WHAT ROLE DID REALLY TIN BRONZE PLAY IN THE ARGARIC SOCIETY?

MILYEN SZEREPET JÁTSZOTT AZ ÓNBRONZ AZ ARGAR TÁRSADALOMBAN?•

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Abstract

The transition from arsenic copper to tin-bronze in ancient metallurgy has long been attributed to the superior physical and mechanical properties of tin-bronze. However, recent archaeometallurgical studies have cast doubt on this theory, suggesting that the functional and productive advantages of tin-bronze over arsenical copper may not be as clear-cut as traditionally thought.

In this paper we present the results of metallographic and microhardness tests conducted on the metallic assemblages from several Bronze Age Argaric sites (Southeast Iberia). Compositional analyses of more than 700 copper-based objects revealed a distinct correlation between the use of tin-bronze for ornaments and arsenical copper for functional objects. This fact suggests that the choice of tin-bronze was influenced by factors beyond mere productivity. The results presented in this paper show that both arsenic copper and tin-bronze could exhibit similar mechanical properties. According to them, their microhardness levels depend on the final processes of their manufacture and the intensity of these processes, rather than on the alloy's composition. This challenges the notion that bronze was adopted solely for its functional efficiency. Therefore, alternative interpretations must be considered to explain the adoption of this new alloy.

Kivonat

Az ősi metallurgiában az arzéntartalmú rézről az ónbronzra való áttérést sokáig az ónbronz jobb fizikai és mechanikai tulajdonságainak tulajdonították. A legújabb archeometallurgiai tanulmányok azonban kétségbe vonják ezt az elméletet, és arra utalnak, hogy az ónbronz funkcionális és termelési előnyei az arzénes rézzel szemben talán nem olyan egyértelműek, mint ahogyan azt hagyományosan gondolták.

Ebben a tanulmányban a bronzkori Argar-kultúra (Délkelet-Ibéria) több lelőhelyéről származó fém leletegyütteseken végzett metallográfiai és mikrokeménységi vizsgálatok eredményeit mutatjuk be. Több mint 700 réz alapú tárgy összetételének elemzése egyértelmű összefüggést mutatott ki az ónbronz díszítésre és az arzéntartalmú réz használati tárgyakra történő felhasználása között. Ez a tény arra utal, hogy az ónbronz választását a puszta termelékenységen túlmutató tényezők is befolyásolták. Az ebben a tanulmányban bemutatott eredmények szerint mind az arzéntartalmú réz, mind az ónbronz hasonló mechanikai tulajdonságokkal rendelkezhet. Mikrokeménységük inkább készítésük végső folyamataitól és azok intenzitásától függ, mint az ötvözet összetételétől. Ez megkérdőjelezi azt az elképzelést, hogy a bronz kizárólag funkcionális hatékonysága miatt terjedt el és alternatív értelmezés szükséges az új ötvözet alkalmazására.

KEYWORDS: BRONZE, ARSENIC COPPER, ALLOY, MICROHARDNESS TEST, METALLOGRAPHY, IBERIAN PENINSULA, EARLY BRONZE AGE.

KULCSSZAVAK: BRONZ, ARZÉNES RÉZ, MIKROKEMÉNYSÉG MÉRÉS, METALLOGRÁFIA, IBÉRIAI-FÉLSZIGET, KORA BRONZKOR

• How to cite this paper: MURILLO-BARROSO, M.; MORENO ONORATO, A.; ARANDA JIMÉNEZ, G.; LACKINGER, A.; CONTRERAS CORTES, F.; MONTERO-RUIZ, I., (2024): What role did really tin bronze play in the Argaric society?, *Archeometriai Műhely* **XXI/3** 237–252.
doi: https://doi.org/10.55023/issn.1786-271X.2024-019

Fig. 1.: The Argaric territory in South-East Iberia

1. ábra: Az Argar-kultúra elterjedési területe Délkelet-Ibériában.

Introduction

Traditionally, societies have been classified by their technology and, therefore, the origin of metallurgy has usually represented a central aspect in historical explanation. From an evolutionary point of view, societies progress as technology is developed. Actually, the chronological division in 'Stone Age, Bronze Age or Iron Age' following Thomsen's scheme of the 'three ages' is not coincidental: this cultural framework underscores the significance attributed to metallurgy in historical interpretation and reflects the belief that technological development drives social change and cultural progress. Oversimplifying, technological advances let communities increase their resources and, hence, population grows, specialists are needed, and societies become more and more complex. However, while it is true that certain technological improvements can qualitatively modify some production processes and have a decisive impact in societies, it cannot be considered a general rule. In fact, this is the case of the first metallurgy in Iberia and the role played by tin-bronze in the Argaric communities.

The Argaric society

Before going into metallurgical questions in some detail, it may be worth presenting here some general information on the Argaric society, which corresponds to South-Eastern Iberia's Early Bronze Age (c. 2250–1550 cal BC) (**Fig. 1**). As a general rule, Argaric sites tended to be strategically located in mountains and hills with natural defensive features and a commanding view of the surrounding area. In addition, some of these sites have impressive stone fortifications.

Differences in settlements size, location and material culture suggest that there was a hierarchical settlement pattern, whereby different sites had specialized strategic, social and/or economic functions (Aranda et al. 2015).

Due to its peculiar nature, one of the most significant features of the Argaric societies is the location of burials within the settlement area, usually under the floors of the dwellings in four main types of containers: ceramic urns, cists, pit-graves and *covachas* (small artificial caves cut into the rock). Funerary ritual mainly consisted in individual, double or, more rarely, triple inhumations that were placed in a flexed position (**Fig. 2**). Argaric communities generally buried their dead with a series of objects that constituted the funerary offering. Grave goods are different in number, type and quality, ranging from none at all in some tombs, to important accumulations of wealth items in others. Ritual offerings can be cluttered in five main groups: pottery vessels; metal weapons such as swords and halberds; tools (axes, daggers/ knives, awls); ornaments (made from copper/ bronze, silver or gold) such as rings, bracelets, earrings, diadems and necklaces (usually made of stone beads); and faunal remains (usually limbs from cattle, sheep and goats) (**Fig. 3**).

Most scholars accept the image of a deeply stratified society, with evidence of ascribed status

HU ISSN 1786-271X; urn: nbn: hu-4106

Fig. 2.: Argaric burial in a cist from the Cerro de la Encina (Monachil, Granada) site (Photo GEPRAN)

2. ábra: Argar sír a Cerro de la Encina-ban (Monachil, Granada) feltárt temetőből. (Fotó: GEPRAN)

due to differences in funerary wares and the first appearance of wealth children graves (for further discussion see Chapman 2008 and Aranda et al. 2015). Recent extensive excavations have provided a wealth of new data which constitutes the basis of current socio-political interpretations of the Argaric society – cf. Castro et al. 1999; Contreras Cortés 2000; Schubart et al., 2000; Aranda & Molina 2006; Aranda et al. 2012; Lull et al. 2021 –; however, the excavations of Louis and Henry Siret, Belgian mining engineers and archaeologists, mainly from the 1880s to the 1886s still provide important evidence for the Bronze Age burials in the region (Siret & Siret 1890).

Argaric metallurgy can be characterized by an important intensification of production. In contrast with the previous Copper Age societies, metal objects presented a 5-fold increase in quantity. Ornaments like bracelets, rings, earrings and diadems were the type of object primarily produced, especially from 1800 BC onwards, amounting for more than 50%, followed by tools (axes, daggers/knives, awls, etc.), and then specialized weaponry in the form of halberds and swords, which make their appearance for the first time in the Iberian Peninsula (Montero Ruiz 1993; 1994; Lull et al. 2017).

Fig. 3.: Argaric grave goods from burial 21 of the Cerro de la Encina (Monachil, Granada) site (Photo GEPRAN)

3. ábra: Argar sírmelléklet a Cerro de la Encina (Monachil, Granada) lelőhelyen feltárt 21-es számú sírból. (Fotó: GEPRAN)

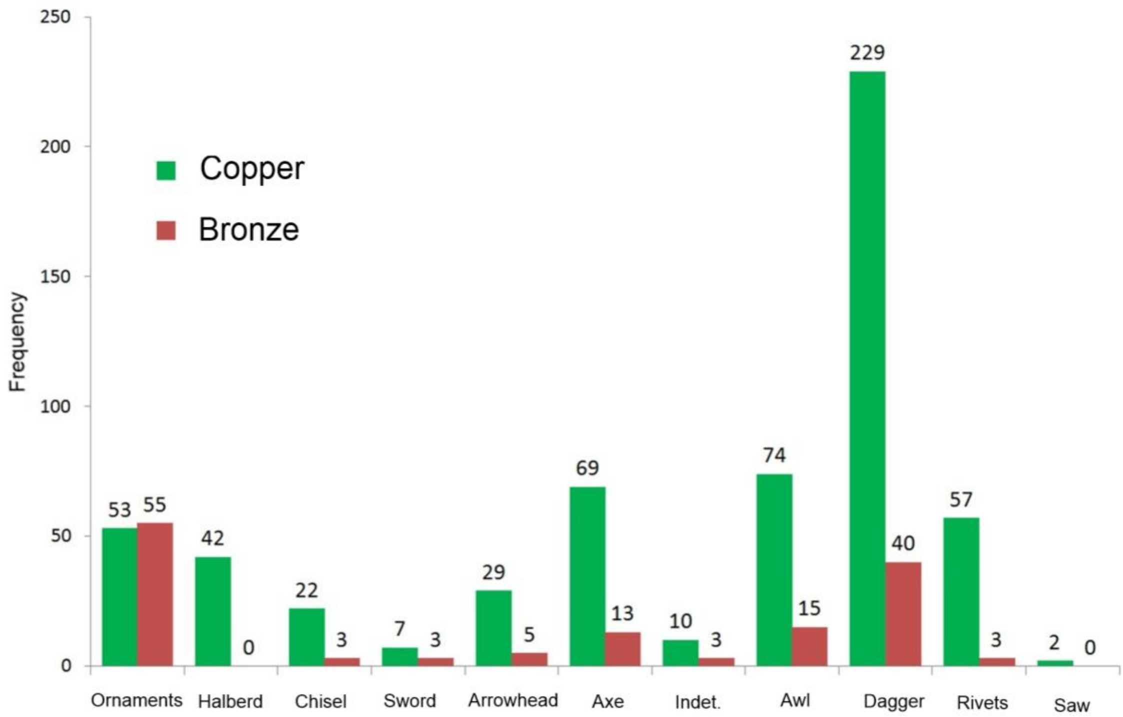

Fig. 4.: Frequency of copper and bronze in Bronze Argaric metals according to their typology (after Montero Ruiz et al. 2019, fig 1)

4. ábra: A réz és a bronz gyakorisága az Argar-kultúra réz alapú fémtárgyai esetében, tárgyfajták szerint (Montero Ruiz et al. 2019, 1. ábra után).

Copper was the primary metal used in Argaric communities from their inception, although gold and, particularly, silver eventually became significant for ornament production. The tin-bronze alloy was not in use when the Argaric society emerged and never became widespread (Montero Ruiz et al. 2019) (**Fig. 4**). Therefore, the adoption of this alloy does not appear to be the origin, or even a significant cause, of the changes that occurred at the beginning of the Early Bronze Age which shaped the Argaric society.

Why were bronze alloys introduced and what role did they play, then? As we will discuss here, one of the most intriguing debates related to bronze production is whether this alloy was chosen for specific tasks by replacing copper or arsenic copper due to its superior mechanical properties, or if it was used because of fashion, its colour, or any other social values.

The adoption of tin-bronze alloy has been the subject of different theories: some archaeologists state that tin-bronze substituted arsenic copper because of its better mechanical properties in terms of hardness and strength, which supposed an increase of productivity (Childe 1944; Kristiansen 1987). Other scholars suggest that tin-bronze is a deliberate alloy while arsenic copper is not, so the amount of tin is better controlled than the amount of arsenic, thus improving the final properties of objects (Tylecote 1976; Pernicka 1998). Even the “healthier” properties of tin technology in contrast with the toxicity of arsenic fumes have been also proposed as an explanation (Charles 1967).

Mechanical and physical properties of copper, arsenic copper and tin bronze

Here we will comment the mechanical and physical properties of copper, arsenic copper and tin bronze in order to understand why the Argaric society adopted this significant technological change.

The first physical property that becomes evident is that certain amounts of either arsenic or tin can reduce the melting point of copper (1083 ºC). It is assumed that under equilibrium conditions, the eutectic point of arsenic copper (i.e. the lowest melting point, 689 ºC) will be reached with an arsenic content of 21.5% (weight percent). However, in the range of Argaric alloys – As average of 2.4% for arsenic coppers and Sn average of 8.0% for tin-bronzes, considering bronze those containing >1% Sn (Montero Ruiz 1994: 244-245) – the decrease of the melting point is not significant and remains similar in both cases.

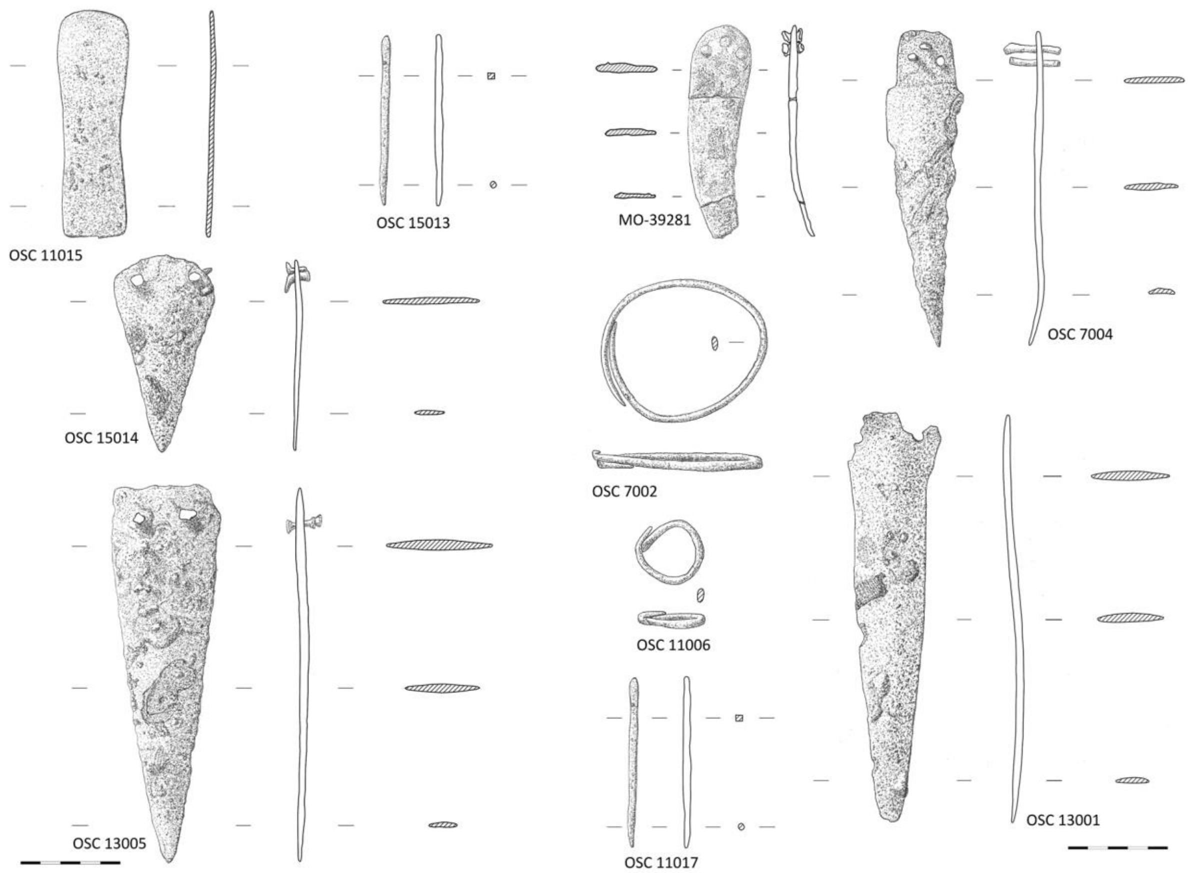

Fig. 5.: Some of the objects analyzed from Cerro de San Cristobal (OSC) and Cerro de la Encina (MO) (After Murillo-Barroso et al. 2015, Figures 4 and 5).

5. ábra: Néhány vizsgált tárgy a Cerro de San Cristobal (OSC) és a Cerro de la Encina (MO) lelőhelyről (Murillo-Barroso et al. 2015 után, 4. és 5. ábra).

According to phase diagrams, for the average value of 2.4% As the melting point will be reduced to c. 1050 ºC, and for the highest arsenic concentration value documented (12.7%, Rovira Llorens et al. 1997), the melting point will be reduced to c. 900 ºC. Those values are similar to the ones obtained with tin-bronze alloys: the average for tin concentration documented in Argaric bronzes is of 8.0% Sn (Montero Ruiz 1994: 244), which would decrease the melting point to c. 1020–1030 ºC, and the maximum amount of Sn measured is of 14.4% (Rovira Llorens et al. 1997), which would reduce the melting point to c. 970 ºC. Although these temperatures correspond to alloys under equilibrium conditions, and those are not usually reached in practice, – according to experimental studies, under practical conditions eutectic segregates can appear with a much lower amount of arsenic, around 2–3% (McKerrel & Tylecote 1972: 211; Lechtman 1996: 486; Mödlinger et al. 2018) –, it seems that in a first instance, both types of alloy reduce the melting point of copper in a similar way, so this might not be the main reason for the adoption of tin-bronze alloy.

Another significant characteristic is the increase in hardness that arsenic and tin provide due to their differences in atomic size compared to copper. In a solid solution alloy, the solute atoms are generally of a different size than the atoms of the metal in which they are dissolved, and the resulting distortion of the crystalline structure contributes to the final hardness of the alloy. The result of the addition of arsenic or tin to copper exemplifies this chemical principle. Arsenic atoms are larger than copper ones, but not as big as tin atoms, therefore tin is expected to increase hardness more than arsenic.

Experimental studies show that hardness and malleability of arsenic copper alloys begin to change with 0.5% or 1% As and 2% Sn (Lechtman 1996; Northover 1989). In all cases, some overlapping of both alloys' properties seems to be detected: Marechal (1958 cf. Lechtman 1996) and more recently Northover (1989) stated that the hardness achieved with an 8% arsenic concentration is very similar to the one achieved with 8% tin (over 250 HV). Scott (1991) points out that with 4% tin or arsenic, for a reduction of 50% of thickness, arsenic copper remains harder than tin bronzes. On the other hand, Lechtman (1996) indicates that tin bronze with a 2% tin content after 75% reduction of thickness, is far harder than a 2% arsenic copper hardened up to the same point. However, Lechtman also concludes that there is a lot of overlap in the mechanical properties of both alloys in the documented concentration ranges.

As a matter of fact, the mechanical properties of arsenic copper and tin-bronzes are sometimes difficult to measure comparatively in experimental studies due to same-conditions concerns that must be considered; even with no thermo-mechanical processes applied to them, there are several features, like cooling rate, which relates to the grain size, or impurities in the metal, that can influence the final result (Sabatini et al. 2020)

As these results are from tests conducted on experimental alloys, metallographic and micro-hardness analyses on 54 archaeological samples (12 bronzes and 42 arsenic coppers) from several archaeological sites were carried out in order to compare and contrast the properties of both types of alloys in original artifacts (**Fig. 5**).

Methodology

Elemental composition was determined by Inductively Coupled Plasma-Mass Spectrometry with a Sector Field (ICP-SFMS) and Energy Dispersive -X Ray Fluorescence (ED-XRF). ICP-SFMS analyses were conducted by Dr. Michael Bode at the Archaeology and Materials Science laboratories of the *Deutsches Bergbau-Museum* (Bochum, Germany) using a spectrometer Thermo Scientific ELEMENT XR (for further method-logical questions about ICP-SMFS cf. Renzi et al. 2014). ED-XRF was conducted at the National Archaeological Museum (Madrid, Spain) using a portable spectrometer INNOV-X serie Alpha. Working conditions, using an X-Ray detector with silver anode, were of 35 kV and 20 µA. Patina and corrosion products were mechanically removed and analyses were conducted on an area of 25 mm^2 of the clean metallic surface (for calibration and further methodological questions about XRF analysis procedure see Rovira Llorens & Montero Ruiz, 2018). All the values of the elements used in the text refers to weight percent (wt%).

For metallographic examination, samples were embedded in epoxy resin, ground and polished to 0.25 µm grit size, following the conventional procedure (Scott, 1991). Samples were etched with an aqueous ferric chloride solution (120 ml H_2O: 30 ml HCl: 10 g $FeCl_3$) and were observed under an optical microscope Leica DMLM. Microhardness tests were carried out using a REMET HX1000 tester. Given results are the average of between 4 and 20 indentations, depending on the sample size. Both types of analyses were conducted at the laboratories of the Institute of History (CCHS-CSIC, Madrid, Spain). Methodological questions on metallographic preparation and microhardness analyses followed the recommendations of Scott (1991) and Rovira Llorens & Gómez Ramos (2003).

Table 1.: Working techniques, average Vickers microhardness and composition of Argaric copper-based artifacts. C=As Cast, A=Annealed, CW=Cold Working. Working techniques in brackets show low intensity; in bold + italics show high intensity. Composition is given in wt%. (nd=not detected, Tr=traces)

1. táblázat: Argar-kultúra réz alapú tárgyainak megmunkálási technikái, átlagos Vickers keménységei és összetételei. C=öntött, A=lágyított, CW=hidegalakított. A zárójelben lévő készítési technikák kis gyakoriságot, a félkövér+dőlt betűk a nagy gyakoriságot jelzik. Az összetételt tömeg%-ban adtuk meg. (nd= nem detektálható, Tr=nyomokban)

Site	ID	Type	Working Techniques	HV	As %	Sn %
Llano de la Gabiarra	PA2984B	Rivet	C		Nd	nd
Cerro de la Virgen	PA0927	Awl	C		1.97	nd
Peñalosa	BE-9533	Rivet	C + A	60	3.9	nd
Hoya de la Matanza	PA2967A	Dagger	C + A + CW		1.25	nd
Peñalosa	PA14034	Dagger Rivet	C + A + CW	205	10.9	0.01
Cerro San Cristobal	OSC 7004_R	Dagger Rivet	C + *CW*	170	Nd	nd
Peñalosa	BE-28882	Awl	C + CW	177	Nd	nd
Peñalosa	PA14032	Awl	C + CW	117	0.66	0.04
Hoya de la Matanza	PA2970	Dagger	C + CW		0.8	nd
Mina Alianza-Herrerías	AA1148B	Halberd Rivet	C + CW		0.94	0.07
Cerro San Cristobal	OSC 13006_60	Staple	C + CW	116	1	Tr
Las Angosturas	PA2433	Dagger	C + CW		1.32	0.02
Peñalosa	PA14048	Awl	C + CW	134	1.41	nd

Table 1., cont.

1. táblázat, folyt.

Site	ID	Type	Working Techniques	HV	As %	Sn %
Peñalosa	PA14053	Awl	C + CW	116	1.45	0.04
Peñalosa	PA14036	Awl	C + CW	153	1.7	nd
Cerro de la Virgen	PA0924	Chisel	C + CW		2.2	0.02
Peñalosa	PA14051	Dagger	C + CW	209	2.37	0.03
Cerro San Cristobal	OSC 11010_R	Rivet	C + CW	158	2.39	nd
Peñalosa	PA 20107	Dagger	C + CW	97	2.6	nd
Peñalosa	BE-10249	Awl	C + CW	173	2.7	nd
Peñalosa	PA13631	Bead	C + CW	135	2.75	nd
Cerro San Cristobal	OSC 15014_R	Dagger Rivet	C + CW	127	2.98	nd
Peñalosa	PA 14049_R	Dagger Rivet	C + CW	205	3.43	nd
Cerro San Cristobal	OSC 15014_H	Dagger	C + CW	151	3.51	nd
Cerro de Enmedio	PA2612L	Awl	C + CW + A		1.28	Tr
Peñalosa	PA14033	Dagger	C + CW + A	106	2.12	0.02
Cerro de la Encina	MO-39261	Bracelet	C + (CW + A)	62	2.52	nd
Peñalosa	PA14047	Dagger	C + CW + A	197	3.55	nd
Peñalosa	PA13632	Axe	C + CW + A + CW	132	0.31	nd
Hoya de la Matanza	PA2967B	Awl	C + CW + A + CW		0.9	nd
Cerro San Cristobal	OSC 13006_37	Nail	C + CW + (A + CW)	146	1.09	Tr
Cerro San Cristobal	OSC 13006_71	Nail	C + CW + (A + CW)	144	1.23	Tr
Cerro San Cristobal	OSC 13006_15	Nail	C + CW + (A + CW)	134	1.29	Tr
Peñalosa	PA14049	Dagger	C + CW + A + CW	145	2.3	0.03
Cerro de la Virgen	PA0922	Awl	C + (CW)		2.3	nd
Hoya de la Matanza	PA2968	Dagger	C + CW + A + CW		2.37	nd
Cerro San Cristobal	OSC 15013	Awl	C + CW + A + CW	200	4.08	nd
Cerro San Cristobal	OSC 13005_R	Dagger Rivet	C + CW + A + CW	142	4.3	nd
Cerro San Cristobal	OSC 13005_H	Dagger	C + CW + A + CW	186	5.05	nd
Cerro de la Encina	MO-39257	Awl	C + CW + A + CW	200	5.28	nd
Cerro San Cristobal	OSC 13001_H	Dagger	C + CW + A + CW	196	6.47	nd
Cerro de la Encina	MO-21292	Dagger	C + CW + A + CW	175	6.73	nd
Peñon de la Reina	PR-PUNZON	Awl	C		Nd	10.7
Cerro de la Encina	MO39264_H	Dagger	C + CW		2.3	4.38
Cerro de la Encina	MO39281_H	Dagger	C + CW		0.09	8.7
Cerro de la Encina	MO-39255	Bracelet	C + CW + A	90	0.13	4.1
Peñon de la Reina	PR-ARET-19	Ring	C + CW + A		nd	4.99
Cerro San Cristobal	OSC 7002	Bracelet	C + CW + A	108	0.42	5.58
Peñalosa	PA 20106	Sword	C + CW + A	149	1.3	9.92
Cerro de la Encina	MO-39260	Ring	C + CW + A		0.24	8.93
Cerro San Cristobal	OSC 11006	Ring	C + CW + A + CW	183	0.01	4.47
Cerro San Cristobal	OSC 11015	'Scraper'	C + (CW + A + CW)	149	0.87	4.91
Cerro San Cristobal	OSC 11017	Awl	C + CW + A + CW	198	0.01	6.61
Peñalosa	PA14050	Dagger	C + CW + A + CW	184	0.4	9.4

HU ISSN 1786-271X; urn: nbn: hu-4106 © by the author (s)

Results and discussion

Table 1. summarizes the results of the analyses. Full data has been previously published in several papers (Rovira Llorens et al. 1997; Murillo-Barroso et al. 2015 or Moreno Onorato & Contreras Cortés, 2015). The average content of arsenic and tin in the assemblage studied (2.7% As, Std. 2.05, and 6.9% Sn, Std. 2.46) is consistent with previous analyses of Argaric metallurgy, featuring low tin bronzes and arsenic coppers with a 2.4% As average (Montero 1994: 245), but in our case also with some high As levels (the rivet with 10% As stands out but is included in the 2.7% As average value).

The arsenic in the metal has been related with the smelting of As-rich copper ores (Rovira Llorens & Montero Ruiz 2013) and it is assumed that its presence is not deliberate, unlike the case of tin bronzes, but the identification of arsenic-rich ores and its selective mining cannot be rejected (Hook et al. 1991; Moreno et al. 2003). This is suggested by the fact that tin is not detected in copper or arsenic copper items, while tin bronzes can contain significant arsenic levels, even higher than 1%.

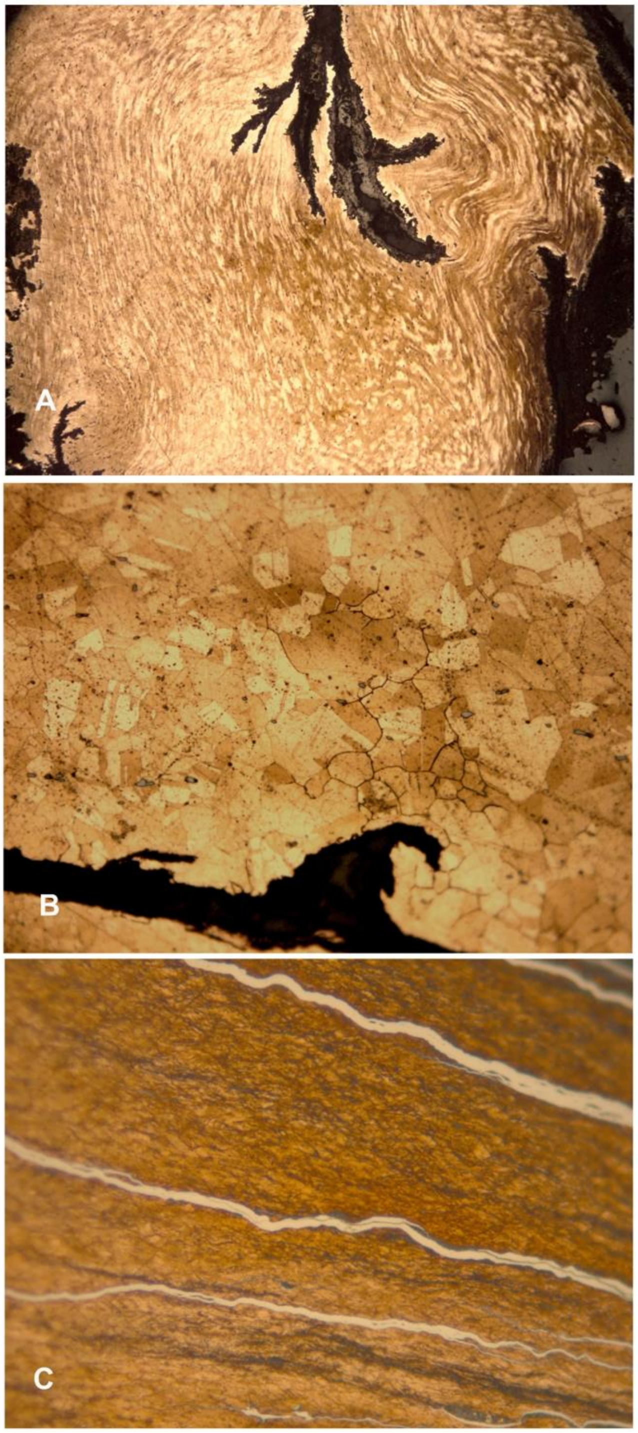

Fig. 6.:
Microstructures of the main *chaîne opératoires* documented in the Argaric assemblages. All samples have been etched with aferric chloride solution.
(A) Rivet OSC15014_R, Cast and Cold Worked. Note the deformation of the dendritic structure, especially on the right side of the rivet (100X).
(B) Bracelet OSC7002. Cast, Cold Worked and Annealed. Note that rectilinear grains have formed as a consequence of annealing. Bands inside the grains indicate that the bracelet was previously hammered (200X).
(C) Dagger OSC13001_H with 6.47% As. Cast, Cold Worked, Annealed and Cold Worked. The dagger was heavily hammered and the microstructure is completely deformed. Note that arsenic has been segregated in bright As-rich bands that can be easily identified (500X).

6. ábra:
Az Argar-kultúrabeli leleteken megfigyelt készítési folyamatok mikroszkópos felvételei. A mintákat vas-klorid oldattal maratták.
(A) Szeg OSC15014_R, öntött és hidegen alakított. Dendrites szerkezet deformációja, különösen a szeg jobb oldalán (100x).
(B) Karkötő OSC7002. Öntött, hidegen alakított, lágyított. Egyenes vonalú szemcsék alakultak ki a lágyítás következtében. A szemcsék belsejében látható sávok arra utalnak, hogy a karkötőt korábban kalapálták. (200X).
(C) Tőr OSC13001_H 6.47 % As-tartalommal. Öntött, hidegen alakított, lágyított és hidegen alakított. A tőrt erősen átkalapálták, így a mikroszerkezete teljesen deformálódott. Az arzén jól elkülöníthető, világos sávokban szegregálódott (500X).

Table 2.: Average hardness of each type of artifacts

2. táblázat: A különböző tárgytípusok átlagos keménységértékei.

Type	Number of samples	Mean HV	StD
Awls	9	163	34.6
Bracelets	3	87	23.3
Daggers	10	165	38.9
Rivets	11	146	40.5

Metal production follows different *chaîne opératoires*, and some objects only prepare the edges with cold working while others use longer actions combining annealing and cold working (**Fig. 6**). The chosen process is essential in the microhardness get in the final stage independently of the elemental composition of each object.

Microhardness averages range from 60 HV to 209 HV. Typologically, daggers and awls are the hardest objects, with an average of 164 HV and 163 HV respectively (**Table 2.**). However, we have to take into account that these are mean values, and daggers usually present a high standard deviation (20–30 Std) while awls have a low one (5–9 Std) due to the fact that daggers have hardened edges while awls have a more homogeneous hardness.

If the highest values for daggers are considered, there is no doubt that this is the group with the hardest HV values. **Fig. 7.** shows this selective working on dagger edges: Vickers values drop as soon as measurements are taken in the inner part of the dagger. Even light cold working can, under the right conditions, produce a significant increase of the hardness, as it is shown in the case of the object OSC11015 (**Fig. 5**). While its dendritic structure was identified at a low magnification (**Fig. 8/A**), the evidence for annealing and cold hammering could only be identified, in the edges, at a higher magnification (**Fig. 8/C**). Annealing and hammering might not be too intense, as the remnant dendritic structure suggests, however it was intense enough to get high hardness values (230 HV) in the edges (**Fig. 7**).

If we compare microhardness values for arsenic or tin alloys, we see that there is not direct correlation between hardness and composition, and that bronzes are not necessarily harder. Average Vickers micro-hardness measurements of both arsenic copper and tin-bronze objects have been graphically plotted in **Fig. 9**. This graph shows how artifacts with low arsenic content could be as hard as tin-bronzes or even harder, and how metals with a similar *chaîne opératoire* have a similar hardness, regardless of their bronze or arsenic composition.

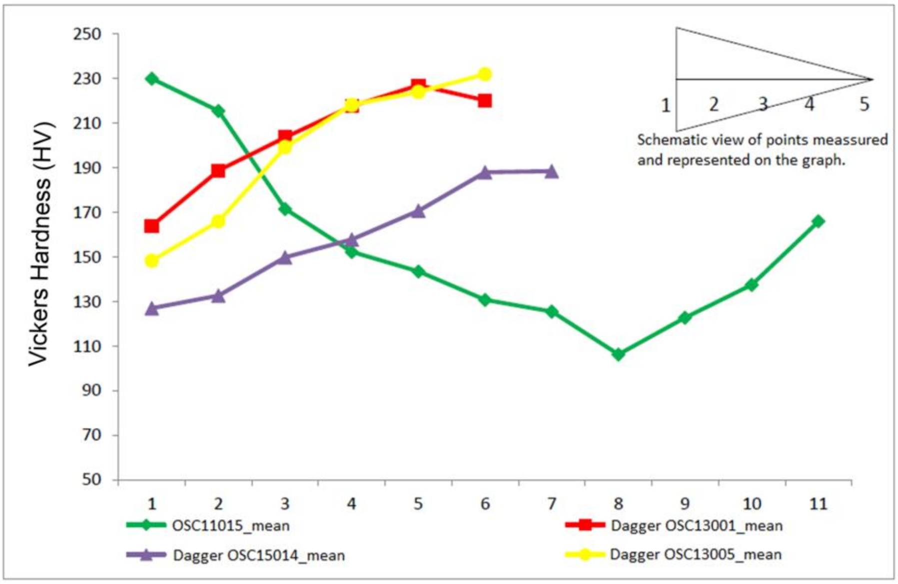

Fig. 7.: Microhardness values of longitudinal axes of some edged objects and the 'scraper' OSC 11015. Hardness has been measured in at least three points on the longitudinal axis, as shown in the schematic drawing.

7. ábra: Éllel ellátott tárgyak, valamint egy „kaparóvas" (OSC 11015) hosszanti tengelye mentén mért mikrokeménységi értékei. Legalább három ponton mértünk keménységet a hosszanti tengely mentén, ahogy ezt a sematikus rajzon is lehet látni.

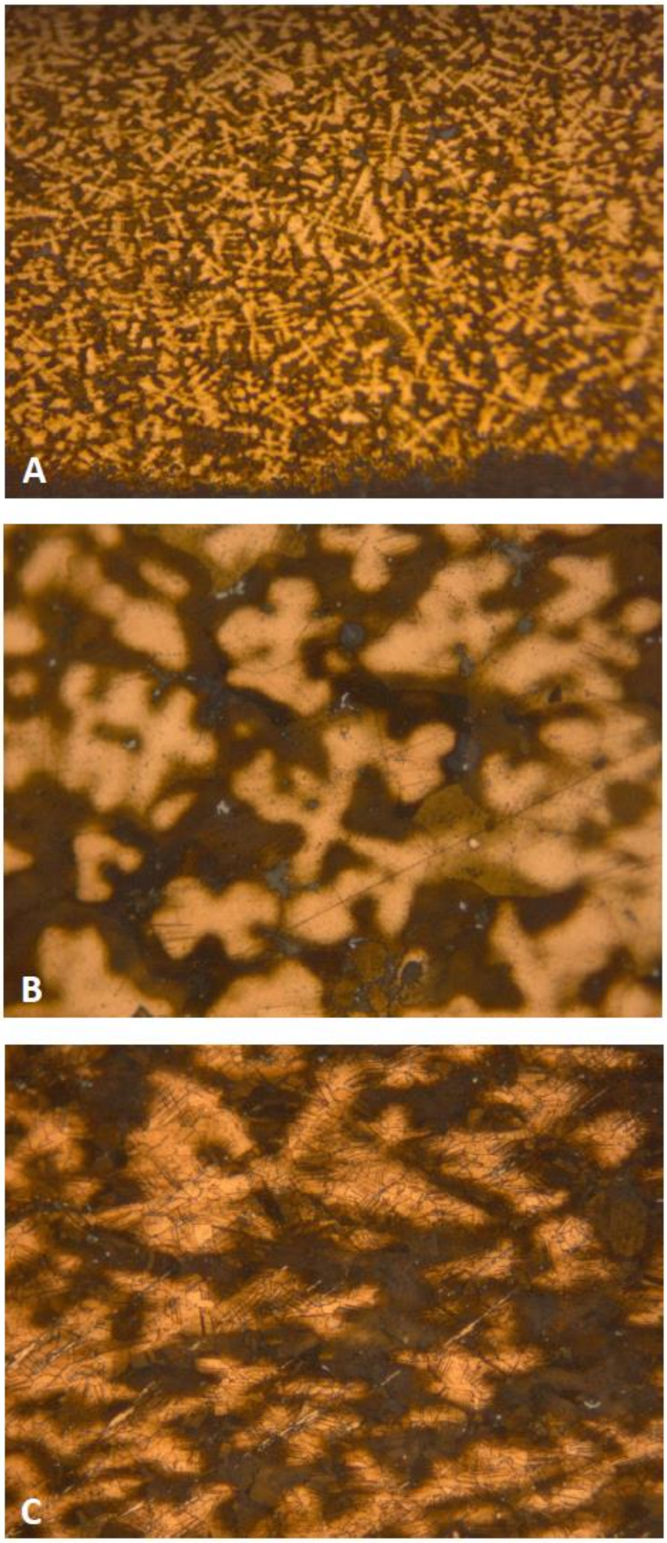

Fig. 8.:
Metallographic section of OSC11015.
(A) General view (50X). Note the dendritic structure as a consequence of slow cooling.
(B) Inner area (500X). Only the dendritic structure can be identified and no grains or slip bands can be recognized.
(C) Edge area (500X). Twinned grains and slip bands can be now recognized in the remnant dendritic structure due to selective cold working.

8. ábra:
Az OSC11015 számú tárgy metallográfiai felvételei.
(A) A minta általános szerkezete (50X). A dendrites szerkezet a lassú hűlés következménye.
(B) A minta belső területe (500X). A dendrites szerkezet nem láthatók szemcsék és nyírási sávok.
(C) Él felé eső terület (500X). A hidegmegmunkálás hatására megmaradt dendrites szerkezetben ikerszemcsék és nyírási sávok láthatók.

This is the case, for example, of two daggers, both cold worked, annealed and cold worked again: the arsenic copper one (5.05% As; OSC13005 in **Fig. 5.**) reaches an average hardness of 186 HV (with a maximum of 232 HV in the edge) while the bronze one (9.4% Sn; PA14050) has an average hardness of 184 HV (with a maximum of 239 HV in the edge). Something similar happens with three awls: the arsenic ones (OSC1503 and MO39257 with 4.08% and 5.28% As respectively) have an average hardness of 200 HV and the bronze one (OSC11017 in **Fig. 5.**, with 6.61% Sn) shows a similar value of 198 HV. Even some copper artifacts with low amounts of arsenic or tin (even <1%) display a hardness similar to that of bronzes. For example, one pure copper awl has a hardness of

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177 HV (BE28882), and one arsenic copper rivet with 2.3% arsenic and a final stage of hammering exhibits a hardness of 209 HV (PA14051), still harder than bronzes with more than 9% tin such as the dagger PA14050 (184 HV). Contrarily, artifacts with a final stage of annealing have a lower hardness: a bronze bracelet (MO39255, with 4.1% Sn) cold hammered and annealed features a hardness of 90 HV, and a rivet, equally hammered and annealed and with 3.9% As shows a hardness of 60 HV (BE9533), while objects with similar amounts of Sn or As but a final stage of hammering have higher microhardness values so far (for example the above mentioned awl with 4% As and 200 HV, OSC15013).

Hence, main differences in microhardness can be established on account of the final stage of the *chaîne opératoire* and its intensity: with one exception in each case, all objects with a final stage of annealing have a hardness below 110 HV, and all artifacts with a final stage of hammering (annealed or not) show a hardness over 115 HV. This is because cold working provides more hardness although at the price of causing the object to eventually become brittle. The process of annealing reduces brittleness while also decreasing hardness, which increases ductility and malleability. This is why, up to a point, annealing is necessary to continue hammering intensely, for example when significantly reducing thickness. Probably this is the reason that explains why almost all the bronzes studied in this article are annealed (either with annealing being the last stage in the *chaîne opératoire* or not). According to Lechtman, 13% tin bronzes become brittle at deformations of about 50% thickness reduction, and annealing is necessary to continue working them, but it is probable that bronzes with c. 7% Sn also require annealing to be extensively cold worked without any risk of cracking (Lechtman 1996).

Therefore, it is the final stage of the *chaîne opératoire*, more than the amount of arsenic or tin in the alloy composition, what seems to determine the final hardness of Argaric artifacts. In order to clarify if prehistoric metalworkers were aware of this fact, these differences in the *chaîne opératoire* have been related with the type of objects, classified in two main categories: body ornaments (mainly rings for different parts: finger, arm or ears) and 'functional' ones (including all objects, mainly tools and weapons, which potentially could have had other purposes than display, even if the possibility of some weapons being used for ostentation rather than for violence is not discarded, see for instance Aranda et al. 2009).

Only 7 body ornaments and 12 tin-bronzes of the assemblage studied were available for sampling, so even if some patterns can be initially deduced, one has to be cautious, as they could change when more analyses are developed. Bearing this in mind, a distinct trend on working techniques can be seemingly inferred: a majority of ornaments have a final stage of annealing (71.5%) while most of functional objects have a final stage of hammering (89.4%).

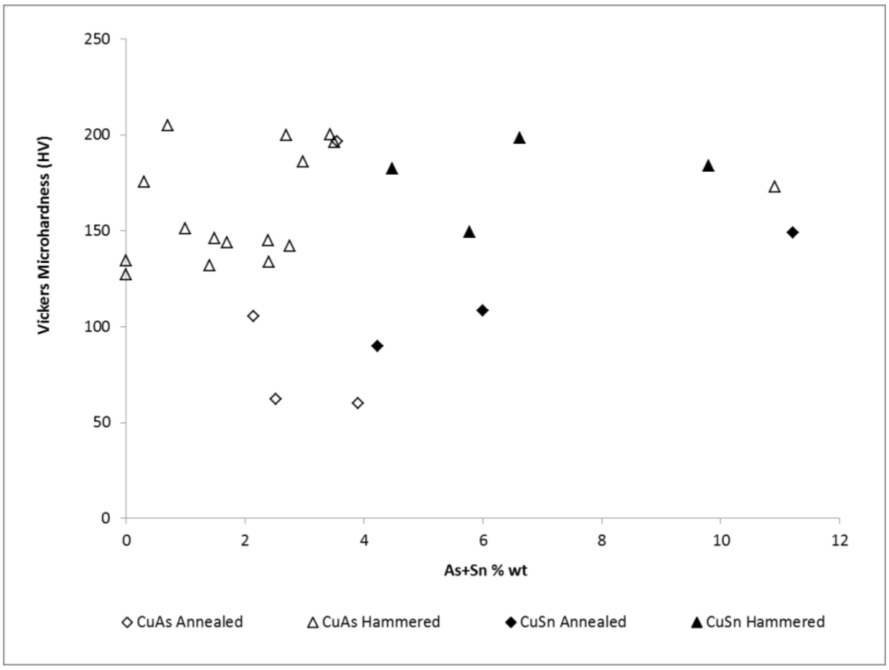

Fig. 9.: Microhardness values of arsenical copper and bronze artifacts. Note how some artifacts with low levels of arsenic are harder than some bronzes.

9. ábra: Az arzéntartalmú réz és bronz tárgyak keménységértékei. Néhány esetben a kis arzéntartalmú tárgyak keményebbek a bronztárgyaknál

This is not surprising, since, as we have seen, a final cold working stage will increase the hardness of metals, a desirable quality in functional items, while annealing will re-homogenize the alloy, increasing its ductility and malleability, a quality probably more desirable for the manufacture of mainly spiral ornaments that in any event do not need hardness as a fundamental mechanical property and that instead it is desirable to preserve the condition of their surface, which would be destroyed by hammering. Although more metallographic analyses are needed, this trend shows some awareness on working techniques and metals properties by Argaric metalworkers.

The fact that different type of artifacts featured different working techniques could be also related with their chemical composition, which is also consistent with the above results. Elemental analyses of c. 700 copper-based Argaric objects have been conducted since Siret´s first study (Montero Ruiz et al. 2019). The relationship between their variables shows a positive correlation between chemical composition and type of artifacts. There seems to be a trend by which most of the ornaments analysed (51%) are in fact bronzes, being tools and weapons mostly made of arsenic copper: 100% of the halberds, 85% of the daggers, 84% of the axes, 83% of the awls and 70% of the swords analysed (Montero Ruiz et al. 2019).

It is in a late phase of the Argaric period (c. 1800 cal BC) that tin bronze appears for the first time, being mainly used in ornament manufacture (Montero Ruiz et al. 2019): this is suggested by some grave goods where copper or arsenic copper tools can be found together with tin-bronze ornaments. This is the case of graves number 1034 from El Argar site (Antas, Almería), 164 and 237 from El Oficio (Cuevas de Almanzora, Almería) or grave number 6 from Cerro de San Cristóbal (Ogíjares, Granada). Other late graves contain tools made with both types of metals (arsenic copper and tin bronze). This is the case, for example, of the grave 21 at Cerro de la Encina (Monachil, Granada) (**Fig. 4.**), with two inhumations dated by AMS at the end of the Argaric period (Beta-230005, 1650–1460 cal BC 2σ and Beta-230006, 1730–1510 cal BC 2σ) (Aranda et al. 2008) what shows that arsenic copper was not substituted by tin-bronze in the Argaric period. We have no knowledge of graves where tools are made of tin-bronze and ornaments with copper or arsenic copper.

With all this, it becomes apparent that copper or arsenic copper was not completely substituted by tin-bronze. The innovation of tin-bronze occurred in a late period of the Argaric society and its adoption and generalization were a slow process, being more related to ornamental and aesthetic motivations than to productive ones. However, most of the metallic assemblages are recovered from the funerary record.

We can also see that some of the copper-tin alloys have also some amounts of arsenic, which could suggest that the same copper ores were used in the production of arsenic copper and tin-bronze objects, while arsenic is not detected in other tin-bronzes, suggesting the exploitation of different ores or some technological issues in the co-smelting of copper and cassiterite which might prevent arsenic to alloy with copper. The recent identification of tin bronzes with a likely provenance from areas outside the Argaric territory (Pedroches, Pyrennes or the Alps) and the identification of some unusual objects (decorated daggers) suggest that an exchange of manufactured metal objects underlies in this demand (Montero Ruiz et al. 2022). However, the option of the ap-pearance of bronze artifacts exclusively as imported objects does not explain the technological change and their adoption. Unfortunately, we do not have archaeological evidence (tin ores, slags or smelting debris) to support the local production of bronze, except the lead isotope analysis confirmation of the involvement of local copper resources in this production.

Conclusion

Although more analyses are needed, some social patterns and issues regarding technological change, such is the adoption of tin-bronze and the later abandonment of arsenic copper in Argaric societies, can be pointed out.

Physical and mechanical properties of both alloys have been discussed, evaluating the possible adoption of bronze due to its superior utilitarian properties. However, although tin bronze objects can be hardened more than arsenic copper ones, especially in alloys over 8% tin, most Argaric bronzes have a low tin content and present a significant overlap with the mechanical properties of arsenic coppers within the recorded compositional ranges for both alloys in the period studied. Under these conditions, it has been shown that the hardness of metal artifacts depends more on the final stage of the *chaîne opératoire* than on their content of arsenic or tin, and that arsenic coppers can be as hard as tin bronze objects or even harder. Moreover, tin bronze alloy was preferably used in the manufacture of body ornaments. All these characteristics of Argaric metallurgy imply that the potential improvements of mechanical and physical properties of this new alloy were not being exploited.

If tin bronze was not chosen because of its mechanical properties, other explanations should be considered. Colour, shining, reflectivity, symbolic and aesthetic values of metals have also been proposed in other cultural contexts and an anthropological view of the use and consumption of metals, more based on sensory and symbolic aspects (Comendador Rey 2010) must be considered. This perspective also affects to the operational technical chain of metal, attending the sensory aspects of metallurgy (smell, sound, colour, etc.) as it has been proposed and related to skills (Kuijpers 2013, 2017).

Leaded high-tin bronzes reflectivity was highly valued in mirror fabrication in China or the Roman Empire (Scott 1991; Mei 2000; Wang 2002) and this quality was also considered when manufacturing bronze drums or bells in India, Southeast Asia or China (Rajpitak 1983; Srinivasan 1997; Srinivasan & Glover 1995; Murillo-Barroso et al. 2010). However, in all these cases, tin values were far higher than those found in the Argaric society.

Metallurgical studies in Latin America have also stressed properties other than "functional" or "utilitarian" in alloys used. The importance of color (especially silver and gold) in the cosmologies and political ideologies in Andean Societies was discussed by Lechtman (1993) with a gender approach; and in Western Mexico sonority and colour of metals seem to have played a role in the selection of the alloys for making bells and other 'status items' (Hosler 1995: 100). In pre-Columbian metal objects of Muisca (Bogotá), alloys of different proportions of copper and native gold have been documented in offering assemblages, although symbolism, rather than color, has been proposed to explain differences in compositions (Uribe Villegas & Martinón-Torres 2012). In Argaric societies, cosmological and gender approaches in the use of silver and gold have also been proposed (Perea 2011), in which silver or silvery objects would be preferably associated to women and gold or golden objects to men.

In recent years, the influence of arsenic and tin on the colour of copper alloys has been intensively studied and described (Fang & McDonnell 2011; Mödlinger et al. 2017; Radivojevic et al. 2018). In the case of copper-based alloys, it is known that high tin will provide a silvery resemblance to copper and the characteristic reddish color of copper is progressively lightened as more tin is added, but the yellowness of the alloy will only decrease with tin contents over 18% and hence the silvery resemblance is only accomplished in alloys between 18–33% Sn. Therefore, in the Argaric bronzes, with an average of 6.9% Sn in this study, colour will surely be modified, and even if the silvery colour was not obtained, the redness of copper would have been significantly lightened to a more golden appearance, so the addition of tin would have been conspicuous. The change of colour depending on the metal could explain the combination in the same grave of body ornaments made of arsenic copper, tin bronze and silver (Montero Ruiz et al. 2019: 22).

The scarcity of tin in the area compared with the easy acquisition of copper and arsenic copper (which occurs abundantly in Southeast Iberia) can be also considered a key issue. Adding a scarce raw material to copper would have increased its social value (Gilman 1981, De Marrais et al. 1996). In this sense, the adoption of tin-bronzes, together with the expansion of copper-based and silver ornaments, chronologically a contemporaneous phenomenon c. 1800 cal BC, can be linked with the ideological and social role played by metals in the visibility and materialization of social power and status of the Argaric elite. In the Argaric society, tin-bronzes would be part of these mechanisms which would have contributed to legitimate and reproduce asymmetrical social relations and political power. We could therefore consider bronzes as a way of wealth accumulation and ostentation.

More analytical studies must be carried out, but up to now all evidence seem to indicate that the adoption of bronze by the Argaric society could respond more to ideological mechanisms of ostentation and status consolidation than to mechanical improvement concerns or productive requirements, and therefore it should be considered more a consequence of social stratification processes than a cause of them.

Contribution of authors

Mercedes Murillo-Barroso Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – Original Draft, Writing – Review and Editing, Visualization, Funding acquisition. **Auxilio Moreno Onorato** Validation, Investigation, Writing – Original Draft, Writing – Review and Editing. **Gonzalo Aranda Jiménez** Investigation, Resources, Writing – Review and Editing, Funding acquisition. **Aaron Lackinger** Formal analysis, Investigation, Writing – Review and Editing, Visualization. **Francisco Contreras Cortés** Investigation, Resources, Writing – Review and Editing, Funding acquisition. **Ignacio Montero-Ruiz** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – Original Draft, Writing – Review and Editing, Visualization.

Acknowledgements

This research has been supported by the R & D Project PID2022-137494NB-I00 (*Redes Sociales y de Intercambio en la Sociedad Argárica)* funded by MICIU/AEI/10.13039/501100011033 and by FEDER, UE and the Research Groups (HUM 065) GEA: *'Material Culture and Social Identity in Recent Prehistory of the Iberian Southeast' and* (HUM 274) GEPRAN: *Group for the Study of the Recent Prehistory of Andalusia*. We are very grateful to Isidro Toro Moyano, director of the Archaeological Museum of Granada, for allowing us to take samples of Argaric metal artifacts, and to Marc Gener, CENIM-CSIC, Madrid, for his comments on the text and editing improvements. Thank you to the reviewers for the useful comments to improve the paper. All errors remain our own.

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