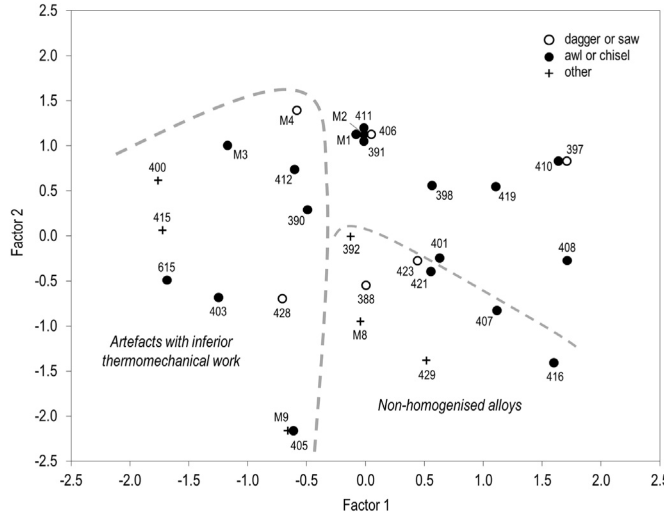

Fig. 8. Factor scores obtained from microstructural variables of MBA arsenical copper artefacts from Torre Velha 3 and Monte da Cabida 3 (artefacts 391, 406, 411, M1 and M2 have the same values).

aesthetics of the dagger by using silver rivets, and did not care with the functionality of the weapon.

Generally there are no major differences between operational sequences applied in the production of MBA arsenical copper and bronze. The combined use of forging and annealing to produce metal artefacts has long been established in the region. For instance, the majority of Chalcolithic artefacts recovered at the settlement of Vila Nova de São Pedro (Azambuja) already shows evidences of hammering and annealing (Pereira et al., 2013). This combination was also identified in the remaining Iberian area, although some regions seem to display an important use of cold forging during the MBA (Rovira, 2004).

Regarding the few Iron Age bronzes, a major difference is the absence of copper sulphidic inclusions, which can indicate the use of purer or more refined raw materials. An increased tendency to use bronze scrap was already established in this southern region of Portugal by metals from the Phoenician settlement of Quinta do Almaraz, Almada (Valério et al., 2012b). Another distinction is the higher density of slip bands in later bronzes. The use of a final hammering has continuously increased since the Chalcolithic till the Iron Age (Rovira, 2004). A more intense hammering will increase the metal hardness, but it takes an expert metallurgist to considerably strain the metal without fracture.

5.5. Hardness of arsenical copper and bronze artefacts

MBA arsenical coppers have a wide range of hardnesses (73–144 HV0.2) that seems to depend mainly on the grain size and slip bands density. For instance, the body of the chisel 615 presents a large grain size (~50–100 μm, see Fig. 3) and no slip bands, hence having a low hardness (73 HV0.2). On the contrary, the awl 407 has a small grain size (~10–20 μm) and a very high density of slip bands resulting in a higher hardness (125 HV0.2). It must be noted that the arsenical coppers with lower hardness were all subjected to "inferior thermomechanical work", as evidenced by the factor analysis of microstructural variables.

The majority of MBA bronzes presents a comparable hardness (110–154 HV0.2) despite having a low density of slip bands (Fig. 5). However, when subjected to a significant final deformation (e.g. awl 418) the bronze alloy attains a considerably higher hardness (221 HV0.2). The same is observed in bronzes belonging to the Iron

Age, whose higher hardness (171–235 HV0.2) mostly results from a strong final deformation.

The initial advantage of bronze over arsenical copper is related with the usually higher amount of the alloying element (e.g. awl 418: ~9 wt.% Sn and awl 407: ~5 wt.% As). In the case of bronzes with low tin content there will be no difference in the hardness of both metals, as evidenced by the low-tin bronzes and arsenical coppers of the Argaric necropolis of Cerro de San Cristóbal, Granada (Aranda Jiménez et al., 2012). However, the disadvantage of the low amount of alloying element on arsenical coppers does not necessarily mean that these would have been worked inefficiently. A saw with hardened teeth and a blade with an increasingly higher hardness from the middle to the cutting edge are good examples that evidence the correlation between function and manufacture of MBA arsenical coppers.

6. Conclusions

The elemental and microstructural characterisation of metallic grave goods of hypogea and cists, in addition to domestic artefacts recovered in the filling of pits, allowed a unique insight on the MBA metallurgy of the southern Portuguese territory and, consequently, on the Southwestern Iberian MBA. The radiocarbon dating of bone samples closely associated with the analysed artefacts allowed narrowing the chronology of these items for a period between c. 1900 and 1300 BC. The emergence of bronze and silver artefacts makes of this chronological interval a period of important technological developments after a millennium of arsenical copper metallurgy.

Despite revealing distinct burial practices, the synchronic hypogea and cists, together with the domestic pits of Torre Velha 3 and Monte da Cabida 3, share the main use of arsenical copper. This MBA metallurgy is somewhat different from the one found in Chalcolithic contexts in the region, which is mostly composed of copper artefacts with low arsenic contents (As < 2 wt.%). The use of mixed silver-copper ore deposits commonly found in Iberia might explain a silver bead with high amounts of copper and a copper awl with high levels of silver. On the contrary, a bronze dagger with silver rivets with high amounts of gold suggest different raw materials or, perhaps more probably, a foreign origin.

Nevertheless, the most significant outcome of this study was the identification of the first bronzes in the Southwestern Iberian MBA, exhibiting a chronology of the second quarter of the 2nd Millennium BC. These MBA tools/weapons present tin contents similar to LBA metals suggesting the importation from another region with a developed metallurgy of bronze. Moreover, the early appearance of this alloy in this region points to an earlier introduction or a more rapid expansion than previously anticipated. Cultural similarities with the Argaric world, such as comparable funerary monuments (hypogea and cists) and similar funerary rituals (meat offerings), do suggest contacts between Southwestern and Southeastern Iberian regions. Nevertheless, there are important technological differences concerning the MBA metallurgy, such as the suitable tin content of the southwestern bronzes compared with the high variability of alloys in Argaric bronzes. Altogether, the origin of these first bronzes, as well as the direction of the spread of the new alloy through Southern Iberia is still unknown.

The bronze dagger with two silver rivets evidences the prestige rather than functional significance attributed by MBA communities to early bronzes. Moreover, the arsenical copper dagger with silver coloured rivets shows the ability of those prehistoric metallurgists to duplicate prestige artefacts using local knowledge and available raw materials.

Finally, the bronze metallurgy do not introduced significant changes in the manufacture of artefacts. Arsenical coppers and bronzes were made with hammering plus annealing cycles and, usually, the procedure was finished with a hammering of varying strength that increases the hardness of the alloy. As expected from a prehistoric technology, the arsenical coppers display variable operational conditions with quite a few presenting second-rate thermomechanical work. Others exhibit a careful manufacture to improve its efficiency, such as the hardened cutting edge of a blade. Nevertheless, it seems that the bronzes could be more hardened than the coeval arsenical coppers, being probably one of the reasons for the disappearance of these during the last quarter of the 2nd Millennium BC.

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