Islamic Copper-Based Metal Artefacts from the Garb Al-Andalus. A Multidisciplinary Approach on the Alcáçova of Mārtulah (Mértola, South of Portugal)

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Abstract

A multidisciplinary approach has been applied to investigate the production technology of a collection of copper-based artefacts found during archaeological excavation campaigns carried out in the Almohad neighbourhood of Mārtulah, the Islamic name of modern Mértola (South of Portugal). In stark contrast to other Islamic materials found in the same site such as common and finely decorated pottery, glass, and bone artefacts, metal objects have received less attention despite the number of artefacts recovered.

This study focuses on the chemical characterisation of 172 copper-based artefacts dating back to the 12th and the first half of the 13th centuries. The artefacts are daily use objects and consist of personal ornaments (earrings, rings, and casket ornaments), tools (spindles, spatulas, and oil lamp sticks) and artefacts with unknown functions. X-ray fluorescence Spectroscopy (XRF) and Scanning Electron Microscopy coupled with Energy Dispersive X-ray Spectroscopy (SEM/EDS), provided information not only about technological issues, as well as on the socio-economic implications of metal consumption at Islamic Mértola. The results revealed that metals were produced with a variety of Cu-based alloys, namely unalloyed copper, brasses, bronzes, and ternary alloys, by mixing Cu, Zn and Sn and Pb without any apparent consistency, as a likely result of recurrent recycling and mixing scrap metals practices or use of minerals available locally.

Introduction

Islamic culture provided a very important contribution to European history and modern science and technology [1, 2]. In fact, since the 7th century AD, scholars from the Muslim world stand out in virtually every field of knowledge, being at the forefront of scientific advance and technological innovation in a wide range of research fields such as astronomy, medicine, mathematics, cartography, and agriculture [3, 4].

Since the arrival of the Muslim army led by Tariq ibn Ziyad (711 AD), who firstly crossed the Strait of Gibraltar from the North African coast, and up to the end of the 13th century, when the Christian Reconquest was completed in the west of al-Andalus, the Iberian Peninsula was gradually and actively involved in this climate of cultural and scientific development, generally known as Islamic Golden Age [5, 6]. Along this period of prosperity that lasted about five centuries, the Iberian Peninsula also played a central role as one of the major points of transmission of Islamic culture and technology to the rest of Europe.

Towns in al-Andalus like Seville, Cordoba or Granada progressively became the centre of social life and political power, also establishing themselves as commercial hubs where manufacturing activities (e.g., pottery, metalwork, tannery) and different forms of art (e.g., textiles, illuminated manuscripts, woodwork, architecture, ceramics, and metalwork) flourished. Within this scenario, Mārtulah, located at the confluence between the Guadiana River and the Oeiras creek, benefited from its strategic location as the last navigable point of the river, functioning as a commercial hub able to link the Atlantic Ocean with Northern Africa, al-Andalus and the Mediterranean Sea [7, 8] (Fig. 1).

The 12th and 13th centuries, i.e., the period when the metal artefacts analysed in this paper were produced and used, was characterized by a great trade development despite a political instability caused both by internal crises within the Islamic community and by increasing external pressure caused by the advancing Reconquest by the Christian Iberian kingdoms. From the beginning of the 2nd millennium, periods of political fragmentation (with the creation of the so-called *Taifas*), alternated with periods of reunification, especially under the Almoravid and Almohad dynasties. The conquest of Mértola by the Portuguese king Sancho II in 1238 was a key moment in the southward advance of the Christian forces that, in 1249, with King Afonso III, put an end to the Islamic presence in southern Portugal by conquering the Algarve region.

The systematic archaeological excavations carried out along more than 40 years in the urban area of the modern Mértola have shed light on this historical period, allowing to reconstruct its history and the evolution of the town since the Iron Age period and across the centuries. Important archaeological vestiges, including monumental buildings, still stand as evidence of Islamic Mārtulah. One of the most relevant excavated areas is the Almohad neighbourhood, located in the Alcazaba of Mértola, i.e., the walled fortification, in an area located on the northern slope of the castle that overlooks the town (Fig. 2).

The area where the Almohad neighbourhood is located stands on an artificial platform built on a late Roman cryptoporticus, already occupied by buildings richly decorated with mosaics during the Late Antiquity (5th -8th centuries). Around the 12th century, it was used for the edification of a neighbourhood that was abandoned soon after the Christian conquest of Mértola. In the following centuries, the area served for different purposes: it was used as a Christian cemetery until the 18th century, as a vegetable garden afterwards and from the beginning of the 20th century onwards as a football pitch. The metals analysed in this paper are from the excavated area, where 15 houses from the Islamic period have been dug [9] (Fig. 2).

Even though Islamic copper-based artefacts have been recurrently found at different excavations carried out at Mértola, this paper present for the first time the results of a large-scale analytical program aimed at characterising the chemical composition of a collection of metals composed of 172 artefacts. In fact, the lack of analytical data on Islamic metallurgy is a common trait to other Iberian regions. Very few studies have been dedicated to this topic so far, with few exceptions, i.e., Madinat al-Zahra (Córdoba) [10], Denia (Alicante) [11, 12], Silves (Faro) [13], and a collection of oil lamps from different Portuguese sites [14].

The analytical strategy adopted in this work was oriented towards the acquisition of information on the composition of copper alloys by means of portable and hand-held X-ray fluorescence spectroscopy (XRF) and scanning electron microscopy (SEM) coupled with energy dispersive X-ray spectrometry (EDS). The aim of the research was to provide an overview of the metal production technology in the site while, at the same time shedding light on social and economic issues related to the use of metal in Mértola during the last phase of Islamic rule.

Materials And Methods

Materials

The collection of objects analysed in this paper represents a selection of the copper-based artefacts recovered to date from the Almohad neighbourhood of Mārtulah. The assemblage includes 172 artefacts that, based on their typology, can be subdivided into three different groups, namely: a) tools (i.e., spindles, oil lamp sticks, and spatulas), b) ornaments (i.e., rings, earrings, buckles, casket ornaments), and c) fragmented object of undetermined function (Fig. 3). Regardless of their typological characteristics, it is important to stress that all the artefacts analysed here come from domestic contexts and can therefore be considered as objects of daily use.

Methods

The XRF equipment used was a Bruker TRACER III-SD handheld spectrometer equipped with a rhodium anode tube and a Silicon Drift Detector with a resolution of 140 eV at Mn Kα FWHM 5.9 keV. The operating conditions were 40 kV and 3 μA current with an Al/Ti filter (304.8 μm aluminium/25.4 μm titanium) and 60 s acquisition time. The spectra were acquired using the Bruker S1PXRF v.3.8.30 software and Bruker ARTAX v.5.3.0.0 software for the first spectra evaluation. For system calibration, analyses of certified copper alloys were used, namely five standards from certified reference materials [15] and three standard reference materials (National Bureau of Standards Standard Reference Material 1107, 1110 and 1113). Quantification was performed using Bruker S1CalProcess v.2.2.33 software to find the concentration of the unknown samples.

A few selected artefacts were also investigated by SEM/EDS using a HITACHI S3700N interfaced with a QUANTAX EDS microanalysis system equipped with a Bruker AXS 5010XFlash 5010 Silicon Drift Detector (129 eV Spectral Resolution at FWHM/Mn Kα). The operating conditions for EDS analysis were set as follows: backscattered electron mode (BSEM), 20 kV accelerating voltage, ~10 mm working distance and 90 μA emission current. Spectra were analysed using Bruker Esprit 1.9 software.

Results And Discussion

Results are summarized in Table 1. Even though we recognise the arbitrary nature of establishing a threshold values system for defining the different types of ancient alloys, to better organize the collected data, the following scheme proposed by Gaudenzi Asinelli et al. has been adopted [16]:

Table 1
Elemental composition of the artefacts from the Almohad neighbourhood of Mārtulah (wt.%); n.d.: not detected.

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
Brass										
AA-01-02	casket ornament	91.90	4.95	1.40	0.75	0.30	0.46	0.07	0.02	0.15
AA-01-04	casket ornament	90.30	6.60	1.60	0.93	0.07	0.20	0.09	0.01	0.20
AA-01-13	casket ornament	86.75	12.40	0.48	0.08	0.19	0.10	n.d.	n.d.	n.d.
AA-01-14	casket ornament	89.35	8.35	0.90	0.40	0.35	0.40	0.09	0.01	0.15
AA-01-16	casket ornament	91.00	7.20	0.33	0.17	0.30	0.90	0.06	0.01	0.03
AA-01-17	casket ornament	89.30	8.75	1.20	0.22	0.02	0.42	0.09	n.d.	n.d.
AA-01-20	casket ornament	88.70	8.45	0.50	0.90	0.30	1.15	n.d.	n.d.	n.d.
AA-02-25	casket ornament	86.45	10.40	0.60	0.95	0.32	0.80	0.48	n.d.	n.d.
AA-02-26	casket ornament	86.91	10.34	0.91	1.15	0.22	0.15	0.32	n.d.	n.d.
AA-02-27	casket ornament	88.12	7.90	1.40	1.55	0.25	0.23	0.26	0.02	0.27
AA-02-28	casket ornament	93.05	4.62	0.70	0.75	0.29	0.15	0.29	n.d.	0.15
AA-02-29	casket ornament	77.31	22.37	n.d.	0.10	0.02	0.13	n.d.	0.07	n.d.
AA-02-30	casket ornament	83.92	14.52	n.d.	1.10	0.26	0.20	n.d.	n.d.	n.d.
AA-02-32	casket ornament	83.56	14.74	1.10	0.15	0.23	0.20	n.d.	0.02	n.d.
AA-02-33	casket ornament	78.31	20.30	1.10	0.05	0.03	0.12	0.06	0.03	n.d.
AA-02-36	casket ornament	86.15	12.07	1.25	0.30	0.10	0.13	n.d.	n.d.	n.d.
AA-02-37	casket ornament	86.55	10.20	1.70	1.02	0.10	0.19	0.07	0.02	0.15

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
AA-02-46	casket ornament	90.46	6.52	1.37	0.95	0.20	0.24	0.07	0.02	0.17
AA-02-54	casket ornament	88.80	9.95	n.d.	0.15	0.08	0.70	0.11	0.17	0.04
AA-03-62	casket ornament	81.93	14.11	1.40	1.35	0.11	0.90	0.20	n.d.	n.d.
AN-01-36	ring	87.51	10.50	0.40	0.55	0.27	0.70	0.07	n.d.	n.d.
AR-01-35	ring	85.22	12.07	1.38	0.80	0.26	0.22	n.d.	0.05	n.d.
BC-01-05	earring	94.32	2.47	1.16	1.40	0.10	0.55	n.d.	n.d.	n.d.
BC-01-06	earring	91.04	7.07	0.70	0.40	0.05	0.66	0.08	n.d.	n.d.
BC-01-07	earring	93.41	4.75	0.65	0.60	0.06	0.15	0.14	0.01	0.23
BC-01-11	earring	89.79	7.98	1.35	0.21	0.03	0.55	0.09	n.d.	n.d.
BC-01-16	earring	88.53	9.41	1.05	0.45	0.10	0.31	0.15	n.d.	n.d.
BC-01-17	earring	82.88	14.12	1.35	0.65	0.20	0.80	n.d.	n.d.	n.d.
BC-01-18	earring	85.47	13.08	0.85	0.15	0.07	0.25	0.13	n.d.	n.d.
BC-01-19	earring	88.67	9.23	0.65	0.22	0.06	1.10	0.07	n.d.	n.d.
BC-01-21	earring	82.98	14.72	1.20	0.25	0.05	0.80	n.d.	n.d.	n.d.
BC-01-26	earring	86.96	11.55	0.75	0.10	0.06	0.50	0.08	n.d.	n.d.
BC-01-30	earring	92.38	4.98	1.16	0.95	0.12	0.40	n.d.	0.01	n.d.
BC-01-42	earring	88.27	8.71	0.90	0.48	0.14	1.50	n.d.	n.d.	n.d.
BC-01-46	earring	91.79	5.76	0.88	0.62	0.10	0.85	n.d.	n.d.	n.d.
BC-01-55	earring	88.69	7.73	1.85	0.85	0.20	0.40	n.d.	0.02	0.26

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
BC-01-61	earring	90.92	5.75	1.35	1.00	0.26	0.35	0.11	0.02	0.24
BC-01-62	earring	87.33	10.32	1.15	0.21	0.03	0.90	0.06	n.d.	n.d.
BC-01-66	earring	90.56	5.61	1.80	0.95	0.25	0.55	0.10	n.d.	0.18
BC-01-72	earring	86.07	11.43	1.30	0.50	0.02	0.60	0.07	0.01	n.d.
BC-01-76	earring	91.58	6.44	0.75	0.60	0.04	0.25	0.15	0.01	0.18
BC-01-89	earring	84.81	12.14	1.63	0.68	0.06	0.35	0.09	0.01	0.23
BC-03-101	earring	83.48	14.74	1.55	0.05	0.03	0.15	n.d.	n.d.	n.d.
BC-03-105	earring	88.35	9.00	1.25	0.60	0.20	0.30	0.09	0.01	0.20
BC-03-106	earring	87.23	9.42	1.70	1.05	0.05	0.30	0.07	0.01	0.17
BC-03-108	earring	83.68	14.32	0.87	0.38	0.10	0.65	n.d.	n.d.	n.d.
BC-03-109	earring	88.55	8.19	1.15	1.40	0.25	0.30	0.16	n.d.	n.d.
BC-03-115	earring	90.60	7.60	0.48	0.60	0.13	0.33	0.08	0.01	0.17
BC-03-97	earring	91.97	5.10	1.30	0.65	0.19	0.70	0.09	n.d.	n.d.
BC-03-98	earring	83.50	15.10	1.17	0.05	0.04	0.08	n.d.	0.06	n.d.
DV-02-44	undetermined	93.28	4.19	0.90	0.25	0.21	0.74	0.43	n.d.	n.d.
DV-02-75	undetermined	89.80	7.65	0.95	0.85	0.09	0.47	0.06	0.01	0.12
DV-02-80	undetermined	89.87	6.45	1.30	1.93	0.12	0.22	n.d.	0.01	0.10
DV-03-113	sword sheath	93.98	2.27	0.80	1.35	0.18	0.60	0.16	0.07	0.59
DV-03-86	undetermined	88.30	8.90	1.55	0.48	0.03	0.48	0.08	0.02	0.16

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
DV-03-89	undetermined	89.46	8.10	0.95	0.47	0.35	0.45	0.07	0.02	0.13
EC-01-05	oil lamp stick	87.86	11.65	n.d.	0.30	0.07	0.12	n.d.	n.d.	n.d.
EC-01-07	oil lamp stick	85.70	14.05	n.d.	0.10	0.07	0.03	0.05	n.d.	n.d.
EC-01-09	oil lamp stick	89.74	6.94	1.65	1.15	0.25	0.21	0.06	n.d.	n.d.
EC-01-12	oil lamp stick	85.54	12.43	0.10	1.07	0.20	0.47	0.14	0.05	n.d.
EC-01-15	oil lamp stick	88.13	10.67	n.d.	0.55	0.03	0.10	0.21	0.31	n.d.
EC-01-16	oil lamp stick	88.89	8.70	0.60	0.38	0.35	0.78	0.14	0.01	0.15
ES-01-01	spatula	85.69	13.70	0.01	0.50	0.03	0.07	n.d.	n.d.	n.d.
ES-01-06	spatula	92.77	5.18	0.30	0.54	0.14	0.90	n.d.	0.02	0.15
ES-01-08	spatula	91.43	4.77	1.38	0.95	0.87	0.31	0.26	0.03	n.d.
ES-01-11	spatula	94.88	2.52	0.49	0.43	0.16	1.30	n.d.	0.01	0.21
ES-01-12	spatula	87.94	7.33	2.02	1.55	0.21	0.58	n.d.	0.03	0.34
ES-01-20	spatula	89.03	8.07	0.85	0.77	0.20	0.71	0.14	0.05	0.18
ES-01-22	spatula	88.14	7.68	1.55	1.70	0.15	0.44	0.12	n.d.	0.22
ES-02-23	spatula	88.66	9.67	0.50	0.51	0.04	0.62	n.d.	n.d.	n.d.
ES-02-27	spatula	89.26	8.41	0.74	0.28	0.10	1.20	n.d.	0.01	n.d.
ES-02-29	spatula	85.99	10.63	1.40	1.10	0.15	0.40	n.d.	0.03	0.30
ES-02-30	spatula	88.77	7.63	0.83	1.80	0.12	0.40	0.20	0.03	0.22
ES-02-54	spatula	93.94	4.49	0.24	0.61	0.21	0.30	0.20	0.01	n.d.

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
PF-01-02	spindle	88.39	7.61	1.41	0.90	0.06	1.35	0.27	0.01	n.d.
PF-01-04	spindle	90.93	5.08	2.03	0.49	0.15	0.50	0.60	0.01	0.21
PF-01-06	spindle	90.36	7.90	0.38	0.62	0.20	0.14	0.15	0.04	0.21
PF-01-100	spindle	93.10	3.67	1.55	0.52	0.35	0.23	0.27	0.01	0.30
PF-01-104	spindle	89.45	8.20	0.61	0.66	0.40	0.40	0.11	0.01	0.16
PF-01-106	spindle	84.99	12.06	0.85	0.88	0.16	1.05	n.d.	0.01	n.d.
PF-01-107	spindle	85.23	14.08	0.03	0.32	0.13	0.14	0.07	n.d.	n.d.
PF-01-16	spindle	88.66	9.23	1.03	0.13	0.15	0.80	n.d.	n.d.	n.d.
PF-01-20	spindle	85.81	11.58	1.42	0.32	0.07	0.80	n.d.	n.d.	n.d.
PF-01-27	spindle	88.54	9.28	0.40	0.65	0.13	0.86	0.14	n.d.	n.d.
PF-01-32	spindle	80.91	15.48	1.90	0.40	0.16	1.15	n.d.	n.d.	n.d.
PF-01-33	spindle	91.05	6.67	0.60	0.20	0.05	1.35	0.08	n.d.	n.d.
PF-01-36	spindle	81.16	15.78	1.66	0.20	0.23	0.83	0.13	0.01	n.d.
PF-01-37	spindle	83.62	13.83	1.00	0.40	0.25	0.79	0.11	n.d.	n.d.
PF-01-47	spindle	93.09	4.90	0.70	0.79	0.28	0.11	0.13	n.d.	n.d.
PF-01-54	spindle	83.75	14.87	0.29	0.17	0.07	0.78	n.d.	n.d.	0.07
PF-01-65	spindle	87.95	9.26	1.20	0.32	0.04	1.15	0.07	0.01	n.d.
PF-01-77	spindle	84.43	11.90	1.45	1.70	0.18	0.30	n.d.	0.04	n.d.
PF-01-81	spindle	92.73	4.39	1.15	0.85	0.19	0.55	0.14	n.d.	n.d.

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
PF-01-85	spindle	89.42	6.98	0.83	0.88	0.58	1.20	0.10	0.01	n.d.
PF-02-114	spindle	94.92	2.68	0.56	1.50	0.09	0.25	n.d.	n.d.	n.d.
PF-02-122	spindle	79.28	19.83	0.11	0.42	0.08	0.28	n.d.	n.d.	n.d.
PF-02-129	spindle	88.29	8.70	1.65	0.84	0.24	0.16	0.12	n.d.	n.d.
PF-02-131	spindle	88.57	8.95	1.70	0.41	0.07	0.19	0.08	0.03	n.d.
PF-02-156	spindle	84.17	13.09	1.10	0.37	0.15	0.88	0.24	n.d.	n.d.
PF-02-179	spindle	91.47	6.86	0.88	0.43	0.03	0.14	n.d.	0.19	n.d.
PF-02-182	spindle	86.72	9.23	1.81	1.72	0.20	0.26	n.d.	0.06	n.d.
PF-02-183	spindle	88.63	9.67	0.63	0.22	0.10	0.67	0.08	n.d.	n.d.
PF-02-185	spindle	84.86	13.43	1.26	0.15	0.06	0.13	0.05	0.06	n.d.
PF-02-223	spindle	90.63	5.92	1.74	0.95	0.05	0.38	0.11	0.02	0.20
PF-02-231	spindle	87.98	8.25	2.06	1.03	0.10	0.14	n.d.	0.44	n.d.
PF-02-232	spindle	82.17	14.86	1.74	0.73	0.20	0.25	n.d.	0.05	n.d.
Leaded brass										
AA-02-56	casket ornament	71.20	14.75	1.75	10.90	0.32	0.42	0.20	0.02	0.44
BC-01-81	earring	85.07	9.78	0.12	3.70	0.13	1.05	0.15	n.d.	n.d.
BC-03-100	earring	86.64	8.63	1.30	2.30	0.55	0.30	0.09	0.01	0.18
BC-03-120	earring	90.50	4.10	0.40	3.75	0.20	0.70	0.35	n.d.	n.d.
DV-03-95	undetermined	86.88	3.06	1.71	6.50	0.58	0.45	0.13	0.15	0.54

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
EC-01-14	oil lamp stick	75.84	19.69	0.07	4.19	0.07	0.12	n.d.	0.02	n.d.
ES-01-10	spatula	89.85	2.10	1.25	6.20	0.08	0.52	n.d.	n.d.	n.d.
PF-01-31	spindle	74.97	20.58	1.10	2.20	0.14	0.52	0.18	0.03	0.28
PF-01-38	spindle	80.97	15.44	0.14	2.90	0.11	0.20	0.24	n.d.	n.d.
PF-01-44	spindle	92.88	2.24	1.14	2.85	0.04	0.25	0.21	n.d.	0.39
PF-02-130	spindle	85.54	5.83	1.93	5.74	0.40	0.36	0.20	n.d.	n.d.
PF-02-230	spindle	91.89	3.33	1.62	2.10	0.30	0.51	0.24	0.01	n.d.
Bronze										
BC-02-70	earring	94.99	0.08	4.25	0.40	0.06	0.20	n.d.	0.02	n.d.
DV-02-47	undetermined	91.32	0.15	5.41	1.16	0.25	0.14	1.56	0.01	n.d.
DV-02-64	undetermined	90.54	0.06	9.09	n.d.	0.11	0.14	0.06	n.d.	n.d.
PF-01-01	spindle	91.34	1.54	4.63	1.10	0.20	0.61	0.17	n.d.	0.41
PF-01-78	spindle	91.94	1.94	2.93	0.79	0.20	0.26	1.94	n.d.	n.d.
Ternary alloy										
AA-01-06	casket ornament	83.75	10.70	2.30	1.47	0.50	0.25	0.85	0.03	0.15
AA-01-08	casket ornament	84.40	11.00	3.00	1.05	0.15	0.17	0.10	0.03	0.10
AA-01-18	casket ornament	88.60	7.40	2.10	1.10	0.05	0.32	0.08	0.15	0.20
AA-02-49	casket ornament	87.92	7.76	3.40	0.34	0.50	0.08	n.d.	n.d.	n.d.
AA-03-79	casket ornament	84.52	10.39	3.30	1.40	0.17	0.20	n.d.	0.02	n.d.

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
AN-01-35	ring	88.52	7.48	2.64	0.50	0.05	0.55	0.08	0.01	0.17
BC-01-09	earring	91.40	5.60	1.95	0.57	0.06	0.15	0.10	n.d.	0.17
BC-01-12	earring	89.71	5.45	3.65	0.60	0.02	0.27	0.09	n.d.	0.21
BC-01-82	earring	90.13	4.81	2.41	1.25	0.50	0.90	n.d.	n.d.	n.d.
BC-03-116	earring	86.52	6.27	4.85	1.15	0.20	1.00	n.d.	0.01	n.d.
EC-01-06	oil lamp stick	87.76	8.35	2.35	0.90	0.07	0.32	0.11	0.01	0.13
EC-01-08	oil lamp stick	82.46	12.62	2.75	1.25	0.05	0.12	0.40	n.d.	0.35
ES-02-26	spatula	86.74	9.39	2.30	0.81	0.04	0.32	0.13	n.d.	0.27
ES-02-36	spatula	88.22	8.05	2.50	0.58	0.25	0.26	0.14	n.d.	n.d.
PF-01-03	spindle	89.18	6.13	2.82	0.78	0.15	0.59	0.10	n.d.	0.25
PF-01-05	spindle	91.77	4.23	2.25	0.86	0.14	0.37	0.37	0.01	n.d.
PF-01-11	spindle	86.88	11.59	0.43	0.25	0.12	0.72	n.d.	0.01	n.d.
PF-01-30	spindle	77.77	17.61	3.68	0.74	0.09	0.11	n.d.	n.d.	n.d.
PF-01-55	spindle	87.74	7.03	3.35	1.10	0.29	0.24	0.25	n.d.	n.d.
PF-01-99	spindle	90.06	4.15	3.80	1.25	0.15	0.18	0.12	0.02	0.27
PF-02-128	spindle	84.22	10.78	2.97	1.15	0.48	0.34	n.d.	0.06	n.d.
PF-02-161	spindle	84.89	11.74	2.19	0.75	0.15	0.26	n.d.	0.02	n.d.
PF-02-178	spindle	87.33	8.06	2.90	1.18	0.15	0.30	n.d.	0.08	n.d.
Leaded ternary alloy										

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
AA-01-21	casket ornament	88.10	3.70	3.05	4.00	0.50	0.37	0.09	0.02	0.17
AA-02-39	casket ornament	84.68	7.33	5.30	2.15	0.24	0.07	0.10	0.01	0.12
AR-01-53	ring	75.13	2.12	9.75	11.05	0.55	0.80	n.d.	0.05	0.55
BC-01-86	earring	68.74	18.08	6.25	6.30	0.09	0.30	0.21	0.03	n.d.
BC-03-118	earring	83.57	10.94	2.34	2.25	0.32	0.50	n.d.	0.08	n.d.
DV-01-31	undetermined	82.25	9.30	4.30	3.60	0.15	0.10	0.09	0.01	0.20
DV-02-63	undetermined	82.71	4.96	2.12	7.85	0.40	0.64	0.20	0.10	1.02
EC-01-01	oil lamp stick	85.62	8.73	2.12	2.75	0.25	0.12	0.14	n.d.	0.27
PF-01-08	spindle	69.34	21.27	2.33	5.90	0.17	0.77	0.15	0.07	n.d.
PF-01-14	spindle	82.98	5.71	4.00	6.60	0.51	0.20	n.d.	n.d.	n.d.
Unalloyed copper										
AA-02-43	casket ornament	99.52	0.08	n.d.	0.21	0.01	0.15	n.d.	0.03	n.d.
AA-03-59	casket ornament	96.45	1.90	0.26	0.65	0.07	0.16	0.15	0.06	0.30
AA-03-66	casket ornament	97.05	0.13	1.30	0.15	0.29	0.65	0.05	0.08	0.30
BC-03-117	earring	96.13	1.58	0.87	0.45	0.23	0.63	n.d.	0.11	n.d.
DV-01-04	undetermined	98.00	0.09	0.10	0.63	0.13	0.09	0.62	0.04	0.30
DV-01-05	undetermined	98.92	0.10	n.d.	0.03	0.25	0.13	0.57	n.d.	n.d.
DV-01-36	undetermined	98.55	0.09	n.d.	0.33	0.10	0.06	0.66	0.02	0.19
DV-02-53	undetermined	97.70	0.10	0.95	0.80	0.02	0.14	0.11	0.02	0.16

ID	Function	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
DV-02-54	undetermined	96.44	1.77	0.85	0.35	0.17	0.24	n.d.	n.d.	0.18
DV-02-79	undetermined	98.07	0.07	0.26	0.80	0.04	0.31	0.25	0.02	0.18
FIV-01-12	buckle	98.95	0.09	n.d.	0.64	0.05	0.04	0.09	0.01	0.13
PR-02-13	nail	98.55	0.25	0.07	0.66	0.23	0.20	n.d.	0.04	n.d.
Leaded unalloyed copper										
AR-01-42	ring	96.63	0.05	0.63	2.30	0.03	0.25	0.11	n.d.	n.d.
DV-01-19	undetermined	96.48	0.09	0.49	2.30	0.20	0.18	0.09	0.09	0.08
PR-02-06	nail	93.62	0.11	0.15	4.85	0.24	0.46	0.16	0.03	0.38

- unalloyed Cu: Cu + <2% Sn and <2% Zn;
- bronze: Cu + >2% Sn and <2% Zn;
- brass: Cu + >2% Zn and <2% Sn;
- ternary alloy (modern gunmetal): Cu + >2% Sn and >2% Zn;
- Leaded alloys: >2% Pb.

According to the adopted criteria, brass was the most frequent alloy in use at Mértola, being represented by 118 artefacts, 12 of which with more than 2 wt.% Pb. Seventeen objects were made of unalloyed Cu, three of which with more than 2 wt.% Pb. Ternary alloys are 36, 10 of which with more than 2 wt.% Pb. Finally, there are six bronzes, none of them with Pb higher than 2 wt.% (Fig. 4A). As for minor elements, i.e., Zn, Sn, and Pb when <2 wt.%, Fe, As, Ag, Ni, and Sb, their content tends to be quite variable, ranging from 0.21 to 3.25 wt.% in total.

At first glance, the results showed Zn to be the principal element to have been mixed with Cu and brass is the main alloy detected (Fig.4 B). At the same time, the data also indicate a seemingly random and unpredictable variability of the major components, i.e., Zn, Sn and Pb.

As the addition of each of these elements is known to alter the specific mechanical properties of a metal, a major archaeological question to address would be to find the reasons behind this variability. In this sense, the presence/absence of standardised compositional patterns can shed light not only on production technology aspects, but also on a range of issues related to different features of past societies. In fact, social strategies, technological choices, and economic constraints may have an impact on any single phase in the manufacturing process, including mining, smelting, melting, and casting processes, and any decisions made along the production chain, whether conscious or irrational, can directly affect the characteristics of the finished objects.

Based on this assumption, and looking at the overall Mértola data, it is quite clear that the high variability of the main elements found in the objects analysed does not allow clear compositional patterns to be identified. Tools, ornaments, and fragments that do not fall into any of the previous categories seem to be produced seamlessly together with alloys containing variable concentrations of Zn, Sn and Pb. Considering the high technological expertise in metal production reached during the Islamic Iberian Peninsula [12, 14, 17], it is not at all likely that the metalworkers who produced the objects found in Mértola were unaware of the mechanical properties of the different copper-based alloys to the point of not taking advantage of them. A detailed analysis of each of the alloys identified at Mértola will be presented with the aim of discussing the probable causes of this elemental variability.

Brass artefacts (Cu-Zn)

Brass is the predominant alloy produced and used at Mértola. It representsalmost69% of the entire assemblage analysed in this work, having been used to produce different types of metals, namely: 37 spindles, 31 earrings, 21 casket ornaments, 13 spatulas, 6 undetermined, 2 rings, and 1 sword sheath. Furthermore, only 12 out of 118 brasses contain more than 2% of Pb (Fig. 5 A).

Overall, Zn ranges from 2.1 to 22.3 wt.%, showing an average of *c.* 9.5 wt.%. Only one of the objects analysed (AA-02-29) has a Zn content higher than 22 wt.%, thus falling into the 22-28 wt.% Zn range, that is the typical interval for brasses produced with the method of cementation [18-20].

Fig. 5 B confirms the lack of correlation between Zn variability and the functionality of the artefacts. In fact, a Zn content in the range of 10-20 wt.% is known to be responsible for a golden yellow colour in the final alloy, making the latter particularly suitable for ornamental objects. When looking at Mértola's data, though, the 40 artefacts that fall in this range are evenly distributed between tools and ornaments, suggesting that the brightness and colour nature of an alloy was probably not considered relevant in producing finished objects with specific forms and functions.

With only few exceptions, almost all the brasses (105 out of 118) contain Zn between 4% and 20% (Fig. 5C), that is the range that Craddock considers as typical for objects produced through the mixing of pristine brass with 22 to 28 wt.% Zn with scrap copper-based alloys with lower Zn content [18].

The overall Zn content, however, is evidence for the use of scrap metals as raw materials to produce new objects. Further evidence in support of this hypothesis is the moderate and apparently random occurrence of Zn. When a brass is remelted, the alloy progressively loses about 10 wt.% of its Zn content, and a 4 to 5 wt.% additional Zn should be added to compensate the melting losses. Considering, for instance, the recycling of an ancient brass produced via the so-called cementation process and containing 28% Zn, Zn content may drop to about 25 wt.% after the first remelting, to 22 wt.% after a further remelting, and so on [21]. This means that the Zn variability observed in Mértola's brass objects is most likely the result of multiple remelting of scrap metal composed, in turn, of alloys with varying Zn content.

Another point to highlight is the presence of a small group of brass artefacts containing just over 20 wt.% Zn. These may be brasses that have undergone two or three remelting cycles or, alternatively, may have been produced by cementation throughout the so-called medieval method [22]. This involves the reaction of zinc vapour with liquid rather than solid copper, at higher temperatures and in open vessels. The use of this method at a temperature of about 1200 ºC makes it possible to produce a brass with about 20 wt.% Zn.

In any case, most brasses from the Almohad neighbourhood of Mārtulah contain a moderate Zn content and an overall low level of impurities. According to Gaudenzi Asinelli et al., this could be suggesting the use of sphalerite ((Zn,Fe)S) as a zinc ore [16]. In the case of the metals from Mértola, this is a relevant data as sphalerite zinc ores are common in the Iberian Pyrite Belt, a metallogenic province located in the SW Iberian Peninsula [23-25], thus suggesting the use of local minerals.

Unalloyed Cu artefacts

Pure coppers are relatively scarce, accounting for about 8.8% of the investigated collection. Of the 15 artefacts in this group, 12 contain less than 2 wt.% Pb. From a typological perspective, these Cu-artefacts include three casket ornaments, one ring, one earring, one buckle, seven undetermined objects, and two nails. The only two nails analysed in this paper are both made of copper, although with differences in terms of Pb content. Due to the small number of artefacts composed of unalloyed copper, the data does not allow however for any further noteworthy comment to be made.

Bronze artefacts (Cu-Sn)

Binary bronzes consist of only six objects (3.5% of the entire collection). Similar to the other alloys discussed so far, also bronze was not used to produce a specific type of object. Altogether, one earring, two undetermined objects, and two spindles were made of bronze, i.e., one ornament, two unknown, and two tools in terms of functionality.

The Sn content is highly variable, ranging from 2.93 to 9.09 wt.% (Fig. 6A). Minor elements like Zn, Pb, Fe, As, Ni, and Sb, range from 0.76 to 5.13 wt.% in total, and no leaded bronzes are present (Fig. 6B). Given their residual occurrence, none of impurities had any significant impact on the mechanical properties of the alloys.

As it is well known, the addition of Sn to Cu lowers the melting temperature of Cu, and improves the mechanical properties of the metal, making the alloy physically more resistant to impacts. In this respect, the mechanical effects that the presence of Sn may have on the finished alloy begin to become evident only at Sn concentrations above 3-4% [26], with the best outcome with Sn % above 10-15 wt.%.

Considering the Sn content found in the bronzes from the Almohad neighbourhood of Mértola, it is quite evident that the addition of fresh Sn during the melting process was not a technological option for the metalworkers that produced these metals. As such, the reduced content of Sn is a further indication that, at that time, the use of recycled scraps as a raw material, instead of alloying Cu and Sn in suitable proportions, was a well-established practice. The reduced amount of Sn in the finished objects is a consequence of the decrease in concentration that this element experiences as a consequence of the recycling process. Each time a tin-bronze is remelted, Sn gradually decreases through volatilization, leading to the production of objects with less Sn content than those used as scrap. The higher the number of remelting episodes, therefore, the lower the amount of tin in the final alloy [18].

When placed in its historical context, the low concentration of tin in the alloys found in Mértola cannot be considered unexpected [27]. In fact, no tin mines have so far been identified in the South of Portugal with the most likely source of tin at the beginning of the 2^{nd} millennium being located in the Iberian Peninsula northwest, where tin had been exploited since antiquity [28]. However, it is very likely that with the Reconquista underway, these tin mines were no longer accessible to Moors as in the first quarter of the 2^{nd} millennium Iberia northwest was already under the firm control of the Christian kingdoms.

The low concentration of tin in the alloys analysed in this paper could therefore be explained by a shortage of Sn supply due to the interruption of the tin trade to southern Portugal. However, it cannot be underestimated that tin, during the Islamic period, was also used for other craft productions, in particular for pottery glazes [29-35]. Thus, it is also possible that the little available tin, given its scarcity, may have been deliberately restricted to productions of greater social and artistic values such as prestige pottery, rather than for metal objects of daily use.

Ternary alloys artefacts (Zn-Sn-Pb)

Ternary alloys represent 18.7% of the entire assemblage, and includes ten spindles, seven casket ornaments, six earrings, three oil lamp sticks, two rings, two spatulas, and two undetermined objects.

All the ternary alloys show very reduced tin concentration, and a variability in composition of both Zn (from 2 to 13 wt.%), and Pb (26 of the 36 ternary objects have less than 2 wt.% Pb, ranging from 0.3 to 11 wt.%) (Fig. 7). A clear pattern linking the variability of major elements and the typology of the artefacts is, once again, not detectable as evidenced by Fig. 8.

The composition of ternary alloys appears to be a further argument in favour of a predominantly scrap-based metallurgy in which fresh ores were not added to the melt. Low levels of Zn and Sn, in particular, confirm the hypothesis already discussed above which regards the use of scrap as raw material to produce new objects as a very common practice of the time.

Impurities

As for impurities, i.e., elements whose concentration is less than 2 wt.%, they most likely derive from minor metallic elements present in the ore and reduced unintentionally during the smelting process that end up incorporated into the finished objects. Their concentration depends on different factors such as the quantity of impurities in the ore or the smelting technology in use. Attempting to address questions concerning to origin of raw materials through the identification of impurity patterns is a controversial issue in archaeology, although it is quite clear that some chemical characteristics of the ore are still detectable in the finished objects. The point here is that the composition of an ore source is not homogeneous, and impurities can vary even at different locations within the same ore (Fig. 9).

Conclusion

This research has shown that a variety of different Cu-based alloys were in use in Mértola during the 12th and the first half of the 13th centuries, i.e., during the Islamic period. Tin bronze artefacts are the smallest group, while brass appears to be the preferred alloy to produce objects of daily use. Moreover, bronzes and brasses were further mixed to produce ternary alloys (Cu+Sn+Zn). Occasionally, Pb was randomly added to the different alloys.

The variability of major elements alloyed to Cu, i.e., Zn, Sn, and Pb, could be due to technological constraints or political restrictions or to the combination of both factors.

On the other hand, data suggest that objects were not produced with well-defined and predetermined composition. In fact, the XRF analysis clearly revealed that no link can be found between the functions or the forms of the artefacts and their composition as similar objects were produced with different alloys, and vice versa, objects with distinct forms and functions were made of alloys with very similar mechanical properties. This data could indicate that metalworker that produced the objects found in the Almohad quarter of Mértola apparently did not possess advanced technical skills or they were not particularly concerned with the final alloy composition of the artefacts, and/or even if they were aware of the advantages linked to the different chemical compositions of the alloys, they chose not to take advantage of this knowledge.

However, it is very likely that political barriers and economic constraints of the time could have deeply influenced the technological options that metalworkers took along the production chain. In this respect, mention has been already made of the great instability experienced in southern al-Andalus during the 12th and the first half of the 13th centuries, characterized by periods of strong political fragmentation, i.e., with the formation of the so-called *Taifas*, and periods of reunification, particularly under the Almoravids and Almohads dynasties. Furthermore, since the beginning of the 2nd millennium, an increasing intensification of pressure on Islamic territories by Christian forces lead to the conquering South of Portugal finally achieved in the mid-13th century. This climate of political instability is very likely to have had a negative effect on the ore minerals trade, with Islamic communities experiencing ever increasing difficulties in the access to ore mineral resources located in territories they did not control. This is especially true for tin, which unlike zinc ores were not available locally and was also used for other types of production, such as glazed pottery. As a result, metal technology was affected by the widespread political insecurity in al-Andalus at the time, and local craftsmen were forced to adapt their production to circumstances beyond their control and to use as raw material local ores or scrap metals they had easier access to.

Declarations

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Authors’ contributions

CB: conceptualisation, methodology, XRF analysis, data collection and interpretation, and writing-original draft preparation. RB: data interpretation and writing editing. MB: XRF analysis, data collection and writing editing, JM: writing editing. SGM and LR: archaeological investigation and writing editing. NS: writing editing. All authors have read and agreed to the published version of the manuscript.

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Availability of data and materials

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests.

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Figures

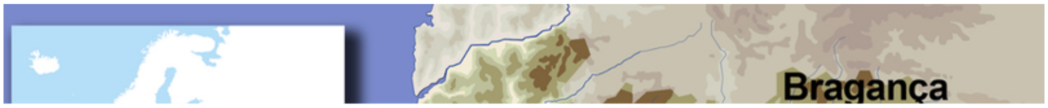

Figure 1

Location of Mértola (South of Portugal).

Figure 2

The Almohad neighbourhood. Aerial view (A), and detail of an archaeological area of the excavation (B).

Figure 3

A selected group of metals analysed in this paper.

Figure 4

Histogram showing the distribution of artefacts within the different kind of alloys found at Mértola (A). Bivariate plot displaying Sn and Zn values for the objects analysed in this paper (B)

Figure 5

Composition of the different types of metals (A), plot displaying the lack of any correlation between the concentration of Zn and the functionality of artefacts (B), and frequency histogram of Zn in the brasses(C)

Figure 6

Frequency of bronze artefacts showing the variability of Sn (A), and Pb (B), within the bronze artefacts.

Figure 7

Variability of Zn (A), Sn (B), and Pb (C) within the ternary alloy artefacts.

Figure 8

Ternary alloys showing the lack of correlation between the variability of Zn, Sn, and Pb concentration in ternary alloys and the function of the artefacts (red dots: undetermined, green dots: tools, black dots: ornament).

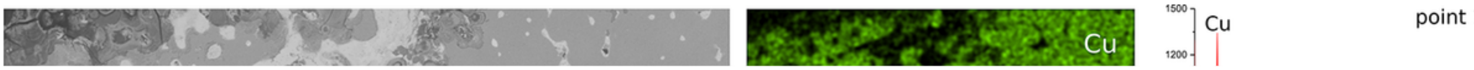

Figure 9

SEM-EDS mapping of the nail PR-02-06, showing Pb-rich inclusions in a pure copper alloy