LATE BRONZE AGE METAL ARTEFACTS FROM AN ORIENTALISING BURIAL (?) AT "FRAGA DOS CORVOS" (MACEDO DE CAVALEIROS – NE PORTUGAL): A FIRST ARCHAEOMETALLURGIAL APPROACH

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ABSTRACT

Several bronze artefacts were found in a rock shelter in the western versant of Fraga dos Corvos archaeological site that suggest the presence of a Late Bronze Age female (?) orientalised burial. From these, a pendant, a needle, a *fibula*, a ring and three bar fragments were studied from an archaeometallurgical point of view. Micro-EDXRF analyses were made in order to obtain data about metal composition. Results show that all the artefacts are bronze alloys (Cu-Sn) with some Pb contents and all, except the pendant, present similar compositions (8.14-12.2% Sn and <2.04% Pb). The microstructure of the three bar fragments and the ring was studied under optical and scanning electron microscopy. Results show that all suffered thermo-mechanical operations that finished either with an annealing heat treatment or with a final mechanical deformation. All microstructures exhibit sulphide inclusions.

KEYWORDS

Archaeometallurgy; Late Bronze Age; NE Portugal; Bronze alloy; Thermo-mechanical operations

INTRODUCTION

Fraga dos Corvos is a Bronze Age habitat site recently studied and located in the north-western versant of Serra de Bornes, Eastern Trás-os-Montes (Macedo de Cavaleiros, Bragança, Portugal). With an altitude of 870m, it rises over the modern hamlet of Vilar do Monte.

The site was already known as an Iron Age fortified settlement. In 2003, agricultural work on its northern platform put in danger part of it and revealed Bronze Age levels which determined the recent archaeological intervention [1,2].

The hilltop of Fraga dos Corvos visually controls the Macedo de Cavaleiros basin. It is in the main traditional pass into and out of this basin that have been found the deposits of halberd copper blades which gave the name to the so called Carrapatas type of the Iberian Early Bronze Age Atlantic halberds (Abreiro, Carrapatas, Vale Bemfeito and Vimioso [3]).

In the summer of 2006 it took place the 4^{th} field campaign of excavations of its northern platform and of a rock-shelter in its western versant. Both of these areas can now be characterized as a habitat belonging to the First Bronze Age. This habitat comprises, in its lowermost stratigraphic phase, four excavated huts and a working area adjacent to one of them, as well as five occupation layers in the rock-shelter.

A later occupation moment in the rock-shelter is documented by several metal artefacts found in disturbed layers at its entrance, recovered during the field seasons of 2005 and 2006. The metal artefacts comprise:

- a double resort *fibula*;
- a flat pendant, decorated in both sides with punctuated lines forming star like motifs;
- a sewing needle;
- a cosmetics *spatula*;
- a wrist-band;
- a ring;
- three little bars.

The first four artefacts have an undeniable Mediterranean character suggesting a Phoenician origin (or at least a strong influence) from the southern areas of the Iberian Peninsula. Even the decoration of the pendant has close parallels in the orientalising graffiti found on Medellin grey ware [4]. This coincidence of possible cultural affiliation (c. 8th to 7th century BC) together with the finding nearby of a probable human premolar strongly suggests the existence of a Late Bronze Age (female?) burial in the disturbed area of the shelter entrance.

The pendant, needle, *fibula*, ring and the three metal fragments (Fig. 1) were studied from an archaeometallurgical point of view.

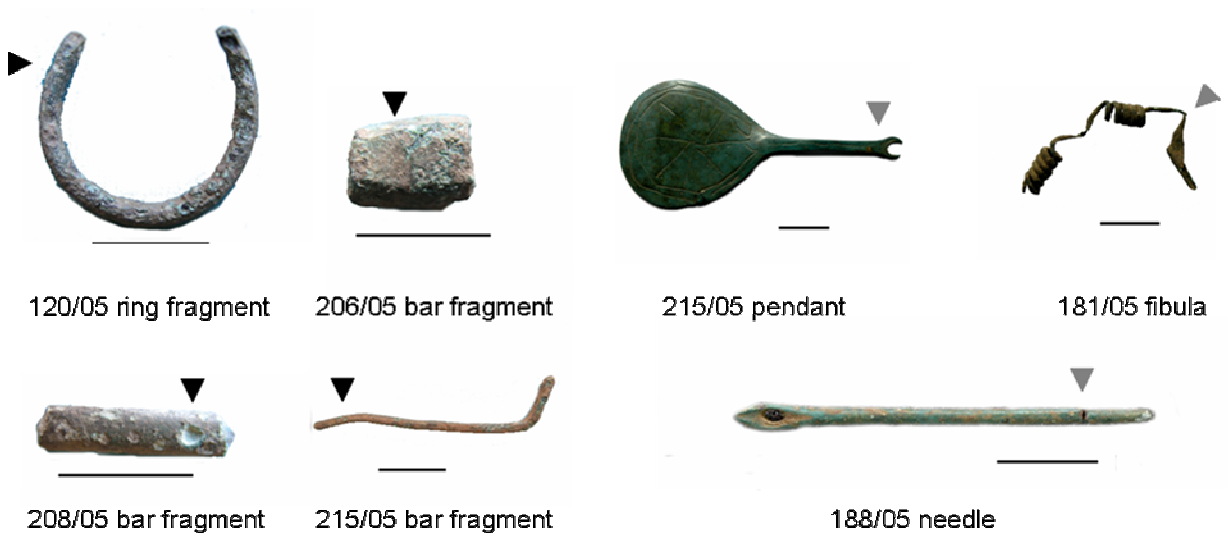

Fig. 1 – Image of the artefacts analysed. Black lines represent one centimetre for each metal artefact. Black triangles indicate where the ring and the three bar fragments were sampled for cross-section analyses. Grey triangles indicate where micro-EDXRF analyses were made with no surface treatment or preparation: in fracture surface of needle and *fibula* and in internal corrosion layer exposed in the pendant.

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1. EXPERIMENTAL DETAILS

The needle (188/05) is complete but has suffered a recent fracture close to the needle point (see Fig.1) separating the needle into two fragments. This allowed the analyses of the uncorroded metal alloy exposed in the centre of the fracture. The pendant (252/05) is complete and shows a uniform green patina over the surface with some small areas where the outer corrosion layer has been lost and an internal corrosion layer is exposed. It shows signs to have been hanging during long periods of time since the perforation is worn out. The *fibula* (181/05) is almost complete, missing only the pin and is composed by a single piece of metal string. Between the spring and the pin rest the *fibula* has a recent fracture, leaving the two parts of the *fibula* united by a small metallic branch that was analysed. The three bar fragments (206/05; 208/05 and 215/05) have distinct sizes and their original function is unknown, as well as the ring (120/05).

In order to maintain the physical integrity of the first three artefacts only non destructive superficial analyses were made with no surface preparation. The three bar fragments and the ring were sampled for metallographic studies in such a way that their cross-sections could be observed.

Energy dispersive micro X-ray fluorescence spectrometry (micro-EDXRF) was used to determine the metal composition of the artefacts. Analyses were performed in: the metal bulk of the polished cross-sections of the three bar fragments and the ring; and in the metal exposed due to the recent fractures of the needle and *fibula*. Since the pendant showed no metal exposed, analyses were made on a small area where the external corrosion layer was missing. EDXRF results are then much closer to metal composition than analysis performed over an external (thicker) corrosion layer.

For the micro-EDXRF analyses it was used an ArtTAX Pro spectrometer that comprises a low-power X-ray tube with a molybdenum anode. The system includes a set of polycapillary lens that generate a micro spot, lower than 100 µm in diameter, of primary radiation [5].

All the mounted samples from the four metal fragments were first polished in SiC abrasive paper from 240 to the 2400 grit size and then polished with a diamond suspension in a rotary polishing wheel (until one-quarter micron size diamond).

The etching was made using an aqueous ferric chloride solution. Samples were examined both in unetched and etched conditions.

A selected sample, cross-section of bar fragment 206/05, was studied under the scanning electron microscope (SEM), Zeiss model DSM 962, with a secondary electrons detector (SE), backscattered electrons detector (BSE) and an energy dispersive spectrometer (EDS) from Oxford Instruments model INCAx-sight with a ultra thin window. The sample was previously sputtered with a thin layer of gold.

2. RESULTS AND DISCUSSION

Results from micro-EDXRF analyses are presented in Table 1 and show that all artefacts are made of a bronze alloy with lead content up to 2%. Tin content varies from 8 to 12% (except for the pendant determinations), and traces of arsenic is detected in two of the bar fragments (208/05 and 215/05) and the ring (120/05). The high tin value obtained for the pendant can be due to the presence of a Sn enriched corrosion layer formed by decuprification phenomena undergone during burial in corrosion layers [6,7]. No nickel is detected and iron is present in contents under 0.04%.

Table 1. Micro-EDXRF analyses performed in artefacts and metal fragments. (n.d. – not detected)

			% wt, normalised					
			Cu	Sn	Pb	As	Fe	Ni
252/05	Pendant	corr[1]	72.2	26.6	1.15	n.d.	<0.04	n.d.
188/05	Needle	metal[2]	86.8	11.8	1.46	n.d.	<0.04	n.d.
181/05	*Fibula*	metal[2]	87.4	11.9	0.64	n.d.	<0.04	n.d.
208/05	Bar frag	metal[3]	88.6	9.76	1.29	<0.1	<0.04	n.d.
206/05	Bar frag	metal[3]	89.2	8.57	2.04	n.d.	<0.04	n.d.
215/05	Bar frag	metal[3]	89.7	8.14	1.78	<0.1	<0.04	n.d.
120/05	Ring	metal[3]	86.8	12.2	0.79	<0.1	<0.04	n.d.

[1] Analyses that refer to internal corrosion layer that was uncovered by loss of superficial corrosion layer
[2] Analyses of metal in fracture zones with no surface preparation
[3] Analyses made over metal bulk in sampled, mounted and polished cross-section

In Fig. 2 are displayed the Sn and Pb elemental variations for each artefact. The low variations ranges in the tin content, from 8 to 12% may indicate that good metallurgical skills and control in making the bronze alloy were known and that availability or supply of copper and tin (or bronze) existed. Lead content in values lower than 2% in all artefacts and fragments analysed may indicate that this element results from ore impurities. The low geometric complexity of the artefacts analyzed and the mechanical work performed after casting seems do not justify an intentional lead addition.

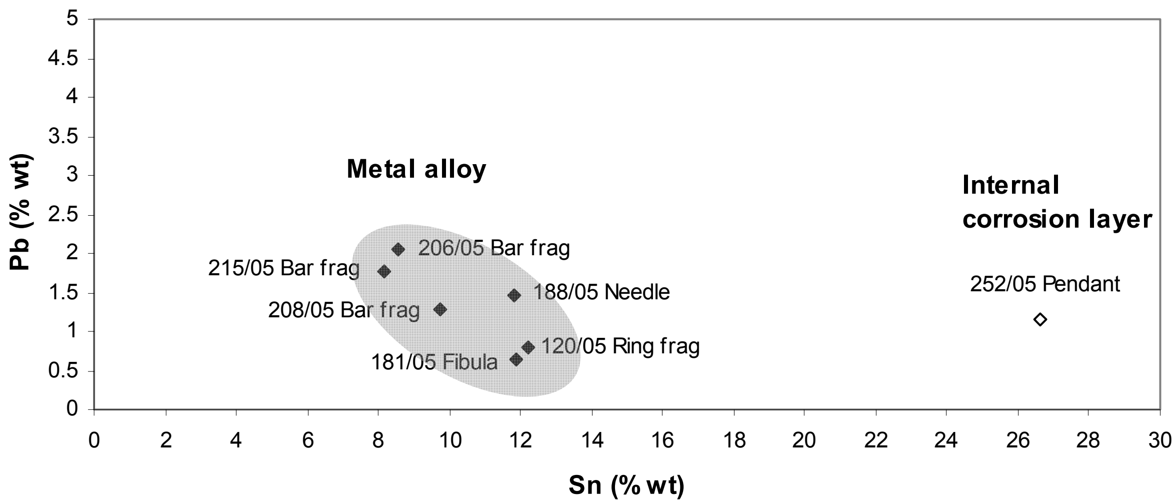

Fig. 2. Lead (Pb) and tin (Sn) content in metal alloy of needle, *fibula*, bar fragments and ring and in internal corrosion layer of the pendant.

Regarding archaeometallurgical data from other related Late Bronze Age contexts a comparison can be done between the metal composition from these artefacts and the metal composition of artefacts from "Baiões/Santa Luzia" cultural group (Viseu, Beira Alta) some 150km south, namely from "Castro da Senhora da Guia de Baiões" and "Castro de Santa Luzia". Regarding the lead content, the metal alloys seem to be different in the two regions. The EDXRF analyses performed over 74 artefacts and fragments from "Castro da Senhora da Guia de Baiões" [8] and "Castro de Santa Luzia" [9] show that Pb content was on a regular basis lower than 0.5% in corrosion layers, which taking into account the observed strong copper leaching [10], it would probably be even lower in the metal core. This could mean that different ores were used in these two regions during Late Bronze Age.

Metallographic examinations made on the cross-sections of the bar fragments and ring show that all were subjected to mechanical work followed by annealing (Table 2). Two bar fragments (120/05 and 215/05) have also suffered mechanical work as a final operation (Fig. 3). The bar fragment 206/05 is the only one that shows an heterogeneous microstructure with areas that were not subjected to mechanical work (Fig. 4) and others that have suffered some mechanical work as the final operation. In all the analysed fragments sulphide inclusions have been observed (Fig. 5). The regular presence of significant quantities of sulphide inclusions has also been observed in the bar fragments from Santa Luzia site, meaning that sulphidic ores were probably used for smelting [11] in these northern regions during Late Bronze Age.

Table 2. Some data provided through the metallographic examination of cross-sections of the bar fragments and ring.

		Section	Thermo-mechanical operation chain			Sulphide inclusions	Pb globules	(α+δ) tin rich phase
			Mechanical work	+ Annealing	+ Final mechanical work			
208/05	Bar frag	oval	Yes	Yes	No	Yes	Yes	No
206/05	Bar frag	quadrangular	Most parts	Yes	Some parts	Yes	Yes	No
215/05	Bar frag	rectangular	Yes	Yes	Yes	Yes	Yes	No
120/05	Ring frag	circular	Yes	Yes	Yes	Yes	Yes	No

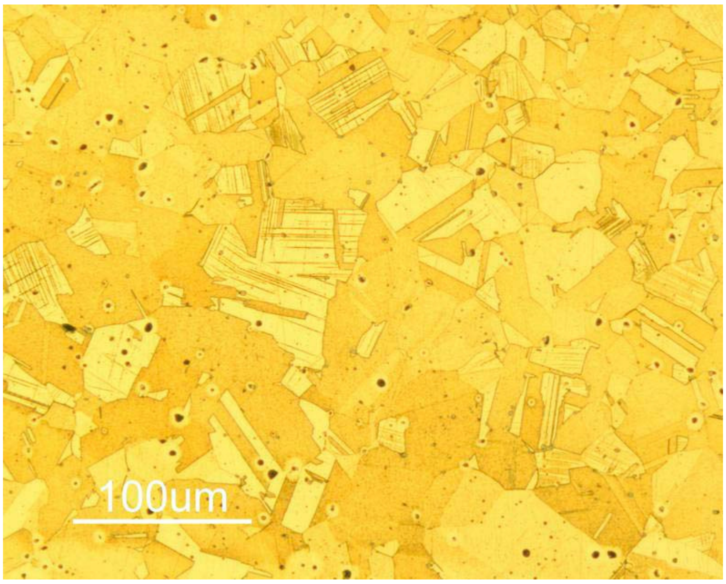

Fig. 3. Detail of cross-section from 120/05 ring. Microstructure showing annealed twins (broad bands) in recrystallized grains and deformation twins (thinner lines on the grains). Black dots are lead inclusions and/or copper sulphide inclusions. Reflected light microscope, bright field, sample etched.

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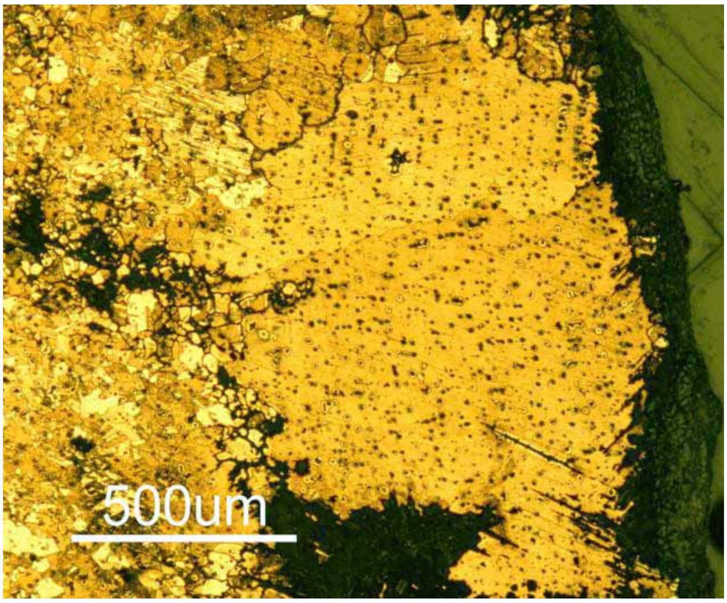

Fig. 4. Microstructure of cross-section from 206/05 bar fragment near the surface of the fragment (at right). The bar fragment shows heterogeneous microstructure: as cast (at right) and small grains with annealing twins (at left). Black dots are lead globules and/or copper sulphide inclusions. Reflected light microscope, bright field, etched sample.

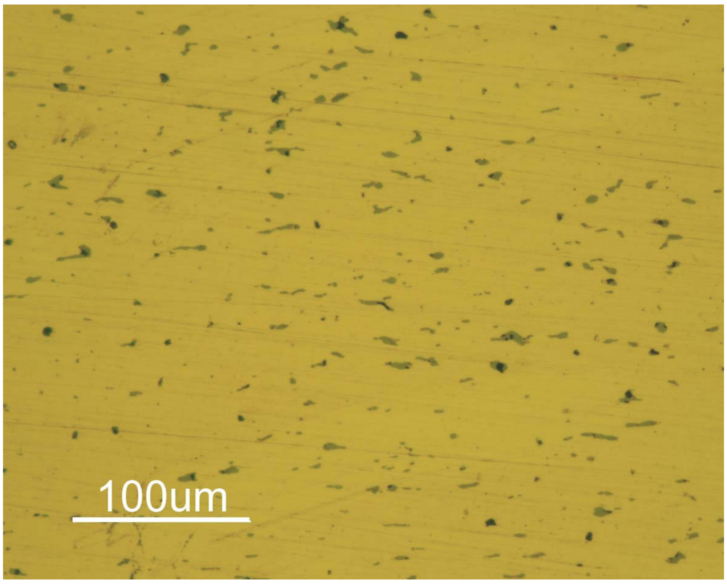

Fig. 5. Detail of cross-section from 208/05 bar fragment. Microstructure showing sulphide inclusions (grey colour) that have a preferable orientation (horizontal) that is perpendicular to the deformation induced by mechanical work. Reflected light microscope, bright field, sample unetched.

The presence of lead globules was observed in all bar fragments and ring, having the bar fragment 206/05 the higher density of lead globules (Fig. 6) which is in agreement with its higher Pb content determined by micro-EDXRF. Lead globules were not observed in the Castro de Santa Luzia bar fragments confirming the low Pb content in the metal used to produce them.

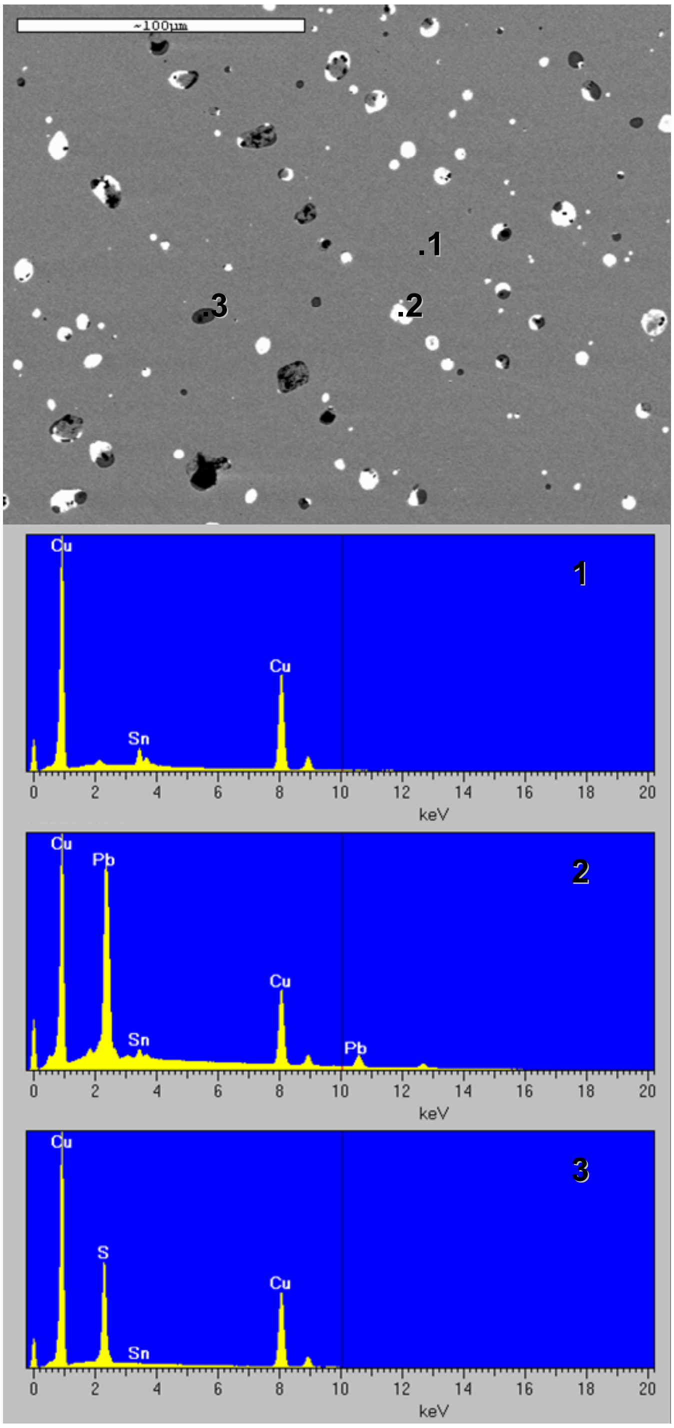

Fig. 6. Detail of the bar fragment 206/05 cross-section. SEM image (BSE) on top with marked points (black spots on left of each number) analysed by EDS (spectra bellow) as (1) α phase, (2) lead inclusions and (3) sulphide inclusions.

Eutectoid (α+δ) phases was not observed in the microstructure of Fraga dos Corvos metal fragments. Usually normal casting cooling rates, as those attained in sand castings, result in a biphasic microstructure. According to the binary phase diagram for copper-tin metal alloys, α copper rich phase is present as the only phase in the microstructure of an alloy with up to 8 to 12% Sn when an homogenizing heat treatment, as annealing, is performed. This heat treatment will improve the bronze mechanical properties, since it would allow an increase in hardness, by a solid solution effect of Sn in the α-Cu phase, resulting in the absence of the brittle δ phase in the final

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microstructure. In the case of a previous mechanical work a posterior heat treatment will help to recover is ductibility, by a recristalization process. A final mechanical work could increase/adjust the final hardness by a strain hardening effect.

This kind of homogeneous microstructure was not observed in the bar fragments previously analysed from Santa Luzia, where a microstructure with (α+δ) eutectoid was present and no final mechanical work was reported.

The amount of thermo-mechanical work that an artefact would need after the casting is related to its final shape and the mechanical properties needed. The differences in the microstructures found in the two sites can be explained through differences in the annealing operations that were carried out. This could be due to different skills of the metalworkers or due to different shapes and/or mechanical properties required for the artefacts. Since the original shape of the artefacts to which the bar fragments were related is not known, no correlation between thermo-mechanical treatments and final shape/mechanical properties required by the artefacts can be made. However, given that the bar fragments that were submitted to a final mechanical work had no (α+δ) eutectoid phases present may attest that a good knowledge of the metal properties and high metallurgical skills were already reached by the Late Bronze Age metal workers from the NE Portuguese regions.

3. CONCLUSIONS

The typological features of the artefacts found in the rock shelter in Fraga dos Corvos archaeological site attributes them to a Late Bronze Age cultural context. The metallographic examination of the bar fragments and ring show that the annealing heat treatment was long enough to homogenize the alloy and solubilise any remains of the δ eutectoid phase resulting in a decrease of brittleness and in a increase of tenacity of the metal. All bar fragments show a homogeneous microstructure over the cross-section except for bar fragment 206/05.
The alloy composition (8-12%Sn and <2%Pb) of the fragments and artefacts is very similar and is in agreement with a Late Bronze Age good quality bronze alloy. This shows that controlled metallurgical procedures and good supply in copper and tin (or bronze) were available.

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