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EARLY IMPORTS IN THE LATE BRONZE AGE OF SOUTH-WESTERN IBERIA: THE BRONZE ORNAMENTS OF THE HYPOGEA AT MONTE DA RAMADA 1 (SOUTHERN PORTUGAL)

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The composition and manufacture of Late Bronze Age metallic artefacts from funerary and domestic contexts of southern inland Portugal was studied. The prevailing trend comprises binary bronzes (10.3 ± 2.1 wt% Sn) showing deformed equiaxial grains, annealing twins and slip bands. The alloy composition is somewhat independent of artefact type, while the manufacture seems to rely on artefact function and the skilfulness of the metallurgist. The technological characteristics were linked with archaeological and chronological features, disclosing some artefacts of uncommon composition, such as low-tin bronze bracelets (4.3–7.1 wt% Sn) associated with ornaments of exotic materials (glass and Egyptian faience beads, and also ostrich egg shell beads). The assemblage testifies to an archaic trade with the Mediterranean region before the establishment of the first Phoenician colonies on the southern Iberian coast.

KEYWORDS: BRONZE ALLOY, COMPOSITION, MANUFACTURE, HARDNESS, SOUTH-WESTERN IBERIA, LATE BRONZE AGE, MEDITERRANEAN TRADE

INTRODUCTION

Radiocarbon dating of secure archaeological contexts has established the emergence of the bronze alloy in south-western Iberia during the second quarter of the second millennium BC (Valério *et al.* 2014). Those early bronzes are daggers and awls, recovered as grave goods from funerary hypogea, a pre- or protohistoric negative structure comprising a lowered atrium with a side entry to the underground burial chamber (Alves *et al.* 2010). Nevertheless, the full implementation of bronze alloys occurred only during the closing centuries of the second millennium BC, with the onset of the Late Bronze Age (LBA, 1170–1050/780–730 cal BC; Mataloto *et al.* 2013). The 11th to 10th century BC metallic hoard dredged in the Ría de Huelva is a typical example of the LBA copper-based metallurgy of south-western Iberia. The collection

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features almost 400 artefacts with a 'standard copper–tin binary composition' and with manufacture commonly including cycles of hammering and annealing (Rovira 1995). At the south-western end of Iberia, a similar metallurgical trait was found in a 10th to 9th century BC metallurgical workshop at Entre Águas 5 (southern Portugal), where crucibles, moulds, prills and a tuyere were recovered, attesting to the local production of binary bronze with ~10 wt% Sn and low amounts of impurities (Valério *et al.* 2013a).

The state of affairs would eventually be altered by the arrival of Eastern Mediterranean traders, the earliest contact evidence concerning metallurgical innovations seeming to be iron artefacts dating to the 11th century BC (Almagro Gorbea 1993; Senna-Martinez 2000; Vilaça 2013). Orientalizing trade contacts culminated with the establishment of Phoenician colonies on the Mediterranean and Atlantic southern coasts of Iberia during the ninth century BC or even just prior to that time, at sites such as Plaza de las Monjas (Huelva) and La Rebanadilla (Málaga) (Arancibia *et al.* 2011; Arruda 2011; Aubet 2008; González de Canales *et al.* 2008). The weapons, tools and ornaments of Phoenician colonies denote the common use of unalloyed copper and leaded bronze, in addition to binary bronze with a low tin content, as has been recorded at Morro de Mezquitilla (Giumlia-Mair 1992), La Fonteta (Renzi 2013), Quinta do Almaraz (Valério *et al.* 2012) and Alcácer do Sal (Schiavon *et al.* 2013). However, the technology transfer to indigenous communities was a rather slow process and local settlements with clear indications of Phoenician contacts (rectangular houses, wheel-turned ware and iron artefacts), such as Castro dos Ratinhos (ninth to eighth century BC—Soares and Martins 2010; Valério *et al.* 2010) or Moita da Ladra (eighth century BC; Valério *et al.* 2016), still show a conservative metallurgy of binary bronzes.

The recent archaeological excavation of two funerary hypogea at Monte da Ramada 1 (MR1, Aljustrel, southern Portugal) with LBA inhumations bearing different kinds of grave goods and funerary rituals, immediately proved to be an unusual and important discovery (Baptista *et al.* 2016). On one hand, these funerary structures of Monte da Ramada 1 constitute the first LBA hypogea ever found in south-western Iberia, thus attesting to the lasting use of this funerary architecture, which had been common since the Late Neolithic to Middle Bronze Age (MBA) (e.g., Alves *et al*. 2010; Valera 2012). On the other hand, it provided an exceptional opportunity to study grave goods of well-defined chronology, some of them having been unknown in LBA contexts until the discovery and excavation of this necropolis.

This work presents an elemental and microstructural characterization of bronze bracelets and necklace beads recovered *in situ* in these two funerary hypogea at Monte da Ramada 1. Other ornaments, tools and weapons from coeval domestic contexts in southern Portugal are also included for comparison with these examples of a funerary nature. The combined approach merging distinct microanalytical techniques has preserved the cultural integrity of the artefacts, and produced new information about the composition and manufacture of the metals present in the life and death of the protohistoric communities that lived inland at the south-western end of Iberia.

THE HYPOGEA AT MONTE DA RAMADA 1 AND COEVAL DOMESTIC CONTEXTS

The archaeological works at Monte da Ramada 1 were part of the mitigation strategy concerning the threatened cultural heritage, implemented during the construction of the Irrigation Network of Ervidel (Beja) integrated into the Alqueva dam project. A large set of archaeological structures were excavated, comprising an extended chronological interval from the Chalcolithic to the Islamic period (Baptista *et al.* 2016). However, only three structures were related to the Bronze

Age, all of which were of a funerary nature. The smallest structure was a circular pit with two burials, having three lithics and one ceramic fragment as grave goods. The other contexts were hypogea (hypogeum 2 and hypogeum 4), each one comprising an atrium and a burial chamber with distinct inhumation sequences, and containing burial offerings (ceramic cups) and several types of personal adornments.

The last individual to be buried in hypogeum 4 of Monte da Ramada 1 had a rich and rare set of personal adornments, including three copper-based bracelets, two gold beads, six ostrich egg shell beads, one glass bead and another bead, possibly of ivory (Fig. 1). The skeleton was radiocarbon-dated to the 10th century BC (see Table S1). Glass beads with a highly probable origin in the Eastern Mediterranean (Towle *et al.* 2001; Henderson *et al.* 2010) are known at

Figure 1 *(a) Hypogeum 4 at Monte da Ramada 1, with a rectangular atrium and a collapsed burial chamber (see the entrance still blocked by large slabs). Grave goods include (b) copper-based bracelets, (c) gold beads, (d) ostrich egg shell beads (top), a glass bead (bottom left) and an ivory(?) bead (bottom right). (e) Hypogeum 2 at Monte da Ramada 1, with a detail of a copper-based bracelet still on an ankle. Grave goods include (f) copper-based bracelets, (g) annular beads, (h) a ring-shaped bead and (i) a faience bead. [Colour figure can be viewed at wileyonlinelibrary.com]*

LBA coastal sites in Iberian Peninsula, suggesting precolonial contacts (see, e.g., Lorrio 2008; Rovira and Port, 1996). The early chronology and exotic materials of the Monte da Ramada 1 high-status burial imply that some vestiges of this maritime commerce were able to arrive in fairly inland regions in south-western Iberia. Contrary to the known LBA Iberian examples, the gold beads of Monte da Ramada 1 are hollow, probably being made of sheet metal, yet they do not display the common decoration elements of the Eastern Mediterranean gold jewellery, such as filigree or granulation (Perea 1991).

The archaeological excavations at hypogeum 2 of MR1 identified a very distinct situation, namely a total of at least 20 individuals, including several primary inhumations (Baptista *et al.* 2016). The grave goods of hypogeum 2 include exotic adornments of foreign origin, specifically two faience beads—also evidence of the trade with the Eastern Mediterranean region—and another bead probably made of jet. Along with those buried individuals, some copper-based ornaments were found, such as open bracelets and small necklace beads, either of annular or ring-shaped typology (Fig. 1). Six of those skeletons were radiocarbon-dated and Bayesian statistics was applied to this radiocarbon data using the known stratigraphic correlation between samples (Table S1 and Fig. S1). The results illustrate that these two hypogea began to be used in the 10th century BC, whereas hypogeum 2 continued to be used during the ninth century BC.

Metallic artefacts included for comparison belong to domestic contexts, some of them radiocarbon-dated, located at several archaeological sites in southern Portugal (Fig. 2), namely Monsaraz (MSZ), Evoramonte (EVM), Pedreira de Trigaches 2 (PT2), Soeiros (SOR), Horta da Morgadinha 2 (HM2), Poço Novo 1 (PN1) and Monte das Pereiras (MPR). The sites of Monsaraz (Reguengos de Monsaraz) and Evoramonte (Estremoz) are LBA settlements located in prominent places in the landscape (Calado *et al.* 1999). The bracelets from Monsaraz (Fig. 2) are open pieces, without any decoration being ascribed to LBA due to the typology of the associated ceramics. Some copper-based tools were recovered at Evoramonte in contexts radiocarbon-dated to the 10th to 9th century BC (Mataloto *et al.* 2013). A unique yet very interesting situation occurs at Pedreira de Trigaches 2 (Beja), involving an awl retrieved with thousands of barley seeds inside a pit lined with cork (Antunes *et al.* 2012). Radiocarbon dating of those seeds indicates a time interval corresponding to the transition from the MBA to the LBA (1380–1130 cal BC; Mataloto *et al.* 2013). The double-looped socketed axe of Soeiros, Arraiolos (Fig. 2) was an isolated find, the typology of which has many parallels in the Portuguese Estremadura, and it has been assigned to the eighth century BC (Coffyn 1985). The remaining artefacts belong to LBA negative structures of dubious functionality, such as a circular pit (a refuse pit?) at Horta da Morgadinha 2 (Serpa), a rectangular pit at Poço Novo 1 (Vidigueira) and a shallow structure (a hut floor?) at Monte das Pereiras (Alvito). A Rocannes-type sickle found at Monte das Pereiras (Fig. 2) stands out in these contexts, showing strengthening ribs on one side and a completely flat surface on the other. This LBA typology presents a wide distribution, comprising the Portuguese Estremadura and Alentejo regions (Coffyn 1985).

METHODOLOGY

Elemental compositions were obtained by micro-EDXRF analyses, using an ArtTAX Pro spectrometer with a 30 W Mo X-ray tube, a focusing polycapillary lens (analysis area diameter of ~70 µm) and an electrothermally cooled Si drift detector (FWHM of 160 eV at 5.9 keV) (Bronk *et al.* 2001). Each sample was analysed in three spots with 40 kV, 600 µA and

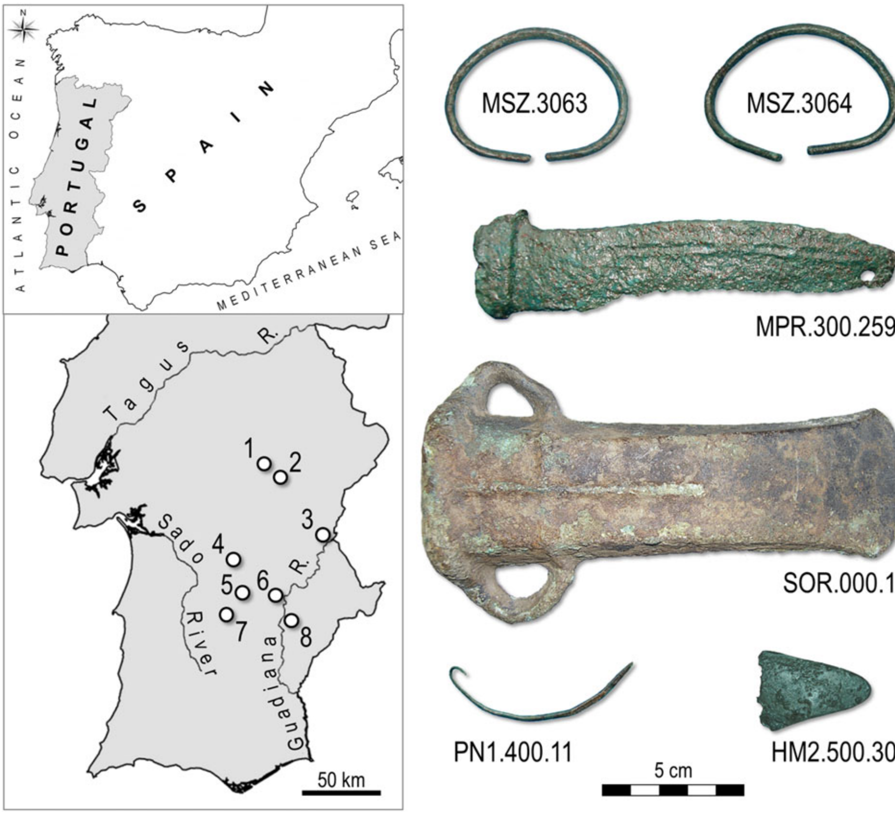

Figure 2 *Left, the location of archaeological sites in southern Portugal: 1, Soeiros; 2, Evoramonte; 3, Monsaraz; 4, Monte das Pereiras; 5, Pedreira de Trigaches 2; 6, Poço Novo 1; 7, Monte da Ramada 1; 8, Horta da Morgadinha 2. Right, some of the artefacts studied, namely the bracelets of Monsaraz, the Rocannes-type sickle of Monte das Pereiras, the double-looped socketed axe of Soeiros, the awl of Poço Novo 1 and the arrowhead of Horta da Morgadinha 2. [Colour figure can be viewed at wileyonlinelibrary.com]*

100 s of live time. Quantifications were made with WinAxil software, including experimental calibration factors obtained with British Chemical Standards Phosphor Bronze 551 and BNF Metals Technology Centre Leaded Bronze C50.01. The relative uncertainty is better than 10%, while the quantification limits are 0.05 wt% Fe, 0.10 wt% Ni, 0.10 wt% As, 0.50 wt% Sn and 0.10 wt% Pb.

Optical microscopy observations were made in a Leica DMI 5000 M inverted microscope with a 10× ocular lens, a 5× to 100× objective lens and distinct illumination modes, namely bright-field, dark-field and polarized light (Figueiredo *et al.* 2013). Microstructural features were enhanced by etching with an aqueous ferric chloride solution, while the ability to automatically assemble digital images of different focus positions enabled the characterization of non-planar surfaces, such as those of non-mounted samples.

The Vickers microhardness of mounted sections of artefacts was measured with a Zwick–Roell Indentec testing machine, using a load of 0.20 kgf for 10 s. The reported hardness is the average of at least three indentations, having a relative standard deviation lower than 10%.

RESULTS AND DISCUSSION

Composition of artefacts from funerary and domestic contexts

Micro-EDXRF analyses of artefacts have identified mostly binary bronze alloys with a low content of common impurities such as Pb, As and Fe (Table 1). The ring-shaped bead (MR1.253.2) has a relatively higher content of nickel (0.24 wt%) that possibly indicates a different origin, because this element can be considered a reliable source indicator, as the Cu/Ni ratio will be the same for the ore and the metal (Tylecote *et al.* 1977). However, the very low iron content of all artefacts points to a common technology characterized by the weak reducing conditions of smelting operations, which hinder the reduction of iron impurities and lessen the incorporation of this element in copper (Craddock and Meeks 1987).

The collection studied does not show any noticeable compositional difference between ornaments and tools/weapons, nor among individual typologies. The average tin content of these LBA binary bronze alloys (10.3 ± 2.1 wt% Sn, $n = 20$) matches the typical value of other collections in south-western Iberia, where some parallels can be found in the vast LBA hoard of the Ría de Huelva (11.0 ± 3.2 wt% Sn, $n = 387$; Rovira 1995) or, geographically closer, in

Table 1 *The composition of artefacts from LBA funerary and domestic contexts in southern Portugal*

	Artefact	*Reference*	*Cu (wt%)*	*Sn (wt%)*	*Pb (wt%)*	*As (wt%)*	*Ni (wt%)*	*Fe (wt%)*
Funerary contexts*								
MR1—hypogeum 2	Annular bead	MR1.234.1	89.3	10.7	n.d.†	n.d.	n.d.	< 0.05
	Annular bead	MR1.239.1	86.2	13.8	n.d.	n.d.	n.d.	< 0.05
	Annular bead	MR1.253.1	90.5	9.5	n.d.	n.d.	n.d.	< 0.05
	Ring-shaped bead	MR1.253.2	90.3	9.4	n.d.	n.d.	0.24	< 0.05
	Bracelet	MR1.201.1	87.7	12.3	n.d.	n.d.	n.d.	< 0.05
	Bracelet	MR1.234.2	87.9	11.9	0.13	n.d.	n.d.	< 0.05
	Bracelet	MR1.260.1	88.1	11.8	n.d.	n.d.	n.d.	< 0.05
	Bracelet	MR1.262.1	88.0	11.8	0.20	n.d.	n.d.	< 0.05
MR1—hypogeum 4	Bracelet	MR1.418.1	92.9	6.8	0.13	n.d.	n.d.	0.09
	Bracelet	MR1.419.1	91.7	4.3	n.d.	4.0	n.d.	< 0.05
	Bracelet	MR1.420.1	92.7	7.1	0.13	n.d.	n.d.	< 0.05
Domestic contexts*								
EVM	Awl	EVM.315	90.5	8.2	0.51	0.69	n.d.	< 0.05
	Chisel(?)	EVM.276	87.4	12.3	0.22	< 0.10	n.d.	< 0.05
	Rod	EVM.238	88.3	11.4	0.14	< 0.10	n.d.	< 0.05
HM2	Arrowhead	HM2.500.30	89.1	10.1	0.30	0.45	n.d.	< 0.05
MSZ	Bracelet	MSZ.3063	87.9	11.4	0.61	< 0.10	n.d.	< 0.05
	Bracelet	MSZ.3064	86.9	10.9	2.1	n.d.	n.d.	< 0.05
MPR	Rocannes sickle	MPR.300.259	88.6	10.3	0.67	0.42	n.d.	< 0.05
	Shapeless	MPR.301.1	88.8	10.6	0.35	0.21	n.d.	< 0.05
PT2	Awl	PT2.989.1	93.2	5.8	0.54	0.42	n.d.	< 0.05
PN1	Awl	PN1.400.11	90.3	8.4	0.34	0.88	n.d.	< 0.05
SOR	Socketed axe	SOR.000.1	87.7	12.2	< 0.10	< 0.10	n.d.	< 0.05

*MR1, Monte da Ramada 1; EVM, Evoramonte; HM2, Horta da Morgadinha 2; MSZ, Monsaraz; MPR, Monte das Pereiras; PT2, Pedreira de Trigaches 2; PN1, Poço Novo 1; SOR, Soeiros.

†n.d., Not detected.

the 10th to 9th century BC settlement of Entre Águas 5 (9.8 ± 3.2 wt% Sn, $n = 10$; Valério *et al.* 2013a), the ninth to eighth century BC settlement of Castro dos Ratinhos (10.1 ± 2.5 wt% Sn, $n = 37$; Valério *et al.* 2010) or a recently analysed set comprising LBA sites in southern Portugal (9.7 ± 2.9 wt% Sn, $n = 18$; Valério *et al.* 2015). This metallurgy with suitable tin contents implies a steady supply of cassiterite, which could come either from central Portugal (Merideth 1998) or the Ossa–Morena Zone (e.g., the Santa Eulália and Cáceres areas—Gonçalves 1973; Cardoso *et al.* 1992; Rodríguez Díaz *et al.* 2013). North-western Iberia is another possible supply area for the tin (see, e.g., Rovira 2003, fig. 3B), in this case strengthening the connections between this inland region and Atlantic Europe. Nevertheless, it must be noted that LBA metallurgists produced bronze by the co-reduction of copper and tin ores (as is the case at Entre Águas 5; Valério *et al*. 2013a) and it seems more likely that tin ores were imported from available nearby sources.

The exceptions to this regularity of binary bronzes with suitable tin contents allow some quite interesting considerations. The low tin content of the awl from Pedreira de Trigaches 2 (PT2.989.1, 5.8 wt% Sn) should be related to the early chronology of this MBA/LBA context (1380–1130 cal BC). The tin content of bronzes varies very irregularly during the MBA, due to the difficulty in obtaining suitable thermal and chemical conditions inside the smelting crucible, and perhaps also because the method that was probably used—the co-smelting of copper and tin minerals of unknown composition—may hinder the control of the alloy composition (Rovira and Montero-Ruiz 2013). For instance, an awl and three bronze prills recovered in MBA contexts at Evoramonte (1500–1300 cal BC) display more variable tin contents (~11–19 wt%) than the LBA collection presented here.

Another exception to the prevailing binary bronzes is the Monsaraz leaded bronze bracelet (MSZ.3064, 2.1 wt% Pb). A LBA metallurgy almost exclusively composed of binary bronzes typifies most of the Iberian Peninsula. On the contrary, a leaded bronze alloy with this chronology relates better to Atlantic Europe, which comprises north-western Iberia, the British Isles and western France (see, e.g., Figueiredo *et al.* 2010, fig. 1). Therefore, the leaded bronze bracelet unearthed at Monsaraz may actually be a product of this Atlantic world, following a possible tin supply route from north-western Iberia. An early interaction with Mediterranean traders should also be considered, since the oldest leaded bronzes recovered in southern Portugal belong to the Phoenician settlement of Quinta do Almaraz (Valério *et al.* 2012). However, such leaded bronzes, as well as the majority of the copper-based artefacts from the Iberian Phoenician colonies, have higher Fe contents, this being considered an indicator of foreign origin (Craddock and Meeks 1987; Giumlia-Mair 1992; Valério *et al.* 2012).

An additional singularity identified is the set of bracelets from hypogeum 4 at Monte da Ramada 1, namely one arsenical bronze (MR1.419.1, 4.3 wt% Sn and 4.0 wt% As) and two bronze alloys (MR1.418.1, 6.8 wt% Sn and MR1.420.1, 7.1 wt% Sn), all with a low tin content when compared with the rest of the collection. Arsenical bronze resembles some early bronze alloys encountered in MBA contexts, such as an awl from Torre Velha 3, southern Portugal (8.7 wt% Sn and 2.2 wt% As; Valério *et al.* 2014) or an axe from Escaroupim, central Portugal (5.9 wt% Sn and 2.2 wt% As; Figueiredo *et al.* 2012). Moreover, bronzes with less than 8 wt% tin only account for 10% of the above-mentioned LBA collections of south-western Iberia (about 450 items), demonstrating that low-tin bronze alloys are uncommon. The existing low-tin alloys do not seem to be related with typology, since examples include both ornaments and tools, such as a bracelet (5.0 wt% Sn) and a needle (5.9 wt% Sn) from Entre Águas 5 (Valério *et al.* 2013a), a knife (4.9 wt% Sn) from Castro dos Ratinhos (Valério *et al.* 2010) and a ring (2.4 wt% Sn) from Outeiro do Circo (Valério *et al.* 2013b). Generally, the use of bronze scrap diminishes the amount of tin due to the preferential oxidation of this element during casting; for example, trials have shown that a few melting cycles can reduce the tin content from 9.5 wt% to 3.1% (Sarabia 1992). However, in the particular case of hypogeum 4, there is evidence attesting against the recycling of scrap, at least in the bracelet with a high arsenic content (MR1.419.1, 4.0 wt% As), because this element is rapidly lost by melting and hot working in oxidizing conditions (see, e.g., McKerrell and Tylecote 1972). The unusual alloy composition of those bracelets, and their association with exotic grave goods (ostrich egg shell and glass beads) inside a LBA burial (1004–853 cal BC), in addition to the presence of Egyptian faience beads in the nearby and coeval hypogeum 2, suggest a foreign origin for those metal ornaments, which most probably belong to the Eastern Mediterranean region. This area displays a diversified metallurgical technology during the early stages of the first millennium BC, but, for instance, a collection of fibulae and pins from Nimrud (Iraq) includes five binary bronzes of analogous composition (3.2–8.0 wt% Sn, 0.20–0.90 wt% Pb, 0.15–0.85 wt% As and 0.02–0.24 wt% Fe; Giumlia-Mair 1992).

Table 2 *Microstructural and compositional features of ornaments and tools/weapons from LBA contexts in southern Portugal*

	*Reference**	*Sn* (*wt%*)	*Phases*	*Grain size* (μm)[†]	*Features*[‡]	*Inclusions*	*Manufacture*[§]	*Hardness* (HV0.2)
Ornaments								
Annular bead	MR1.253.1	9.5	α	20	t, sb	Cu–S	H + A + FH	186
Ring-shaped bead	MR1.253.2	9.4	α	60	t, sb	Cu–S	H + A + FH	–
Bracelet	MR1.201.1	12.3	α	60	t	Cu–S	H + A	120
Bracelet	MR1.234.2	11.9	α	60	t	Cu–S	H + A	111
Bracelet	MR1.260.1	11.8	α	100	t, sb	Cu–S	H + A + FH	–
Bracelet	MR1.262.1	11.8	α	60	t	Cu–S	H + A	–
Bracelet	MR1.418.1	6.8	α	60	t, sb↑	Cu–S↓	H + A + FH↑	175
Bracelet	MR1.419.1	4.3	α	20	t, sb	Cu–S	H + A + FH	–
Bracelet	MR1.420.1	7.1	α	60	t	Cu–S↓	H + A	87
Bracelet	MSZ.3063	11.4	α	20	t, sb↓	Cu–S	H + A + FH↓	–
Bracelet	MSZ.3064	10.9	α	20	t, sb↓	Cu–S	H + A + FH↓	–
Tools/weapons								
Awl	EVM.315	8.2	α	20	t, sb	Cu–S	H + A + FH	129
Awl	PN1.400.11	8.4	α	20	t	Cu–S	H + A	102
Awl	PT2.989.1	5.8	α	60	t, sb↑	Cu–S	H + A + FH↑	182
Arrowhead	HM2.500.30	10.1	α	60	t, sb	Cu–S	H + A + FH	–
Chisel(?)	EVM.276	12.3	α, δ	60	t	Cu–S	H + A	106
Rocannes sickle	MPR.300.259	10.3	α	20	t, sb	Cu–S	H + A + FH	–
Rod	EVM.238	11.4	α, δ	–	Coring	Cu–S↓	'As-cast'	105

*MR1, Monte da Ramada 1; EVM, Evoramonte; HM2, Horta da Morgadinha 2; MSZ, Monsaraz; MPR, Monte das Pereiras; PT2, Pedreira de Trigaches 2; PN1, Poço Novo 1.

[†]Approximate average value.

[‡]t, Annealing twins; sb, slip bands.

[§]H, hammering; A, annealing; FH, final hammering.

The manufacture of ornaments, tools and weapons

Optical microscopy observations have identified recrystallized microstructures with deformed equiaxial grains, annealing twins and slip bands (Table 2). A chisel and a cylindrical rod of unknown functionality (EVM.276, 12.3 wt% Sn; and EVM.238, 11.4 wt% Sn) have the $\alpha + \delta$ eutectoid, demonstrating relatively fast cooling of the castings (the α phase can dissolve up to 14 wt% Sn at the eutectoid temperature of 530°C; Hanson and Pell-Walpole 1951) and mild thermomechanical cycles (in the case of the chisel) that were insufficient to dissolve the excess δ phase. Apart from these production features, all artefacts show Cu–S inclusions arising from smelted ores. Overall, no significant differences were found in the manufacture and composition of ornaments retrieved from funerary and other contexts (i.e., bracelets from Monte da Ramada 1 and Monsaraz), reinforcing the idea that the first group are customary ornaments taken to the final resting place of their owner. Moreover, the supposedly imported bracelets from hypogeum 4 show a similar manufacture to local parallels from hypogeum 2.

A uncharacteristic microstructure was found in the cylindrical rod (EVM.238), showing generally a cored dendritic structure typical of 'as-cast' artefacts, but some minute areas with twinned grains of very small size (< 10 µm, Fig. 3) evidence an initial thermomechanical working. The post-casting work of the remaining artefacts included one or more hammering and annealing cycles, whereas about two thirds of both tools/weapons and ornaments had a clear final hammering operation, resulting in strain hardening. For some typologies, such as arrowheads, the need to increase the hardness seems to be obvious. For other types such as awls, the strain-hardening intensity was variable and had a decisive effect on the hardness, as shown by the direct relation between the hardness and the density of the slip bands (e.g., PN1.400.11, EVM.315 and PT2.989.1; Fig. 3).

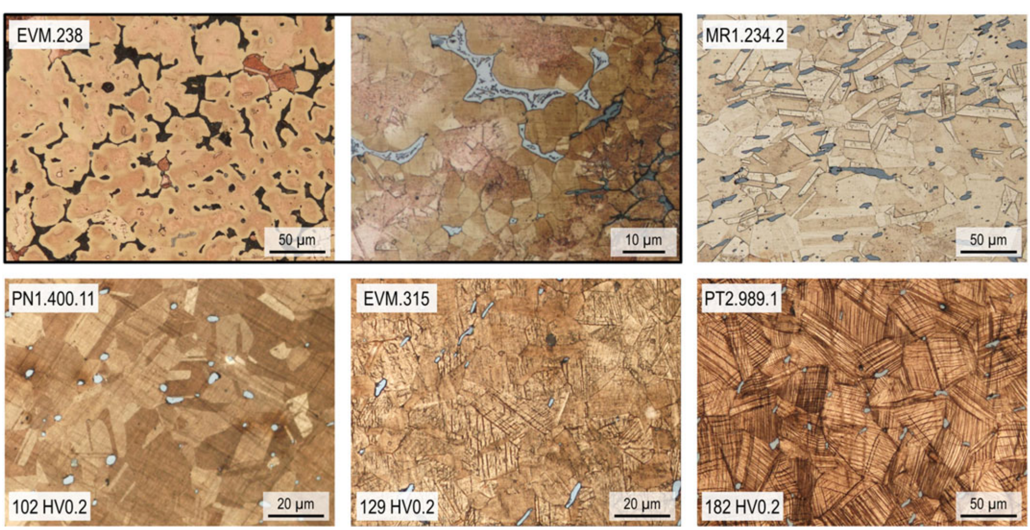

Figure 3 *Optical microscopy images (bright-field, etched) of LBA artefacts of southern Portugal, namely a cylindrical rod (EVM.238), a bracelet (MR1.234.2) and awls (PN1.400.11, EVM.315 and PT2.989.1) (the light blue (light gray) is the $\alpha + \delta$ eutectoid, the darker blue (dark gray) areas are Cu–S inclusions and the pink (darker) areas in EVM.238 are α phase regions that are richer in copper). [Colour figure can be viewed at wileyonlinelibrary.com]*

The greater thickness of the Cu–S inclusions in some bracelets (e.g., MR1.234.2; Fig. 3) suggests less deformation and/or fewer thermomechanical cycles. Although not having a noticeable effect upon the hardness of those artefacts, the grain size supports this hypothesis, as ornaments generally show a higher grain size than tools. It seems feasible that some ornaments such as bracelets were cast closer to the final shape, thus requiring less post-casting processing than tools and, consequently, having a lower degree of grain refining.

The variable hardness of different bracelets (87–175 HV0.2; Table 2) brings up to light the uneven manufacture of such similar ornaments, the intended function of which was apparently the same. Contrary to bracelets, the hardness variability of awls (102–182 HV0.2; Table 2) can be related to the multipurpose usage of these tools, the various functions either benefiting or not benefiting from a higher strength. Moreover, the hardness profiles on selected artefacts suggest that some awls are more hardened on the edge (e.g., the hardness reaches about 200 HV0.2 on the edge of awl PT2.989.1; Figures 4 (a) and 4 (b)).

The comparison of the composition, manufacture and hardness of prehistoric bronzes from southern Portugal underlines the main role of the final hammering operation in obtaining a superior strength (this work; see also Valério *et al.* 2010, 2013a,b, 2016). In this collection, the average increase in hardness due to strain hardening is about 50% (annealed artefacts, 97 ± 13 HV0.2, $n = 16$; strain-hardened artefacts, 143 ± 26 HV0.2, $n = 34$). Despite the similar

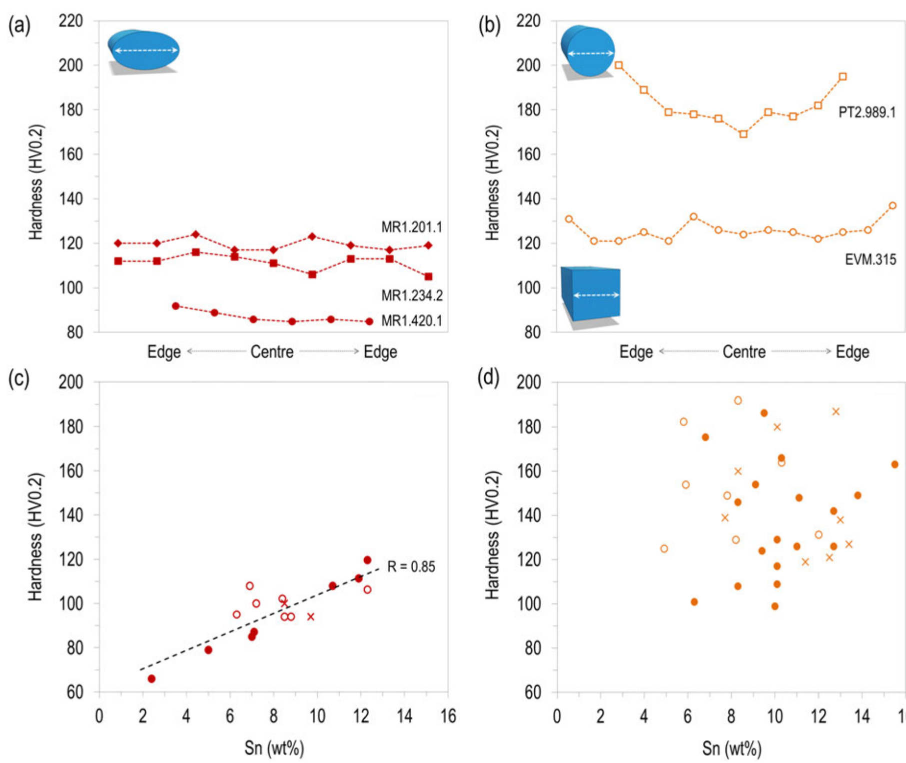

Figure 4 *Hardness profiles along the cross-sections of (a) annealed bracelets and (b) strain-hardened awls of southern Portugal (the distance between indentations is about 250 µm). The hardness versus tin content of (c) annealed artefacts and (d) strain-hardened artefacts of southern Portugal: solid circles, ornaments; open circles, tools; other, unknown functionality (this work and Valério et al. 2010, 2013a,b, 2016). [Colour figure can be viewed at wileyonlinelibrary.com]*

behaviour of ornaments and tools concerning hardness, it should be emphasized that the latter often could not be analysed in key areas, such as the tip of awls and chisels. Another significant result concerns the increase in hardness of annealed artefacts with the increasing amount of tin (Fig. 4 (c)). As the grain size does not vary significantly and the δ phase is almost absent, the increase corresponds mostly to solid-solution hardening: an increase in hardness of about 30% should be expected by increasing the amount of tin from 6 to 12 wt%. A study with cast alloys has found an analogous hardening of about 32% when comparing 7 and 13 wt% Sn bronzes (Lechtman 1996). Additionally, it is very interesting to verify that solid-solution hardening is much less significant than skilful strain hardening, which can reach up to 70–80% for a 10 wt% Sn bronze (Fig. 4 (d)).

Finally, the ornaments in the above-mentioned collection of prehistoric bronzes from southern Portugal have slightly higher tin contents than the tools (9.7 ± 2.9 wt%, $n = 25$ and 8.1 ± 2.1 wt%, $n = 15$, respectively; see Figures 4 (c) and 4 (d)). Although a larger number of cases is needed to reinforce (or disprove) this trend, it suggests that tin was not used to increase the hardness of tools and weapons. On the contrary, its preferential use in ornaments implies a relation with the golden hue of high-tin bronzes, which would be highly valued for prestige artefacts, such as a 15.5 wt% Sn ring recovered at the LBA settlement of Entre Águas 5 (Valério *et al.* 2013a).

CONCLUSIONS

LBA artefacts from funerary and domestic contexts in southern Portugal characterize a metallurgy featuring binary bronzes with a suitable tin content (~10 wt%) implying a stable supply of cassiterite and reliable technological knowledge during the opening centuries of the first millennium BC. Those ornaments, tools and weapons were worked with cycles of hammering plus annealing, and were sometimes strain hardened, depending to some extent on the artefact function and the skilfulness of the metallurgist.

Despite the apparent monotony of the LBA metallurgy in southern Portugal, there are a few artefacts that show an uncommon composition and imply interesting new evidence. First, the early bronzes with a low tin content, such as the awl from Pedreira de Trigaches 2, highlight the difficulties faced by local metallurgists in producing this new alloy, probably due to the poor temperature and reducing conditions of co-smelting operations. Second, if typological features such as a double-looped socketed axe already suggest regional contacts betwen local communities and the Estremadura region, only the technological features now identified, such as bronze alloys with a low tin content, indicate trade over long distances. In fact, these unusual bronze alloys, linked with glass and faience beads and also ostrich egg shell beads, disclose an archaic Orientalizing trading network during the 10th century BC.

Finally, it should be emphasized that through the combined use of microanalytical techniques, we were able to preserve the cultural integrity of the artefacts, while yielding significant archaeometallurgical conclusions. However, additional analytical studies comprising precisely dated contexts are essential to continue to increase our knowledge about the bronze metallurgy at this south-western end of Iberia during those ancient times of multiple foreign influences.

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SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

Figure S1. Modelled calibrated dates of the burial sequence of hypogeum 4 (stratigraphic unit 422) and hypogeum 2 (stratigraphic units 259, 246, 245, 218, 203 and 209) at Monte da Ramada 1, using the calibration curve IntCal13 (Reimer *et al.* 2013), the Oxcal calibration program (Bronk Ramsey 2013) and a Bayesian statistics considering a Sequence model (Bronk Ramsey 2008, 2009).

Table S1. Radiocarbon dates of stratigraphic units in hypogea 4 and 2 at Monte da Ramada 1. Calendar dates were determined using the calibration curve IntCal13 (Reimer *et al.* 2013), the Oxcal calibration program (Bronk Ramsey 2013) and a Bayesian statistics considering a Sequence model (Bronk Ramsey 2008, 2009).