ARTÍCULO

Trabajos de Prehistoria, 81 (2)
Julio-Diciembre 2024, 963
ISSN-L: 0082-5638 – eISSN: 1988-3218
https://doi.org/10.3989/tp.2024.963

Non-invasive analysis of faience necklace beads and scarabs from Late Bronze Age and Early Iron Age necropolises in southern Portugal

Análisis no invasivo de cuentas de collar y escarabeos de fayenza de necrópolis de la Edad del Bronce Final y de la Primera Edad del Hierro en el sur de Portugal

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Submitted: 18-10-2023; accepted: 27-09-2024; published online: 04-02-2025

Abstract: In this work, non-invasive techniques (XRD and microPIXE) were used to study 14 necklace beads associated with human inhumations from the Late Bronze Age (10th-9th century BC) hypogea of Monte da Ramada 1 (Aljustrel), from the Early Iron Age (7th-6th century BC) necropolises of Palhais (Beja), Montinhos 6 (Serpa) and Corte Margarida (Aljustrel), and a blue bead from the Orientalising settlement (8th century BC) of Quinta do Almaraz (Almada). Two scarabs were also analysed, one found in Palhais and the other at Corte Margarida. The results show that most of these small, high-value ornaments have a body of ground quartz covered by glaze. They are consequently classified as faience artefacts. In addition, specific colorants of the glazing mixture were recognised, namely metal ions such as those of copper for the blue/green glazes and of iron for the red/brown glazes.

This kind of glazed artefacts from Portuguese protohistoric contexts has rarely been identified and characterised by archaeometric techniques. Moreover, its occurrence in a necropolis located far inland in southern Portugal, accurately radiocarbon-dated to the Late Bronze Age, testifies to an Orientalising trade in luxury products before or just at the time of the foundation of the first Phoenician settlements on the Iberian Peninsula's coasts.

Keywords: Orientalising trade; ornaments; scarabs; XRD; microPIXE.

Resumen: *En este trabajo se han caracterizado, mediante técnicas no destructivas (DRX y microPIXE), 14 cuentas de collar asociadas a inhumaciones humanas de la Edad del Bronce Final (ss. X-IX a. C.) en hipogeos de Monte da Ramada 1 (Aljustrel) y de la Primera Edad del Hierro (ss. VII-VI a. C.) en las necrópolis de Palhais (Beja), Montinhos 6 (Serpa) y Corte Margarida (Aljustrel), así como una cuenta azul encontrada en el poblado orientalizante (s. VIII a. C.) de Quinta do Almaraz (Almada). También se analizaron dos escarabeos, uno encontrado en Palhais y otro en Corte Margarida. Los resultados muestran que la mayor parte de estos pequeños elementos de adorno de alto valor tienen un cuerpo de granos de cuarzo recubierto por un vidriado. Se han identificado, en consecuencia, como adornos de fayenza. También se han determinado colorantes específicos de la mezcla de glaseado, a saber, iones metálicos como los de cobre para los azules/verdes y de hierro para los glaseados rojo/marrón.*

Este tipo de artefactos vidriados encontrados en contextos protohistóricos de Portugal rara vez han sido identificados y caracterizados mediante técnicas arqueométricas. Además, su aparición en una necrópolis situada en el interior del sur de Portugal y fechada con precisión por radiocarbono en el Bronce Final apunta a un comercio orientalizante de productos lujosos con anterioridad o contemporáneamente a la fundación de los primeros asentamientos fenicios en las costas de la península ibérica.

Palabras clave: *comercio orientalizante; adornos; escarabeos; DRX; microPIXE.*

Cómo citar / Citation: Soares, A. M. M., Gonçalves, A. P., Alves, L. C., Valério, P., Santos, F., Soares, R. M. and Deus, M. de (2024). "Non-invasive analysis of faience necklace beads and scarabs from Late Bronze Age and Early Iron Age necropolises in southern Portugal". *Trabajos de Prehistoria,* 81 (2): 963. DOI: https://doi.org/10.3989/tp.2024.963

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1. INTRODUCTION

In the Iberian Peninsula, necklace beads and small ornaments with a macroscopic glazed appearance have been sporadically recorded since the Bronze Age. They are more frequently associated with Early Iron Age (henceforward EIA) funerary contexts. However, their identification is very incomplete, as only a relatively scarce number of archaeometric analyses have been performed so far (Martínez Mira and Vilaplana Ortego, 2014a, 2014b; Costa *et al*., 2019, 2022; Zilhão *et al*., 2021; Vilaça and Gil, 2023). For this reason, the terminology used in the archaeological literature is often incorrect. They are broadly classified as being fashioned from glass or vitreous paste, when in fact they are made of faience (Egyptian faience). Recent archaeological fieldwork carried out in the south of Portugal as part of the measures to mitigate the impact on cultural heritage caused by the construction of the Alqueva dam irrigation network, has led to the recording and collection of many of these artefacts, i.e., necklaces with dozens or even hundreds of small beads, allowing a systematic scientific study to be carried out to characterise and, consequently, accurately identify these small objects.

Faience generally consists of a body of ground quartz or quartz sand that is aggregated and coated with a soda-rich glaze made by mixing an alkali flux, lime and colorants (Kaczmarczyk and Hedges, 1983; Tite *et al*., 2007, 2009; Tite and Shortland, 2008; Toffolo *et al.*, 2013). Three different glazing techniques can be used in the production of faience artefacts - application glazing, efflorescence glazing and cementation glazing, the latter being the most common in the manufacture of small objects like necklace beads (Toffolo *et al.*, 2013). Regardless of the technique used, faience artefacts are constituted by a structure consisting of three microstructural levels: a quartz-rich core in which the grains are held in place by interparticle glass, an outer, generally thin, quartz-free glaze, and between these two layers a very thin intermediate layer in which the quartz grains are immersed in the glass matrix (Tite *et al*., 2007). Under burial conditions, the vast majority of faience artefacts are subject to severe weathering, which causes the glaze to undergo a selective leaching with removal of sodium and potassium and a dissolution of the silicate network (Costa *et al.,* 2022, p. 3). As a result, many faience artefacts collected from archaeological excavations no longer have the outer glaze layer, or this layer is severely degraded. Therefore, it is usually easy to verify the existence of a quartz core by identifying the crystalline grains of this mineral by X-ray diffraction (XRD). Unlike the XRD analysis of faience, the diffractograms obtained for glass will essentially indicate an amorphous, non-crystalline structure, making it easy to distinguish between these two types of materials. XRD analysis is a non-invasive technique that represents a fast and reliable method for the identification of archaeological faience artefacts, namely the tiny faience necklace beads that are so common in EIA funerary contexts in the Iberian Peninsula.

Two recently published studies have aimed to characterise the glass-like scarabs and necklace beads found in the EIA necropolis of Vinha das Caliças 4 (Beja) using a multi-analytical approach to “shed some light on their manufacture and provide insights into their probable provenance” (Costa *et al.*, 2019, 2022). They combine variable-pressure scanning electron microscopy coupled with energy-dispersive X-ray spectrometry (VP-SEM-EDS), micro X-ray diffraction (μXRD and handheld X-ray fluorescence spectroscopy (p-XRF) in the case of the scarabs, and laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS), VP-SEM-EDS and μXRD in the case of the beads. Since faience artefacts are very heterogeneous and this condition increases during burial, it is difficult to reliably interpret the results of techniques such as LA-ICP-MS, p-XRF, and VP-SEM-EDS, without sacrificing some of the samples to achieve a more consistent analysis (Costa *et al.*, 2022, p. 2). Nevertheless, they managed to obtain some data on the manufacture of the artefacts and assigned their provenance to the Mediterranean Levant. A third recent study on a single glass-like bead recovered in a Holocene context at Gruta do Caldeirão (Tomar) (Zilhão *et al.*, 2021), has successfully applied the same methodology presented here. In fact, this faience bead was originally integrated in the set of beads whose results are now published.

The use of non-invasive techniques is very important in archaeometric studies, as artefacts are frequently unique, often small, and damaging them is unacceptable. In our study, 14 necklace beads, some of them possibly made of faience, associated with human inhumations in the Late Bronze Age (henceforward LBA) hypogea of Monte da Ramada 1 (Aljustrel) and in the EIA necropolises of Palhais (Beringel, Beja), Montinhos 6 (Serpa), and Corte Margarida (Aljustrel) (Fig. 1), were analysed using non-invasive techniques such as XRD and micro-particle induced X-ray emission spectrometry (microPIXE) in order to identify and characterise the material from which they were made. Two scarabs, one from the Palhais necropolis and the other from a burial cist at Corte Margarida, and a blue cogwheel bead found in the Phoenician settlement of Quinta do Almaraz (Almada) were also analysed using the same methodology.

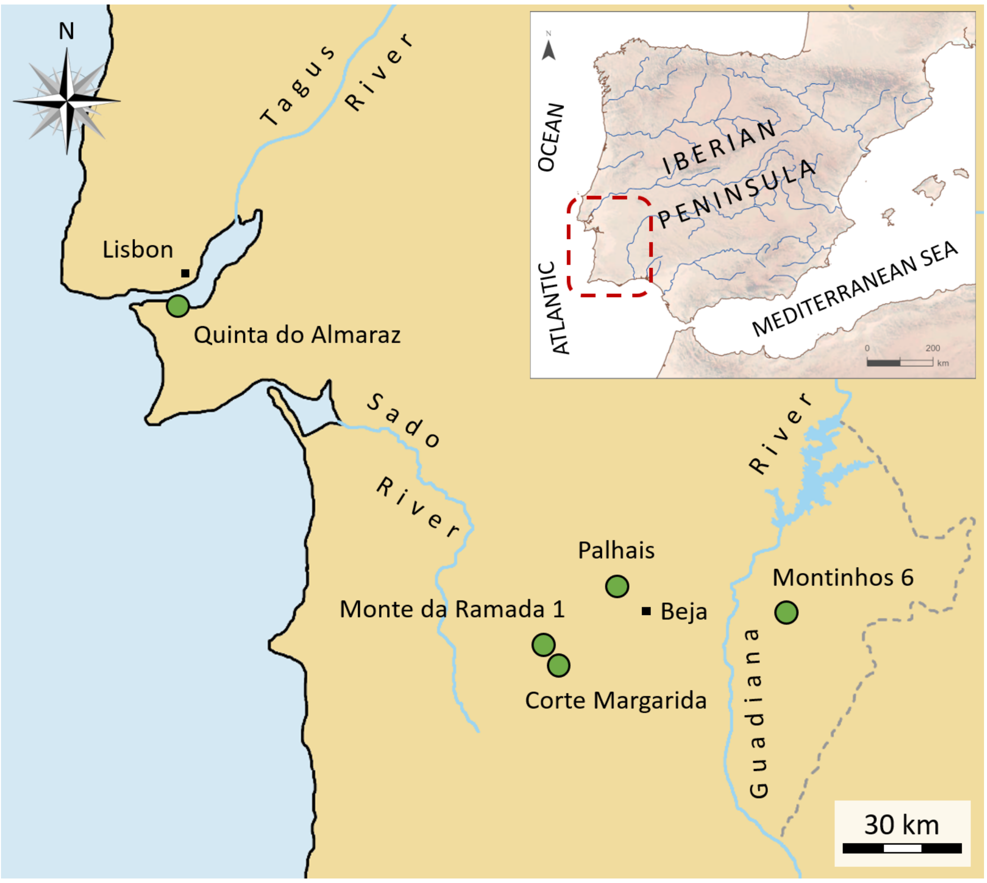

Fig. 1. Location of archaeological contexts (green circles) from where samples were analysed.

2. ARCHAEOLOGICAL SITES AND CONTEXTS

2.1. Monte da Ramada 1

The archaeological site of Monte da Ramada 1, situated in the inland region of southern Portugal (Fig. 1), encompasses a diverse array of archaeological structures and contexts, exhibiting a range of different morphologies and chronologies. These span a considerable chronological range, from the Chalcolithic to the Islamic period (Baptista *et al*., 2018). Two hypogea (hypogeum 2 and hypogeum 4) have been identified among the remains. Each one is composed of a funerary chamber, closed by slabs, with an atrium and presents distinct inhumation sequences. They have been assigned to the southwestern LBA. In the context of hypogeum 4, the slabs were found to be wrapped in a greasy black clayey earth, which is likely to have had waterproof properties due to the presence of an organic component (a fatty material) that would have sealed the chamber (Ribeiro and Soares, 1991; Frade *et al*., 2012). Several individuals buried in these graves were radiocarbon-dated (Soares *et al*., 2020), providing an exceptional opportunity to study rituals and grave goods with a well-defined chronology (Valério *et al*., 2018; Soares *et al*., 2021). Actually, the funerary structures of Monte da Ramada 1 are the first LBA hypogea ever found in southwestern Iberia, substantiating the enduring prevalence of this funerary architecture, which had been in use from the Late Neolithic to the Middle Bronze Age (Alves *et al*., 2010; Valera *et al*., 2008).

The chamber floor of the hypogeum 4 revealed a primary burial of an individual, accompanied by an ossuary on its north side, that consisted of the bone remains of two individuals. The last individual buried in hypogeum 4 had a rich and uncommon set of personal ornaments, including three bronze bracelets and several beads (two of them of gold, six crafted from ostrich eggshell, one presumed to be glass and another made of ivory or bone). The skeleton was radiocarbon dated to the 10th century BC (Soares *et al*., 2020). The presence of such exotic materials and their early chronology point to the existence of pre-colonial contacts with the eastern Mediterranean (Soares *et al*., 2021; Valério *et al*., 2018).

Hypogeum 2 presented a very complex stratigraphic sequence, with a succession of osteo-archaeological deposits that corresponded to multiple phases of funerary usage. The recorded sequence of anatomical connections, isolated or mixed with disarticulated parts, indicates an intense reuse of the funerary structure within a relatively short period of time. In fact, the presence of these anatomical connections reveals that the oldest burials still had soft tissue when they were remobilized. The minimum number of individuals was calculated based on the conjunction of articulated skeletons and disarticulated bone remains, resulting in a total of 20 individuals (Baptista *et al*., 2018). This number is unique in peninsular Bronze Age funerary contexts. Grave goods include exotic ornaments, perhaps of foreign origin, specifically two tiny cylindrical beads, likely made of faience, and a ring, likely made of jet or amber. Six of the skeletons were radiocarbon dated. The results show that the hypogeum was in use during the 10th century BC and continued to be used during the 9th century BC (Soares *et al.,* 2020).

In addition, grave goods include ceramics and bronze artefacts (most of them with an average content of *c.* 10 wt% Sn) that align with the typologies known from the southwestern LBA. However, some of the metal artefacts, such as the set of three bracelets from hypogeum 4, present an uncommon composition, *i.e.*, one arsenical bronze (4.3 wt% Sn - 4.0 wt% As) and two bronze alloys (6.8 wt% Sn - 0.13 wt% Pb - 0.09 wt% Fe and 7.1 wt% Sn - 0.13 wt% Pb) all with a low tin content when compared with the rest of the collection (Valério *et al.*, 2018). The unusual alloy composition and their association with exotic grave goods, either in hypogeum 4 or in the coeval hypogeum 2, suggest a foreign origin for those metal ornaments. It is most probable that they originate from the eastern Mediterranean, where a diversified metallurgical technology emerged during the early stages of the first millennium BC. For instance, a collection of fibulae and pins from Nimrud (Iraq) includes five binary bronzes of analogous composition (3.2–8.0 wt% Sn - 0.15–0.85 wt% As - 0.20–0.90 wt% Pb - 0.02–0.24 wt% Fe; Giumlia-Mair, 1992). The bracelets' early chronology indicates that pre-colonial contacts occurred in some places on the Iberian coast at that time (Soares *et al.*, 2021; Valério *et al.*, 2018).

2.2. Palhais

The Palhais necropolis is part of a distinctive type of EIA funerary structures specific to the Beja region. The funerary architecture and rituals of the EIA in the south and southwest of Portugal are the result of the Orientalising process that occurred in this region between the 7th and 5th centuries BC. Three sub-regions, corresponding to three distinct types of funerary architectural structures, can be identified. The coastal area is characterised by inhumations in stone cists, in accordance with the indigenous southwestern Bronze Age tradition. The inland area of Ourique displays stone *tumuli*, which appear to be associated with the ritual of incineration. The nucleus of Beja was only recently identified and characterised due to the archaeological works related to the Alqueva irrigation network. In this nucleus, the ritual of inhumation prevails, and the architectural structures are characterised by open (U-shaped) or closed orthogonal funerary enclosures that are limited by ditches. A central tomb, a pit with a rectangular shape, was placed within each enclosure, while other individual graves, also with a rectangular shape similar to that one of the central tomb, were located on the limits of these enclosures or in their immediate vicinity. The grave goods, which included ceramics, metal, and exotic raw materials, generally exhibited an Orientalising character (Arruda *et al*., 2016; Soares *et al*., 2016).

The fieldwork conducted at the Palhais necropolis was undertaken as a rescue excavation. The archaeological intervention did not exceed an area of 37 m^2 (Santos *et al.,* 2009). Along with other EIA contemporary necropolises in the Beja region with similar architectural features and contexts, *i.e.*, Monte do Marquês 7 (1300 m to the southwest from Palhais) and Vinha das Caliças 4 (2300 m to the northeast) (Arruda *et al.* 2016), Palhais stands out for being one of the first known Iron Age necropolises with this characteristic typology of the Beja region, and also for the richness and diversity of grave goods that have been recorded therein (Santos *et al*., 2016). In the excavated area, several structures were identified, namely two partially overlapping funerary enclosures. Inside enclosure 1, two negative structures were recorded: a severely damaged burial tomb (grave 4) and another structure, which may have been votive or related to a funerary cult, containing a number of ceramic vases (Santos *et al.* 2016, pp. 242-249). Among them, four vases *à chardon* with red paintings stand out, one of which is crowned by nine ornithomorphs. Furthermore, two additional graves were identified (graves 1 and 2), each featuring a rectangular shaped plant. Each grave held the remains of a single female individual accompanied by grave goods of a personal nature (Santos *et al*., 2016). In grave 1, a necklace with 438 tiny discoid beads and one fusiform bead (perhaps the central element of the necklace) was recovered, along with a small iron dagger with a falcata-shaped blade, a fibula, and a set of instruments for body grooming. These items were all made of copper-based metal (Santos *et al.,* 2009, fig. 6; Santos et al., 2016, fig. 7). Analyses of these artefacts show that they were produced as functional tools using bronze alloys with low tin content (Valério *et al*., 2013). Grave 2 yielded a bronze belt-lock (type 4c, following Cuadrado and Brito, 1970), with an exceptional scroll decoration of palm leaves, a motif characteristic of the eastern Mediterranean region. A small iron falcata-shaped knife and a bronze toilet set, of a lesser quality than the one from grave 1, were also identified. Other grave goods include a silver acorn-shaped pendant and five hollow beads of the same metal (Valério *et al.,* 2013). All of them would originally belong to a necklace, along with several beads of different shapes and apparently made from assorted materials, including glass beads, a decorated circular ceramic pendant and a scarab bearing an incised inscription of Amon-Ré (Santos *et al*., 2009, fig. 8; Santos *et al*., 2016, figs. 9, 10). The most distinctive feature of the copper-based grave goods from Palhais is the alloy composition - bronzes with a low tin content and a relatively high amount of iron impurities (2.0–8.3 wt% Sn - 0.34–0.92 wt% Fe). These compositions clearly fall within the parameters of Orientalising metallurgy (Valério *et al.,* 2013). It should be noted that the composition of these metals agrees with the coeval metallurgy in the region, as evidenced by analyses of metal grave goods from other similar EIA funerary contexts in the Beja region (Valério *et al.,* 2021). A different question is whether the items were manufactured abroad or locally. The presence of uncommon metallic typologies, such as the toilet sets, together with the association with clearly imported materials, such as the necklace beads and scarab made of exotic raw materials, suggest the potential trade with other Mediterranean regions. Furthermore, the iron content of the analysed bronze alloys is also of considerable significance. The importance of a low amount of iron (<0.05 wt%) in ancient bronze alloys was first acknowledged through a comparative analysis between LBA and Phoenician bronzes from Iberian sites. This observation was regarded as an indicator of smelting with a poor reducing atmosphere (Craddock and Meeks, 1987). In the Iberian Peninsula, the low iron contents of EIA bronzes are attributed to the continued use of smelting crucibles throughout the Iron Age, as opposed to the utilization of true furnaces (Rovira and Montero Ruiz, 2013; Valério *et al*., 2016). Additionally, in regard to the copper-based metallic artefacts unearthed in Morro de Mezquitilla (Málaga) and Tejada la Vieja (Escacena del Campo, Huelva), which date to the 7th-6th centuries BC and 6th-4th centuries BC, respectively, Giumlia-Mair states that “the relatively high iron content of the analysed items indicates the use of copper smelted in furnaces with facilities for tapping the slag and therefore close contact with the civilizations of the eastern Mediterranean” (Giumlia-Mair, 1992, p. 114). As far as we know, the earliest copper-based metallurgical workshop in EIA contexts of southwestern Iberian Peninsula is that of Cabeço Redondo (Moura), ascribed to the 5th century BC. This workshop used copper obtained in slag-tapping furnaces, which were likely imported from abroad (Valério *et al.*, 2022). Therefore, the bronze artefacts recovered in the Palhais necropolis and other coeval necropolises in southern Portugal are most likely local copies of foreign models or imports, particularly those with a high iron content.

2.3. Montinhos 6

The site of Montinhos 6, located on the left bank of the Guadiana river (Fig. 1), was also surveyed in the framework of the irrigation network linked to the Alqueva dam. However, the area covered by the archaeological excavation was considerably larger than that of Palhais. Approximately 250 archaeological contexts were identified and recorded, of which around 200 were assigned to a prehistoric chronology. This consisted of hypogea and pits dated to the Chalcolithic and the Bronze Age. The remaining contexts are from the post-Roman period or of an undetermined chronology (Baptista *et al*., 2012). An unusual EIA grave was recorded. It contained the burial of an adult female individual, deposited in a long and narrow pit that seemed to have been in existence prior to the burial. It was radiocarbon dated to a period between the 8th and the first half of the 6th century BC (Soares *et al*., 2016, fig. 2, p. 133). With regard to grave goods, only a necklace made up of 39 whitish and/or bluish-green discoid beads, some with a vitreous appearance, as well as a drop-shaped glass pendant and two brownish tubular beads of the same material, were recovered. All of these items were located in the neck region of the deceased (Soares et al., 2016, fig. 3).

2.4. Corte Margarida

Two stone cists were excavated in the geological substrate (schist) at the necropolis of Corte Margarida. Their architecture is very similar to that of graves from the southwestern Bronze Age, *i.e.*, a stone box with a rectangular shaped plant made up of four slabs placed vertically. The interior of grave 1 features a triangular compartment, situated within one of the cist’s corners, which contained a ceramic vase, potentially a funerary urn. However, no evidence of bones or ashes was found within this vessel or within the remaining space of the cist. The burial gifts consist of a ceramic cup, a necklace comprising approximately 20 beads, the majority of which are ring-eye black glass beads, and three small amber pendants. Similarly, grave 2 also lacks bone remains, but the grave goods are more varied. These include a necklace with 48 beads, some of which are ring-eye black glass beads and others of different shapes and raw materials; a scarab with an inscription on the base referring the name "Pedubaste"; a fragment of a silver ring; a probable shark tooth; and two bird-shaped clay figurines. An EIA chronology (6th century BC) has been attributed to this necropolis (Deus and Correia, 2005), which displays architectural features that are not affiliated with the typical EIA typology observed in Beja funerary necropolises, but rather with the funerary world of the coastal regions of Algarve and Alentejo (Arruda, 2009), although it is located not far from Beja (Fig. 1). It is important to note, however, that there seem to be a common symbology and mythology shared by these EIA communities, in which birds, bulls, and felines, among others, assume a central role. The ceramic ornithomorphs and bovids from funerary contexts in the Beja region, like those from Corte Margarida and Palhais necropolises, as well as the ceramic figurines representing felids and bovids from necropolises of the Ourique region, seem to translate a similar ideological discourse of Mediterranean inspiration, most likely with an Orientalising origin.

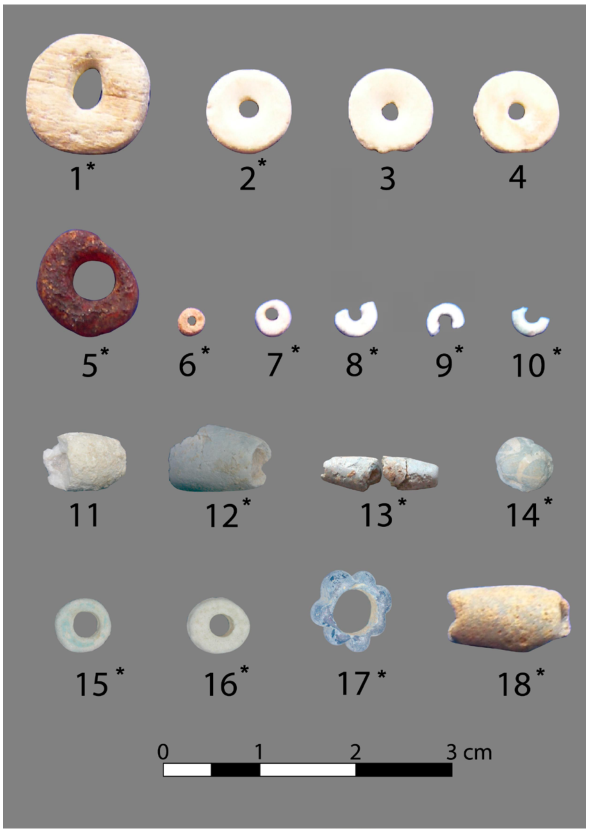

Fig. 2. Protohistoric necklace beads from southern Portugal (* - beads analysed in this work). 1-6 - Monte da Ramada 1 (Aljustrel); 7-14 - Palhais (Beringel, Beja); 15-16 - Montinhos 6 (Serpa); 17 - Quinta do Almaraz (Almada); 18 - Corte Margarida (Aljustrel).

2.5. Quinta do Almaraz

Archaeological excavations carried out in the estuarine settlement of Quinta do Almaraz have revealed a diverse material culture, in which Phoenician and/or Orientalising artefacts occupy an important place. However, the presence of materials with chronologies spanning a considerable length of time suggests that the occupation of the site began in the Late Neolithic. While an exhaustive study of the artefacts recovered during the various archaeological surveys has yet to be published, the radiocarbon dating of several samples indicates that the EIA occupation started at the end of the 9th, beginning of the 8th century BC (Barros and Soares, 2004; Soares and Arruda, 2018). Furthermore, the site, situated on the left bank of the Tagus River, has yielded a plethora of Orientalising archaeological finds, some of which are rare and even unique within the Portuguese territory. These include alabaster vases (Cardoso, 1995), a scarab dated between the 7th and

Fig. 3. Scarabs analysed: A - Palhais; B - Corte Margarida.

6th centuries BC (Almagro Gorbea and Torres Ortiz, 2009), Middle Corinthian pottery, a Greek import from the archaic period dated between 600 and 575 BC (Cardoso, 1995) which is very scarce in Portuguese sites (Arruda, 2007), some Phoenician red slip ware dishes with a flattened rim of small width (Barros *et al.,* 1993) and a diversified copper-based metallurgy that diverges significantly from the indigenous metallurgy inherited from the LBA (Valério *et al.,* 2012).

In fact, the metallurgy introduced by Orientalising populations is clearly discernible in the Phoenician settlement of Quinta do Almaraz (Almada). The metal collection from this coastal site, dating from the 9th to 7th century BC, comprises a considerable number of copper artefacts and some leaded bronzes. Nevertheless, binary bronzes remain the most common alloy, yet are much poorer in tin compared to those from LBA (5.4 ± 2.0 wt% Sn, n=15, Valério *et al.*, 2012), exhibiting an average value that is approximately half of the LBA indigenous collections. The iron impurities are also much higher (0.15 to 1.3 wt% Fe) than the observed in the 'indigenous' collections (mostly <0.05 wt% Fe). A comparable diversification of metallurgy has been identified in other coeval areas of the southwestern Iberian Peninsula, suggesting strong links with the prevailing Mediterranean metallurgy of the period.

3. MATERIALS AND METHODS

Until recently, little use was made of instrumental analytical methods to identify and characterise the materials employed in the manufacture of archaeological artefacts recorded in LBA and EIA contexts from the Iberian Peninsula, with the notable exception of research on archaeological pottery or metals. In the course of our research, we selected 15 necklace beads and 2 scarabs from protohistoric contexts (Fig. 1). The main purpose of this investigation is to facilitate the differentiation of glass or the so-called vitreous paste materials from faience (Egyptian faience), in an

Context	Artefact	Reference*	Colour	Shape	D	Th/L	P
Monte da Ramada 1 necropolis (Late Bronze Age)							
Hypogeum 2, UE 213	Bead #2	6	Brown	Cylindrical (tiny)	2.8	1.9	0.7
Hypogeum 4, UE 421	Bead #1	5	Dark red/ brown	Cylindrical	10-12	6.5	5.0
Hypogeum 4, UE 416	Bead #2	1	White	Discoid	18	3	3-5
Hypogeum 4, UE 416	Bead #3	2	White	Discoid	9	2	1.7
Palhais necropolis (Early Iron Age)							
Grave 1	Bead #18a	10	Bluish-green	Discoid (tiny)	4	1.5	2.0
Grave 1	Bead #18b	7	White	Discoid (tiny)	4	1.0	1.3
Grave 1	Bead #18c	9	Greenish hue	Discoid (tiny)	4	1.5	1.5
Grave 2	Bead #5	14	Pale blue with circular and oval white patches	G lobular	6	5	1.5
Grave 2	Bead #8	-	White with greenish stains	Spherical	9	8	2.9
Grave 2	Bead #13	13	Pale blue	Barrel (fragmented)	8	25	2.9
Grave 2	Bead #16b	12	White	Barrel (fragmented)	7	~16	1.7
Grave 2	Scarab #2	(Fig. 3A)	White with greenish stains	-	-	15×9**	-
Montinhos 6 grave (Early Iron Age)							
UE 3303	Bead #28	15	Bluish-green	Discoid (tiny)	7	2-3	2.4
UE 3303	Bead #31	16	White	Discoid (tiny)	5.5	2.5	2.1
Corte Margarida necropolis (Early Iron Age)							
Cist 2	Bead #40	18	Brown	Barrel (fragmented)	7	13	4.0
Cist 2	Scarab #1	(Fig. 3B)	White with some dark green patches	-	-	11×7**	-
Quinta do Almaraz settlement (Early Iron Age occupation)							
ALZ 434	Bead #35	17	Blue	Cogwheel with 7 flutes	9	3-5	5

Tab. 1. Dimensions, shapes and colours of protohistoric beads and scarabs analysed by XRD and micro-PIXE (measurements are provided in millimetres; D - outer diameter of the bead (maximum outer diameter for barrel beads); Th - thickness of beads (except barrel beads); L - length of barrel beads; P - diameter of the perforation; *reference in Fig. 2; **length × width).

expedited way, through the utilisation of non-invasive and therefore non-destructive techniques, namely XRD and microPIXE. We took advantage of the availability of these techniques at our research centre (C^2TN) to also attempt to identify or confirm the identification of the raw material (mineral or organic) used in other beads usually found in association with the aforementioned ones, which were of dubious identification through simple macroscopic observation. Fig. 2 illustrates most of the beads included in the analysed set, while Fig. 3 shows the two scarabs. Tab. 1 presents the references associated with the analysed items, along with the dimensions, shapes and colours.

XRD can be used in a non-destructive manner, providing valuable insights into the crystal structure and the composition of the specimens under study. This enables the straightforward differentiation between Egyptian faience and glass. Similarly, the non-invasive character of microPIXE spectrometry renders it an optimal choice for the analysis of cultural artefacts, enabling the determination of their elemental composition and the inference of the colorants used in the glazed mixtures of faience items. MicroPIXE spectrometry also allows to differentiate amorphous organic materials from inorganic ones, thereby complementing the data obtained through XRD.

In this research, the X-ray diffraction system consisted of an X'Pert PRO Panalytical diffractometer with a Bragg-Bentano assembly. The scans were performed at room temperature in a reflection mode using monochromatic Cu-Kα radiation (1.54056 Å) and under set conditions of 45 kV and 35 mA. The data were measured with a 2θ step size of 0.04° in a 2θ-range of 15°-65° and a counting time of 40s per step, in order to obtain good and reliable statistical data. Phase identification was derived from the X-ray diffraction data using the PowderCell programme (Nolze and Krauss, 2000).

Elemental mapping analyses of the artefacts were conducted using microPIXE with an Oxford Microbeams type set-up. One or two different experimental conditions were employed, namely a 2.0 MeV proton beam generated by a 2.5 MV Van de Graaf accelerator and a 1.0 MeV proton beam also generated by the same accelerator. The X-rays emitted by the sample elements were collected by a Si(Li) detector with a resolution of 150 eV. Beam currents of 100 pA were used for all spectra, and the beam spatial resolution was kept at $3 \times 4\ \mu m^2$. The 1.0 MeV proton beam was used to determine low atomic number elements, whereas the 2.0 MeV proton beam was utilised to ascertain the presence of trace elements with high Z, for which it has greater sensitivity. PIXE and Rutherford Backscattering Spectrometry (RBS) spectra were accumulated simultaneously. With the spatial resolution of the beam kept at $3 \times 4\ \mu m^2$, scans of areas up to dimensions of $3 \times 3\ mm^2$ were carried out to obtain elemental distribution maps. Zones or spots from them were chosen for further elemental quantitative or semi-quantitative determinations. The samples were highly heterogeneous, both due to their intrinsic nature and the strong degradation they underwent while buried. Consequently, several spots and/or areas were analysed in each of them to determine their original elemental composition and/or identify the elements that were used as colorants or opacifiers. Furthermore, this approach facilitated an understanding of the manufacturing process. Also, RBS (which is routinely performed simultaneously with PIXE analysis through the use of a particle detector) can provide information on the presence of light elements like C or O based on the elastic recoil of the impinging particles when interacting with the nucleus of the sample constituents. This may offer insight into the presence of oxides or, for instance, carbonates or compounds of organic origin (containing carbon) when analysing samples.

4. RESULTS AND DISCUSSION

4.1. Mineralogical and chemical characterization

XRD was performed on all necklace beads and scarabs (Tab. 1), with the exception of beads #2 and #3 (Fig. 2 - 1 and 2) from hypogeum 4 of Monte da Ramada 1. They were only analysed by microPIXE, as it became evident through simple macroscopic analysis that their manufacture involved the use of raw materials of organic origin.

The diffractograms of the XRD analysis revealed that the majority of the specimens are faience (Tab. 2), with the exception of three beads. Bead #1 of hypogeum 4 of Monte da Ramada 1 (Fig. 2 - 5) and bead #5 of grave 2 of the Palhais necropolis (Fig. 2 - 14) were manufactured with an amorphous raw material. This result may initially be interpreted as corresponding to glass, although this requires confirmation or refutation by subsequent analysis by PIXE. Bead #16b (Fig. 2 - 12) of the same grave is made of calcite. The remaining specimens display a predominantly quartz composition, though they may also contain other minerals, such as coesite [SiO_2], lutecite or moganite [SiO_2], calcite [$CaCO_3$], gehlenite Ca_2Al [$AlSiO_7$] or genthelvite [$Be_3Zn_4(SiO_4)_3S$] (Tab. 2). Fig. 4 presents five XRD diffractograms, which demonstrate the diverse range of minerals or mineral mixtures identified through XRD analysis.

It should be noted that coesite and lutecite or moganite are polymorphs of quartz (SiO_2). These polymorphs are formed at pressures and temperatures

Context	Artefact	Reference*	Colour	Minerals	Raw material
Monte da Ramada 1 necropolis (Late Bronze Age)					
Hypogeum 2, UE 213	Bead #2	6	Brown	Quartz + Coesite	Faience
Hypogeum 4, UE 421	Bead #1	5	Dark red/ brown	*Amorphous*	Glass (?)
Hypogeum 4, UE 416	Bead #2	1	White	n.a.	Bone(?)/Ivory (?)
Hypogeum 4, UE 416	Bead #3	2	White	n.a.	Ostrich eggshell
Palhais necropolis (Early Iron Age)					
Grave 1	Bead #18a	10	Bluish-green	Quartz + Lutecite	Faience
Grave 1	Bead #18b	7	White	Quartz	Faience
Grave 1	Bead #18c	9	Greenish hue	Quartz + Coesite	Faience
Grave 2	Bead #5	14	Pale blue with circular and oval white patches	*Amorphous*	Glass
Grave 2	Bead #8	-	White with greenish stains	Quartz + Coesite + Gehlenite + Genthelvite	Faience
Grave 2	Bead #13	13	Pale blue	Quartz + Calcite	Faience
Grave 2	Bead #16b	12	White	Calcite	Calcite
Grave 2	Scarab #2	(Fig. 3A)	White with greenish stains	Quartz	Faience
Montinhos 6 grave (Early Iron Age)					
UE 3303	Bead #28	15	Bluish-green	Quartz	Faience
UE 3303	Bead #31	16	White	Quartz	Faience
Corte Margarida necropolis (Early Iron Age)					
Cist 2	Bead #40	18	Brown	Quartz	Faience
Cist 2	Scarab #1	(Fig. 3B)	White with some dark green patches	Quartz	Faience
Quinta do Almaraz settlement (Early Iron Age occupation)					
ALZ 434	Bead #35	17	Blue	Quartz	Faience

Tab. 2. Mineralogical contents of protohistoric beads and scarabs analysed by XRD (n.a. - not analysed).

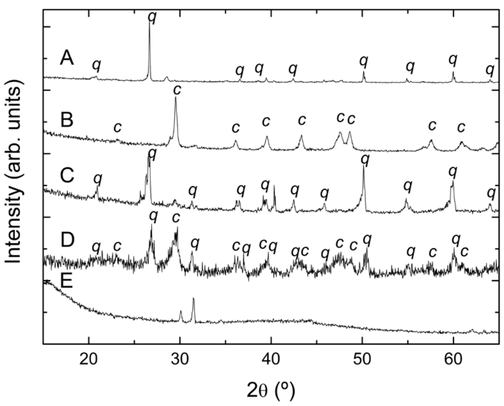

Fig. 4. X-ray diffractograms of: A - Palhais, grave 1, #18b; B - Palhais, grave 2, #16b; C - Palhais, grave 1, #18c; D - Palhais, grave 2, #13; and E - Monte da Ramada 1, UE421, #1. The presence of calcite (c) can be seen in samples B and D, quartz (q) was detected in samples A, C and D, while E has an amorphous structure. Polymorphs of quartz are detected in samples C and D.

different from those involved in the formation of quartz. This evidence indicates that the small crystalline particles that constitute the core of the artefact were obtained from quartz sand and not by the fragmentation of a quartz block or pebble. Genthelvite may be also indicative of the use of this type of sand, while gehlenite is the result of the decomposition of calcite at temperatures between 700-850 ºC, followed by a reaction with aluminosilicates present in faience. This process is similar to the one observed during the firing of clays for the manufacture of common ceramics (Trindade, 2007; Ringdalen, 2015). The presence of gehlenite may be indicative of the use of beach sand, as this kind of sand contains fragments of shells which are, as is known, made up of calcite (or aragonite).

For the PIXE results calculations presented here, all detected elements (those heavier than Na in the case of combining 1.0 MeV and 2 MeV proton beam spectra) were considered to be in their oxide form, with final concentrations normalised to 100 wt%.

4.1.1. Scarab #2 from grave 2 of Palhais

Within area A (Fig. 5), but outside the hieroglyphs depicted on the base of the scarab, there is a slightly greenish tint, which does not appear in the hieroglyphs or on the back of the scarab. There is also a small fracture in area A, apparently recent, with a wavy stain and the appearance of a brownish concretion. This brownish stain, that is easily recognized in the Ca map and the RBS spectrum (not shown), suggests that it

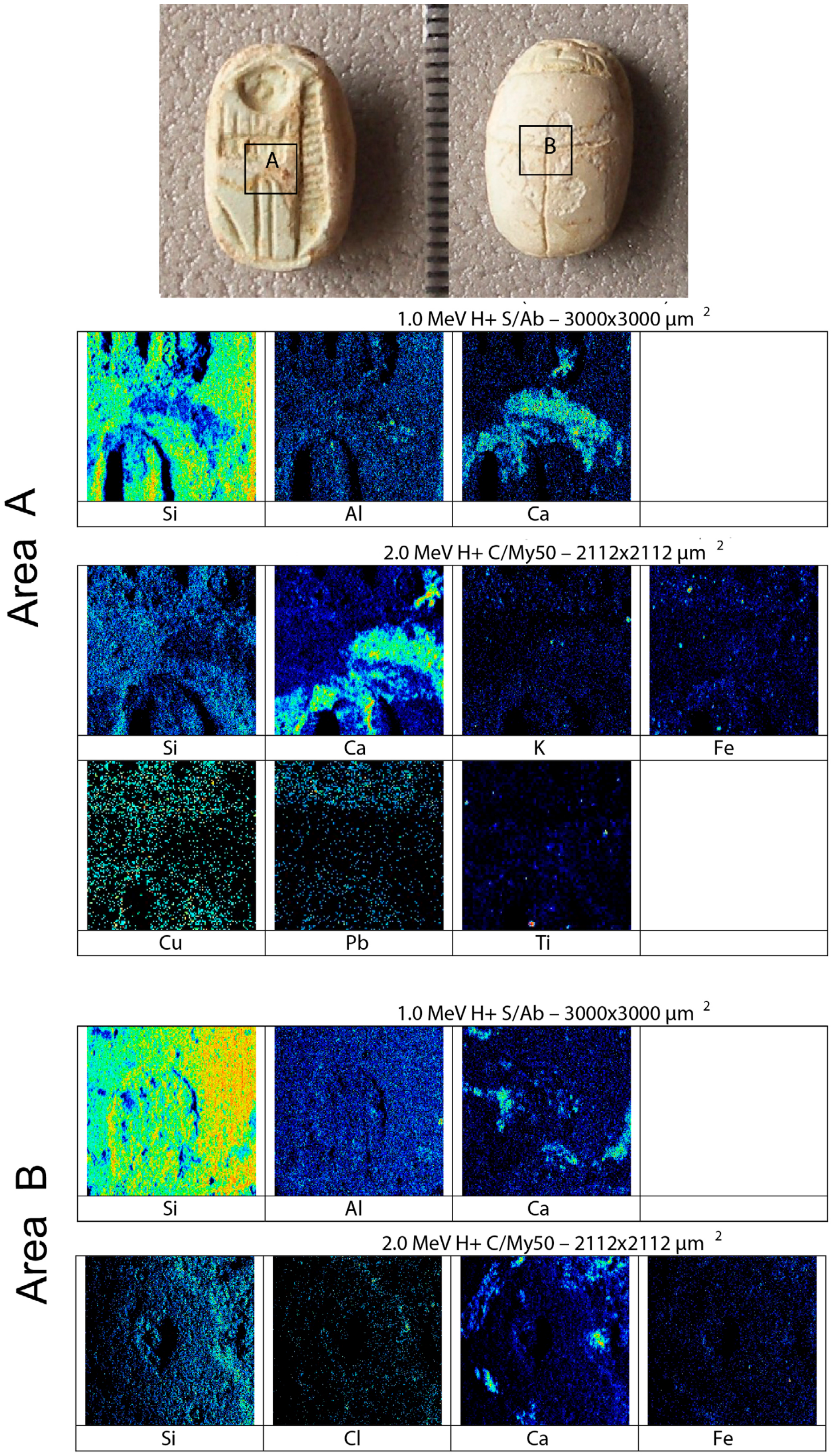

Fig. 5. MicroPIXE elemental distribution maps of areas A and B of the scarab from grave 2 of the Palhais necropolis (colour range from blue to red, representing low to high intensity).

is calcium carbonate. The remaining area in this map, although presenting significant values of Ca, indicates that the predominant component will be silica (Fig. 5 – Si map). On the other hand, there is no marked contrast between the high silica content zone and the more recent fractured zone. Furthermore, it is not possible to associate the greenish tint with any particular elemental composition. With regard to the maps relating to area A obtained with the 2.0 MeV proton beam, although the third map is labelled as corresponding to K, the analysis of the X-ray spectrum indicates that it is Sn or a combination of Sn and K. The energies of the K and Sn X-ray lines have similar values and their contribution to the elemental distribution maps may be difficult to separate. However, their presence can be ascertained by point spectrum deconvolution and indeed the punctual intense zones correspond essentially to K and the extensive ones to Sn. Zones with larger amounts of Cu, Pb and Sn are associated with the deep carved zones of the hieroglyphs, although there is a correlation between these distributions and those of Fe and Ti.

There is also a fracture on the back of the scarab, in the area depicting the chest and wings of the dung-beetle, but this looks old. The distribution maps of major elements (Fig. 5 - maps of area B) show no significant change in elemental content in the fracture zone compared to the rest of the area, except for small agglomerates of Ca (probably $CaCO_3$) on the fracture.

Tab. 3 shows the quantitative results obtained by microPIXE of scarab #2 from grave 2 of Palhais. It appears that the concretions deposited in the fractures are essentially composed of carbonates, which are abundant in the calcareous soil in which the Palhais necropolis is located (Santos *et al*., 2016). It should also be noted that zones of the scarab with high $CaCO_3$ contents also have quantifiable P contents (*cf*. Tab. 3), most likely due to funerary context with which the artefact was associated. The Fe and Ti contents may also partly correspond to soil contamination. The punctual analysis concerning the spot with a high Fe content indicates that it is really a granule of an Fe oxide (goethite? haematite?). On the other hand, there is a correlation between Cu and Sn, which may indicate

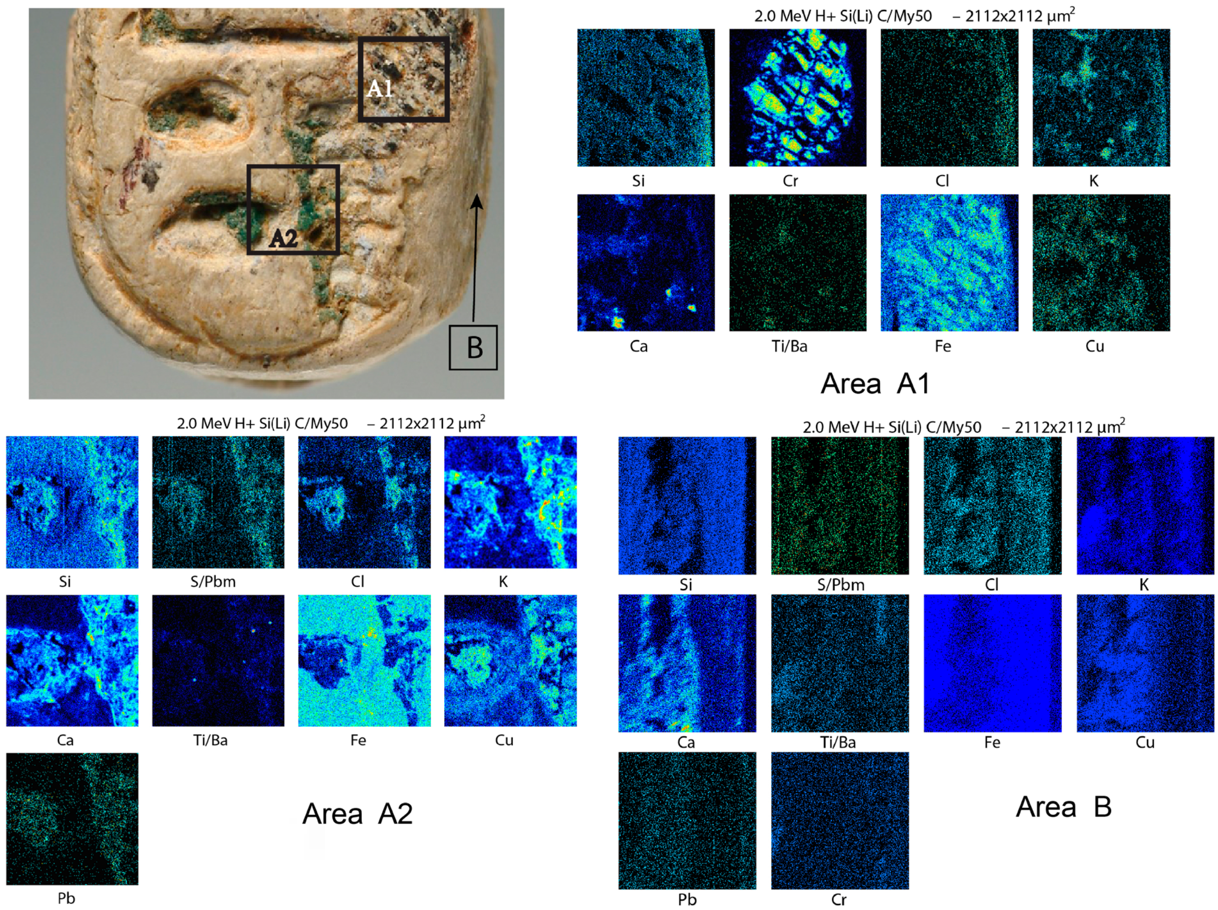

Fig. 6. MicroPIXE elemental distribution maps of areas A1, A2 and B of the scarab from cist 1 of the Corte Margarida necropolis (colour range from blue to red, representing low to high intensity).

that the former, used as a colorant, was added in the form of bronze filings, as has already been detected in beads and scarabs from the coeval necropolis of Vinha das Caliças 4 (Costa *et al*., 2019, 2022). In terms of alkali elements, the low Na and K contents are indicative of the degradation suffered by the scarab. The glaze coating has completely disappeared and the remaining glaze remnants, which are not macroscopically visible, are just those that aggregate the quartz grains that make up the scarab core. However, these low and similar contents also indicate the use of an alkaline flux, the composition of which includes sodium and potassium. A similar situation occurs in the Iron Age faience beads found in Ashkelon, Israel, where, in addition to these low levels of alkali elements, the amount of silica ranges from *ca*. 80 wt% to 99 wt% (Toffolo *et al*., 2013, Tab. 3), which is of the same order of magnitude as that determined for our sample (85-89 wt% SiO_2).

4.1.2. Scarab #1 from cist 2 of Corte Margarida

Three areas of this scarab were subjected to detailed microPIXE analysis using a 2.0 MeV proton beam (Fig. 6). The areas analysed, A1 and A2, are on the base of the scarab and include parts of the hieroglyphs. Area B is located on the side of the artefact that had suffered an extensive superficial fracture during the excavation of the stone cist in which it was found. The carved motifs in A1 and A2 show remains of a very dark green or green-black glaze that would have originally coated the entire scarab. The silica content is generally lower than that of the scarab found at Palhais (Fig. 6 and Tab. 3), while other elements such as K, Cu, Sn, Pb and Ca tend to be higher. This fact may have one of two different interpretations or a combination of both. Differences in elemental content could result from the better preservation of the Corte Margarida scarab, where remnants of the dark green glaze that covered it can still be seen. Alternatively, these two artefacts may have been made in different workshops. The dark green colour of the glaze remains is due to the strong presence of Cr and Fe (see maps of area A1 in Fig. 6). The presence of high levels of Cr (associated with high levels of Fe) has very rarely been detected in analyses of archaeological glass or glazes and has been taken as an indication that the glass (or glaze) is of modern manufacture (Freestone and Bimson, 2003). However, very recent research has allowed the identification of glass beads from the region of present-day Syria dating

	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	S	Cl	K_2O	CaO	TiO_2	Cr_2O_3	MnO	Fe_2O_3	CoO	NiO	CuO	ZnO	Br	Ag_2O	SnO_2	Sb_2O_5	PbO
Scarab #2 / Grave 2 / Palhais (see Fig. 5 for elemental distribution maps)																						
A: Ca-rich area	0.30	0.57	1.9	20.1	0.19	0.19	0.13	0.15	74.6	0.54	n.d.	n.d.	1.24	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
A: Si-rich area	0.58	0.42	2.4	85.1	n.d.	0.13	0.09	0.24	6.5	0.82	0.05	n.d.	1.12	n.d.	n.d.	0.81	n.d.	n.d.	n.d.	0.89	n.d.	0.81
A: area with a greenish tint	0.65	0.17	1.5	88.6	n.d.	0.13	0.24	0.30	5.7	0.42	n.d.	0.01	0.49	n.d.	n.d.	0.63	n.d.	n.d.	n.d.	0.63	n.d.	0.55
A: Fe-rich spot	n.a.	n.a.	n.a.	0.24	n.d.	n.d.	0.02	0.07	0.8	0.10	n.d.	n.d.	98.14	n.d.	n.d.	0.26	n.d.	n.d.	n.d.	n.d.	n.d.	0.42
B: Ca-rich area	0.33	0.66	2.5	23.0	0.25	0.10	0.09	0.24	70.1	0.62	n.d.	n.d.	1.75	n.d.	n.d.	0.15	n.d.	n.d.	n.d.	n.d.	n.d.	0.19
B: Si-rich area	0.73	0.25	2.0	89.1	n.d.	0.15	0.12	0.28	2.9	1.42	n.d.	n.d.	0.96	n.d.	n.d.	0.54	n.d.	n.d.	n.d.	1.18	n.d.	0.36
Scarab #1 / Cist 1 / Corte Margarida (see Fig. 6 for elemental distribution maps)																						
A1: global area	n.a.	n.a.	n.a.	65.3	0.31	0.13	0.38	0.97	1.8	0.15	12.49	0.18	15.3	n.d.	0.24	1.7	0.19	n.d.	0.09	0.5	n.d.	0.2
A1: Fe+Cr-rich spot	n.a.	n.a.	n.a.	3.9	n.d.	0.06	0.16	0.15	0.2	0.08	59.4	0.59	34.6	n.d.	0.06	0.3	0.62	n.d.	n.d.	n.d.	n.d.	n.d.
A1: outside Fe+Cr-rich area	n.a.	n.a.	n.a.	79.1	n.d.	0.15	0.42	0.60	1.9	0.10	1.31	n.d.	11.7	n.d.	0.50	3.3	n.d.	n.d.	n.d.	0.8	n.d.	n.d.
A2: global area	n.a.	n.a.	n.a.	76.7	0.19	0.04	0.57	1.32	3.9	0.23	0.06	0.02	6.7	n.d.	0.25	5.4	n.d.	0.14	0.41	2.1	0.4	1.5
A2: outside Cu+Ca-rich area	n.a.	n.a.	n.a.	81.0	n.d.	0.15	0.53	0.84	1.2	0.10	0.13	n.d.	11.4	n.d.	0.54	2.8	n.d.	n.d.	n.d.	n.d.	n.d.	1.3
A2: Cu-rich spot	n.a.	n.a.	n.a.	73.2	n.d.	n.d.	1.77	1.18	4.0	0.16	0.10	n.d.	2.9	n.d.	0.14	9.7	n.d.	n.d.	0.12	2.7	0.7	3.4
B: global area	n.a.	n.a.	n.a.	77.9	0.28	0.13	0.71	1.83	2.1	0.23	0.26	0.03	7.9	n.d.	0.28	4.5	n.d.	0.20	0.30	1.5	n.d.	1.9
B: Cu-rich spot	n.a.	n.a.	n.a.	64.4	0.14	0.08	1.48	1.50	4.3	0.20	n.d.	n.d.	3.0	0.05	0.07	15.0	n.d.	n.d.	0.20	2.0	2.6	5.0
B: Fe-rich spot	n.a.	n.a.	n.a.	73.1	0.46	0.19	0.77	9.42	1.5	1.54	0.06	0.04	11.8	n.d.	0.13	0.9	n.d.	n.d.	0.10	n.d.	n.d.	n.d.

Tab. 3. Composition of scarabs from Palhais and Corte Margarida (values in wt% and normalized to 100%; the relative error of the analyses is less than 5% for major elements; n.d. - not detected; n.a. - not analysed).

to the beginning of the first millennium BC with high Cr and Fe contents, likely due to the use of heavy metal-rich black sands found in the coastal regions of that country (Reade *et al.*, 2009, table 1, p. 51). It is also verified that in the majority of the analyses made on the Corte Margarida scarab, there is a positive correlation between the Sn contents and those of Cu. This is even more expressive than in the Palhais scarab and, therefore also bronze filings must have been added in the glaze production for this artefact. Finally, it is worth noting the presence of a low Ag content in most of the zones analysed, which could indicate that the scarab was embedded in a silver ring, as was common for this kind of artefact at the time.

4.1.3. Tiny brown cylindrical bead #2 from Monte da Ramada 1

One of the two very small Egyptian faience beads recovered in hypogeum 2 of Monte da Ramada 1 was included in our study. It was analysed by microPIXE using the 1.0 MeV proton beam (Fig. 7 and Tab. 4). It is worth highlighting the Fe content, which is certainly due to the brownish colour of the glaze that this tiny bead presents. The elemental contents determined for the global area, namely those of Si, Na, K, Mg, Al and Ca, are the result of a relatively good preservation of the artefact and indicate the use of a soda lime flux in its manufacture.

4.1.4. Tiny white discoid bead #31 and tiny bluish-green discoid bead #28 from the Montinhos 6 grave

The necklace associated with the EIA female burial at Montinhos 6 is almost entirely made up of tiny discoid faience beads, some white and others with greenish or bluish tones. Two, one white and the other bluish-green, were selected for analysis (Fig. 7 and Tab. 4).

The results suggest that the colorant used may have been a mixture of Cu and Fe, since the content of these elements is higher in the bluish-green bead than in the white one. However, when the Fe content in the glaze is lower than 2 wt%, as it seems to be in these beads, it would not have a significant effect on the glaze colour, according to Gu *et al.* (2014). Conversely, the high levels of Cu in these beads correspond to measurable levels of Sn, suggesting the use of bronze filings

	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	S	Cl	K_2O	CaO	TiO_2	Cr_2O_3	MnO	Fe_2O_3	CoO	NiO	CuO	ZnO	Br	Ag_2O	SnO_2	Sb_2O_5	PbO
Bead #2 / Hypogeum 2 / Monte da Ramada 1 (see Fig. 7 for elemental distribution maps)																						
Global area	1.48	2.78	11.30	70.66	n.d.	0.28	0.06	0.62	3.31	0.28	n.d.	n.d.	8.88	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Si-rich area	2.69	0.40	2.96	84.18	n.d.	0.04	n.d.	0.16	0.98	0.15	n.d.	n.d.	8.46	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Al-rich area	0.28	3.84	17.61	51.97	n.d.	0.03	0.04	0.87	3.58	0.43	n.d.	n.d.	21.08	n.d.	n.d.	n.d.	n.d.	n.d.	0.29	n.d.	n.d.	n.d.
Ca-rich area	0.27	3.00	9.92	33.57	0.36	n.d.	n.d.	0.71	32.84	0.73	n.d.	n.d.	38.51	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
White bead / Montinhos 6 (see Fig. 7 for elemental distribution maps)																						
Global area	n.d.	n.d.	n.d.	98.72	n.d.	n.d.	n.d.	0.06	0.45	0.03	n.d.	0.04	0.31	n.d.	n.d.	0.56	n.d.	n.d.	n.d.	n.d.	n.d.	0.03
Si-rich area	n.d.	0.14	0.81	97.49	n.d.	0.03	n.d.	n.d.	0.49	n.d.	n.d.	n.d.	0.56	n.d.	n.d.	0.54	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Ca-rich area	n.d.	0.15	0.70	96.49	0.32	0.03	n.d.	n.d.	0.99	n.d.	n.d.	n.d.	0.49	n.d.	n.d.	0.86	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Si-rich spot	n.d.	n.d.	0.32	99.30	n.d.	n.d.	n.d.	n.d.	0.19	n.d.	n.d.	n.d.	0.16	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Cu-rich area	n.d.	0.23	0.55	37.80	n.d.	n.d.	n.d.	n.d.	2.90	0.38	n.d.	n.d.	14.58	n.d.	n.d.	41.46	n.d.	n.d.	n.d.	2.06	n.d.	n.d.
Bluish-green bead / Montinhos 6 (see Fig. 7 for elemental distribution maps)																						
Global area	n.d.	n.d.	n.d.	64.13	n.d.	0.23	0.14	1.03	29.52	0.57	n.d.	0.03	2.12	n.d.	0.02	1.87	0.08	n.d.	n.d.	n.d.	n.d.	0.38
Cu-rich area	n.d.	0.44	1.59	88.00	n.d.	n.d.	0.63	0.10	1.75	0.03	n.d.	n.d.	0.84	n.d.	n.d.	5.17	n.d.	n.d.	n.d.	0.26	n.d.	1.28
Si-rich area	n.d.	0.19	0.92	94.37	0.13	0.05	0.02	0.08	3.09	0.02	n.d.	0.06	0.43	n.d.	n.d.	0.36	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Bluish-green bead #18a / Grave 1 / Palhais (see Fig. 7 for elemental distribution maps, 1 MeV)																						
Cu-rich area	0.41	0.69	2.68	86.3	0.19	0.09	0.73	0.02	4.1	0.09	n.d.	0.03	1.13	0.09	n.d.	2.77	n.d.	n.d.	n.d.	0.22	n.d.	n.d.
Si-rich area	0.24	0.26	1.37	94.9	n.d.	0.03	0.04	n.d.	0.91	0.04	n.d.	n.d.	1.35	n.d.	n.d.	0.33	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Ca-rich area	0.37	0.95	3.89	81.5	n.d.	0.08	0.90	0.12	3.9	n.d.	n.d.	n.d.	2.22	n.d.	n.d.	5.46	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Bluish-green bead #18a / Grave 1 / Palhais (see Fig. 7 for elemental distribution maps, 2 MeV)																						
Cu-rich area	n.d.	n.d.	n.d.	90.1	n.d.	n.d.	1.34	0.03	2.2	0.03	n.d.	0.07	0.53	0.17	n.d.	5.40	0.04	n.d.	n.d.	n.d.	n.d.	n.d.
Si-rich area	n.d.	n.d.	n.d.	97.7	n.d.	n.d.	0.33	n.d.	0.47	0.01	n.d.	0.02	0.20	0.03	n.d.	1.02	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Ca-rich spot	n.d.	n.d.	n.d.	65.0	n.d.	0.18	0.22	0.26	25.5	0.33	n.d.	0.10	6.20	0.10	n.d.	1.62	n.d.	n.d.	n.d.	0.45	n.d.	n.d.

Tab. 4. Composition of faience beads from Monte da Ramada 1 (Late Bronze Age), Montinhos 6 and Palhais (Early Iron Age) (values in wt% and normalized to 100%; the relative error of the analyses is less than 5% for major elements; n.d. - not detected).

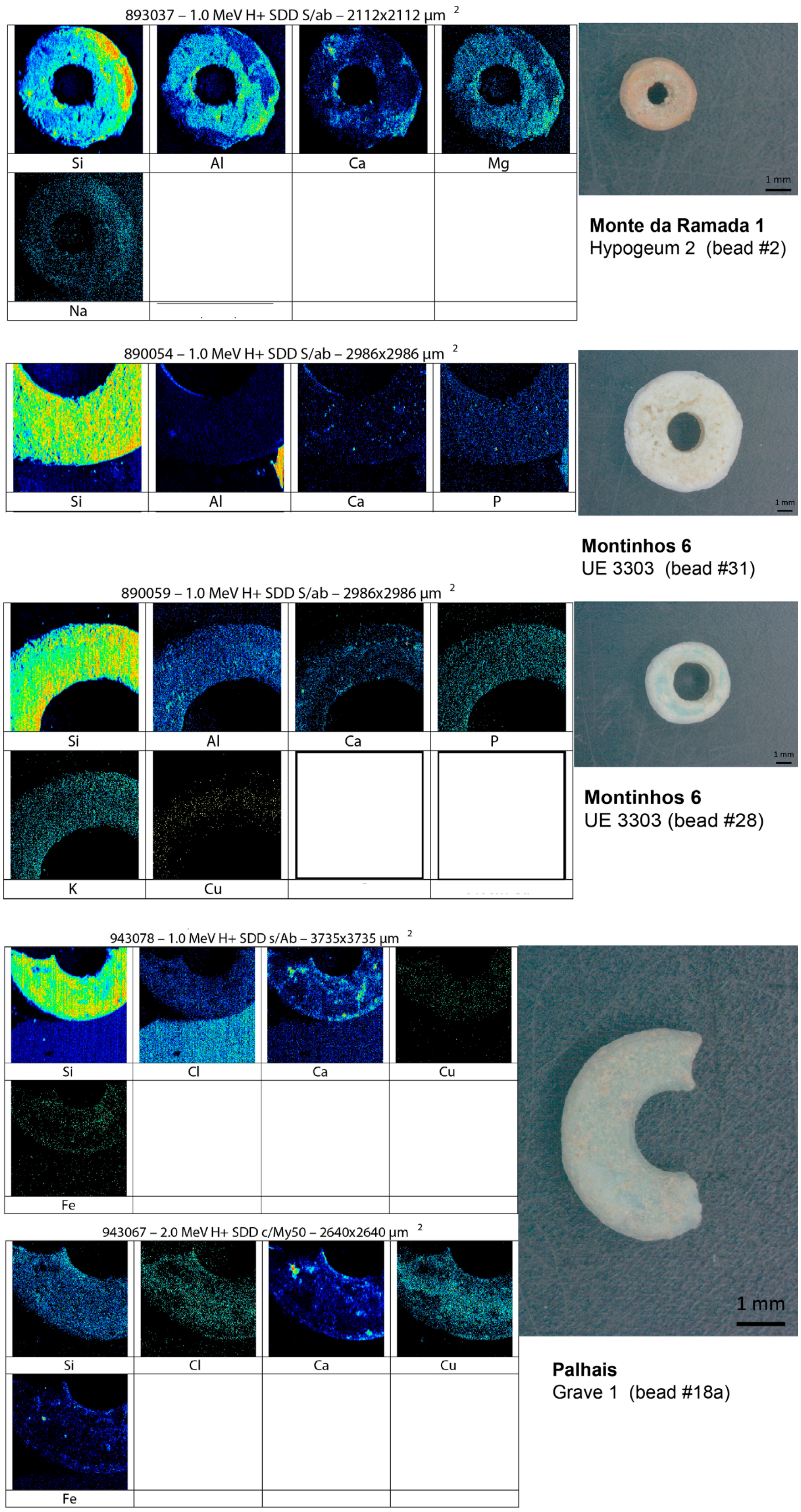

Fig. 7. MicroPIXE elemental distribution maps from discoid faience beads of the hypogeum 2 of Monte da Ramada 1, grave of Montinhos 6 and grave 1 of Palhais (bead number in brackets; colour range from blue to red, representing low to high intensity).

in the manufacture of the glaze, as is the case for the Palhais and Corte Margarida scarabs. Remnants of the glaze coating are only preserved on the bluish-green bead, which still has a faint vitreous appearance. This corresponds to slightly higher levels of K, Mg and Al in some spots compared to the white bead. However, sodium was not detected in any of the beads, indicating a high degree of degradation.

4.1.5. Tiny bluish discoid bead #18a of grave 1 from Palhais necropolis

A number of faience beads were collected from the two graves excavated in the Palhais necropolis, seven of which were analysed. The results of the microPIXE analysis of a slightly bluish discoid bead from grave 1 (Fig. 2 - 10), using the two experimental conditions mentioned above, are shown in Fig. 7 and Tab. 4. The faint bluish colour of this bead would result from the presence of Cu, although the apparent association between Cu and Sn contents identified in other faience artefacts analysed does not seem to exist. Note the absence of measurable levels of Pb and the very high content of Si, which must be due to the severe weathering of this bead, also evident from simple macroscopic observation.

4.1.6. Other analysed necklace beads

The beads from Corte Margarida (#40) and Quinta do Almaraz (#35) are the ones that, unlike most of the faience beads analysed in this work, still have a vitreous lustre due to the preservation of some of the coating glaze. Hence, they have the lowest levels of silica (SiO_2) and the highest levels of alkali metals (Na, K), associated with an equally high content of Ca, the values of which suggest the use of a soda-lime-potash flux in their manufacture. The high iron content (Fe_2O_3: 3.6 wt%) of the brown bead from Corte Margarida indicates that this was the colourant used. Additionally, the bluish bead from Quinta do Almaraz will have this colour resulting from the presence of copper in the glaze (Table 5).

	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	S	Cl	K_2O	CaO	TiO_2	MnO	Fe_2O_3	CoO	NiO	CuO	ZnO	Ag_2O	SnO_2	Sb_2O_5	PbO	Material
Bead #40 / Cist 2 / Corte Margarida																					
Global	5.62	1.03	10.90	67.02	n.d.	0.14	0.54	2.26	5.46	0.36	n.d.	3.62	n.d.	n.d.	0.35	n.d.	n.d.	n.d.	2.76	n.d.	Faience
Bead #35 / Quinta do Almaraz																					
Global	5.60	0.46	9.80	75.1	n.d.	n.d.	0.81	0.73	6.0	0.11	0.02	1.80	0.11	n.d.	0.18	n.d.	n.d.	0.20	n.d.	0.03	Faience
Fe-rich area	n.d.	n.d.	n.d.	73.1	n.d.	0.12	0.62	2.87	6.3	0.97	0.04	14.96	0.08	0.01	0.20	n.d.	n.d.	n.d.	n.d.	0.05	
Bead #18b / Grave 1 / Palhais																					
Global	n.d.	n.d.	0.04	85.7	n.d.	n.d.	0.11	0.12	9.2	0.27	0.02	4.10	n.d.	n.d.	0.05	n.d.	n.d.	0.23	n.d.	n.d.	Faience
Ca-rich area	n.d.	n.d.	n.d.	18.2	n.d.	1.18	n.d.	n.d.	77.9	0.13	0.09	1.56	n.d.	0.12	0.06	n.d.	n.d.	n.d.	n.d.	n.d.	
Bead #18c / Grave 1 / Palhais																					
Global	0.22	0.18	1.04	96.4	n.d.	n.d.	0.02	n.d.	1.0	0.01	n.d.	0.31	0.06	0.01	0.13	n.d.	n.d.	0.63	n.d.	n.d.	
Al-rich área	2.2	3.0	16.5	67.0	0.25	0.09	0.09	0.28	6.3	0.22	n.d.	3.95	n.d.	n.d.	n.d.	n.d.	n.d.	0.16	n.d.	n.d.	Faience
Fe-rich area	n.d.	n.d.	n.d.	64.1	n.d.	n.d.	n.d.	n.d.	0.32	n.d.	0.05	22.59	8.92	1.78	0.97	n.d.	n.d.	0.92	n.d.	n.d.	
Bead #8 / Grave 2 / Palhais																					
Global	n.d.	n.d.	0.70	89.2	n.d.	n.d.	0.10	n.d.	2.9	0.10	n.d.	0.60	n.d.	n.d.	0.50	n.d.	n.d.	n.d.	0.90	4.50	
Ca-rich area	n.d.	n.d.	n.d.	48.8	n.d.	n.d.	0.14	0.11	43.3	0.34	0.05	2.58	n.d.	n.d.	0.45	n.d.	n.d.	0.44	n.d.	3.24	Faience
Si-rich area	n.d.	n.d.	n.d.	90.0	n.d.	n.d.	0.06	n.d.	2.6	0.13	n.d.	0.56	n.d.	n.d.	0.48	n.d.	n.d.	n.d.	0.85	4.09	
Bead #13 / Grave 2 / Palhais																					
Global	0.10	0.30	0.92	78.4	n.d.	n.d.	0.19	n.d.	17.4	0.03	0.06	0.47	0.05	0.01	n.d.	n.d.	n.d.	0.12	n.d.	n.d.	
Fracture section	n.d.	n.d.	n.d.	87.1	n.d.	0.45	0.19	n.d.	11.0	0.04	0.07	0.50	0.07	0.01	0.51	n.d.	n.d.	0.12	n.d.	n.d.	Faience
Si-rich area	n.d.	n.d.	n.d.	95.6	n.d.	0.17	0.15	n.d.	3.0	0.03	0.04	0.46	0.04	0.01	0.30	n.d.	n.d.	n.d.	n.d.	n.d.	
Ca-rich area	n.d.	n.d.	n.d.	50.5	n.d.	1.36	0.09	n.d.	46.8	0.06	0.04	0.70	0.02	0.03	0.14	n.d.	n.d.	0.30	n.d.	n.d.	
Bead #1 / Hypogeum 4 / Monte da Ramada 1																					
(see Figure 8 and interpretation of the data represented therein) Ambar (?)																					
Bead #2 / Hypogeum 4 / Monte da Ramada 1																					
Spot 1	0.69	1.31	0.47	1.71	32.92	0.18	0.14	n.d.	61.73	0.04	n.d.	0.14	n.d.	n.d.	0.47	n.d.	n.d.	n.d.	n.d.	n.d.	Bone (?) Ivory (?)
Spot 2	0.78	1.25	4.20	0.51	31.67	0.11	0.14	n.d.	60.50	n.d.	n.d.	n.d.	n.d.	n.d.	0.63	n.d.	n.d.	n.d.	n.d.	n.d.	
Spot 3	0.63	1.29	0.06	0.13	29.77	0.13	0.14	n.d.	67.24	n.d.	n.d.	n.d.	n.d.	n.d.	0.43	n.d.	n.d.	n.d.	n.d.	n.d.	
Bead #3 / Hypogeum 4 / Monte da Ramada 1																					
Spot	0.29	2.78	1.35	5.19	2.34	0.08	0.04	n.d.	85.35	0.15	n.d.	0.44	n.d.	n.d.	0.16	n.d.	n.d.	n.d.	n.d.	1.76	Ostrich eggshell

Tab. 5. Composition of remaining beads from the Early Iron Age necropolises of Corte Margarida, Quinta do Almaraz and Palhais, and the Late Bronze Age necropolis of Monte da Ramada 1 (values in wt% and normalized to 100%; the relative error of the analyses is less than 5% for major elements; n.d. - not detected; not analysed: bead #5 (Grave 2, Palhais) – glass, and bead #16b (Grave 2, Palhais) – calcite).

In the four samples (#18b, #18c, #8, and #13) from the Palhais necropolis, the coated glaze no longer exists (SiO_2: 78-96 wt%), so that any inference from the determined content values is uncertain. Thus, the colorant used could be iron in the case of beads #18b and #18c or copper in beads #8 and #13 (see also Fig. 2). The high calcium values obtained for these last two beads could be due to concretions observed on their surfaces, resulting from the carbonated geological soil in which they were buried.

Five other beads (#5 and #16b from Palhais, and #1, #2 and #3 from hypogeum 4 of Monte da Ramada 1) were found to have been manufactured with different types of raw material (see Tabs. 2 and 5). Two (Palhais #5 and Monte da Ramada 1 #1) would have been made of glass, based on the non-crystalline raw material identified by XRD. Concerning the globular bead #5 with circular and oval white patches (a ring-eye blue necklace bead) recorded from grave 2 of the Palhais necropolis, the conclusion on its manufacture offers no doubts, also due to its typology. However, the cylindrical dark red/brown bead #1 from the southwestern LBA necropolis of Monte da Ramada 1 raises some doubts due to its colour and external appearance. An attempt was made to better characterise its manufacture through microPIXE analysis. The PIXE spectrum allows the detection of several elements: Na, Mg, Al, Si, P, S, Cl, K, Ca, Fe, Cu and Pb (Fig. 8 - right). This, together with its amorphous character (diffractogram E in Fig. 4), may lead to the assumption that it consists of a glass bead. However, the RBS spectrum (Fig. 8 - left) reveals large amounts of C and O, estimated to account for ~98 wt% of the total matrix composition (C 86 wt% and O 12 wt%), leaving a total amount of ~2 wt% for the elements detected by PIXE. Although these results show its organic origin, further analysis need to be performed to accurately establish the manufacturing material. Due to the colour and texture of this bead, amber is a strong possibility for the raw material used (see Fig. 2 - 6). In this regard, it is interesting to note that in the other funerary hypogeum of Monte da Ramada 1, a ring was found, also of organic origin, very dark in colour, and possibly made of jet (or amber?).

The two remaining beads analysed from Monte da Ramada 1 (#2 and #3) were macroscopically identified as made of ivory or bone and of ostrich eggshell, respectively. The microPIXE analysis of the first, with a P_2O_5 content of 30-33 wt% and a CaO content of 61-67 wt%, confirms this possible attribution, although a more precise identification, bone or ivory, requires other techniques. Regarding bead #3, a CaO content value of *ca*. 85 wt% points to an artefact composed mainly by calcium carbonate (calcite). This confirms that it is made of eggshell. Given its thickness, it would most certainly be an ostrich eggshell (Kandel and Conard, 2005; Stewart *et al.*, 2013). Finally, bead #16b from Palhais was identified as calcite by XRD. It was not analysed by microPIXE, as it is essentially composed of this mineral (Fig. 4B).

5. CONCLUSIONS

Two non-invasive instrumental techniques for structural and elemental analysis, namely XRD and microPIXE, were used to identify and characterise the raw materials used in the manufacture of small ornaments (beads and scarabs) found in LBA and EIA

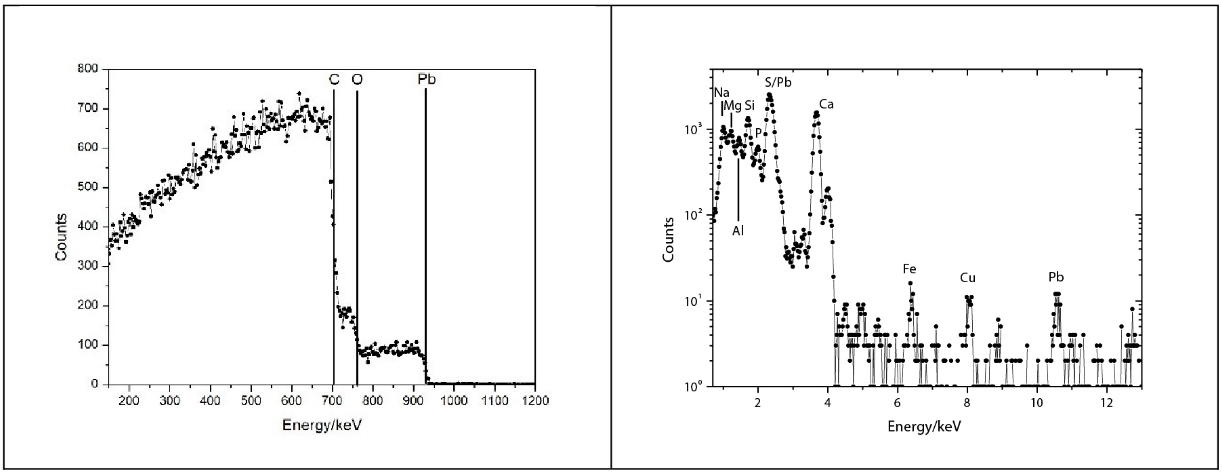

Fig. 8. 1.0 MeV proton beam PIXE and RBS spectra obtained by analysing an area of bead #1 of hypogeum 4 of Monte da Ramada 1. In the PIXE spectrum (right) the detected X-ray lines corresponding to Na or to elements heavier than Na (Mg, Al, Si, Ca, Fe, Cu and Pb) are shown. The RBS spectrum (left) shows the position of the surface barriers of the elements C, O and Pb. The height of these elemental barriers is proportional to the corresponding elemental content (although weighted by a Z^2 ratio).

funerary contexts from southern Portugal. XRD was used to identify the raw materials, while microPIXE was used to characterise their chemical composition. It was found that the vast majority of the artefacts analysed were manufactured from faience, although necklace beads of glass, calcite, ostrich eggshell, and probably jet/amber and ivory or bone were also identified. The archaeological record suggests that, in contexts with these chronologies, most of these small elements of adornment would have been made from vitreous materials, with no distinction being made between glass and faience as the raw material used. However, archaeometric analysis shows that although glass is a common material in the making of many of these artefacts, the predominant material is often not glass but faience.

In the case of the faience objects, which make up most of the set analysed, we must take into account that they are highly degraded, with most, if not all, of the coating glaze having disappeared due to the dissolution of the alkaline metals present during burial. Despite the weathering of these small faience artefacts, it was still possible to draw some conclusions about their manufacture and, occasionally, about their provenance. For example, it has been demonstrated that the core of the analysed artefacts does not only consist of pure quartz grains, but also of crystalline grains of polymorphs of this mineral, such as coesite and/or lutecite. As these polymorphs were formed at pressures and temperatures different from those of quartz formation, this implies that the core of these artefacts would have been manufactured with quartz sand, rather than grains resulting from the fragmentation of a quartz block or pebble. Furthermore, the presence of gehlenite in some samples may indicate the use of beach sand.

The results of the elemental analysis suggest the use of a soda-lime-silica flux in the manufacture of most or all of the faience analysed, as is usual in this kind of artefact. Copper and iron were used as colourants, resulting in the observed blue-green or brown tints, respectively. The microPIXE technique (performed in an RBS mode) allowed to differentiate a non-crystalline raw material of mineral origin from a raw material of organic origin, as shown by bead #1 from the LBA hypogeum 4 of Monte da Ramada 1. Despite its macroscopic appearance resembling glass, it turned out to be of organic origin, possibly amber. This bead and the ostrich eggshell beads, as well as the set of bronze bracelets from the same hypogeum and the faience beads from hypogeum 2, point to a pre-colonial trade with Mediterranean origins. Of particular interest are the remnants of a dark green glaze that can still be seen on the scarab from Corte Margarida. This colour results from the presence of high contents of chromium and iron, which would indicate the use of black sands with a high level of heavy metals, a kind of sand found in deposits scattered from Croatia, across the Balkans, Cyprus, Anatolia and coastal Syria (Reade *et al*., 2009, p. 51), where EIA glassmaking centres are known. However, the name "Pedubaste" is engraved on the base of this scarab, and until now it was thought that scarabs with this name were manufactured in Naucratis, a Greek site in the Nile Delta. The evidence provided by this faience scarab therefore calls into question the traditional provenance attributed to these remarkable amulets with the name "Pedubaste". Yet, the possibility that the scarab originated in Naucratis cannot be completely ruled out, as the black glass material used to glaze the scarab could have been imported from the coastal region of Syria.

In terms of provenance, only the results for the Corte Margarida scarab were somewhat conclusive. Its origin in an eastern Mediterranean region seems very likely. Although, it cannot be excluded that all the other artefacts, including glass, ostrich eggshell and ivory beads, have a similar provenance, *i.e.*, at least a Mediterranean trade origin. This is because they are certainly the result of the Phoenician or Phoenician-Punic trade that existed in those protohistoric times. Moreover, as mentioned above in the sections on the sites of Monte da Ramada 1, Palhais and Almaraz, archaeometric analyses carried out on metallic sets from the same contexts as the necklace beads and scarabs have allowed us to verify that these metallic assemblages have diversified bronze alloys with low tin and high iron content, very different from the copper-based metallurgy of the LBA, which is the oldest testimony to the presence of Orientalising copper-based metallurgy in southern Portugal. Actually, the tin and iron contents of the EIA artefacts clearly distinguish the indigenous metallurgy of the southern Portuguese territory in the tradition of the LBA from the Orientalising metallurgy. The reduction of tin content in bronze alloys is generalised to other Iberian areas with significant Phoenician influence, namely Andalusia, Levante and the southern Meseta. In fact, these unusual bronze alloys, associated with glass and faience beads, scarabs and also ostrich eggshell beads, reveal an ancient Mediterranean trade network since the 10th century BC, attested by the grave goods of the hypogea of Monte da Ramada 1, and extending throughout the EIA.

ACKNOWLEDGEMENTS

The authors would like to express their gratitude to the two anonymous reviewers whose comments and suggestions contributed to improve this paper.

DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

FUNDING SOURCES

The work was financed by national funds from FCT – Fundação para a Ciência e a Tecnologia, I.P., in the scope of the C²TN Project UIDB/04349/2020.

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