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ORIGINAL PAPER

New evidence on Iron Age bronze metallurgy in southwestern Iberian Peninsula: ingots and artefacts from Cabeço Redondo (Portugal)

Pedro Valério[1] · Rui M. Soares[2] · Rui J. C. Silva[3] · António M. Monge Soares[4] · Maria Fátima Araújo[1]

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Abstract
Archaeological works at Cabeço Redondo (southern Portugal), located in the Guadiana River basin, identified the remains of a monumental building with an architecture, ceramics and metal artefacts in line with post-Orientalizing contexts of the sixth–fifth century BC in southwestern Iberian Peninsula. Metal debris connected to the copper-based metallurgy include a significant collection of ingots, lumps and small prills, with emphasis to a massive plano-convex ingot, while artefacts comprise tools, small implements, ornaments and rods. A chemical and microstructural study involving micro-EDXRF, optical microscopy and SEM–EDS provided some answers about foundry activities and identified the composition and post-casting manufacture of artefacts. Most metal debris were composed of pure copper (> 99 wt% Cu), although some examples attest the use of bronze and leaded bronze at Cabeço Redondo metallurgical workshop. Apart from a few copper items, the artefact collection mainly shows low-tin bronze alloys (7.6 ± 3.9 wt% Sn) and leaded bronze alloys (7.7 ± 4.4 wt% Sn and 6.0 ± 3.4 wt% Pb). The manufacture of copper and binary bronze artefacts included hammering and annealing, while leaded bronzes were usually not subjected to post-casting work, implying a well-defined relation among function, composition and manufacture. The features of metal debris and artefacts were then compared with the ones of coeval sites of this region to integrate the metallurgical evidence of Cabeço Redondo into the technological pattern of southwestern Iberian Peninsula during the middle of the first millennium BC.

Keywords Bronze · Ingots · Chemical composition · *Chaîne opératoire* · Iron Age · Iberian Peninsula

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

[1] Centro de Ciências e Tecnologias Nucleares (C2TN), Departamento de Engenharia e Ciências Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Campus Tecnológico e Nuclear, Estrada Nacional 10 (km 139,7), 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 Lisbon, Portugal

[3] CENIMAT/i3N, Department of Material Science, NOVA School of Science and Technology, NOVA University of Lisbon, 2829-516 Caparica, Portugal

[4] Centro de Ciências e Tecnologias Nucleares (C2TN), Instituto Superior Técnico, Universidade de Lisboa, Campus Tecnológico e Nuclear, Estrada Nacional 10 (km 139,7), 2695-066 Bobadela LRS, Portugal

Introduction

The Phoenician endeavour in Western Mediterranean triggered important cultural and technological improvements in Iberian Peninsula since the ninth–eighth century BC giving rise to the so-called Orientalizing Period (Arruda 2019). Phoenician seashore settlements displayed a diversified non-ferrous metallurgy including binary and ternary bronze alloys, in addition to unalloyed copper and copper-lead alloys (Giumlia-Mair 1992; Renzi et al. 2009; Valério et al. 2012). The inland communities of southern Portugal mirrored this metallurgical pattern, as it is evidenced by the funerary items of local necropolises from the seventh to sixth century BC (Valério et al. 2021). Then, the collapse of Eastern Mediterranean influence during the sixth century BC gave rise to modifications in the settlement pattern, burial practices and economic activities in Iberian areas exposed to the Orientalizing stimulus (Neville 2007).

The Iron Age metal production in Iberian Peninsula seems to follow the long-established crucible technology, mainly due to the absence in the archaeological record of smelting furnaces and corresponding slags, the so-called tap-slags. However, the increasingly common occurrence of large copper ingots, namely of plano-convex specimens, suggests an evolved technology like the smelting in slag-tapping furnaces (Renzi and Rovira 2015). So far, there is no evidence of the local production of those ingots, but there are many proofs of an important trade network of small and large ingots along the Mediterranean since the 2nd half of 2nd millennium BC (Emanuel 2019; Sabatini and Lo Schiavo 2020; Yahalom-Mack et al. 2014).

The discovery of a significant collection of metal debris and artefacts during archaeological works at the late sixth–fifth century BC site of Cabeço Redondo, located in the Middle Guadiana River basin (southern Portugal), provided an excellent opportunity to widen the knowledge on metal production and use in SW Iberian Peninsula. Metal debris include ingots (a 6.4 kg plano-convex fragmented specimen, a small plano-convex ingot and an irregular-shaped fragment still with a flat surface), lumps and prills. Artefacts comprise ornaments, tools, small implements and rods. Additionally, a significant amount of metallurgical waste was recorded (a ceramic crucible/mould, tuyeres and slags) indicating the existence of a local workshop (Valério et al. 2015). The chemical and microstructural characterisation of metal debris involved micro-EDXRF and SEM–EDS analyses, while the alloy composition and manufacture of artefacts were determined by micro-EDXRF and optical microscopy. The results on metal production and use were then integrated into the knowledge about the Orientalizing and post-Orientalizing Iron Age metallurgical pattern of SW Iberian Peninsula.

Archaeological framework

The post-Orientalizing site of Cabeço Redondo is located inland in southern Portugal (Fig. 1), in the Middle Guadiana River basin, as with most culturally and chronologically similar places in southwestern Iberian Peninsula. This type of archaeological sites is believed to have been part of a process that began during the eighth/seventh century BC with the disappearance of the large Late Bronze Age settlements and the development of small rural occupations along the banks of the Guadiana River. From the end of the sixth century BC/beginning of the fifth century BC, some monumental building complexes arise, resulting from the growth of those rural occupations. This development had its heyday at the 2nd half of the fifth century BC, when the productive capacity would be much higher than at the foundation time, as it is evidenced by a substantial volume of surplus production. Moreover, large amounts of amphorae and other containers, in addition to an abundant domestic fauna, allow inferring the importance of agriculture and grazing in the economy of these places (Soares and Soares 2016).

During 1942, Cabeço Redondo was identified and classified as an artificial hill (Lima 1988), but it was forgotten until 1990, when the construction of an irrigation structure led to the destruction of a large part of the archaeological contexts with the scatter of the artificial mound over the surrounding area. Some materials were collected and transported to the Moura Museum, namely ceramics, lithics and a large leaded bronze pivot (CR004) of a potter's wheel.

The site was subjected to an archaeological survey during 2011, identifying adobe walls with a stone base, compacted clay floors, fireplaces and a possible ditch, which suggest the existence of a monumental building (Cardoso and Soares 2013; Soares 2012, 2017, 2021; Soares et al. 2013; Soares and Soares 2016). Moreover, the shapes and decoration styles of pottery from Cabeço Redondo point to a chronology of the late sixth–fifth century BC, showing many parallels in coeval contexts of southwestern Iberian Peninsula, namely at Castro da Azougada, Cancho Roano, La Mata, El Castañuelo, Herdade da Sapatoa (Redondo) and Castelão da Horta das Nogueiras (Borba) (Mataloto 2004). A collection of complete ceramic containers fragmented in situ stands out, suggesting that the occupation of the building came to an abrupt end. The abandonment of everyday objects in such context dated to the end of the fifth century BC is indicative of an intentional abandonment of the building and shows parallels in coeval post-Orientalizing SW Iberian sites. The best-known examples are Castro da Azougada (Moura) (Antunes 2009; Soares 2017), El Castañuelo (Aracena) (Amo 1978; Soares 2017), Casas del Turuñuelo (Badajoz) (Celestino Pérez and Rodríguez González 2019), Cancho Roano (Badajoz) (Celestino Pérez 1996; Celestino Pérez and Jiménez Ávila 1993) and La Mata (Badajoz) (Rodríguez Díaz 2004) (Fig. 1).

Metallurgical waste recovered at Cabeço Redondo during the first survey comprise a ceramic crucible/mould fragment with copper and tin oxides (delafossite, cuprite and cassiterite), a tuyere with copper prills, a small rod (CR001), a moon-crescent bronze pendant ("xorca" pendant, CR002) still with the casting core and a brazier handle (CR003) (Valério et al. 2015). During 2017, a second archaeological field campaign confirmed the existence of an architecture similar to other large buildings of the sixth–fifth century BC in the Middle Guadiana River basin, of which Cancho Roano and La Mata are perhaps the best-known examples. The archaeological survey included a metal detector prospecting on sediments scattered by the destruction of the artificial hill, having identified a large set of metal debris that indicate the existence of important metallurgical activities at Cabeço Redondo. Overall, the 2017 archaeological field campaign recovered a total of 53 metallurgical remains and copper-based

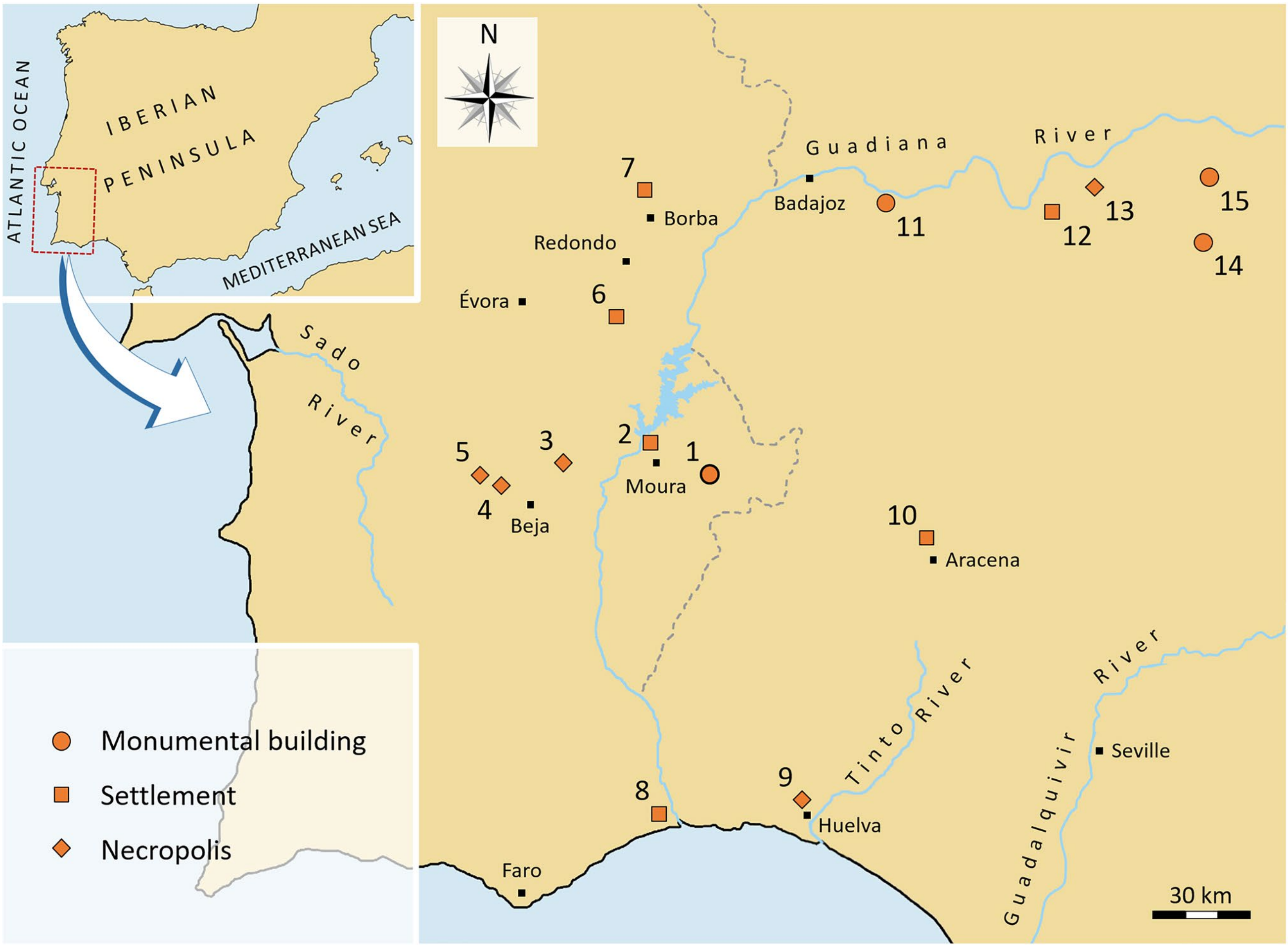

Fig. 1 The location of Cabeço Redondo and related archaeological sites in southwestern Iberian Peninsula (1: Cabeço Redondo; 2: Castro da Azougada; 3: Esfola; 4: Monte do Bolor 1/2; 5: Palhais; 6: Herdade da Sapatoa; 7: Castelão da Horta das Nogueiras; 8: Castelo de Castro Marim; 9: La Joya; 10: El Castañuelo; 11: Casas del Turuñuelo; 12: El Palomar; 13: Medellin; 14: Cancho Roano and 15: La Mata)

artefacts, some of them being highly corroded small fragments that were not studied. The highlight goes to a massive fragment of a plano-convex ingot (CR370: 6.4 kg) with an ellipsoidal shape (*c.* 30 cm × 22 cm), a maximum thickness of about 3 cm and an estimated total weight close to 10 kg (Fig. 2). Additionally, there are a small plano-convex ingot (CR394: 162 g) and an ingot fragment still with a flat surface (CR386: 359 g), in addition to several lumps and prills (specimens with less than 10 g were classified as prills) with highly variable shapes and sizes. The metallurgical waste was updated with a small fragment of a ceramic tuyere, possibly having a rounded shape and an internal orifice with about 1–1.5 cm diameter (CR[55]7171, Fig. 2: C), features similar to the ones of the tuyere fragment recovered in 2011 (Fig. 2: B). Additionally, there are several small and very porous light slag samples, some with greenish stains and vitrified ceramic or clay remains (Fig. 2: 1–7). Some of these slag samples show the presence of copper, but only future analytical studies will be able to relate them to local smelting or, most likely, melting operations.

Regarding the ingots, it is worth mentioning a lower occurrence in protohistoric contexts of the Iberian Peninsula when compared to other Mediterranean regions. During the Bronze Age, very large oxhide ingots weighing between 10 and 37 kg are known in the Central and Eastern Mediterranean (Montero-Ruiz et al. 2010-2011). Plano-convex ingots are also well known, as for instance from the Uluburum wreck dated to the end of the thirteenth century BC and showing copper ingots weighing between 3 and 10.5 kg (6.2 kg average) and 20 to 25 cm diameter (Pulak 2000). In the Iberian Peninsula, the existence of smaller plano-convex copper ingots has been known since the Bronze Age, especially in the northern region, with specimens varying in weight between 1.4 and 1.7 kg, and more rarely reaching 3.8 kg (Gómez Ramos 1993; Montero-Ruiz

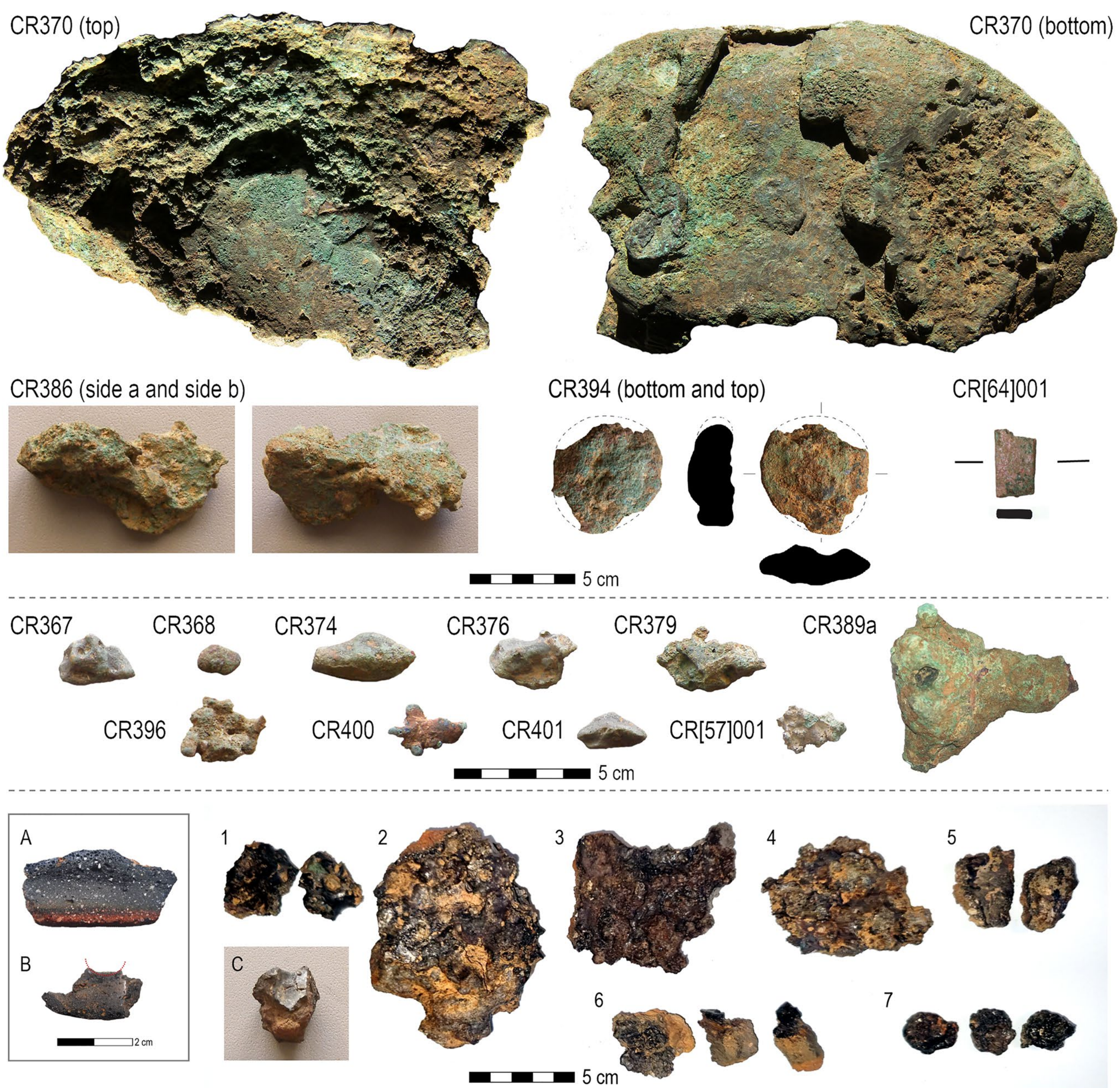

Fig. 2 Ingots, lumps, prills and metallurgical waste of Cabeço Redondo (*ingots*: plano-convex fragment CR370, irregular-shaped fragment CR386, plano-convex fragment CR394 and rod/ingot CR[64]001; *lumps*: CR367, CR374, CR376, CR379, CR389a, CR396 and CR401; *prills*: CR368, CR400 and CR[57]001; and *metallurgical waste*: (A) crucible/mould and (B) tuyere from 2011 survey; (C) tuyere CR[55]7171 from 2017 survey; and slag samples: 1: CR[0]7128, 2: CR[55]7153, 3: CR[55]7167, 4: CR[59]7159, 5: CR[61]7144, 6: CR[61]7151 and 7: CR[61]7157)

et al. 2010-2011). During the Iron Age, the ingots generally continue to correspond to fragments of small weight (Montero-Ruiz et al. 2010–2011, p. 119, Fig. 8). Some exceptions include a context of somewhat uncertain finding and chronology at Lamela (Pontevedra), from where García Alen (1968) described an irregular trapezoidal-shaped ingot with about 5 kg (Montero-Ruiz et al. 2010-2011). Additionally, from Castro da Azougada, near Cabeço Redondo, a fragmented copper ingot, apparently with a sub-trapezoidal shape and weighing 3.2 kg is currently under study by us. In the close vicinity of Iberian Peninsula, the Rochelongue underwater site (seventh–sixth century BC, France) stands out, showing an ingot assemblage with specimens weighing more than 6 kg (Aragón et al. 2022).

The metallic collection of Cabeço Redondo also includes artefacts, although most of them are very fragmented. A

Fig. 3 Copper-based artefacts of Cabeço Redondo (on the left, items retrieved by previous fieldworks: moon-crescent pendant CR002 (still with casting core), brazier handle CR003 and pivot of potter's wheel CR004 (14.5 cm in diameter); on the right, artefacts recovered by 2017 survey: Hathor figurine CR372, rosette figurine CR385, tweezers CR[59]085, lead weight CR397, curved blade CR381/CR382, weighing pan CR371, fragmented box CR388 (with two rivets) and brazier handle CR384)

small weighing pan (CR371, Fig. 3) with four holes for suspension cords, probably evidences the local trade of precious materials. The same can be inferred by a discoidal lead weight (with 9.9 g), the only lead piece recorded so far at Cabeço Redondo (Fig. 3). Weighing pans are common in such Iron Age sites and regional parallels were found at Castro da Azougada (Antunes 2017, p. 909) and Cancho Roano (García-Bellido 2003, p. 136). Other highlight goes to two small figurines depicting a hathoric mask (CR372) and a rosette (CR397) (Fig. 3). These items are used to decorate the Type I bronze braziers dated to the seventh century BC, having a matching pair in a brazier of La Joya necropolis (Huelva) (Jiménez Ávila 2002, p. 445). The orifice in those figurines indicates the attaching by riveting to the brazier and, interestingly, two brazier handles were recorded (CR003 and CR384, Fig. 3). The existence of these archaic objects at Cabeço Redondo and similar archaeological contexts can be explained by the desire to preserve items from generation to generation given their rarity and intrinsic or cultural value, or just by the recycling activities likely practised in those production sites. The remaining artefacts are tools, small implements and rods, such as a needle (CR[55]230), tweezers (CR[59]085), vessels fragments (CR380 and CR387), a fragmented box (CR388) and a very interesting and rare curved blade with cutting edges (sickle/halberd/razor?) (CR381/CR382) (Fig. 3).

Methodology

The preparation of samples for elemental and microstructural analyses involved the cutting of a small section, which was mounted in epoxy resin and polished with SiC papers (P1000, P2500 and P4000 grit size) and diamond pastes (3 µm and 1 µm grit size). In the case of artefacts that could not be sampled (e.g. figurines 372 and 385), a small surface area (*c.* 5 mm diameter) was cleaned and polished to expose the metallic surface. The process efficiency was ascertained with optical microscopy observations using a Zeiss Discovery V20 stereomicroscope. Once the analytical studies were completed, the exposed metallic area of artefacts was protected with a corrosion inhibitor (benzotriazol, 3% m/v in ethanol) and an acrylic polymer (Paraloid B-72, 10% m/v in ethanol). Finally, the coloration of the surrounding patina was replicated with a mixture of pigments dissolved in the acrylic polymer solution.

Micro-energy dispersive X-ray spectrometry

Micro-EDXRF analyses were performed with an ArtTAX Pro spectrometer 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 a spot size on sample surface of about 70 µm in diameter (Bronk et al. 2001). To account for chemical heterogeneities metal debris and artefacts were analysed by a bidimensional scan ($500 \times 500\ \mu m^2$ area and 100 µm step size) using 40 kV of potential difference, 600 µA of current intensity and 20 s of live time on each spot. Samples with smaller cleaned areas (CR380, CR393, CR398c and CR400) were analysed in 4 points with 40 kV, 600 µA and 120 s. The concentrations given are the average values of all analyses in each sample. Quantification involved WinAxil software with experimental calibration factors calculated with reference materials Phosphor Bronze 551 (British Chemical Standards) and Leaded Bronze C50.01 (BNF Metals Technology Centre). The accuracy is better than 10% and quantification limits 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).

Optical microscopy and scanning electron microscopy with X-ray microanalysis

Optical microscopy observations were made with an Olympus BX60M reflected light microscope (50–1000× magnification range) coupled with a Canon EOS M100 digital camera. Prepared samples were observed unetched and etched with aqueous ferric chloride solution to enhance microstructural features. SEM–EDS analyses involved a Zeiss DSM 962 scanning electron microscope coupled with an Oxford Instruments INCAx-sight energy dispersive spectrometer (EDS). The microscope comprises secondary electron and backscattered electron imaging modes, while the EDS spectrometer has an ultrathin window allowing the detection of low atomic number elements. Experimental conditions consisted of 20 kV of accelerating voltage, approximately 3 A of filament current, 70 µA of emission current and 25 mm of working distance. Gold sputtering and carbon conductive bridge on the epoxy resin mounted sample prevented charge accumulation and compositions were determined with ZAF factors.

Results and discussion

Chemical composition of metal debris

Micro-EDXRF analyses of ingots, lumps and prills show distinct contents in the Cu-Sn–Pb ternary system (Table 1). Furthermore, these metal debris display low contents of metallic impurities such as iron and, sometimes, arsenic and antimony (except for lump CR373 with 6.1 wt% Fe). First, it should be emphasised that ingots and, perhaps, larger lumps provide an accurate assessment of the metal or alloy being used in the artefact manufacture, while prills are more dubious due to the irregular conditions of ancient metallurgical operations. In view of this, the debris set is dominated by items composed of pure copper (Cu > 99 wt%), namely two ingots and half of the lumps (6 out of 12). The last ingot is composed of a binary bronze alloy with a tin content (CR394: 14.3 wt% Sn) much higher than remaining binary bronze items (two lumps, CR366: 7.3 wt% Sn and CR373: 5.2 wt% Sn, and three prills, CR368: 2.0 wt% Sn, CR383: 9.4 wt% Sn and CR398a: 5.7 wt% Sn). It is possible that this primary alloy was imported or produced at Cabeço Redondo workshop with an increased amount of tin to account for oxidation losses during subsequent casting operations. The leaded

Table 1 Chemical composition of metal debris of Cabeço Redondo (values in wt%; *n.d.*, not detected)

Type	Reference	Weight (g)	Cu	Sn	Pb	As	Sb	Fe
Ingot (plano-convex)	CR370	6 400	99.8	n.d.	0.20	n.d.	n.d.	< 0.05
Ingot (irregular shape)	CR386	359	99.7	n.d.	0.21	n.d.	n.d.	< 0.05
Ingot (plano-convex)	CR394	162	85.4	14.3	n.d.	< 0.10	n.d.	0.24
Lump	CR366	22	91.3	7.3	1.4	< 0.10	n.d.	< 0.05
Lump	CR367	14	99.9	n.d.	< 0.10	n.d.	n.d.	< 0.05
Lump	CR373	12	88.5	5.2	0.14	n.d.	n.d.	6.1
Lump	CR374	22	87.3	8.0	4.7	< 0.10	n.d.	< 0.05
Lump	CR376	20	87.7	8.6	3.2	n.d.	n.d.	0.47
Lump	CR379	29	99.8	n.d.	< 0.10	0.11	n.d.	< 0.05
Lump	CR389a	175	99.7	n.d.	0.13	n.d.	n.d.	0.13
Lump	CR392	33	99.8	n.d.	0.13	n.d.	n.d.	< 0.05
Lump	CR396	11	83.5	12.5	3.3	n.d.	n.d.	0.61
Lump	CR398c	12	90.8	5.5	3.2	n.d.	n.d.	0.48
Lump	CR401	10	99.4	n.d.	0.51	n.d.	n.d.	< 0.05
Lump	CR[55]234	22	99.2	n.d.	0.73	n.d.	n.d.	< 0.05
Prill	CR368	5	96.5	2.0	1.3	n.d.	n.d.	0.11
Prill	CR369	0.7	98.7	1.0	< 0.10	0.10	n.d.	0.11
Prill	CR375	7	98.4	0.90	0.11	n.d.	n.d.	0.53
Prill	CR383	6	89.1	9.4	0.64	n.d.	n.d.	0.83
Prill	CR389b	9	99.6	n.d.	< 0.10	0.14	0.20	< 0.05
Prill	CR390	1	99.4	n.d.	0.39	n.d.	0.12	< 0.05
Prill	CR395b	2	83.2	7.3	9.5	n.d.	n.d.	< 0.05
Prill	CR398a	3	94.2	5.7	< 0.10	n.d.	n.d.	< 0.05
Prill	CR399	7	95.9	1.2	2.4	n.d.	n.d.	0.50
Prill	CR400	7	99.6	n.d.	0.17	n.d.	n.d.	0.20
Prill	CR[57]001	9	99.4	n.d.	0.59	n.d.	n.d.	< 0.05
Prill	CR[63]001	2	99.5	n.d.	0.27	< 0.10	n.d	0.16

bronze alloy is also present, namely in 4 lumps and 1 prill showing 5.5–12.5 wt% Sn and 3.2–9.5 wt% Pb. Finally, there is a Cu-Pb prill with low lead and low tin contents (CR399: 2.4 wt% Pb and 1.2 wt% Sn).

Overall, the composition of metal debris of Cabeço Redondo shows the existence of copper, bronze and leaded bronze (Fig. 4). The histogram of tin contents evidences the dominance of copper items, but it also shows a cluster comprising samples with 4–10 wt% Sn, probably corresponding to the usual composition range of coeval bronze artefacts. The histogram of lead contents shows a continuous increase towards the low contents suggesting the presence of lead as an impurity in the majority of local metallurgical productions. It should be noted that the two copper ingots have lead as an impurity (*c.* 0.2 wt% Pb) and 10 out of 24 analyses (lumps and prills) show lead contents below this value. On the other hand, it must be noted that no lead ingots, nor pure lead prills or lumps, have been recorded at Cabeço Redondo. Nevertheless, the lead content of a few bronze lumps (e.g. CR374: 4.7% Pb and CR376: 3.2% Pb) suggest the addition of lead to improve the alloy castability. A possible bronze ingot (rod CR[64]001) with 13.0% Pb might indicate the addition of lead using this type of lead-rich bronze alloy (see discussion ahead).

Some questions arise about the actual metallurgical processes taking place at Cabeço Redondo, for instance concerning the possible local production of bronze alloys. The co-smelting of copper ore and cassiterite produces prills with extremely variable composition, i.e. some prills contain hardly any tin, while others have contents above 80 wt% Sn (Rovira et al. 2009). A large compositional disparity (0–30 wt% Sn) was also identified in prills of cementation crucibles from the Iron Age site of El Castru, Asturias, NW Spain (Farci et al. 2017). Conversely, the much smaller range of tin contents of prills and lumps of Cabeço Redondo (up to 12.5 wt% Sn) suggests the alloying of copper and tin metals, being possibly the ingot CR394 an intermediate product to produce those alloys. However, no traces of pure tin or pure lead were found, which makes it difficult to identify the local methods of bronze production. For example, an alternative to the apparent absence of tin ingots would be to use high-tin bronze (e.g. ingot CR394) diluted with pure copper to produce lower tin-content bronze alloys. In southern Iberia, the production of bronze by co-melting of metallic

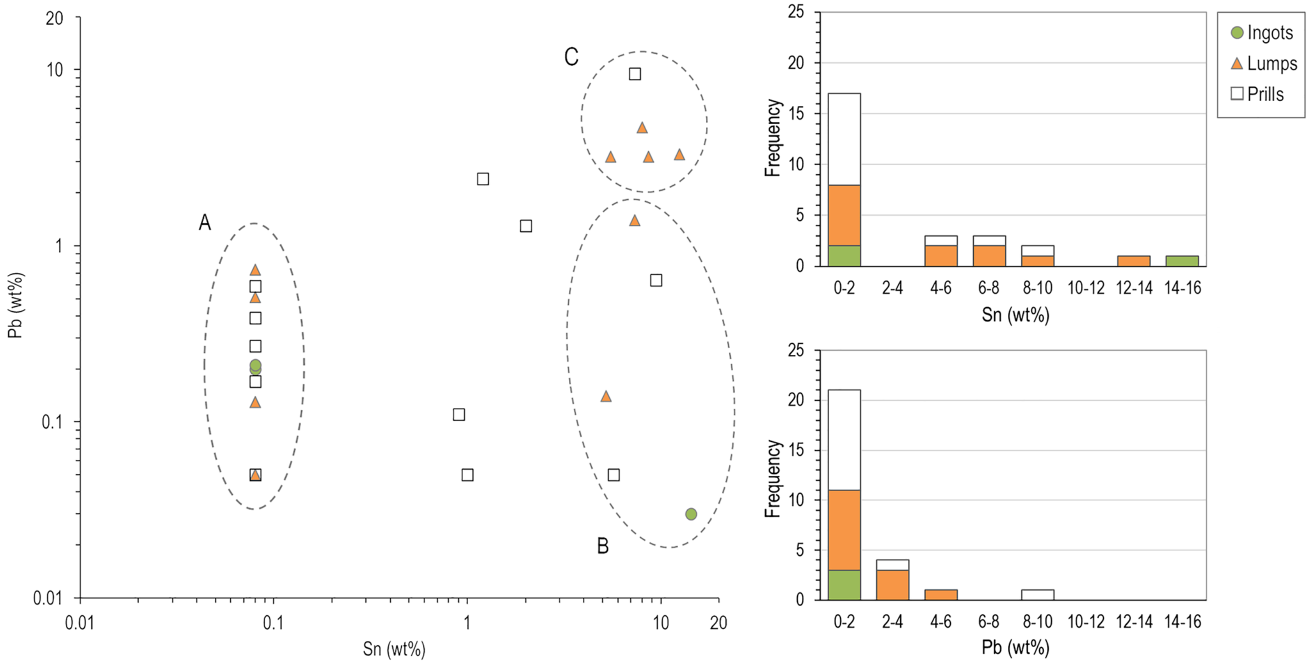

Fig. 4 Plot of tin versus lead contents and histograms of tin and lead contents of metal debris of Cabeço Redondo (A: samples related to copper production; B: samples related to binary bronze production and C: samples related to leaded bronze production)

copper and tin during the Iron Age has been identified at Carmona, Seville (Rovira 2007) and La Fonteta, Alicante (Renzi 2013), although co-smelting and cementation were also used at the latter site.

Microstructural composition of metal debris

SEM–EDS analyses of the large plano-convex ingot CR370 identified a copper matrix with about 1.6 wt% sulphur (Table 2). This element occurs in Cu–S inclusions due to a low solubility in molten copper (Chernykh et al. 1998), so the relatively high amount of sulphur in ingot CR370 implies a high density of Cu_2S inclusions (Fig. 5). Moreover, an uncommonly large Cu–S inclusion is composed by a Cu_2S matrix with an intergranular Cu–S-O compound (78.2 wt% Cu, 16.4 wt% S and 5.4 wt% O) indicating a microdomain with poor reducing conditions. Copper oxides are thought to be common in ingots from refining of smelting lumps (Wang et al. 2018), but these oxides are absent in ingot CR370. Additionally, the nonexistence of iron in copper sulphide inclusions indicates that the ores were richer in copper sulphides (e.g. covellite, CuS and chalcocite, Cu_2S) rather than in copper-iron-sulphide minerals (e.g. chalcopyrite, $CuFeS_2$ and bornite, Cu_5FeS_4). Primary smelting products such as oxhide and plano-convex ingots of the Uluburun shipwreck (Hauptmann et al. 2002) and Nuragic Sardinia (Begemann et al. 2001) often show a high porosity that is absent in our ingot. However, it was suggested that the high porosity of oxhide ingots could be intentional, since such raw copper is more brittle and easier to break into small pieces later used for casting (Hauptmann et al. 2016). Ultimately, the very large size of the plano-convex ingot CR370, in addition to a high density of Cu_2S inclusions and the absence of copper oxides and porosity suggest the smelting of copper ores in a slag-tapping furnace. However, it should be stressed that these metallurgical structures are unknown in the Iron Age archaeological record of Iberian Peninsula, thus suggesting an extra-peninsular origin for the copper used at Cabeço Redondo.

The fragment of the irregular-shaped copper ingot (CR386) shows compositional and microstructural characteristics analogous to the plano-convex ingot CR370 (Table 2 and Fig. 5). Therefore, the ingot CR386 was probably originated by copper smelting in a similar furnace, although the original dimensions of the ingot are unknown. Some of the smaller metal lumps (e.g. lump CR392 with Cu_2S inclusions (*c.* 1.2 wt% S) and Pb inclusions, Table 2 and Fig. 5) show microstructural and compositional features compatible to those ingots. These lumps may have been lost during any metallurgical operation involving such ingots, such as the pouring of the molten metal into a mould.

The plano-convex bronze ingot CR394 shows a dendritic structure with a cored α phase and a high amount of the $\alpha+\delta$ eutectoid (Table 2 and Fig. 5). These features indicate a fast cooling of such metallic mass (162 g), thus suggesting a secondary operation such as alloying. The small impurity

Table 2 SEM–EDS results of metal debris of Cabeço Redondo (values in wt%; *Cl = 19.3 wt%)

Sample	Cu	Sn	Pb	O	S	Fe	As	Se	Sb
Ingot CR370									
Bulk	98.0	-	-	0.4	1.6	-	-	-	-
Cu–S inclusion 1	76.8	-	-	1.9	21.3	-	-	-	-
Cu–S inclusion 2	76.3	-	-	1.3	22.4	-	-	-	-
Cu–S inclusion 3	78.0	-	-	-	22.0	-	-	-	-
Cu–S (matrix)	77.2	-	-	-	22.8	-	-	-	-
Cu–S (intergranular)	78.2	-	-	5.4	16.4	-	-	-	-
Metallic inclusion	60.1	-	13.3	8.0	12.1	1.8	4.7	-	-
Ingot CR386									
Bulk	98.3	-	-	-	1.6	-	-	-	-
Cu–S inclusion 1	79.9	-	-	-	20.1	-	-	-	-
Cu–S inclusion 2	79.5	-	-	-	20.5	-	-	-	-
Metallic inclusion	7.4	-	59.5	11.0	-	-	1.6	-	20.5
Lump CR392									
Bulk	98.8	-	-	-	1.2	-	-	-	-
Cu–S inclusion	78.8	-	-	-	21.2	-	-	-	-
Pb inclusion	15.3	-	78.0	6.7	-	-	-	-	-
Ingot CR394									
Bulk	84.6	15.4	-	-	-	-	-	-	-
α phase	90.2	9.8	-	-	-	-	-	-	-
α + δ eutectoid	71.6	28.4	-	-	-	-	-	-	-
Pb-Fe inclusion	68.4	17.3	10.9	-	-	3.7	-	-	-
Fe-S-Se inclusion	48.0	1.4	-	-	28.8	19.0	-	7.4	-
Prill CR368									
Bulk	98.4	1.6	-	-	-	-	-	-	-
α phase (darker)	98.9	1.1	-	-	-	-	-	-	-
α phase (brighter)	96.5	3.5	-	-	-	-	-	-	-
Pb inclusion	21.3	-	78.7	-	-	-	-	-	-
Sn–O inclusion	-	71.1	-	28.9	-	-	-	-	-
Prill CR398a									
Sn–O inclusion 1	-	71.7	-	28.3	-	-	-	-	-
Sn–O inclusion 2	-	71.4	-	28.6	-	-	-	-	-
Lump CR374									
α phase	90.8	9.2	-	-	-	-	-	-	-
α + δ eutectoid	70.2	29.8	-	-	-	-	-	-	-
Pb inclusion	4.7	-	88.9	6.4	-	-	-	-	-
Cu–S inclusion	79.6	-	-	-	20.4	-	-	-	-
Prill CR395b									
α phase	94.1	5.9	-	-	-	-	-	-	-
Pb inclusion 1	7.3	-	90.2	2.5	-	-	-	-	-
Pb inclusion 2*	14.9	-	61.1	4.7	-	-	-	-	-
Cu–S inclusion 1	78.6	-	-	-	21.4	-	-	-	-
Cu–S inclusion 2	78.2	-	-	-	21.8	-	-	-	-

pattern comprises Pb-Fe and Fe-S-Se inclusions associated with the α + δ eutectoid (the last phase to solidify). Selenium has a high affinity for sulphide phase and such inclusions are considered a strong proof of not remelted metal (Rehren 1991), which agrees with the high tin content of the CR394 ingot (14.3 wt% Sn). These microstructural features support the above-mentioned premise of the bronze production by mixing copper and tin metals.

Two bronze prills (CR368 and CR398a) show a cored α phase despite a low amount of tin (2.0 wt% Sn and 5.7 wt% Sn, respectively) due to a very fast cooling (Table 2 and Fig. 5). These prills also have oxidised tin inclusions

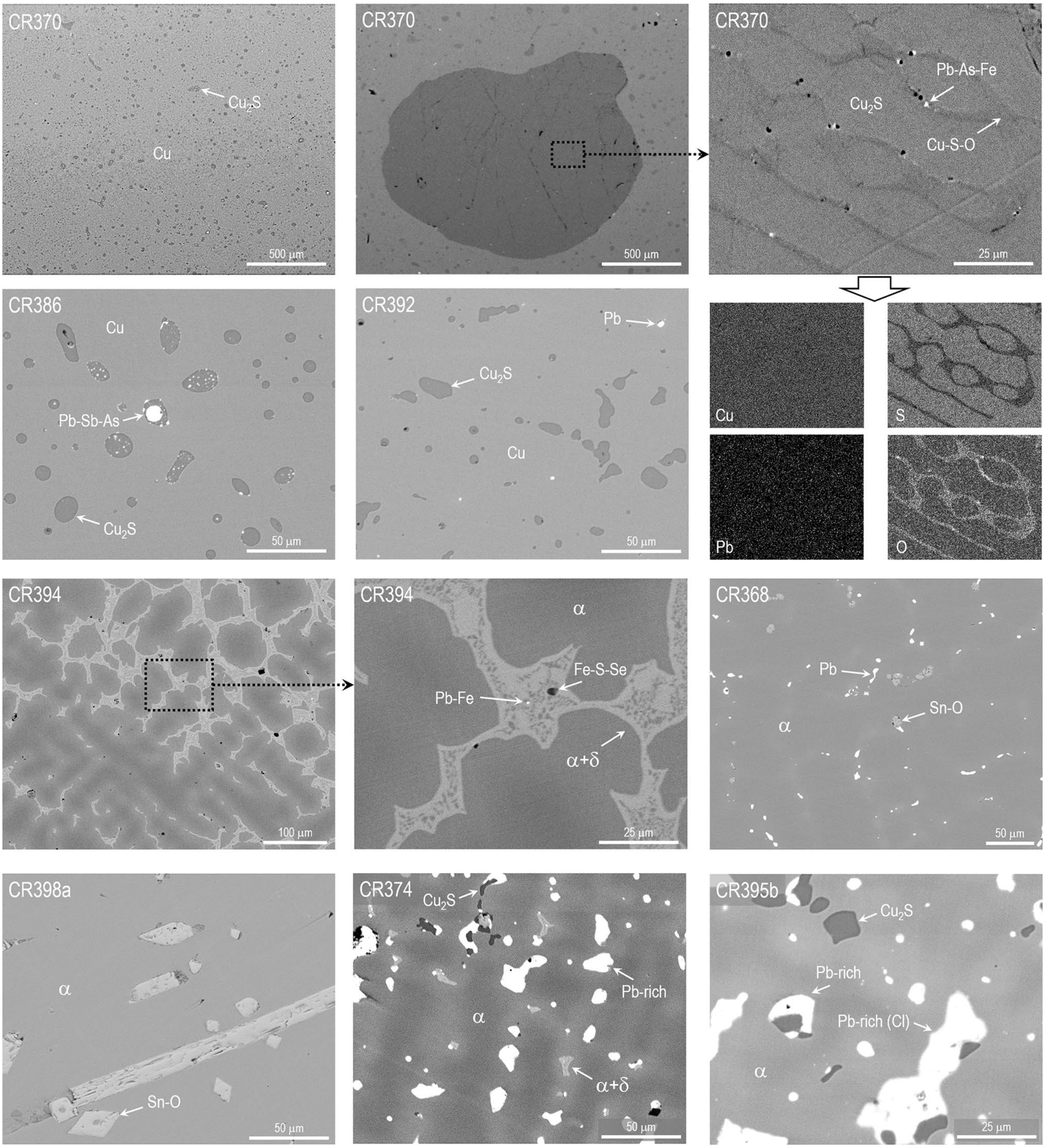

Fig. 5 SEM–EDS characterisation of metal debris of Cabeço Redondo (copper: plano-convex ingot CR370, ingot CR386 and lump CR392; binary bronze: plano-convex ingot CR394, prill CR368 and prill CR398a; leaded bronze: lump CR374 and prill CR395b)

with rhomboidal and acicular shapes. Rademakers and Farci (2018) showed that distinct tin oxide morphologies originate from tin oxidation and recrystallization, but can be formed in any process involving bronze production with oxidising conditions. Moreover, these tin oxides do not completely reduce after several remelting cycles, as their stability is dependent on oxygen availability and dwell time (Vernet et al. 2019). Some larger and more irregularly shaped tin oxide inclusions can be interpreted as mineral cassiterite, but these shapes are absent at Cabeço Redondo, which is in line with the already mentioned inference excluding a bronze production by cementation or co-smelting of copper and tin ores.

A leaded bronze lump (CR374) has a dendritic microstructure with cored α phase, α + δ eutectoid, Cu_2S inclusions and oxidised Pb-rich inclusions (Table 2 and Fig. 5). Another leaded bronze prill (CR395b) shows a similar composition, but some Pb-rich inclusions have a high chlorine content indicating post-depositional corrosion processes (Table 2 and Fig. 5). These microstructures indicate relatively fast cooling, likely pointing to spills from alloying or casting.

Chemical composition of artefacts

The collection includes artefacts composed of copper, binary bronze and leaded bronze with low contents of iron and, in a few cases, arsenic and nickel (Table 3). To this collection, we can add results of the few artefacts previously recovered at Cabeço Redondo, which evidence the use of bronze and leaded bronze (Table 4). Considering the overall set, the leaded bronze alloy is the more common type (44% frequency) showing variable lead contents (6.0 ± 3.4 wt% Pb). Binary bronzes are also quite common (39%), while copper artefacts correspond to a minority (17%). The enhanced castability of leaded bronze alloys was certainly useful in some items, such as the massive pivot (CR004) and the two figurines (CR372 and CR385). Leaded copper and leaded bronze were also commonly used in some figurines of the seventh century BC Phoenician settlement of Alcácer do Sal (Schiavon et al. 2013). On the contrary, the use of this less strong alloy in the curved blade CR381/CR382 points to a ceremonial object or to economic factors concerning the sparse use of tin. The latter could be corroborated by a somewhat low tin content of these leaded bronzes (7.7 ± 4.4 wt% Sn). Binary bronzes also show low tin contents (7.6 ± 3.9 wt% Sn) and, usually, very low amounts of lead (the brazier handle CR384 has 1.6 wt% Pb, but remaining items show values below 0.31 wt% Pb). This fact indicates that leaded bronze scrap was not commonly used to produce tools and small implements. The collection is closed by four copper

Table 3 Chemical composition of artefacts of Cabeço Redondo (values in wt%; *n.d.*, not detected)

Type	Reference	Cu	Sn	Pb	As	Ni	Fe
Brazier handle	CR384	94.1	3.1	1.6	n.d.	n.d.	1.2
Curved blade	CR381/CR382	86.3	6.5	7.2	n.d.	n.d.	< 0.05
Figurine (*Hathor*)	CR372	87.0	9.2	3.7	n.d.	n.d.	0.06
Figurine (rosette)	CR385	93.4	2.9	3.4	0.12	n.d.	0.10
Needle (?)	CR[55]230	99.1	n.d.	< 0.10	0.13	n.d.	0.75
Tweezers	CR[59]085	92.8	6.4	< 0.10	< 0.10	n.d.	0.72
Vessel (fragment)	CR380	96.0	3.7	0.14	n.d.	n.d.	0.10
Vessel (fragment)	CR387	83.4	7.5	8.4	n.d.	0.51	0.15
Weighing pan	CR371	99.6	n.d.	0.17	n.d.	n.d.	0.22
Fragmented box	CR388	75.3	15.1	9.3	0.11	n.d.	0.17
Rivet (from 388)	CR388r	80.5	14.4	4.9	0.15	n.d.	< 0.05
Rod	CR377	99.4	n.d.	0.11	n.d.	n.d.	0.41
Rod	CR393	95.8	3.7	n.d	n.d.	n.d.	0.40
Rod	CR395c	94.7	3.2	2.0	n.d.	n.d.	< 0.05
Rod	CR398d	99.0	n.d.	0.97	n.d.	n.d.	< 0.05
Rod	CR[55]231	89.0	10.5	0.24	n.d.	n.d.	0.23
Rod	CR[55]232	88.3	11.5	< 0.10	< 0.10	n.d.	0.13
Rod	CR[55]233	91.5	2.8	5.4	n.d.	n.d.	0.22
Rod (ingot?)	CR[64]001	79.4	7.2	13.0	< 0.10	n.d.	0.30

Table 4 Chemical composition of artefacts previously analysed of Cabeço Redondo (values in wt%; *n.d.*, not detected; CR001 = CR[2]54; CR002 = CR[38]5; CR003 = CR[9]14 and CR004 = CR364, following Valério et al. 2015, Table 3)

Type	Reference	Cu	Sn	Pb	As	Sb	Fe
Rod	CR001	90.0	9.2	0.31	< 0.10	n.d.	0.46
Pendant	CR002	85.0	14.1	0.10	< 0.10	n.d.	< 0.05
Brazier handle	CR003	93.1	6.4	0.12	< 0.10	n.d.	0.31
Pivot	CR004	88.1	8.4	2.8	n.d.	0.66	< 0.05

artefacts with emphasis to a weighing pan (CR371) made with a very thin sheet. It is easier to shape a copper sheet since this metal is less brittle than bronze. On the opposite side of the spectrum, there is the moon-crescent pendant CR002 with a high tin content (14.1 wt% Sn), ensuing a more golden colour to such prestige ornament. Nevertheless, the presence of the casting core in this hollow pendant suggests that it is a deficient casting that was discarded as scrap metal for later use.

Generally, the artefact collection of Cabeço Redondo compares with the local metal debris since it also shows copper, bronze and leaded bronze alloys (Fig. 6). It should be emphasised that the site was object of treasure hunter activities after its destruction during 1990, which very likely have subtracted the best-preserved artefacts and leave behind lumps and prills. Hence, the artefact collection constitutes a somewhat skewed sample of the objects that actually existed until the destruction of the site in the 1990´s. Nevertheless, it is interesting to note that leaded copper artefacts are absent at Cabeço Redondo, despite occurring in coeval sites in SW Iberian Peninsula such as Cancho Roano (Montero-Ruiz et al. 2003) and El Palomar (Rovira et al. 2005). These leaded copper artefacts show relatively high tin contents (0.89–1.97 wt% Sn) suggesting the melting of bronze scrap instead of a primary production of the Cu-Pb alloy. At Cabeço Redondo, the additional absence of pure lead waste suggests that the leaded bronze alloy was imported from elsewhere, either as ingots or as finished artefacts.

Microstructural composition of artefacts

Copper artefacts show recrystallised microstructures with deformed grains and a high number of annealing twins (CR371, CR377, CR398d and CR[55]230, Fig. 7). These features indicate the use of hammering and annealing operations to manufacture the artefact. The deformation was especially intense in needle CR[55]230 giving rise to a much finer grain microstructure. Apart from the rod CR398d showing a high density of Pb inclusions (black inclusions in Fig. 7), the remaining examples are relatively free from other impurities such as Cu_2S inclusions, which suggests an intermediate stage comprising the refining of the copper ingots showing a high density of those Cu_2S inclusions (see Fig. 5).

Bronzes display different microstructural traits evidencing distinct manufacturing procedures (CR380, CR384, CR393, CR[55]321 and CR[55]232, Fig. 7). The vessel CR380 has extremely large grains indicating a strong annealing of the as-cast piece. The absence of hammering makes sense to manufacture this type of artefact. The remaining bronzes comprise deformed grains with annealing twins indicating a manufacture similar to the one of copper artefacts, namely of hammering followed by annealing. However, the rod CR[55]231 has also a high density of slip bands implying a strong final hammering, which provides a higher hardness to the bronze alloy.

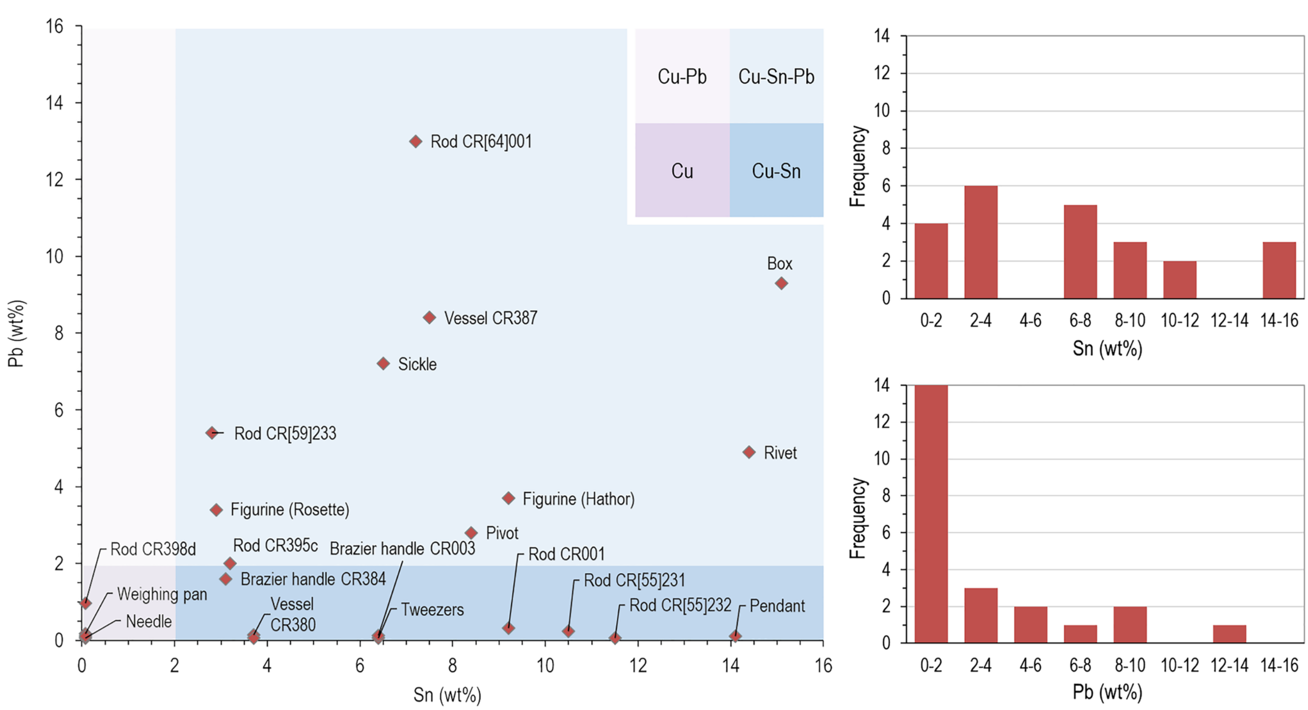

Fig. 6 Plot of tin versus lead contents and histograms of tin and lead contents of artefacts of Cabeço Redondo

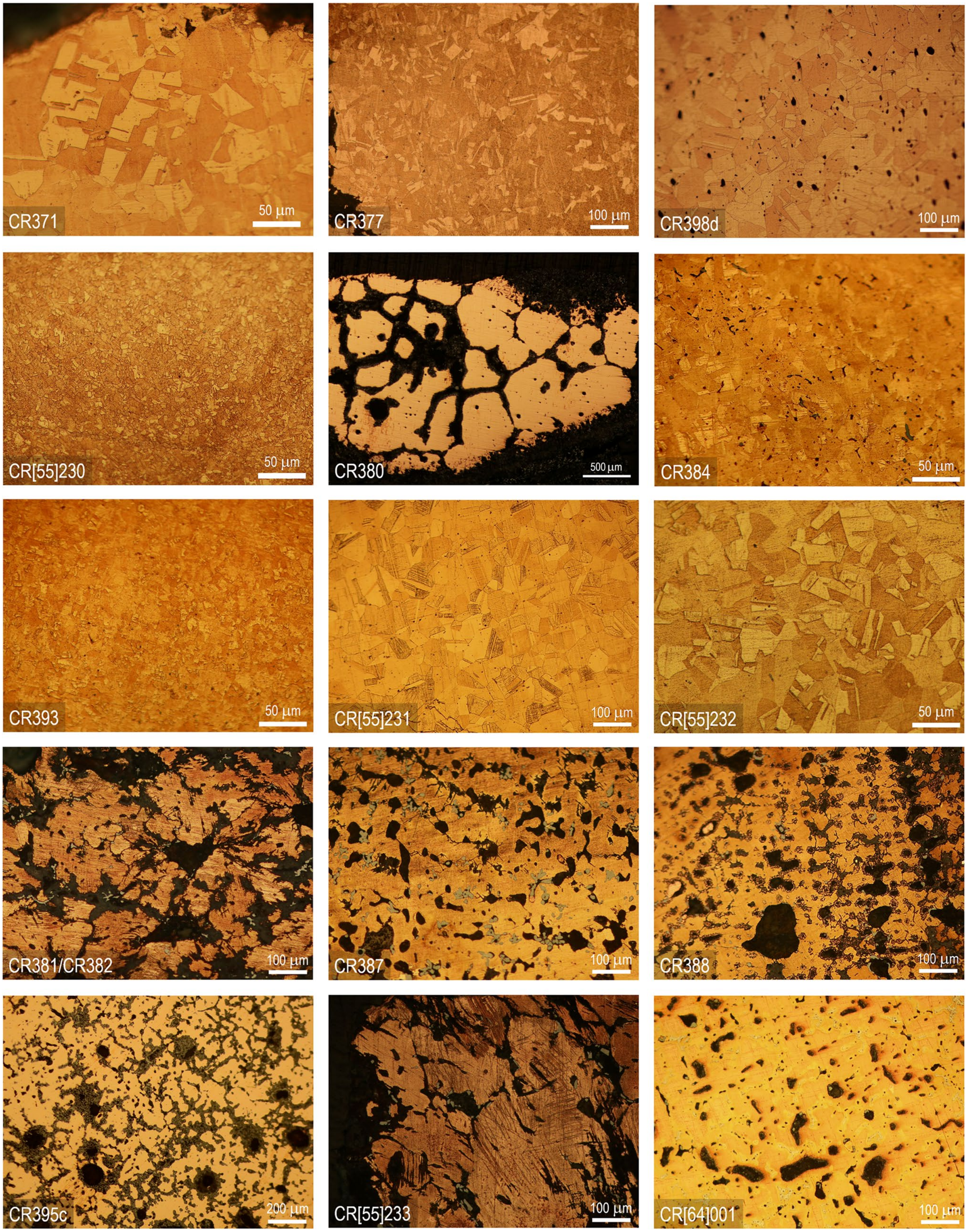

Fig. 7 Optical microscopy characterisation of artefacts of Cabeço Redondo (copper: weighing pan CR371, rod CR377, rod CR398d and needle CR[55]230; binary bronze: vessel CR380, brazier handle CR384, rod CR393, rod CR[55]231 and rod CR[55]232; leaded bronze: curved blade CR381/CR382, vessel CR387, fragmented box CR388, rod CR395c, rod CR[55]233 and rod CR[64]001)

Leaded bronzes show dendritic microstructures and Pb inclusions (CR381/CR382, CR387, CR388, CR395c, CR[55]233 and CR[64]001, Fig. 7). The lack of post-casting operations agrees with the functionality of some of these items, namely the vessel CR387 and the box CR388. The curved blade CR381/CR382 and the rod CR[55]233 also show slip bands from hammering the as-cast artefact, a procedure that may have been intended to obtain the final shape. The rod CR395c shows large pores suggesting a deficient casting. Additionally, some leaded bronzes show a higher density of Cu–S inclusions (CR387 and CR388: dark blue inclusions in Fig. 7). It is possible that these as-cast leaded bronzes used less refined copper, comparable to the CR370 and CR386 ingots. Wang et al. (2018) also detected higher sulphur contents in LBA ingots of Salcombe (SW England) when compared to local artefacts.

The rod CR[64]001 (Fig. 2) seems to be a particular case, as it shows an as-cast microstructure with coring, Pb and Cu–S inclusions and a high amount of the $\alpha+\delta$ eutectoid (Fig. 7), characteristics quite similar to the ones of the lump CR374 (see Fig. 5). Considering also its uncommon shape (rectangular section with 16×4 mm^2) and very high lead content (13.0 wt% Pb), it can be hypothesised that this rod is in fact an ingot fragment intended to produce leaded bronze artefacts. This might explain the lack of lead ingots and adds to the hypothesis mentioned before of the use of high-tin or high-lead instead of pure tin or pure lead ingots, to produce the different bronze alloys of local artefacts. Similar bronze rods from LBA sites in the Iberian Peninsula have been interpreted as ingots (Baptista et al. 2019-2020). Moreover, the utilisation of a leaded bronze ingot with comparable chemical characteristics is a good justification for the higher level of copper sulphides observed in some of the leaded bronze artefacts of Cabeço Redondo.

The production and use of metal at Cabeço Redondo

The significant presence of copper-based debris (about 7 kg), including the two copper ingots, discloses the high importance of such metallurgy at Cabeço Redondo. The large size and microstructural characteristics of the two copper ingots (high amount of Cu_2S impurities, especially when compared to artefacts, low porosity and absence of copper oxides) are indications of the smelting of copper ores in slag-tapping furnaces, thus suggesting a provenance outside the Iberian Peninsula. Regarding the production of bronze, the fast cooling microstructure of the small plano-convex ingot points to a secondary activity such as the alloying of copper and tin metals, while its high tin content could indicate a primary alloy richer in tin to deal with later oxidation losses during casting. Nevertheless, as already mentioned, it is unknown if this plano-convex ingot was locally made or imported. Concerning the bronze prills, the absence of high-tin specimens and the fast cooling microstructures identified also suggest processes unrelated to co-smelting or cementation, instead probably being the result of alloying or casting operations.

Artefacts show a predominance of binary and leaded bronzes, being interesting to realise that the average tin content of binary and leaded bronze alloys is similar (overall average of 7.7 ± 4.0 wt% Sn) and comparable to the cluster of bronze lumps recovered on site (*c.* 4–10 wt% Sn). This correspondence does not necessarily mean that all recorded artefacts were locally produced, but at least it suggests that some of them were manufactured at Cabeço Redondo, which integrates the fifth century BC production model of the Guadiana River basin, i.e. a metallurgical technological pattern comprising the common use of copper, binary bronze and ternary bronze. The relation among function, composition and manufacture is often clear, as for instance the as-cast leaded bronzes (vessel CR387 and box CR388) or the thermomechanically worked tools and implements (needle CR[55]230 and weighing pan CR371). Copper and binary bronzes were manufactured differently from leaded bronzes, as the latter were left as-cast, eventually suffering some cold work, while others usually comprised a longer *chaîne opératoire*. Some leaded bronzes also show a higher density of sulphide inclusions, which may be associated with the use of less refined metal, as these items would not be subjected to post-casting works.

Regarding the composition of Iron Age collections from analogous sites in SW Iberian Peninsula, one should underline the closer similarity with Cancho Roano (Fig. 8). This coeval monumental building showed a collection of bronzes with comparable tin contents (overall average of 8.6 ± 5.3 wt% Sn), although with a higher preponderance of leaded bronzes due to a numerous amount of casting objects (Montero-Ruiz et al. 2003). As already mentioned, after the destruction of the artificial mound in 1990, the site of Cabeço Redondo has suffered the action of metal detectorists, which selectively collected artefacts and left behind lumps and prills. Therefore, it is not surprising the few artefacts recorded so far by archaeological surveys, neither the smaller amount of leaded bronzes among this collection. On the other hand, the different type of artefacts from sites of distinct nature and chronology can explain the divergence of Cabeço Redondo with local necropolises. For instance, the seventh–sixth century BC necropolises of Esfola, Monte do Bolor 1/2 and Palhais displayed mostly ornaments constituted mainly of binary bronze alloys with 6.7 ± 2.5 wt% Sn (Valério et al. 2013, 2021). Other seventh–sixth century BC sites like the settlement of El Palomar and the neighbouring Medellin necropolis share the dominance of binary bronzes, although with alloys richer in tin (12.6 ± 6.2 wt% Sn, Rovira et al. 2005 and 11.7 ± 7.5 wt% Sn, Rovira 2008,

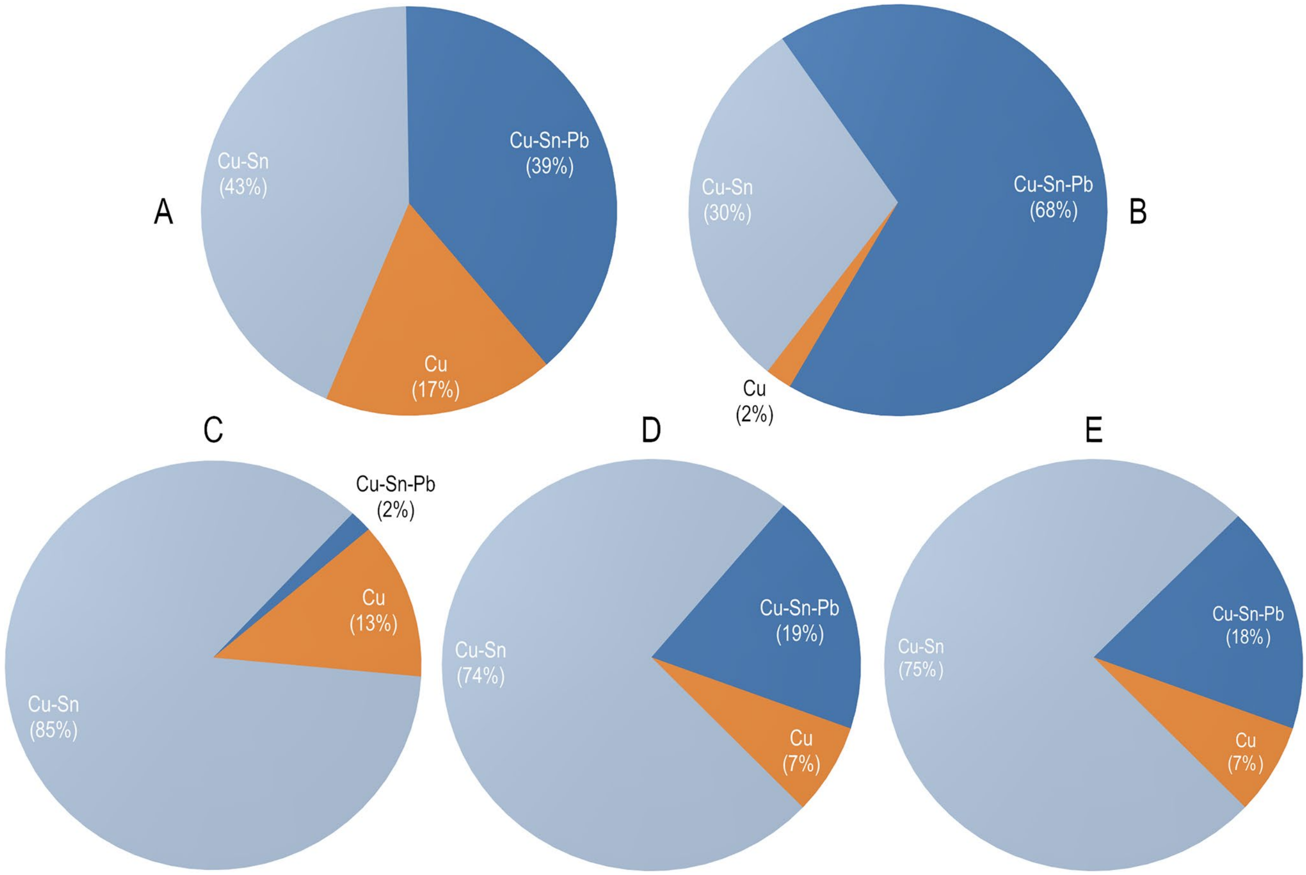

Fig. 8 Distribution of copper (Cu), bronze (Cu-Sn) and leaded bronze (Cu-Sn–Pb) in artefact collections of Iron Age contexts in southwestern Iberian Peninsula (A: Cabeço Redondo (23 items), B: Cancho Roano (114 items), C: Esfola (28 items), Monte do Bolor 1/2 (21 items) and Palhais (7 items), D: El Palomar (57 items) and E: Medellin (78 items))

respectively). This compositional diversity suggests that SW Iberian workshops of the 2nd quarter of the 1st millennium BC used different recipes and, probably, distinct methods of bronze production due to technological, cultural or economic constrains.

Conclusions

The research on metal debris recovered at Cabeço Redondo provides good evidence of the important metallurgical activities that took place at this late sixth–fifth century BC site. In addition to agriculture and pastoralism, which would produce significant surpluses evidenced by the large and numerous ceramic containers, millstones and domestic fauna remains, the metallurgical workshop is another sign of the wealth produced at Cabeço Redondo. This monumental building would certainly control other small rural sites in a rich agricultural area, thus being the central site of a network of Iron Age local communities in this area of the left bank of the Guadiana River basin.

Compositional and microstructural characteristics of metal debris and metallurgical waste recorded at Cabeço Redondo suggest the existence of one workshop carrying out different activities, perhaps involving bronze alloying and, very likely, artefact casting with the manufacture of objects made of copper, bronze and, perhaps, leaded bronze. Concerning the huge plano-convex ingot, the results suggest that it was imported from outside the Iberian Peninsula since it was most likely produced in a slag-tapping furnace. On the contrary, the small bronze ingot may have been produced locally, which would indicate the importation of tin ingots to Cabeço Redondo. Additionally, a fragment of a leaded bronze rod was acknowledged as a possible ingot for the local production of ternary bronzes. Considering the high level of destruction of the site, it is difficult to understand the full range of local metallurgical operations, but imported ingots show that Cabeço Redondo was integrated into the Mediterranean trade system. Therefore, the availability of tin ingots was a possibility, enabling the bronze production by co-melting at Cabeço Redondo. Besides, the high-tin bronze ingot and the high lead-bronze ingot suggest the "dilution" of these with pure copper. These different methods could be selected based on the availability of raw materials (metals, alloys, charcoal), costs and benefits of each process (time and labour consumption) and demand requirements (Montes-Landa et al. 2020). Nevertheless, these matters will be clarified by upcoming studies involving the characterisation of slags and the Pb isotope analysis of metal debris and artefacts.

The artefact collection of Cabeço Redondo shows the use of copper, low-tin bronze alloys and leaded bronze alloys, a pattern in line with the elemental compositions of metal debris recorded in this post-Orientalizing archaeological site. Distinct compositions correspond to different types and diverse post-casting manufactures, namely the leaded bronze artefacts, particularly ornaments, without post-casting works, in opposition to copper and bronze tools and implements with hammering and annealing operations. Apparently, the metallurgical production was no longer a domestic work, as it occurs during prehistoric times, but rather a specialised activity taking place in the monumental building complexes of post-Orientalizing communities in this Guadiana River region.

Finally, the compositional resemblance with copper-based artefacts of Cancho Roano and similar sites in the Middle Guadiana River basin strengthens the similarity of Cabeço Redondo with these Iron Age contexts, showing a similar architecture and material culture including pottery with comparable shapes and decoration styles. Cabeço Redondo can therefore be integrated among the communities of the middle of the 1st millennium BC in this region of SW Iberian Peninsula. These characteristic sites feature a distinctive architecture represented by monumental and singular buildings located in rural areas, in which agricultural and commercial activities, already with an industrial embryo (metallurgy and pottery), took place, giving rise to a wealthy development of these proto-historic communities, which seems to end abruptly by the end of the fifth century BC.

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Data availability All relevant data are within the manuscript.

Declarations

Conflict of interest The authors declare no competing interests.

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