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ORIGINAL PAPER

Early Iron Age copper-based funerary items from southern Portugal

Pedro Valério[1] · Maria Fátima Araújo[1] · António M. Monge Soares[1] · Luís C. Alves[1] · Rui M. Soares[2] · Linda Melo[3] · Lídia Baptista[4]

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Abstract
The recently excavated necropolises of Esfola (Beja) and Monte do Bolor 1/2 (Beja) display a characteristic architecture and distinctive funerary items that refer to the seventh to sixth century BC. The work comprises a microanalytical study of copper-based artefacts, some of them symbols of status, recovered in these necropolises (Tartesic belt buckles, body grooming instruments and different types of fibulae, bracelets and rings). The alloy composition was determined by micro-EDXRF and micro-PIXE analyses, while the colour distinction was estimated using the composition of different alloys. These funerary items disclose a metallurgy centred in low-tin bronze alloys (7.1 ± 2.4 wt% Sn, n = 41) with minor contents of lead, arsenic, nickel and iron. There are a few exceptions composed of copper or leaded bronze (*c.* 3.0 wt% Sn and 2.7 wt% Pb), in addition to arsenical copper (*c.* 5.6–6.5 wt% As), which is particularly remarkable due to the rarity of this last alloy in coeval contexts. A Tartesic belt buckle with rivets of distinct composition from the remaining components is other exception, as most composite artefacts have components of similar composition (e.g. pin, spring and axle of fibulae and body and decoration bead of bracelets and rings). Additionally, it was assessed that only a handful of examples were perceived as having a distinct colour and these outliers were mostly small body grooming instruments. Generally, the seventh to sixth century BC funerary items of southern Portugal disclose an indigenous metallurgy altered by earlier Phoenician stimuli, but the possibility to obtain different colours by changing the alloy composition was not commonly used.

Keywords Bronze alloy · Arsenical copper · Colour · Phoenician · Indigenous metallurgy · Iberian Peninsula

Introduction

The composition of copper-based artefacts can be used as an indicator of the technological expertise of ancient communities (Craddock 1995). The research can evaluate whether ancient metallurgists took advantage of the properties induced by the addition of distinct alloying elements. For instance, the addition of lead to bronze is ideal for casting massive objects, as it improves the castability of the alloy. In the particular case of ornaments, the colour could also be an important feature. In prestige symbols, such as fibulae and bracelets, the golden colour of bronze alloys with higher tin content would indeed be a valued characteristic.

Most regions of the Iberian Peninsula experienced important cultural and technological changes following the establishment of Phoenician colonies in Mediterranean and Atlantic coastal areas during the ninth to eighth century BC (Arruda 2019; López Castro 2019). Early Iron Age (EIA) indigenous communities readily adopted the novelties brought from distant regions, such as iron weaponry and

✉ Pedro Valério
pvalerio@ctn.tecnico.ulisboa.pt

1 Centro de Ciências e Tecnologias Nucleares (C2TN), Departamento de Engenharia e Ciências Nucleares, Instituto Superior Técnico, Campus Tecnológico e Nuclear, Universidade de Lisboa, Estrada Nacional 10 (km 1397), 2695-066 Bobadela LRS, Portugal

2 Centro de Arqueologia da Universidade de Lisboa (UNIARQ), Faculdade de Letras, Universidade de Lisboa, Alameda da Universidade, 1600-214 Lisboa, Portugal

3 Centro de Investigação em Antropologia e Saúde (CIAS), Departamento de Ciências da Vida, Universidade de Coimbra, Calçada Martim de Freitas, 3000-456 Coimbra, Portugal

4 Centro de Estudos de Arqueologia Artes e Ciências do Património (CEAACP), Arqueologia & Património Lda., Rua do Chouso 434, Santa Cruz do Bispo, 4455-804 Matosinhos, Portugal

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exotic ornaments (e.g. Egyptian faience and glass beads), but other advances requiring specialised technological expertise took longer to be integrated in local communities. A diversified bronze technology was among the latter, as evidenced by the conservative metallurgy present in ninth to eighth century BC indigenous settlements such as Castro dos Ratinhos, Moura (Valério et al. 2010) and Moita da Ladra, Vila Franca de Xira (Valério et al. 2016). The exclusive use of binary bronze alloys was inherited from the Late Bronze Age (LBA) and strongly contrasts with the metallurgy of copper, bronze and leaded bronze, existing in Phoenician sites of Iberian Peninsula, such as Morro de Mezquitilla, Málaga (Giumlia-Mair 1992), La Fonteta, Alicante (Renzi et al. 2009), Quinta do Almaraz, Almada (Valério et al. 2012) and Alcácer do Sal (Schiavon et al. 2013).

In southern Portugal, the early evidence of an Orientalizing copper-based technology in indigenous contexts belongs to the seventh to sixth century BC necropolises of Palhais (PL) (Valério et al. 2013) and Monte do Bolor 1/2 (MB) (Valério et al. 2017). On the other hand, recent archaeological works in another seventh to sixth century BC necropolis — Esfola (EF) — disclosed a new collection of copper-based funerary items integrated in a characteristic material culture, including ceramic vessels, iron weapons and glass beads (Melo et al. in press). Therefore, this was an excellent opportunity to expand the knowledge on the use of copper-based alloys during the centuries following the arrival of Phoenician innovations, especially because only a handful of artefacts had so far been analysed.

Our work comprises the chemical characterisation of copper-based funerary items of Esfola and Monte do Bolor 1/2. Artefacts of Esfola necropolis were analysed by micro-EDXRF, while a few objects not previously studied of Monte do Bolor 1/2 were characterised by micro-PIXE. These microanalytical techniques produce comparable results in the determination of the chemical composition of metallic artefacts (see, for example, Lyubomirova et al. 2015). Thereafter, the composition of funerary items was used to identify which of them were perceived by the human eye as having a distinguishing colour, as the appearance of a noble metal was certainly an important characteristic of prestige artefacts. The colour difference was estimated with measurements made previously of the CIELAB colour space of analogous copper-based alloys (Fang and McDonnell 2011; Mecking 2020; Radivojević et al. 2018). Finally, the alloy composition and colour of those items were discussed to establish the technological knowledge of seventh to sixth century BC communities in southern Portugal.

The necropolises of Esfola and Monte do Bolor 1/2

The archaeological sites of Esfola and Monte do Bolor 1/2 are located relatively close to each other (15 km apart) in the inland region of southern Portugal (Fig. 1). These sites were subject to rescue excavations between 2014 and 2015 to mitigate the impact on cultural heritage due to the construction of the São Matias and the Beringel-Beja irrigation systems, respectively.

Fieldworks carried out at Esfola have identified an EIA necropolis comprising two rectangular enclosures and nine

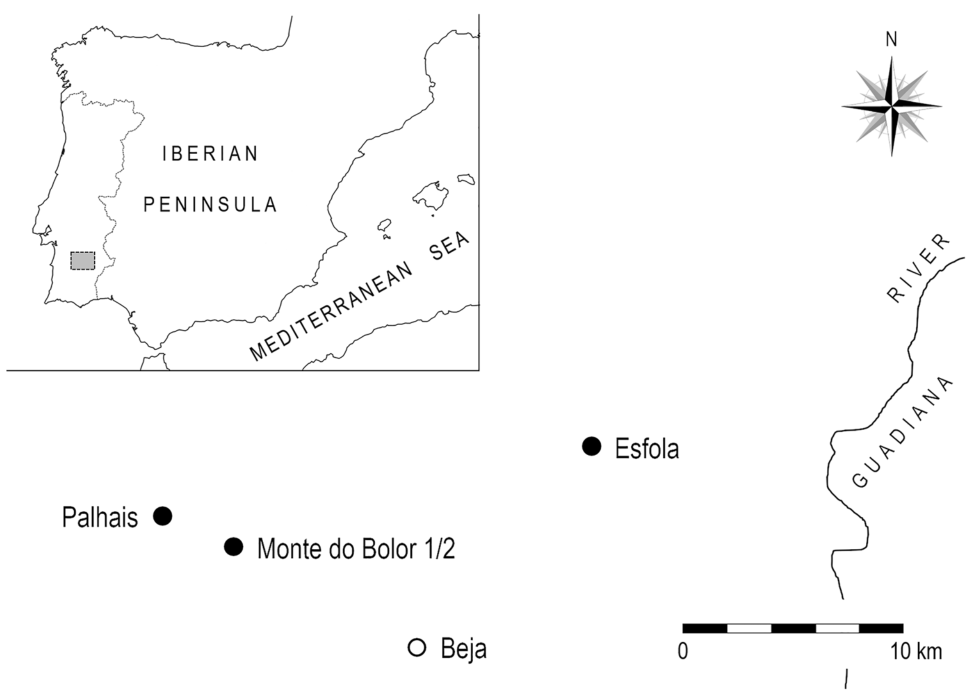

Fig. 1 The inland location in southern Portugal of the necropolises of Esfola (Beja), Monte do Bolor 1/2 (Beja) and Palhais (Beja)

subrectangular graves excavated in the soil (Melo et al. in press). These funerary enclosures display a similar configuration with a central burial and a second grave at the edge of the compound enclosure (Fig. 2). Burials inside the second funerary enclosure clearly stand out due to a higher amount of grave goods: the central tomb (grave 4) contained a male adult with an iron spear, an iron knife and an Alcores fibula (EF-063), while the second burial (grave 7) contained an adult individual with a Tartesic belt buckle (EF-087/088). Wealthier burials are often located inside funerary enclosures (Soares et al. 2017), but scattered graves around the funerary enclosure 1 also contained valuable items. For instance, grave 11 exhibited a young individual with a large set of beads made of glass, marine shells and animal teeth, in addition to copper-based items, namely, three bracelets (EF-144/145/196) and three rings (EF-143/178/208).

Archaeological works at Monte do Bolor 1/2 also identified an EIA necropolis with two rectangular enclosures and 16 subrectangular graves (Soares et al. 2017). These funerary enclosures surround several structures (Fig. 2), being the grave 4914 one of the richest burials with a ceramic vessel; a large number of beads made of gold, silver, amber and glass; an iron knife with a decorated bone handle, in addition to a copper-based fibula of Ponte 9a/1 type (MB-4913); two copper-based bracelets (MB-4911–1/2); and a copper-based ring (MB-4904–1). Grave 5511 is another example of a burial of a high-status individual, in this case displaying a ceramic vessel, two silver bracelets, several beads made of silver, marine shells and glass, an ivory/bone scaraboid, an iron belt buckle, a copper-based hairpin (MB-5507–1) and three copper-based body grooming instruments (MB-5509–1/6/8).

Table 1 presents a synopsis of the funerary items recovered at Esfola and Monte do Bolor 1/2. Copper-based ornaments and instruments constitute the more common prestige symbols, followed by weapons and belt buckles made of iron. Curiously, the ceramic vessels and exotic ornaments are much more common at Monte do Bolor 1/2, while gold and silver jewels are absent from Esfola. Generally, burials in the former necropolis had fewer items, but the fact that some contexts were disturbed hinders a secure conclusion about the wealth of those individuals. Regarding the different types of grave goods, the male individuals frequently display iron weapons and copper-based fibulae, while metal bracelets and beads made of exotic or noble materials often

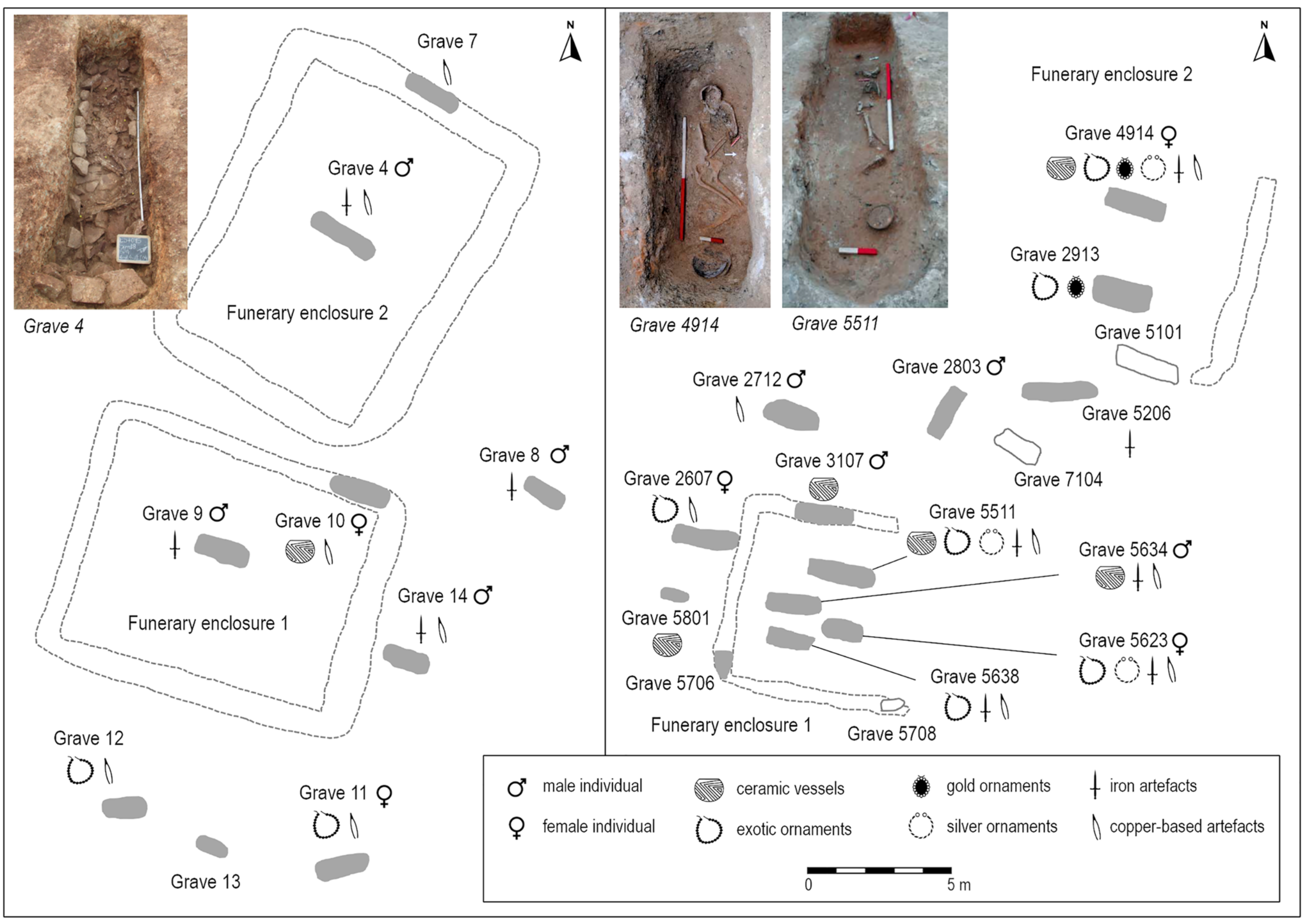

Fig. 2 Schemes of necropolises of Esfola (left) and Monte do Bolor 1/2 (right) with gender of buried individuals and prestige items identified inside each grave (graves 5101, 5708 and 7104 had no skeletal remains nor funerary items)

Table 1 Funerary structures at Esfola (EF) and Monte do Bolor 1/2 (MB) with sex and age at death of buried individual and type of associated grave goods (exotic ornaments comprise glass, Egyptian faience, ivory, amber, bone, marine shell, animal teeth and carnelian; artefact types: bb, belt buckle; bd, bead; bg, body grooming instrument; br, bracelet; f, fibula; h, hairpin; r, ring; and w, weapon)

Grave	Buried individual (sex and age)	Ceramic vessels	Exotic ornaments	Gold ornaments	Silver ornaments	Iron artefacts	Copper-based artefacts
EF-4	Adult male	-	-	-	-	w	f
EF-7	Adult individual	-	-	-	-	-	bb
EF-8	Adult male	-	-	-	-	w	-
EF-9	Adult male	-	-	-	-	w	-
EF-10	Adult female	+	-	-	-	-	f
EF-11	Young female	-	+	-	-	-	br; r
EF-12	Adult individual	-	+	-	-	-	bb
EF-13	Young individual	-	-	-	-	-	-
EF-14	Adult male	-	-	-	-	w	f
MB-2607	Adult female	-	+	-	-	-	bd
MB-2712	Adult male	-	-	-	-	-	f
MB-2803	Young male	-	-	-	-	-	-
MB-2913	Adult individual	-	+	+	-	-	-
MB-3107	Adult male	+	-	-	-	-	-
MB-4914	Adult female	+	+	+	+	w	br; f; r
MB-5206	Adult individual	-	-	-	-	bb	-
MB-5511	Adult individual	+	+	-	+	bb	h; bg
MB-5623	Adult female	-	+	-	+	w	br
MB-5634	Adult male	+	-	-	-	w	bb; f
MB-5638	Undetermined	-	+	-	-	w	bd; bg; f
MB-5706	Adult individual	-	-	-	-	-	-
MB-5801	Young individual	+	-	-	-	-	-

depict the high status of female individuals. A distinction of prestige symbols according to gender is a common characteristic of EIA necropolises in southern Portugal (Figueiredo and Mataloto 2017).

The chronological attribution of these necropolises to the Early Iron Age results from the characteristic funerary architecture and distinctive set of grave goods of this period (Soares et al. 2017). Some funerary items recovered at Esfola and Monte do Bolor 1/2 are particularly important to narrow the chronology of both sites, namely, Tartesic belt buckles and particular types of fibulae and bracelets (Fig. 3). Tartesic belt buckles (EF-087/088, EF-316/317 and MB-5609–1) are attributed to the seventh to sixth century BC (Cerdeño Serrano 1981). Geographically close parallels can be found at the EIA necropolises of Palhais (Santos et al. 2009) and Olival do Senhor dos Mártires (Gomes 2015). The typologies of fibulae recovered at Esfola and Monte do Bolor 1/2 are characteristic of the same chronological period (Ponte 2006), namely, the types Alcores (EF-063), Acebuchal (EF-320), a possible Ponte 3a (MB-5610–1/2), Ponte 3b (MB-2721–1) and Ponte 9a/1 (MB-4913–1). Heart-shaped bracelets (EF-144 and EF-145; two heart-shaped silver bracelets were also recovered in grave 5623 of Monte do Bolor 1/2) also belong to the seventh to sixth century BC. These bracelets show parallels at the EIA necropolises of Vinha das Caliças 4, Beja (Arruda et al. 2016), Talavera la Vieja, Cáceres (Jiménez Ávila 2006) and Medellín, Badajoz (Almagro Gorbea et al. 2008). Finally, the bracelets and rings decorated with a cylindrical bead (EF-196, MB-4911–1, MB-4911–2, EF-208 and MB-4904–1) reinforce the relation and contemporaneity of the two necropolises. This particular type of ornament has a very close parallel in the coeval necropolis of Favela Nova, Ourique (Dias and Coelho 1983).

Methodology

The study comprised a microanalytical methodology to prevent further damage to those cultural items already heavily corroded by the burial environment. The different components of composite artefacts were analysed whenever possible, as the preparation of items for elemental analyses involved the removal of the corrosion layer on a small area (*c.* 5 mm diameter) avoiding decorated surfaces and other significant areas. Metal surfaces were cleaned with SiC papers (P1000, P2500 and P4000 grit size) and polished with diamond pastes (3 µm and 1 µm grit size). The efficiency of

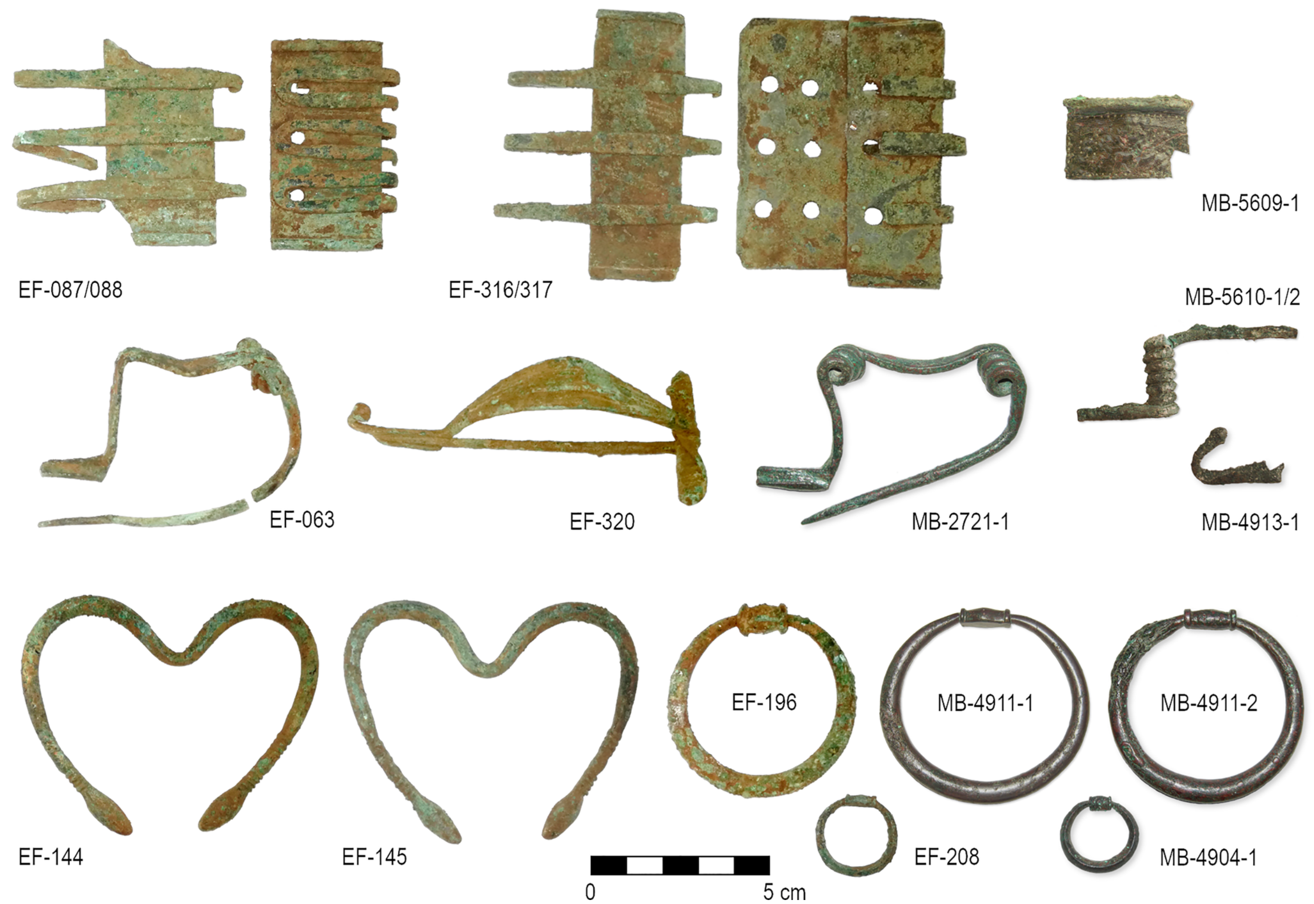

Fig. 3 Copper-based funerary items of necropolises of Esfola and Monte do Bolor 1/2 (Tartesic belt buckles, EF-087/088, EF-316/317 and MB-5609–1; fibulae, EF-063, EF-320, MB-2721–1, MB-5610–1/2 and MB-4913–1; bracelets, EF-144, EF-145, EF-196, MB-4911–1 and MB-4911–2; and rings, EF-208 and MB-4904–1)

the cleaning process was ascertained by optical microscopy observations in a Zeiss Discovery V20 stereomicroscope.

The elemental composition of most artefacts was determined by micro-energy dispersive X-ray spectrometry (micro-EDXRF). The ArtTAX Pro spectrometer utilised is equipped with a 30 W Mo X-ray tube, an electro-thermally cooled Si drift detector (FWHM of 160 eV at 5.9 keV) and focusing polycapillary lens enabling the analysis of an area with *c.* 70 µm diameter (Bronk et al. 2001). Artefacts were analysed in three spots with optimum measurement conditions of 40 kV of potential difference, 600 µA of current intensity and 120 s of live time. The quantification was made with WinAxil software including experimental calibration factors calculated with the analysis of two reference materials: British Chemical Standards Phosphor Bronze 551 and BNF Metals Technology Centre Leaded Bronze C50.01. The quantification of the British Chemical Standards Phosphor Bronze 552 evaluated the technique accuracy as better than 10%. The quantification limit (L_Q) was considered equal to $10 \times \sigma_Q$, where the σ_Q was estimated as equal to $background^{1/2}$ (Currie 1999). Therefore, the quantification limits of interest elements are 0.05 wt% Fe, 0.10 wt% Ni, 0.10 wt% As, 0.50 wt% Sn and 0.10 wt% Pb (Valério et al. 2015).

An additional set of artefacts non-previously analysed (MB-4913–1, MB-5610–1, MB-5610–2, MB-5611–2 and MB-5507–1) was characterised by micro-particle-induced X-ray emission (micro-PIXE). Analyses were conducted in the external microbeam line of the 2.5 MV Van de Graaff accelerator from IST, and samples were irradiated with a 2 MeV proton beam. The external beam end-stage comprises an OM150 triplet quadrupole lenses able to focus the 2 MeV proton external beam in a spot on object surface of $60 \times 70\ \mu m^2$ (Corregidor et al. 2011). The external microbeam was used for scanning the sample surface over an area of about $1 \times 1\ mm^2$, which is imposed by the exit nozzle silicon frame window dimensions and that is covered with a 100-nm-thick silicon nitride membrane (Corregidor et al. 2011). The induced X-rays were collected by a 30 mm^2 SDD detector with 180 keV resolution positioned at 135° with the beam direction while using a 50-µm-thick mylar filter in front. Artefacts were analysed in three cleaned spots selected inside the scanned area (i.e. spots with less amount of low Z elements) and the elemental quantification performed by GUPIXWIN software without self-normalisation (Campbell et al. 2010). As mentioned earlier, the technique accuracy for this type of sample is comparable to the one of the micro-EDXRF technique, especially for the discussed elements (i.e. elements with medium or high atomic number).

After the analytical characteriation, the exposed metal area of artefacts was protected with a benzotriazole corrosion inhibitor (3% m/v in ethanol) and a Paraloid B-72 acrylic polymer (10% m/v in ethanol). Finally, the coloration of the surrounding patina was replicated with a mixture of pigments dissolved in the mentioned acrylic polymer solution.

Results of funerary items from Esfola

Artefacts are composed by copper–tin alloys with minor amounts of lead, iron and, sometimes, arsenic (Table 2). The average tin content of these copper–tin alloys (6.5 ± 2.2 wt% Sn) points to low-tin bronzes, and only the ring EF-178 has a "suitable" amount of alloying element (11.8 wt% Sn). The belt buckle EF-316/317 rivets are on the lower end with very low amounts of tin (2.8–3.1 wt% Sn) being a distinctive characteristic of this Tartesic belt buckle because the remaining components have higher tin contents (5.7–7.5 wt% Sn). Conversely, all components of the Tartesic belt buckle EF-087/088 are composed of comparable bronze alloys (6.4–8.4 wt% Sn). The same is true for individual components of other artefacts from Esfola, namely, the Alcores fibula EF-063 (4.1–4.6 wt% Sn), Acebuchal fibula EF-320 (8.3–8.7 wt% Sn) and fibula EF-113 (8.5–9.7 wt% Sn).

Generally, the distinct artefact types at Esfola do not display a clear compositional difference, although the belt buckles have bronze alloys in the mid-range (*c.* 6–8 wt% Sn), while the bracelets feature low-range tin contents (*c.* 4–6 wt% Sn). In this regard, it should be emphasised that the enhanced castability of high-tin bronzes would be an advantage for casting large or massive artefacts. However, the Tartesic belt buckle EF-316/317 is the only item suggesting a

Table 2 Composition of copper-based artefacts from Esfola (values in wt%; n.d., not detected; AP, active piece; PP, passive piece; PPE, passive piece extension)

Artefact	Component	Reference	Cu	Sn	Pb	As	Ni	Fe
Belt buckle (Tartesic)	Plate (AP)	EF-087a	90.5	8.4	0.24	n.d	n.d	0.25
	Hook (AP)	EF-087b	92.3	7.0	0.17	n.d	n.d	0.27
	Rivet (AP)	EF-087c	91.6	7.6	0.16	n.d	n.d	0.28
	Plate (PP)	EF-088a	92.7	6.5	0.14	n.d	n.d	0.28
	Hook (PP)	EF-088b	92.2	6.9	0.18	n.d	n.d	0.27
	Rivet (PP)	EF-088c	91.9	7.3	0.17	n.d	n.d	0.27
Belt buckle (Tartesic)	Plate (AP)	EF-316a	91.1	6.9	0.73	n.d	n.d	0.79
	Hook (AP)	EF-316b	92.4	5.7	0.68	n.d	n.d	0.72
	Rivet (AP)	EF-316c	95.9	3.1	0.32	n.d	n.d	0.26
	Plate (PP)	EF-317a	90.4	7.4	0.75	n.d	n.d	0.79
	Hook (PP)	EF-317b	91.3	6.5	0.83	n.d	n.d	0.75
	Rivet (PP)	EF-317c	96.2	2.8	0.33	n.d	n.d	0.26
	Plate (PPE)	EF-317d	91.0	6.8	0.89	n.d	n.d	0.76
	Rivet (PPE)	EF-317e	96.2	2.8	0.30	n.d	n.d	0.26
Fibula (Alcores)	Main body	EF-063a	93.5	4.6	0.54	n.d	n.d	0.59
	Pin	EF-063b	94.7	4.1	0.33	n.d	n.d	0.31
Fibula (Acebuchal)	Main body	EF-320a	89.9	8.7	0.15	n.d	n.d	0.60
	Axle	EF-320b	90.7	8.3	0.13	n.d	n.d	0.24
Fibula (undetermined)	Pin	EF-113a	89.4	9.7	< 0.10	n.d	n.d	0.11
	Spring	EF-113b	90.7	8.5	< 0.10	n.d	n.d	0.13
	Axle	EF-113c	90.2	9.0	< 0.10	n.d	n.d	0.14
Bracelet (heart-shaped)	Main body	EF-144	92.1	5.4	0.63	< 0.10	n.d	0.25
Bracelet (heart-shaped)	Main body	EF-145	93.8	5.0	0.43	< 0.10	n.d	0.21
Bracelet (decorated)	Main body	EF-196	94.7	4.5	< 0.10	n.d	n.d	0.42
Ring (decorated)	Main body	EF-208	94.6	4.1	< 0.10	n.d	n.d	1.0
Ring	Main body	EF-143	90.6	8.1	0.25	n.d	n.d	0.50
Ring	Main body	EF-178	87.2	11.8	0.24	< 0.10	n.d	< 0.05
Rod	Main body	EF-321	93.7	4.6	0.36	< 0.10	n.d	1.0

possible selection of bronze alloys to manufacture components with distinct functions.

Results of funerary items from Monte do Bolor 1/2

The core of the collection was previously analysed and presented to the *2nd Congress of the Portuguese Archaeologists Association* (Valério et al. 2017, in Portuguese). However, only now it was possible to analyse the remaining set made up of a handful of other artefacts (MB-4913–1, MB-5610–1, MB-5610–2, MB-5611–2 and MB-5507–1). The overall collection shows a high compositional diversity with copper, copper–arsenic (As > 2 wt%), copper–tin (Sn > 2 wt%) and copper–tin–lead alloys (Sn > 2 wt% and Pb > 2 wt%) (Table 3). Most items are composed of copper–tin alloys with a low average tin content (8.4 ± 2.2 wt% Sn), highlighting the spatula MB-5509–6 made of a high-tin bronze (14.3 wt% Sn). The leaded bronze alloy is represented by a unique specimen (stylus MB-5603–5) with 2.7 wt% Pb. In southern Portugal, leaded bronzes emerge during the EIA with examples in the ninth to seventh century BC Phoenician settlement of Quinta do Almaraz (Valério et al. 2012), seventh century BC site of Alcácer do Sal (Schiavon et al. 2013) and fifth century BC settlement of Cabeço Redondo (Valério et al. 2015). The higher lead content enhances the castability, thus improving the production of complicated shapes or larger objects. The spatula of Monte do Bolor 1/2 does not fit this pattern, so its composition could simply result from an unintended use of a lead-rich raw material. A similar situation might explain the tin and lead contents of the single example of copper: the bracelet MB-4911–1 (1.7 wt% Sn and 1.0 wt% Pb) and corresponding bead MB-4911-1a (1.5 wt% Sn and 0.69 wt% Pb). The low lead and tin contents of this decorated bracelet seem to suggest the recycling of a leaded bronze raw material.

The remaining items from Monte do Bolor 1/2 are five small beads composed of arsenical copper alloys (5.6–6.5 wt% As). In southwestern Iberian Peninsula, the copper with high arsenic content is common in Chalcolithic and Middle Bronze Age contexts but become almost absent among later contexts of Late Bronze Age and Early Iron Age (Hunt-Ortiz 2003). Therefore, the use of a distinct raw material (e.g. arsenic-rich copper ore or ancient artefact) and/or a foreign origin must be taken in consideration. The recycling of arsenical copper objects dating to the Early and Middle Cypriot period has been suggested to explain the presence of arsenic in LBA metallurgical ceramics from Cyprus (Ioannides et al. 2016). On the other hand, an importation from the Mediterranean region is another hypothesis due to the necklace of glass beads also recovered in the grave of that female individual. In the Mediterranean region, the trade of glass ornaments subsists since the LBA (Henderson et al. 2010), while long distance contacts are well-known

Table 3 Composition of copper-based artefacts from Monte do Bolor 1/2 (values in wt%; n.d., not detected; MB-4913–1, MB-5610–1, MB-5610–2, MB-5611–2 and MB-5507–1 refer to new results; remaining data from Valério et al. 2017)

Artefact	Component	Reference	Cu	Sn	Pb	As	Ni	Fe
Fibula (3b type)	Main body	MB-2721–1	94.5	5.0	0.18	< 0.10	0.12	0.15
Fibula (9a/1 type)	Pin rest	MB-4913–1	90.3	7.0	0.28	0.39	n.d	1.8
Fibula (“3a” type)	Spring	MB-5610–1	92.4	6.8	n.d	< 0.10	< 0.10	0.37
	Axle	MB-5610–2	91.8	7.5	n.d	0.15	n.d	0.17
Fibula (undetermined)	Pin	MB-5611–2	88.8	9.5	n.d	< 0.10	< 0.10	0.43
Bracelet (decorated)	Main body	MB-4911–1	96.9	1.7	1.0	n.d	n.d	0.37
	Bead	MB-4911-1a	97.3	1.5	0.69	n.d	n.d	0.41
Bracelet (decorated)	Main body	MB-4911–2	91.7	7.6	0.15	n.d	n.d	0.51
	Bead	MB-4911-2a	91.9	7.6	0.16	n.d	n.d	0.53
Ring (decorated)	Main body	MB-4904–1	90.1	9.3	0.14	n.d	n.d	0.49
	Bead	MB-4904-1a	91.9	7.8	0.15	n.d	n.d	0.11
Tweezers	Main body	MB-5509–1	90.0	9.2	< 0.10	0.16	0.25	0.34
Spatula	Main body	MB-5509–6	85.1	14.3	0.10	n.d	n.d	0.42
Ring	Main body	MB-5509–8	90.9	8.4	0.23	0.25	0.16	< 0.05
Stylus	Main body	MB-5603–5	93.9	3.0	2.7	0.19	n.d	0.08
Hairpin	Main body	MB-5507–1	86.9	9.7	n.d	< 0.10	< 0.10	0.14
Bead	Main body	MB-2605–1	94.3	n.d	n.d	5.6	n.d	< 0.05
Bead	Main body	MB-2605–2	93.2	n.d	0.19	6.5	n.d	0.08
Bead	Main body	MB-2605–3	94.0	n.d	0.11	5.8	n.d	0.05
Bead	Main body	MB-2605–4	93.9	n.d	0.15	5.9	n.d	< 0.05
Bead	Main body	MB-2605–5	94.2	n.d	n.d	5.8	n.d	< 0.05

in EIA southwestern Iberian Peninsula (see, for instance, Murillo-Barroso et al. 2015).

Composition of funerary items from southern Portugal

Apart from Esfola and Monte do Bolor 1/2, the site of Palhais is the only other EIA necropolis in southern Portugal with analysed metals. The necropolis of Palhais comprised two burials with a distinctive set of EIA items such as a Tartesic belt buckle, an Alcores fibula and two sets of body grooming instruments (Santos et al. 2009). The composition of these funerary items resembles the Esfola collection, being entirely composed of low-tin bronze alloys (4.4 ± 2.4 wt% Sn) with minor contents of lead, arsenic and iron (Valério et al. 2013). Therefore, the collection of copper-based funerary artefacts of southern Portugal (56 items) is mostly constituted by binary bronzes (85.7% frequency), while the remaining types are only minimal, namely, the copper–arsenic alloys (8.9%), copper (3.6%) and copper–tin–lead alloys (1.8%). The tin content is unrelated with artefact type; for instance, fibulae components range from 4.1 to 9.7 wt% Sn, while body grooming instruments show an even higher span with bronzes from 2.0 to 14.3 wt % Sn (Fig. 4). Similar research considered that the variable bronze alloy composition of Iron Age religious/ritual artefacts could either indicate the use of uncontrolled alloying processes or that this particular type of objects was not subjected to a specific recipe (Oudbashi and Hessari 2017; Oudbashi and Hasanpour 2018).

Binary bronze alloys of EIA funerary items of southern Portugal show a Gaussian-like distribution of tin contents centred in the average value of 6.7 ± 2.5 wt% Sn, n = 48 (Fig. 5). The higher number of samples in the low-tin section implies the relative importance of recycling activities, as the remelting of scrap decreases the tin content of the alloy. Recently, a few crucibles with recognisable imprints from fragments of twisted wire, fibulae and plates established the remelting of bronze scrap in Northwestern Europe during the Iron Age (Huisman et al. 2020), but such fortunate evidence is yet unknown in southern Portugal. The ninth to eighth century BC indigenous settlement of Castro dos Ratinhos displayed exclusively binary bronzes with "suitable" tin contents (10.1 ± 2.5 wt% Sn, n = 37), while the ninth to seventh century BC Phoenician settlement of Quinta do Almaraz showed a diversified metallurgy composed by copper, leaded bronze and binary bronze, the former with low-tin contents (5.4 ± 2.0 wt% Sn, n = 15). Therefore, the pattern evidenced by those seventh to sixth century funerary items seems to be a combination of local tradition with Phoenician knowledge. Apart from the production of low-tin bronzes, Phoenician advances include the use of a more diversified metallurgy with copper and leaded copper, similarly to what

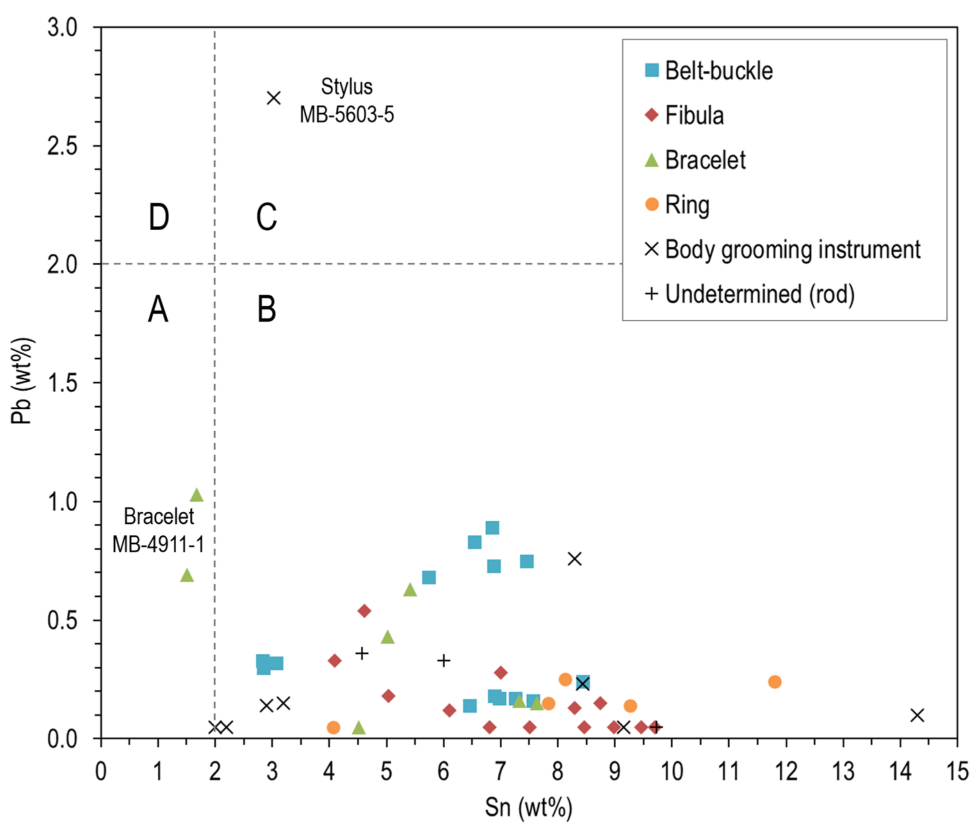

Fig. 4 Plot of tin content versus lead content in funerary items of Esfola, Monte do Bolor 1/2 and Palhais (A, copper; B, copper–tin alloy; C, copper–tin–lead alloy; D, copper-lead alloy)

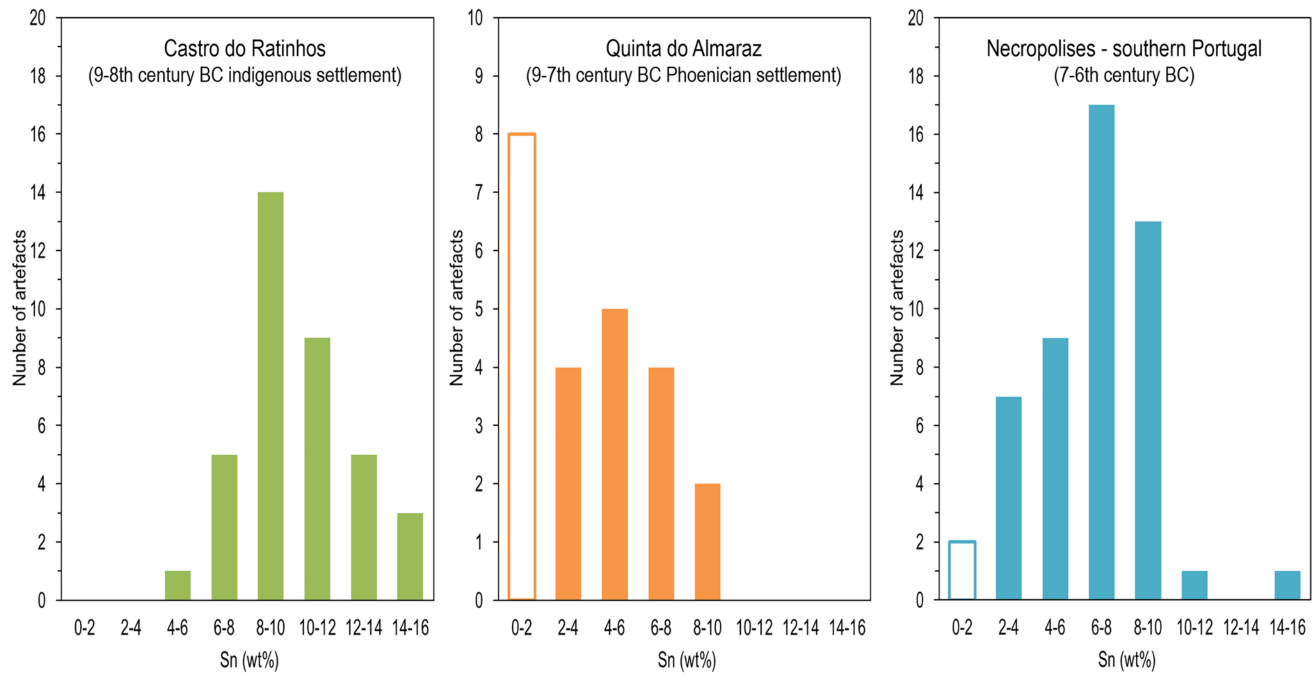

Fig. 5 Histograms of tin contents in collections of Castro dos Ratinhos (Valério et al. 2010), Quinta do Almaraz (Valério et al. 2012) and EIA necropolises in southern Portugal (Esfola, Monte do Bolor 1/2 and Palhais)

was identified in those necropolises. For the moment, a comparison with artefacts of coeval domestic contexts is unfeasible due to the absence of analysed artefacts from the few known coeval settlements in southern Portugal.

Another evidence of Phoenician technology concerns the iron content of artefacts. Craddock and Meeks (1987) were the first to acknowledge the distinct iron compositional patterns in artefacts of LBA and Phoenician communities of Iberian Peninsula. This feature was later considered a technological indicator of the metallurgy in the Mediterranean region (Ingo et al. 2006; Jiménez Ávila 2015). The increased iron content was attributed to the highly reducing atmosphere of Iron Age smelting operations. However, it is thought that these efficient smelting operations would be carried out in furnaces, while during such period, the smelting in ceramic crucibles running with moderately reducing conditions has been recorded in the Iberian Peninsula (Rovira and Montero-Ruiz 2013; Farci et al. 2017). An alternative explanation concerns the pickup of iron from metallurgical tools made of iron. The iron incorporation in metallic copper originates a raw metal that could be refined down to iron contents of about 0.5 wt%, but additional purification would be hard and would not bring mechanical advantages (Northover 2004). If the origin of iron is still a matter of debate, there is definitively a consistent increase in artefacts from later contexts. Some examples of LBA collections with low iron contents (<0.05 wt% Fe) include Baiões (Figueiredo et al. 2010), Freixanda (Gutiérrez Neira et al. 2011), Casais da Pedreira, Moinho do Raposo and Vila Cova de Perrinho (Bottaini et al. 2012). The feature is replicated in EIA artefacts of indigenous contexts such as Moita da Ladra (Valério et al. 2016) and Castro dos Ratinhos (Valério et al. 2010). On the contrary, the iron content is systematically higher (up to 2 wt% Fe) in metals of Mediterranean contexts such as the Phoenician sites of Quinta do Almaraz (Valério et al. 2012) and Morro de Mezquitilla (Giumlia-Mair 1992). EIA funerary items of southern Portugal fit well into the pattern of increased iron content, with a few exceptions with especial emphasis for the arsenical copper beads of Monte do Bolor 1/2.

The colour of funerary items from southern Portugal

The colour could be a differentiating characteristic for highly valued artefacts such as funerary items used to portray the status of buried individuals. The CIELAB (L*a*b*) colour space is a widely used system, where the L* value represents the lightness, the a* value defines the red and green ratio, and the b* describes the yellow and blue proportion (McLaren 1976). These variables can be used to evaluate the perception by the human eye of a colour difference between two alloys with the ΔE value (Eq. 1). The colour distinction starts at ΔE values of 2, a clear difference occurs at a value of 3.5 and for values higher than 5 the two colours are perceived as being different (Mokrzycki and Tatol 2011):

$$\Delta E = \sqrt{\left(L_1^* - L_2^*\right)^2 + \left(a_1^* - a_2^*\right)^2 + \left(b_1^* - b_2^*\right)^2} \quad (1)$$

Fang and McDonnell (2011) studied the effect of tin in the colour of bronze by measuring the L*, a* and b* values in a useful range of copper–tin alloys. Mecking (2020) also measured the CIELAB colour space of a comprehensive series of binary and ternary copper alloys. Both studies came to similar conclusions regarding the perception by the human eye of a colour distinction between two alloys, which can be used to evaluate the colour scheme of our funerary items. Considering as the starting point the measured alloy closer to the average tin content of our collection, the L*, a* and b* measurements of both studies show a linear variation of the ΔE value for both low-tin and high-tin alloys (see Supplementary Table 1). The average variation of the ΔE can be used to estimate the colour distinction with reference to the average tin content of our set of funerary items (ΔE = 0 for 6.4 wt% Sn, Fig. 6). From these plots it can be estimated that, for bronzes with tin contents below the average value of the analysed set, the colour distinction begins at about 4 wt% Sn (ΔE = 2) and becomes evident bellow *c.* 2.5 wt% Sn (ΔE = 3.5). For bronzes with higher tin contents, the colour change is less pronounced, starting to be perceived from *c.* 11 wt% Sn and becoming evident above *c.* 14 wt% Sn.

Thereafter, the linear colour deviation with reference to the average tin content of the collection (red lines in Fig. 6) was used to estimate the ΔE value of each artefact based on the tin content, establishing that most of EIA copper-based funerary items of southern Portugal would have an analogous colour, while the higher number of exceptions comprises body grooming instruments (Fig. 7). The Tartesic belt buckle EF-316/317 is the only composite artefact showing components (the rivets) with a distinguishing colour. This ornamental trait was not used in bracelets and rings decorated with a bead, suggesting that colour was not a valued feature among copper–tin ornaments.

Other examples in our collection can reveal a different pattern, namely, the leaded bronze stylus (MB-5603–5, 3.0 wt% Sn and 2.7 wt% Pb) and the arsenical copper necklace beads (MB-2605–1/2/3/4/5, 5.6–6.5 wt% As). The overall effect of lead in the colour of bronze was considered to be minimal (Mecking 2020), although there are some deviations for low-tin alloys since the ΔE value equals 5.3 for a 2.0 wt% Pb addition to a 2 wt% Sn alloy (Supplementary Table 1). Moreover, colorimetric measurements in the Cu–As–Sn ternary system (Radivojević et al. 2018) allow to estimate a ΔE value of about 9 when comparing a Cu-6wt%As alloy with a Cu-6wt% Sn alloy (Supplementary Table 1). Accordingly, these distinct copper-based alloys would certainly have a distinguishing colour among the usual EIA copper-based funerary items. Finally, it is worth noting that these considerations should be regarded with caution as daily-life objects were not used in pristine conditions and a colour change over time is to be expected due to polishing, corrosion and wear. Moreover, the manufacturing operations such as casting and annealing also have a small influence in the colour of copper alloys (Fang and McDonnell 2011).

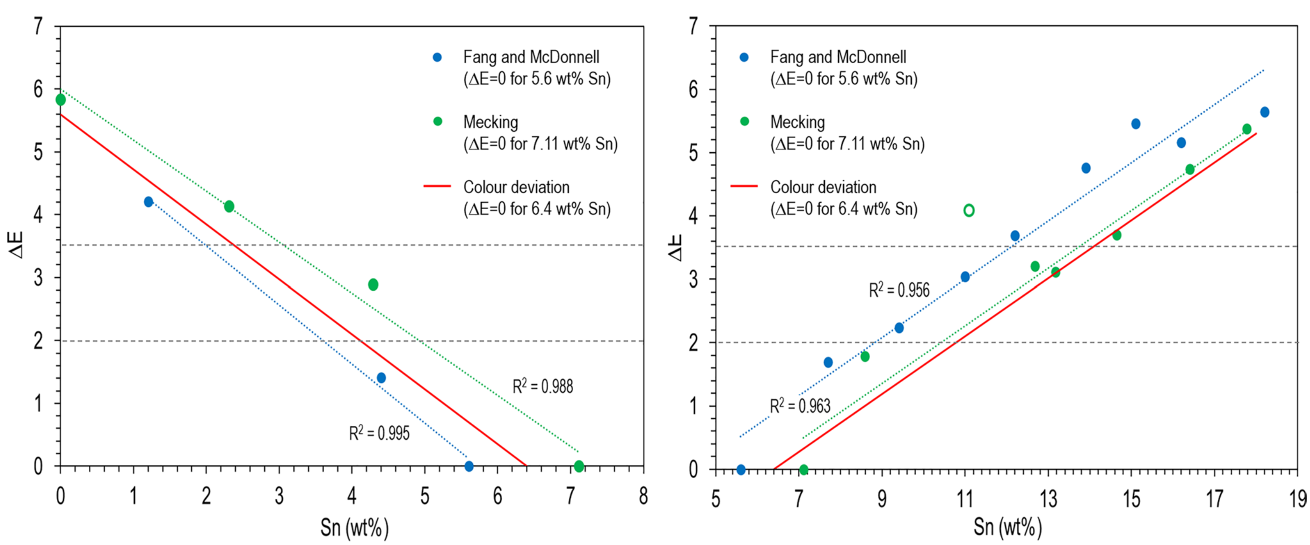

Fig. 6 Plots of tin content versus ΔE value displaying the evolution of the colour deviation from the average tin content of the set of EIA funerary items of southern Portugal (L*, a* and b* data from Fang and McDonnell (2011) and Mecking (2020))

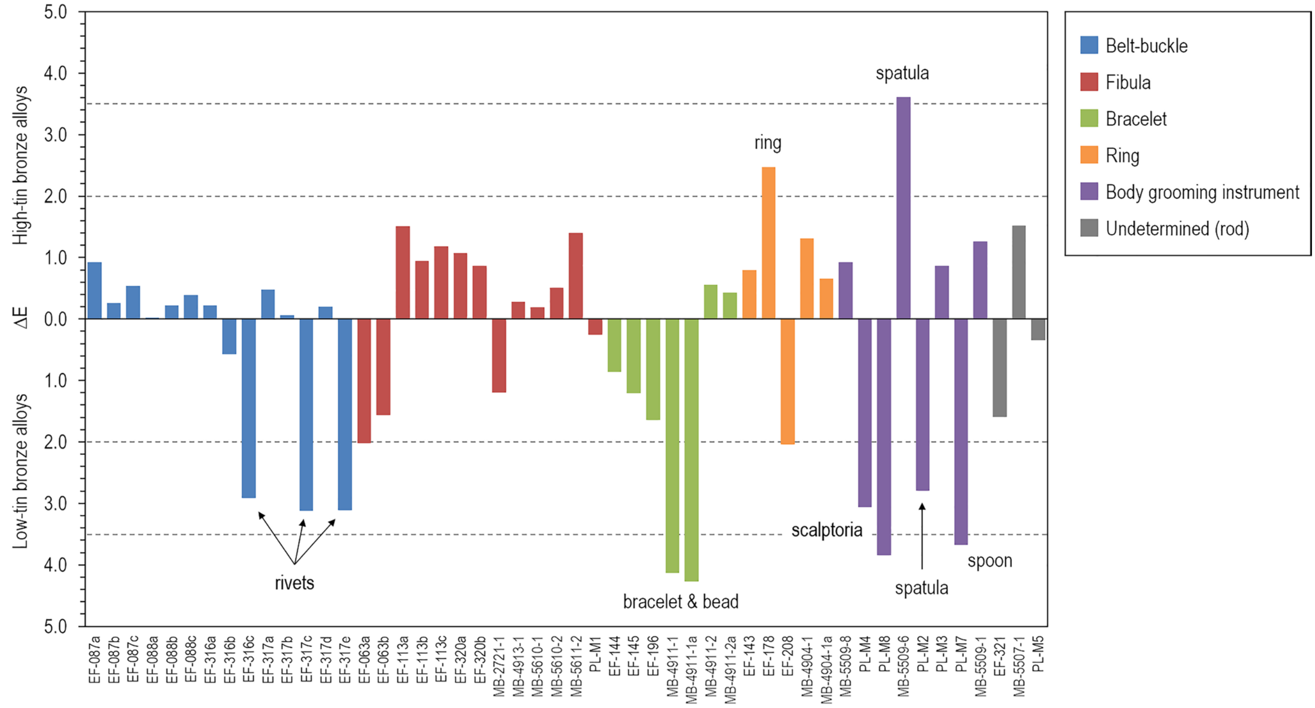

Fig. 7 Plot of ΔE value for the set of EIA funerary items of southern Portugal (ΔE calculated with the colour deviation from the average tin content of the set of EIA funerary items of southern Portugal (ΔE = 0 for 6.4 wt% Sn, see Fig. 6)

Conclusions

The copper-based funerary items of EIA necropolises in southern Portugal disclose numerous symbols of status such as distinctive belt buckles, fibulae, bracelets and rings, in addition to instruments related with the Mediterranean tradition of body grooming. These grave goods compete with other prestige symbols, such as iron weapons and jewels made of exotic or noble materials, all of them establishing a characteristic material culture that unites communities inhabiting the region during the second quarter of the first millennium BC.

Those funerary items reveal a metallurgy centred in low-tin bronze alloys, but with some exceptions made of copper, leaded bronze and arsenical copper. The last category is especially rare in EIA contexts of Iberian Peninsula; thus, those small beads made of arsenical copper clearly stand out from the rest. On the other hand, the common use of low-tin bronzes for funerary items follows the pattern introduced by Phoenicians in the ninth to seventh century BC domestic contexts, but a comparison with coeval domestic sites requires further archaeological research.

The identified compositional model lacks a clear relation between alloy and artefact type. Moreover, the distinct components of composite objects show a similar composition, except for the rivets of a Tartesic belt buckle that may evidence a unique case of raw material selection. Apart from the arsenical copper beads and a leaded bronze stylus, most of the remaining set would have an indistinguishable colour. The few examples that could have been perceived as being more reddish (lower tin content) or more golden (higher tin content) are mostly body grooming instruments. However, there is no evidence of intentionality in the production of such items, and some low-tin bronze alloys may result from the recycling of bronze scrap.

Generally, these funerary items reveal that EIA metallurgists did not take advantage of the possibility of obtaining different colours. Other metals such as gold and silver probably filled this role, constituting important symbols of a differentiated hierarchy among the community. All those luxury items, locally made or imported from distant regions, constitute an evidence of an increasingly hierarchical society that followed the establishment of Orientalising contacts in southern Portugal during the ninth century BC.

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Data availability All relevant data are within the manuscript.

Declarations

Conflicts of interest The authors declare no competing interests.

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