Mediterranean Archaeology and Archaeometry, Vol. 18, No 1, (2018), pp. 237-253

DOI: 10.5281/zenodo.1165362

ARCHAEOMETRIC CHARACTERIZATION OF THE CERAMICS FROM TWO CELTIBERIAN HILLFORTS: PRELIMINARY RESULTS

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Received: 05/07/2017
Accepted: 18/02/2018
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ABSTRACT

In this work, we present the preliminary results of the archaeometric analysis of several ceramic and clay samples from two Celtiberian hillforts of the Iron Age from the Spanish Central Plateau: El Ceremeño and its cemetery (Early and Middle Iron Age) and the oppidum of Los Rodiles (Late Iron Age) including La Rodriga, a potter's workshop contemporary to Los Rodiles. Clay samples were collected from all sites in order to carry out a provenance analysis and to determine if the ceramic production of the proposed archaeological sites was local or foreign. Mineralogical analysis was performed by Thin-Layer Petrography (TLP) and X-Ray Diffraction (XRD), whereas chemical analysis was done by X-Ray Fluorescence: Semi-quantitative (XRF) and Trace Elemental analysis (XRF-t). Moreover, to complete the study a thermal analysis was carried out by a dilatometer (DLT). Although the number of samples evaluated was limited, in all the studied cases, the analyzed pottery was clearly found to be made with the clay from the surroundings of the archaeological sites.

KEYWORDS: Celtiberian Culture, Iron Age, ceramic production, provenance analysis

1. INTRODUCTION

Archaeometric analysis of ceramics has benefited from the development and application of chemical, structural and micro-structural techniques, exceeding the capabilities of the morphological description of the artefacts. Despite the interest in these techniques, the Spanish archaeological community did not employ archaeometric analysis until mid-nineties, when the I Iberian Congress of Archaeometry (Capel, 1999) was organized. More recently, the number of researchers in this field has significantly increased, especially the based in ceramics (Montero et al. 2007; Cordero et al. 2006; García Heras, 2003a, etc.).

In several areas of the inner Iberian Peninsula, where historical Celtiberia was located, García Heras started to carry out structural analysis of ceramics that were considered ground-breaking. García Heras characterized ceramics from the sites of *Numantia* (1999a; 1999b and 2003), *Segontia Lanka* (2003b), Castilterreño (1994 and 2003b) and El Palomar de Aragoncillo (González et al. 1999). Moreover, he analysed several other materials from these sites, including glass (García Heras et al. 2003; García Heras, 2008, etc.). Following these initial studies, few projects have investigated the structural properties of ceramics in this region, including the Celtiberian potter's workshop (Igea et al. 2008 and Saiz et al. 2010) and some archaeological sites of the Celtiberia from the Spanish Central Plateau such as La Coronilla, La Yunta, El Torrejón, etc. (Sánchez Climent, 2015). The interesting findings of these studies motivated us to continue on this research line.

Recently published previous studies have applied characterization technologies to prehistoric pottery with the aim to reinterpret the significance of the ceramic provenance in the Neolithic from the Mondego Plateau in Portugal by Jorge et al. (2013), and the case of the Neolithic ceramics from the Central Plateau of Iran (Marghussian et al. 2017). In the last publication the same techniques were used to stablish a gradual evolution of the pottery from the Sialk I to Sialk II periods due to relative similarity of compositions and homogenous structures, and also the presence of high-temperature phases demonstrated a high specialization in the fabrication of the ceramics. Relevant work that is usual practice in archaeometry from earlier times to later antiquity is the recent ones from the central and other side of Mediterranean (Zeinab Javanshah, 2018; Nagwa. S. Abdel Rahim, 2016).

In the case of the Iron Age, the work from colleagues of the University of Salamanca is very interesting. They studied the Second Iron Age ceramic from the north-western of the Iberian Peninsula (Reyes De Soto et al. 2014), whose results showed differences between local and foreign ceramics related to the origin of the raw materials. The work of Krueger and Brand herm (2016) about archaeometry and chronology of the Early Iron Age pottery from the south-western Iberia is also of interest. In other parts of Europe, it is important to highlight the archaeometric characterization of the ceramics from Oropos (Mazarakis & Vlachou 2014). In this work, the authors carried out archaeometric analysis to several ceramic samples from the archaeological site to determine the local or the Euobean production.

The aim of this work was to carry out a provenance analysis of ceramics from two Celtiberian archaeological hillforts: El Ceremeño (and some ceramic samples from its cemetery) and Los Rodiles. Furthermore, we have collected some clay samples from the surroundings of the hillforts and from La Rodriga potter's workshop, an archaeological site with the same chronology to Los Rodiles. In order to perform this study, common analysis techniques have been used such as X-Ray Diffraction and Thin-Layer Petrography for mineralogical analysis, and X-Ray Fluorescence (Semi-quantitative and Traces) for the chemical study. To complete this archaeometric characterization, we used a dilatometer to determine firing temperatures.

2. METHODOLOGY: SAMPLES AND TECHNIQUES

The archaeometric studies presented in this work were applied to 17 ceramic samples from three Celtiberian archaeological sites of Guadalajara (Spain) (Fig. 1): El Ceremeño hillfort, its cemetery in Herrería, and Los Rodiles oppidum. Furthermore, some clay samples were collected from the surroundings of the mentioned sites and from La Rodriga potter's workshop (Fuentelsaz) that is contemporary to Los Rodiles site. Mineralogical, thermal and chemical analysis were carried out using the most popular techniques for the ceramic characterization. Because the techniques used in this work are destructive, it was not possible to analyse every sample by all listed methods. Table 1 shows the techniques used for each sample, together with the chronology, the archaeological site and the description of the sample.

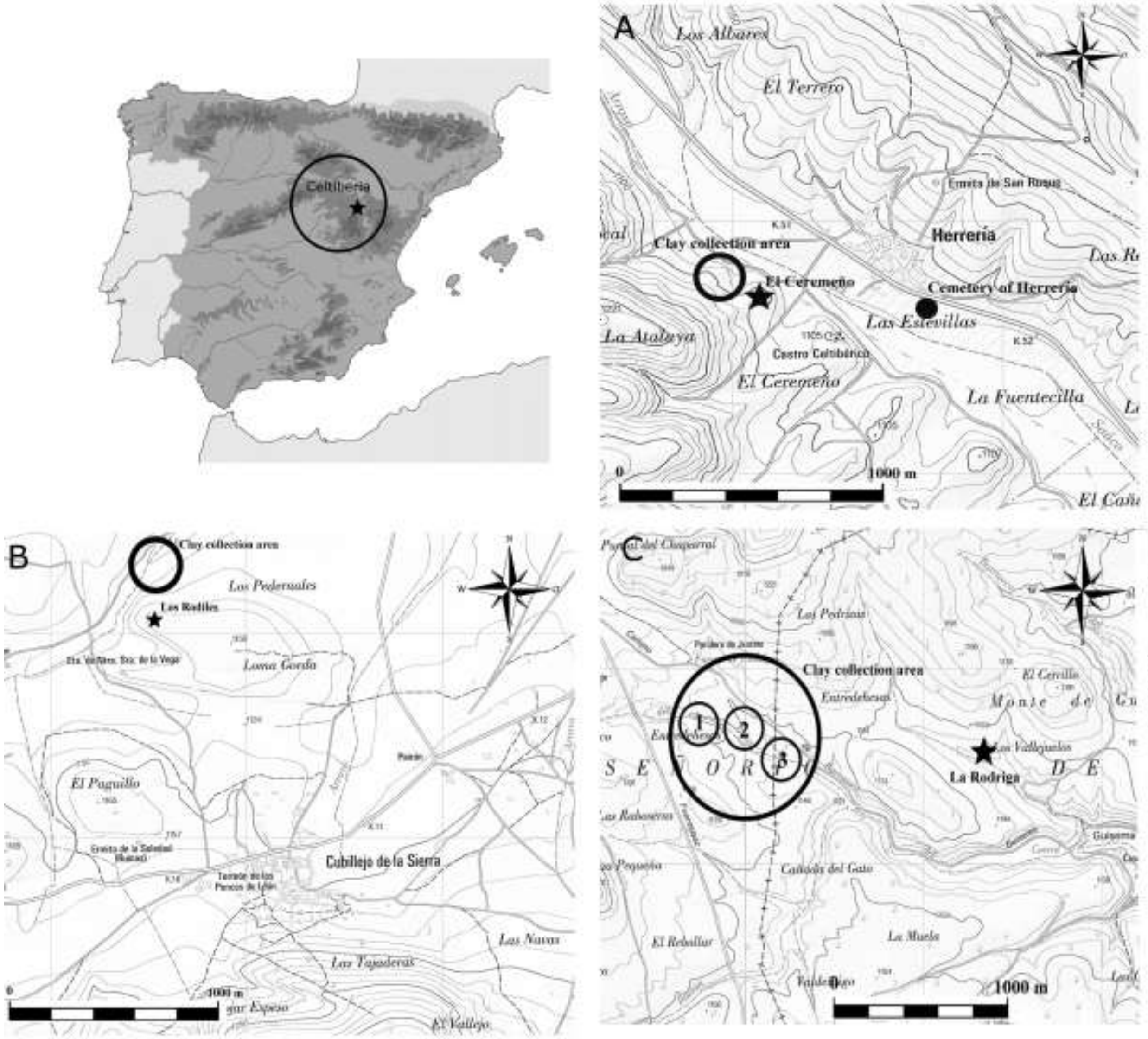

Figure 1. Location of the archaeological sites and clay collection areas in Celtiberia (Spain): (A) El Ceremeño and its cemetery Herrería, (B) Los Rodiles and (C) La Rodriga potter's workshop.

Table 1: Summary of the characteristics of the analysed samples and techniques used.

Id	Sample	Site	Chronology	Description	Technique*
1	CEI-VivA-1	El Ceremeño I	VII-VI BC	Handmade	XRD, XRF, XRF-t and DLT
2	CEI-VivA-2	El Ceremeño I	VII-VI BC	Handmade	XRD
3	CE92-I-VivB-UE9-sector 1/2	El Ceremeño I	VII-VI BC	Wheel	XRD, XRF and XRF-t
4	CE92-II-UE4-VivIII	El Ceremeño II	V BC	Wheel	XRD, XRF and XRF-t
5	CE92-UE28-VivIII (1)	El Ceremeño II	V BC	Wheel	XRD, XRF and XRF-t
6	CE92-UE28-VivIII (2)	El Ceremeño II	V BC	Wheel	XRD, XRF and XRF-t
7	NMO05-15b-N1-P3	Herrería III	VII-VI BC	Handmade	XRD and DLT
8	HRRIII-T353A	Herrería III	VII-VI BC	Handmade	XRD
9	NMO02-24sup-1632	Herrería IV	V-IV BC	Handmade	XRD, XFR, XFR-t and DLT
10	RO09-31e-4014	Los Rodiles I	III-½II BC	Handmade	TLP
11	RO09-31e-4023	Los Rodiles I	III-½II BC	Wheel	TLP

12	RO09-31f-4014	Los Rodiles I	III-½II BC	Wheel	XRD and DLT
13	RO09-27G-1205	Los Rodiles II	½II-I BC	Wheel	TLP
14	RO09-27G-1206	Los Rodiles II	½II-I BC	Wheel	TLP
15	RO09-31f-4020	Los Rodiles II	½II-I BC	Roman	TLP
16	RO09-3F-2002-1	Los Rodiles II	½II-I BC	Wheel	XRD, XRF, XRF-t and DLT
17	RO09-3F-2002-2	Los Rodiles II	½II-I BC	Wheel	XRD, XRF, XRF-t and DLT
18	El Ceremeño clay sample	El Ceremeño	---	Red	XRD, XRF, XRF-t
19	Los Rodiles clay sample	Los Rodiles	---	Red-brown	XRD, XRF, XRF-t
20	La Rodriga clay sample (x3)	La Rodriga	---	Grey	XRD, XRF, XRF-t

*XRD: X-Ray Diffraction; XRF: X-Ray Fluorescence (semi-quantitative); XRF-t: X-Ray Fluorescence (trace elements); DLT: Dilatometry; TLP: Thin-Layer Petrography.

2.1. *Mineralogical analysis*

Two techniques were used for the mineralogical analysis: X-Ray Diffraction (XRD) and Thin-Layer Petrography (TLP).

The mineralogical characterization by XRD consisted of the analysis of crystalline particles from the diffraction of the X-Ray according to Bragg's Law. It was carried out in IRICA by using a diffractometer (Philips X'Pert MPD) in which the angular range was set between 3 and 75º with increases of 0.05º. The sample was introduced in the instrument after being ground with an agate mortar for 10 minutes. The final diameter of the sample particles ranged between 50 and 100 μm. For the provenance analysis, all the clay samples were measured in loose clay format. The quantitative mineralogical composition of the samples analysed by XRD is shown in Table S1. This analysis was carried out using a reflectance powder method. The reflective factors have been taken from some specialist authors such as Schultz (1964) and Barahona (1974).

The analysis by TLP was performed in the HSC (Human Science Complex) of the University of Toronto. The samples were cut using pliers, placed in sample cups, and heated up to 50 ºC in a drying oven until the samples were completely dry. Afterwards, an EpoFix epoxy was added to the cup and, immediately, the cup was placed again in the drying oven for 2 minutes. The samples were transferred to a vacuum chamber and the air inside the epoxy was removed through several vacuum cycles at 800 mbar. The samples were then heated in the oven at 35 ºC for 24 hours and removed from the sample cups. A razor blade was used to cut the sample with the epoxy and, after optimization, it was placed between two glass slides using UV curing glue (Loctite 358 Adhesive IDH No.135414). A polarizing transmitted light microscope (Nikon Photolab 2 POL) was used to analyse the sample. In Fig. S1 some examples of the observations with the microscope are presented.

2.2. *Thermal analysis*

As ceramics are being manufactured, the clay undergoes some changes in its structure during the firing process. For example, there is a dilatation process of the mineral particles. If the changes in the dilatation are measured, it is possible to know the temperature at which the ceramics was baked. In this work, a dilatometer (Misura ODHT 1400 50) located in the AITEMIN Technological Centre (Toledo, Spain) was used. This equipment measures the length variation as a function of temperature for a ceramic sample. The only preparation of the sample required was to have it cut in a cylindrical shape to fit in the instrument. Diagrams like the ones shown in Fig. S2 were obtained with this technique.

2.3. *Chemical analysis*

An X-Ray Fluorescence spectrometer (Philips MagiX PRO) located in IRICA (Instituto Regional de Investigación Científica Aplicada, University of Castilla-La Mancha, Spain) was used to analyse the chemical composition of the ceramic and the clay samples. A pre-treatment of the sample was carried out before the analysis with the spectrometer. First, it was ground to a diameter less than 53 μm (36 μm for the trace analysis). Then, 2 ml of a solution of n-butyl methacrylate in acetone (5%) were added to 8 g of the ground sample and they were well mixed then left to dry until the solvent was evaporated. Boric acid was added to the sample and a tablet 4-mm width was made with the mixture using a 200kN press during 30 s. This tablet was finally introduced in the instrument. Two kind of measurements were done: semi-quantitative (XRF) and trace analysis (XRF-t). The first made possible to detect the oxides of all the possible elements, giving a result in percentage (%). The second analysis only detected 27 programmed elements and the results were given in parts per million (ppm).

A basic statistical analysis was performed to compare the chemical composition of the different samples. The elemental information obtained by XRF

(Table S2) and XRF-t (Table S3) for each ceramic sample was divided by the same elemental information for the clay. The closer the result of the ratio is to 1, the more similar the composition of that element is for the two samples. Finally, for each ceramic sample, an average was done considering the results of the division carried out for each element. Again, if the ratio is close to 1, the ceramic sample will share more chemical features with the clay, and there will be higher probability that the ceramics were made using that clay.

3. RESULTS AND DISCUSSION

3.1. El Ceremeño and its cemetery

El Ceremeño (Herrería, Guadalajara) is a small hillfort located on a hilltop near to Saúco River (Fig. 1-A). Two very well differenced occupational levels have been documented: Ceremeño I (Early Iron Age, 7th-6th centuries cal. BC) and Ceremeño II (Middle Iron Age, 5th century cal. BC). Due to the obtained archaeological information, this archaeological site is considered to be one of the most significant Celtiberian sites in the recent years (Fig. 2), becoming a representative site of the Celtic Hispania in the recently opened remodelled Spanish National Archaeological Museum. From the point of view of the ceramic materials, it is important to emphasize that the majority of the ceramics from the first occupational level (Ceremeño I) were handmade. Ceramics produced using a wheel were in minority proportion, considered to be imported ceramic from the Eastern Iberian culture (Cerdeño and Juez, 2002: 77-78). On the other hand, in the second occupational level (Ceremeño II), the amount of handmade ceramic decreased whereas wheel ceramic became the principal ceramic production in the hillfort.

The land surrounding the site is very clayey, specifically ferruginous, with clays of good quality and suitable for ceramic production. Six ceramic samples were collected from the site (2 handmade and 4 wheel) from the two occupational levels (Table 1). In addition to this, a clay sample (sample 18) was collected from the surroundings of the hillfort to determine the mineralogical and chemical similarities to the selected ceramics. The appearance of the clay was reddish, very rich in iron oxides and composed by small and hard blocks. Furthermore, some ceramic samples were gathered from the Herrería cemetery, very close to El Ceremeño, because its occupational levels III and IV were contemporary to the two levels of the hillfort. The main goal of the analysis was to determine if the origin of the ceramics was local or foreign for El Ceremeño and its cemetery.

Figure 2. Aerial picture of El Ceremeño hillfort.

When the samples were mineralogically analysed by XRD (Fig. 3-A), it was found that all of them contained illite. This mineral is a phyllosilicate (or laminar), very similar to muscovite, and it was found to be abundant in all samples, except in sample 5, in which there were only traces of this material. It is interesting to point out that, in the diffractogram, the peak that corresponded to illite was shifted in samples 5 and 6 of Ceremeño II and sample 3 of Ceremeño I. This is related with the granulometry, i.e. the grind made to the clay in the ceramic production. In this particular case, the shift indicates that it was much depurated, in agreement with the clay used in wheel ceramics.

The most remarkable feature of the clay sample (sample 18) is the presence of kaolinite. This is a two-layer phyllosilicate clay mineral that was not found in any ceramic sample. This might question if the ceramics had the same mineralogical origin than the clay. However, kaolinite disappears at temperatures higher than 550ºC which is its thermal deshydroxylation point (the dilatometry shows the firing temperature in all of the ceramic samples are over 700ºC). For that reason, according to the XRD, the ceramics reached this thermal deshydroxylation point of the kaolinite during the firing process, what would explain the absence of this mineral in the ceramic samples despite its presence in the clay. However, the melting point of illite is circ. 900ºC (the thermal deshydroxylation point of the illite is at 700ºC, but the mineral maintains the crystalline structure), so its presence in El Cerameño ceramics indicate that the firing process was carried out between 550 and 900ºC. This is supported by the dilatogram of sample 1 (Fig. S2), in which the measured firing temperature was between 850 and 875ºC.

The samples from the cemetery of Herrería were also analysed (Table 1). The diffractogram (Fig. 3-B) showed again that the ceramics and the clay were very similar. It must be pointed out that samples 8 and 9, in contrast with the clay, did not present illite, because during the firing process the temperature was higher than 900ºC. On the other hand, in the diffractogram of sample 7 there was a clear peak corresponding to that mineral. As can be seen in the dilatometry results (see Fig. S2), it is confirmed that firing temperature for sample 9 was between 1000 and 1050ºC, whereas for sample 7, it was between 700 and 750ºC, explaining the presence of illite in this sample. Sample 7 also contained calcite and dolomite, which are two minerals that were not found in the rest of the ceramic samples. There are two hypotheses that can explain this: either the ceramic was not produced from the analysed clay (sample 18), or these minerals were secondary depositions, considering that clay and ceramic are mineralogically similar enough.

From the mineralogical point of view, all the samples are similar as confirmed by the diffractograms. This similarity between the samples might indicate that the ceramics were made with clay from the surroundings of the hillfort, and that El Cerameño was a local ceramics producer. However, since quartz, feldspar, and phyllosilicates are very abundant in nature, they do not fully confirm that the ceramics were made with that clay. To confirm this, it was necessary to perform the chemical analysis by XRF and XRF-t proposed in this work.

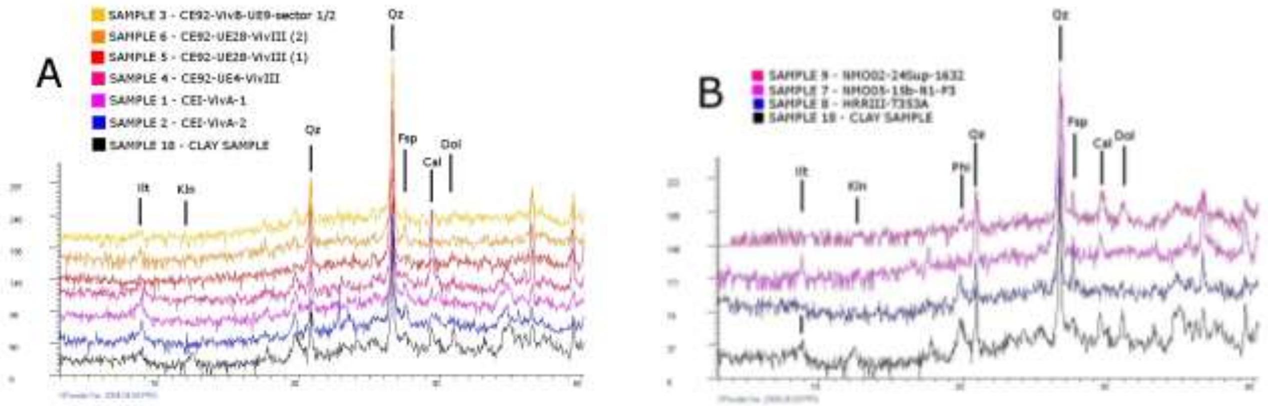

Figure 3. XRD diffractograms showing the comparison between the clay and the ceramics from (A) El Cerameño and (B) the cemetery of Herrería.

Chemical Analysis by XRF and XRF-t was carried out to evaluate the similarities between the selected samples. From pre-stablished chemical elements detected by the instrument, it was possible to know the chemical composition of the samples. The XRD study showed several similarities between ceramics and clay from the mineralogical point of view. In this section, the chemical analysis can confirm these aspects.

It is very important to take into account that the chemical composition in nature is not homogenous, i.e. the percentage of each element found in all the samples will not be exactly the same. Ceramics can suffer some alterations in the composition during the fabrication process: when tempers are added, due to washing and firing processes, and even if different clays are mixed. Clay can also suffer contamination (due to farming), runoff, etc. that could modify its chemical composition. Therefore, the accuracy when comparing ceramics and clay is not so important. We only have to take into account what elements were similar in amount and in which proportion they were close.

Trace elements analysis

The results obtained by this technique following the statistical analysis (see Methodology section) are shown in Table 2. The only element with a ratio significantly greater than 1 was cesium, Cs, with a ratio greater than 2 in samples 3 and 6, and greater than 3 in samples 1 and 9. This indicated that the ceramics contained more Cs than the clay, but this does not necessarily mean that the clay was not used to make those ceramics, as it is discussed below. All the ceramic samples showed similar proportions of Cs (see Table S3), suggesting that they shared the same origin and fabrication process.

For other chemical elements, the calculated ratios were close to 1, except for some elements, such as, barium, (Ba), lead, (Pb), thorium, (Th), tungsten, (W), and cerium, (Ce). Sample 9, the cemetery ceramic sample, was the most chemically different sample. In this particular case, there was a high ratio, over 3, of Ce and neodymium, Nd. Only the average of ratios could indicate the degree of similarity between the ceramics and the clay. As we can observe in Table 2, the determined averages were very similar between all the ceramics. The average in all the hillfort samples was between 1.2 and 1.3, except for sample 9 (Herrería IV) that was 1.6, being slightly different.

In summary, the resulting averages obtained by XRF-t indicated that the ceramics of El Ceremeño and Herrería have high possibilities of coming from El Ceremeño clay (or some other similar clay from the surroundings). As we have seen, the only sample that deviated from the expected ratio of 1 was the ceramic from the cemetery. However, its average (1.6) is not significantly different from the other hillfort samples to consider a foreign production, which is the reason why, although numerically it has a higher ratio, it could also be considered as a local ceramic.

Table 2. Comparison ratios between the results obtained by XRF-t for the ceramics from El Ceremeño and Herrería and the clay from the hillforts surroundings.

Sample	1	3	4	5	6	9
Sc	0.97	1.30	1.02	1.02	0.92	1.42
V	1.02	0.80	0.93	0.77	0.76	0.84
Cr	1.10	0.92	1.15	0.84	0.68	0.80
Co	1.31	0.66	1.13	0.76	0.12	0.94
Ni	1.03	0.39	1.17	0.45	0.17	0.66
Cu	1.81	0.73	1.65	0.71	0.51	0.73
Zn	1.61	0.63	1.88	0.80	0.54	0.69
Ga	1.22	1.73	0.99	1.39	1.50	1.48
As	1.40	0.70	0.94	0.63	0.57	0.47
Rb	1.38	1.32	1.16	1.06	1.35	1.43
Sr	0.59	0.19	0.24	0.24	0.18	0.11
Y	1.13	1.06	1.20	1.82	1	2.54
Zr	0.87	1.51	1.12	1.87	1.79	1.29
Nb	1.06	1.50	1.06	1.52	1.33	1.39
Mo	0.90	---	0.81	0.45	---	---
Sn	---	---	---	---	---	---
Cs	3.04	2.51	1.64	1.90	2.45	3.41
Ba	1.85	1.04	2.15	0.83	0.81	0.53
La	1.17	1.15	0.98	1.46	2.02	2.60
Ce	0.98	0.94	0.90	1.42	1.63	3.10
Hf	1	1.86	1.37	2.11	2.48	1.57
Ta	---	---	---	---	---	---
W	1.16	2.20	1.04	2.64	2.48	3.16
Pb	2.55	2.52	2.16	2.55	4.14	2.38
Th	1.18	2.13	1.57	2.18	2.56	2.11
U	1.75	1.66	1.95	1.70	1.91	2.20
Nd	0.97	0.70	0.88	1.67	1	3.76
AVERAGE	**1.32**	**1.26**	**1.24**	**1.31**	**1.37**	**1.65**

Semi-quantitative analysis

The results of the XRF Semi-quantitative showed that, almost all the ceramic samples had a similar ratio between the concentration in the ceramic and the clay, ranging between 0.1 and 3.0 (Table 3). It is very significant that for Na_2O in sample 1 and CaO in sample 4, the ratio was higher than 2.0, which is far from our ideal ratio of 1. Surprisingly, CaO in the rest of samples showed a ratio much lower than 1. It is also noteworthy that MgO and SO_2 showed ratios always below 0.6 in all the samples.

Like in the Trace Elements analysis, the most interesting result is the average of the ratios for the different elements, which ultimately indicates if the ceramics and the clay were similar or not. Based on the Trace Elements results, sample 4 (with a ratio of 0.98) was the most likely ceramic to be made from the analysed clay. The other ceramics varied slightly from our ideal ratio of 1, with low probabilities of coming from the clay. Samples 3, 5, 6 and 9 presented ratios higher than 0.7, but sample 1, the handmade ceramic, showed an average of 1.23. It is interesting that this value was very similar to that obtained by XRF-t, which suggests that its origin was from the analysed clay or from the surrounding area.

Table 3. Comparison ratios between the results obtained by XRF for the ceramics from El Ceremeño and Herrería and the clay from the hillforts surroundings.

Sample	1	3	4	5	6	9
Na_2O	2.96	1.25	0.65	0.79	1.44	1.17
MgO	0.59	0.17	0.42	0.16	0.17	0.17
Al_2O_3	0.81	1.51	1.05	1.39	1.64	1.57
SiO_2	1.03	1.09	0.92	1.29	1.27	1.23
P_2O_5	2.49	0.48	1.43	0.66	0.62	0.59
SO_3	0.43	0.13	0.46	0.21	0.57	0.14
K_2O	1.18	0.54	0.69	0.49	0.60	0.70
CaO	0.64	0.19	2.39	0.48	0.16	0.16
TiO_2	0.98	1.15	1.04	1.41	1.04	1.28
Fe_2O_3	1.13	0.58	0.70	0.64	0.44	0.68
AVERAGE	**1.23**	**0.71**	**0.98**	**0.75**	**0.80**	**0.78**

We can conclude from the chemical analysis of the ceramics and the clay from El Ceremeño and its cemetery that all the ceramics were locally produced, since the average ratios are close to our ideal value of 1. The mineralogical composition was also very similar when the ceramics and the clay were compared, which corroborated the hypothesis that they had a local origin.

Previous studies discussed the presence of the wheel ceramic in the first stages of the Iron Age in the Spanish Central Plateau, because this type of ceramic was considered a very novel artefact that could indicate that there were contacts between Celtiberi and the Iberian people from the Levant (Cerdeño y Juez, 2002: 77-78). Therefore, until now, the wheel ceramic was considered to be imported, whereas the handmade ceramics were thought to be locally produced. Since we observed a great similarity in the composition between the handmade and the wheel made ceramics from El Ceremeño I, we can conclude that all these ceramics were produced using the clay from the surroundings of El Ceremeño, either in the hillfort, or in an unknown potter's workshop close to the hillfort. This hypothesis is confirmed when the ceramics from El Ceremeño I and El Ceremeño II were compared and found to be similar, since the latter had always been considered to be locally made. The same conclusion can be deduced with the ceramics from the cemetery. Since these samples were mineralogically similar, we believe that they could have been produced using clay from the surroundings.

The present study showed, for the first time, that the ceramics from El Ceremeño I were made with local clay. This analysis would indicate that the potter's wheel arrived to the Spanish Central Plateau between the 7th and 6th centuries BC. Although in this work it was possible to establish some hypothesis about the origin of the production of the ceramics, it has to be pointed out that the number of samples used in this work was limited, so more analysis should be done in the future with a greater number of samples of ceramics and clays. This work can be considered as the key for further studies related to this hypothesis.

3.2. Los Rodiles

The Celtiberian Los Rodiles *oppidum* is a good example of the final stages of the Celtiberian culture (Fig. 4) at Late Iron Age. Similarly to the previous study, the ceramic characterization was focused on the mineralogical and chemical analysis of some ceramic samples from the two occupational levels (see Table 1): Rodiles I (3rd-½2nd centuries cal. BC) and Rodiles II (½2nd-1st centuries cal. BC). Due to the size of this archaeological site, we aimed to determine if the *oppidum* was a ceramic production centre. For this reason, we collected a clay sample from the riverbank near the site (see Fig. 1-B). The appearance of this clay was very similar to the clay from El Ceremeño, with a reddish colour typical of ferruginous clays.

Clay from La Rodriga potter's workshop (Fuentelsaz, Guadalajara) was also collected since it is located 15 km from the site and it is dated between 3rd-2nd centuries BC (Arenas, 1991-92: 225), similar chronology to Los Rodiles archaeological site. This clay had a very good quality and it was ideal for the ceramic production. The clay was sedimentary, originated in the Jurassic and Cretaceous, and composed by dolomites, loams and limestone. In fact, it was completely different from Los Rodiles clay. The clay from La Rodriga was grey coloured since it contained more calcium carbonate. Clay was collected from three different points in a location known as Fuente de Rodriga (Fig. 1-C), that is 800 m from the potter's workshop. Some ceramics were found in this site, and they were studied from a mineralogical and chemical point of view (Igea et al. 2008). These authors reported great compositional similarities between the samples, showing that it was possible that they share the same origin. In the present work, we compared the ceramics of Los Rodiles with the clays collected near the archaeological site and in the potter's workshop to determine if there was any correspondence between the samples.

Figure 4. Aerial picture of Los Rodiles.

The results obtained in the mineralogical analysis by TLP and XRD were very interesting. The ceramic samples from Los Rodiles presented mineralogical similarities with the two clays analysed: from the site and from the potter's workshop. All of them contained minerals typically found in nature, such as quartz and muscovite.

Unlike XRD, using TLP is possible to observe non crystalline elements or the porosity of the ceramic paste. In that sense, it is interesting to highlight that sample 10 from Rodiles I was found to contain, other abundant minerals such as quartz and phyllosilicates, grog and basalt as tempers, and that the ceramic had great porosity. The presence of porosity and tempers may be have been intentional because a very porous ceramic creates a humid environment that is ideal for storage. In the case of sample 10, since it was handmade ceramic, it was probably used for cooking purposes. This was corroborated by the fact that it contained abundant tempers that reduced the thermal shock when the ceramic was placed in the kiln.

The rest of samples, which were wheel made, were analysed either by TLP (see Fig. S1), or XRD (see Table S1). The results obtained by both techniques showed negligible differences in the mineral composition. All these samples presented high levels of quartz, calcites (micrites), feldspars and silicates (muscovite and illite). In particular illite was found to be very abundant in all the samples. It is worth noting that in samples 12 and 16, the peak corresponding to illite in XRD was shifted when compared to the other samples (Fig. 3). The presence of this particular mineral indicated that the firing temperature was always below 950ºC. This was confirmed by the dilatometry assays on samples 12, 16 and 17 that showed a temperature range between 800 and 900ºC (see Fig. S2).

When the ceramics from Los Rodiles were compared with the clay from the surroundings of the site (Fig. 4-A), no kaolinite was observed in any sample. There are two hypothesis that can explain this: (1) the clay used to produce these ceramics was from Los Rodiles surroundings, or (2) the ceramics contained kaolinite initially but it disappeared when the temperature in the kiln reached the melting point of this mineral (550 ºC), a condition confirmed by dilatometry.

The ceramics were also compared to the three samples of clay from La Rodriga (see Fig. 4-B). In the diffractogram, a good similarity was observed between this clay and the clay from Los Rodiles: both presented high levels of quartz and feldspar. In La Rodriga clay samples, the major component was calcite and dolomite, although sample 20-2, which was collected at the riverbank, presented more carbonate than the other samples. Lower amounts of illite and phyllosilicate were observed in these clay samples than in the ceramics. Sample 20-1 presented kaolinite, which was negligible in the other clay samples.

From the mineralogical point of view, the ceramics presented significant similarities to Los Rodiles clay and the clay from the potter's workshop. However, this was not enough evidence to confidently state that the ceramics were locally produced, so we decided to conduct the chemical analysis to confirm the origin of these ceramics and to distinguish the provenance of the clay in the ceramic production.

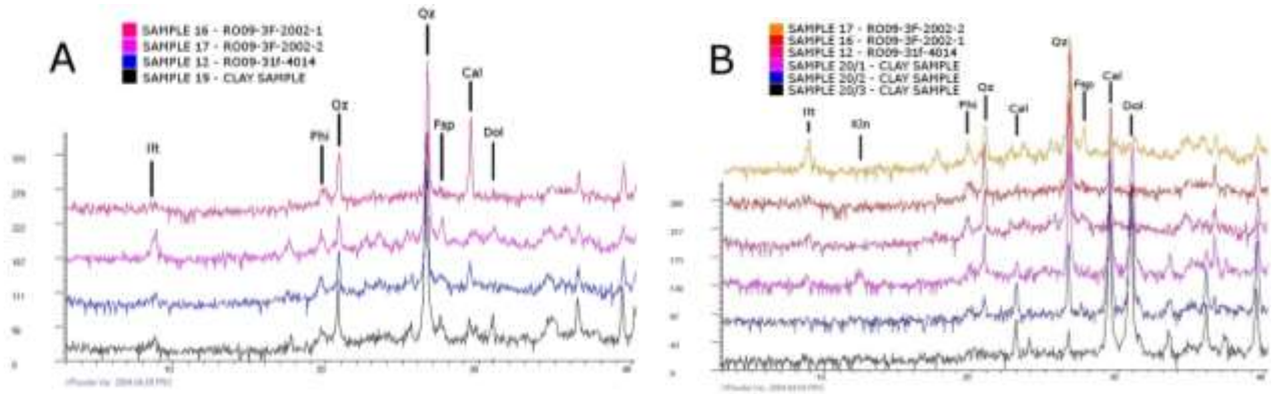

Figure 4. XRD diffractograms showing the comparison between the ceramics from Los Rodiles and the clay from (A) the hillfort surroundings and (B) La Rodriga potter's workshop.

In the chemical study of Los Rodiles samples, we applied the same statistical method used for El Cеremeño (see methodology): the ratio between the elemental results for the ceramics and the results for the different clays was determined, an average was reported, and then used to determine the degree of similarity between the samples.

Trace Elements

The ratios obtained when the ceramics were compared with the four available clays are presented in Table 4. In this table, a better agreement (i.e. the ratios are closer to 1) is observed when the ceramics are compared to Los Rodiles clay (sample 19). In this case for most of the elements, the ratio was around 1, except for rubidium, (Rb), Cs, zirconium, (Zr), W, Pb and Th, for which the ratio was higher than 2 in sample 16, and between 1.2 and 1.6 in sample 17. Gallium, (Ga), was also an exception since its ratio was approximately 2.4 in both samples.

When the ceramics were compared to La Rodriga clay, the ratio of each element was always further from 1, indicating less similarity between the samples. Better agreement was found when the samples were compared to clay 20-1, for which the ratios were closer to 1 for sample 16. The only two elements for which the ratios were much higher than 1 were Cs and barium, (Ba).

The resulting averages showed that the ratio between the ceramics and Los Rodiles clay was close to 1: 1.6 for sample 16 and 1.7 sample 17. This indicated that these samples were similar to Los Rodiles clay. On the other hand, the ratios from La Rodriga clays were further from 1. Sample 20/2 was the most similar to the ceramics, although its ratio was much higher than the result for Los Rodiles. The two other clays from La Rodriga, samples 20/1 and 20/3, presented final averages above 3.5 and 5, respectively. In summary, the results obtained by XRF-t suggest that the ceramics have a higher probability of being locally produced than coming from La Rodriga potter's workshop.

Table 4. Comparison ratios between the results obtained by XRF-t for the ceramics from Los Rodiles and the clay from different sources (Los Rodiles surroundings and La Rodriga potter's workshop).

Sample	16	17	16	17	16	17	16	17
Clay	Clay sample 19		Clay sample 20/1		Clay sample 20/2		Clay Sample 20/3	
Sc	1.44	1.74	2.27	2.74	1.17	1.41	3.40	4.11
V	1.40	2.25	4.05	6.51	1.93	3.10	5.56	8.92
Cr	1.12	1.74	2.82	4.38	2.13	3.32	3.11	4.83
Co	1.01	0.61	2.29	1.38	0.90	0.54	1.56	0.94
Ni	1.04	1.94	2.65	4.93	1.30	2.43	3.64	6.79
Cu	1.31	3.52	2.62	7.05	1.09	5.13	1.05	2.82
Zn	1.44	1.54	1.45	1.55	1.78	1.90	2.03	2.17
Ga	2.38	2.45	5.4	5.55	2.89	2.97	7.59	7.81
As	0.88	1.72	1.88	3.68	1.43	2.80	1.67	3.26
Rb	2.33	1.20	6.46	3.34	4.05	2.09	10.26	5.31
Sr	0.81	0.63	1.14	0.88	1.64	1.27	1.05	0.82
Y	1.79	1.17	3.20	2.09	1.68	1.10	5.22	3.40
Zr	2.52	1.65	5.33	3.5	1.76	1.16	14.48	9.51
Nb	2.01	1.62	2.97	2.40	2.11	1.71	3.76	3.05
Mo	0.5	3	0.6	3.6	0.75	4.5	0.27	1.63
Sn	---	---	---	---	---	---	---	---
Cs	0.79	0.87	---	---	4.17	4.58	---	---
Ba	1.57	6.10	5.82	22.55	4.28	16.57	8.14	31.51
La	1.62	1.36	5.82	4.84	1.89	1.59	6.25	5.25
Ce	1.53	1.01	5.37	3.57	2.08	1.38	6.39	4.25
Hf	2.02	1.25	4.57	2.84	1.58	0.98	6.69	4.15
Ta	---	---	14.5	2	---	---	9.66	1.33
W	2.73	1.46	1.64	0.88	2.73	1.46	10.25	5.5
Pb	2.01	1.44	0.67	0.48	1.53	1.09	0.87	0.62
Th	2.70	2.07	6.17	4.75	2.08	1.60	---	---
U	1.60	1.25	3.21	2.50	1.8	1.4	4.5	3.5
Nd	1.22	0.71	3.44	2.01	1.74	1.02	3.24	1.89
AVERAGE	**1.59**	**1.77**	**3.85**	**4**	**2.05**	**2.68**	**5.03**	**5.14**

Semi-quantitative

The analysis by XRF corroborated the results obtained by XRF-t (see Table 5). Similar to the results of trace elements analysis, the chemical composition of the ceramics is similar to Los Rodiles clay. The ratios for most of the elements remained around 1 only for the clay sample 19.

The same ceramic samples showed a lesser degree of similarity when compared to La Rodriga clay, as few compounds had a ratio close to 1. Some examples with ratios far from the ideal value 1 are Fe_2O_3 in sample 17 compared to clay 20/1 and 20/3, with values above 4, and Al_2O_3 and SiO_2 in samples 16 and 17 compared to clay samples 20/1 and 20/3, especially in the latter clay. For clay sample 20/2, ratios are closer to 1, but still far from this ideal value.

By considering at the averages obtained (last row in Table 5), it is concluded that the ceramics were more likely made with Los Rodiles clay (average = 0.9-1.1) than the clays from La Rodriga (average = 1.5-4.4), although sample 20/2 showed some similarities.

Table 5. Comparison ratios between the results obtained by XRF for the ceramics from Los Rodiles and the clay from different sources (Los Rodiles surroundings and La Rodriga potter's workshop).

Sample	16	17	16	17	16	17	16	17
Clay	Clay sample 19		Clay sample 20/1		Clay sample 20/2		Clay Sample 20/3	
Na_2O	1.12	0.94	2.53	2.12	2.02	1.7	2.31	1.94
MgO	0.52	0.53	0.35	0.36	0.32	0.33	0.31	0.32
Al_2O_3	1.56	1.55	4.16	4.13	1.90	1.89	5.70	5.66
SiO_2	0.95	0.8	4.66	3.90	2.15	1.80	9.22	7.73
P_2O_5	0.85	2.76	0.86	2.79	1.35	4.37	2.03	6.56
SO_3	0.17	0.20	0.10	0.12	0.3	0.34	0.006	0.007
K_2O	1.09	0.62	6.16	2.96	3.89	1.87	9.87	4.74
CaO	0.47	0.62	0.05	0.06	0.07	0.10	0.04	0.06
TiO_2	1.70	1.39	3.60	2.94	1.79	1.46	8.62	7.05
Fe_2O_3	1.01	1.92	3.95	7.51	1.43	2.72	5.45	10.34
AVERAGE	**0.94**	**1.12**	**2.64**	**2.69**	**1.52**	**1.66**	**4.35**	**4.44**

The obtained results suggested that the selected ceramics were more likely made of the clay from Los Rodiles surroundings due to the similarities observed for samples 16 and 17 from both the mineralogical and the chemical analysis perspectives. Although the results by XRF for sample 20/2 that showed possible similarities with the ceramics, the XRF-t analysis confirmed that it was very unlikely that the ceramics were made using clay from the potter's workshop area. According to the obtained results, the size and the magnitude of Los Rodiles, it is possible that the *oppidum* was a ceramic production centre.

All the ceramic samples from Los Rodiles were very interesting, but we would like to highlight two of them: the handmade ceramic (sample 10) that revealed exciting technological information as reported above, and the Roman black-glazed ceramic (sample 15) that presented a mineralogical composition very similar to rest of the analysed ceramics that were Celtiberian. This may confirm that black-glazed ceramic was really a local production (showing all the same origin), being an interesting case of imitation of Roman production.

4. CONCLUSIONS

The samples analysed in this work established provisional conclusions about the origin of the ceramic production in the evaluated archaeological sites. The diffractograms made it possible to observe great mineralogical similarities between the ceramics and the clays from the surroundings. Due to the crystalline elements of the ceramics samples and clays are very common in nature, the most relevant results were obtained by XRF and XRD.

The chemical composition varied among the samples, which was useful in determining the origin of the ceramics. The chemical composition showed significant differences between La Rodriga clay and the clays from El Ceremeño and Los Rodiles (these two were found to be mineralogically and chemically very similar). These differences were clearly observed in their appearance since clays from El Ceremeño and Los Rodiles were reddish coloured because of their high level of iron oxides, whereas La Rodriga clay was grey coloured due to the presence of calcium carbonate. For the ceramics, all of them were found to be similar in composition and to have a local origin, as they were made with clays from the surroundings and distributed later to different hillforts. This fact was most evident in the case of Los Rodiles.

El Ceremeño site and its cemetery were the most interesting examples of ceramics that were closely related to the clay of the surroundings. In both occupational levels, we found a compositional correspondence between handmade and wheel made ceramics. This, together with the similarities with the clay, indicated that the potter's wheel should have arrived in the Spanish Central Plateau earlier than researchers previously suspected, i.e. in the Early Iron Age (7th-6th centuries BC).

Moreover, this work shows for the first time that ceramics were produced imitating Iberian ceramics which confirmed the exchange of craft techniques or, according to Miller, "cross-crafts" (Miller, 2007: 237). The same can be said for the black-glazed ceramic found in Los Rodiles, which showed a similar mineralogical composition than the other ceramics found in the site.

Other relevant information obtained in this work was the related to the technological purposes of the ceramics, such as the handmade ceramic of Los Rodiles (sample 10). This sample showed the intentional addition of tempers to the clay that could be related to its thermal shock resistance. This fact confirmed its use as cooking pottery.

As shown above, the results obtained in this work were interesting and they encourage us to work in this direction in the future by analysing a greater number of samples. This will make it possible to confirm the hypotheses presented in this work with greater confidence. With this study we intended to improve the characterization studies of the Celtiberian ceramics that until now were not well studied.

ACKNOWLEDGEMENTS

This work could not be possible without the collaboration of several colleagues and friends. We would like to thank Prof. Heather M.-L. Miller and Dr. Greg Braun from the Anthropology Department at University of Toronto (Canada) for their assistance during the preparation of the TLP of some ceramic samples from Los Rodiles. We want also to thank Jorge Velasco from AITEMIN Technological Centre of Toledo (Spain) for the DLT applied to several ceramic samples. Also we would like to thank Carlos Cabanillas, laboratory technician at Instituto Regional de Investigación Científica Aplicada (IRICA) of University of Castilla-La Mancha (Spain) for his assistance during the analysis by XRD and XRF. Finally, we appreciate the collaboration of María Antiñolo and Katie Badali for their assistance in the elaboration and English translation of the paper.

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SUPLEMENTARY INFORMATION

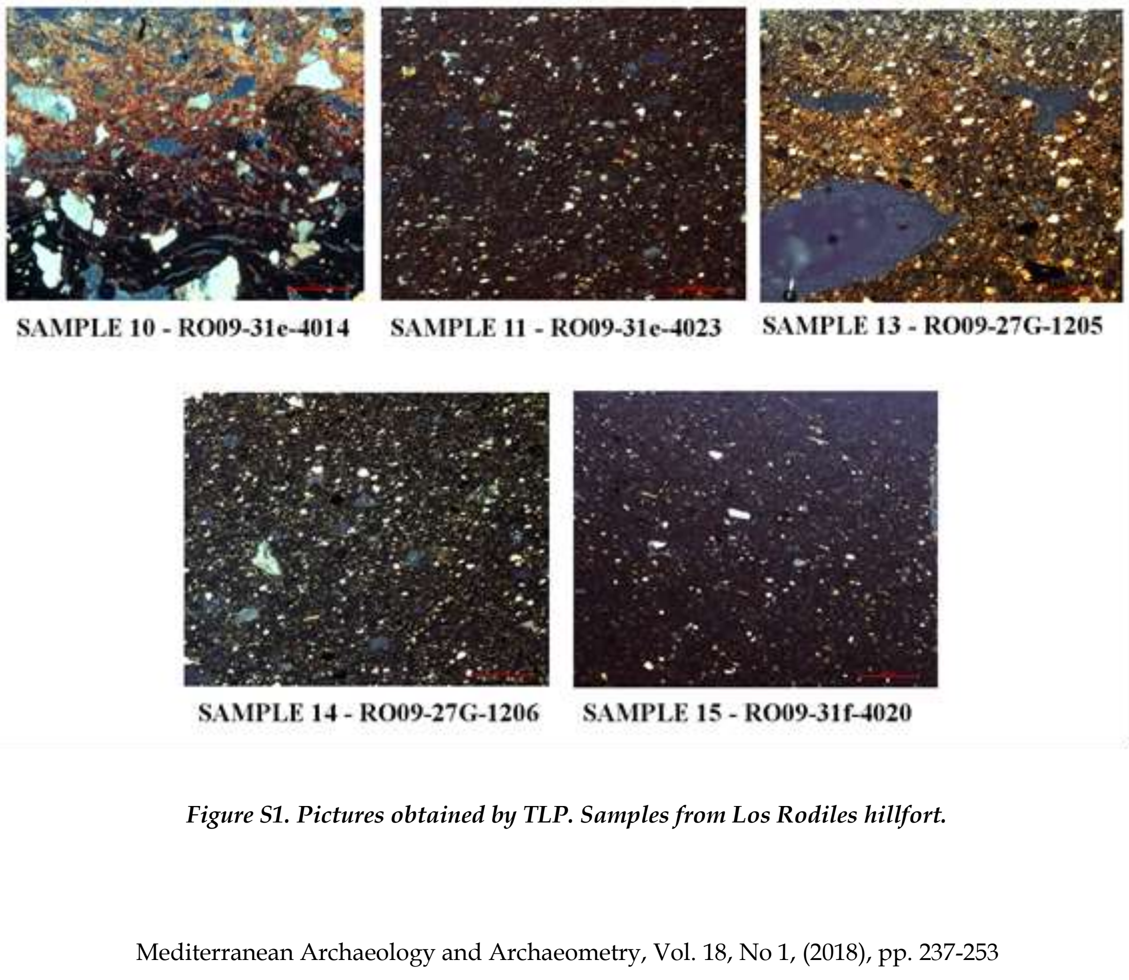

Figure S1. Pictures obtained by TLP. Samples from Los Rodiles hillfort.

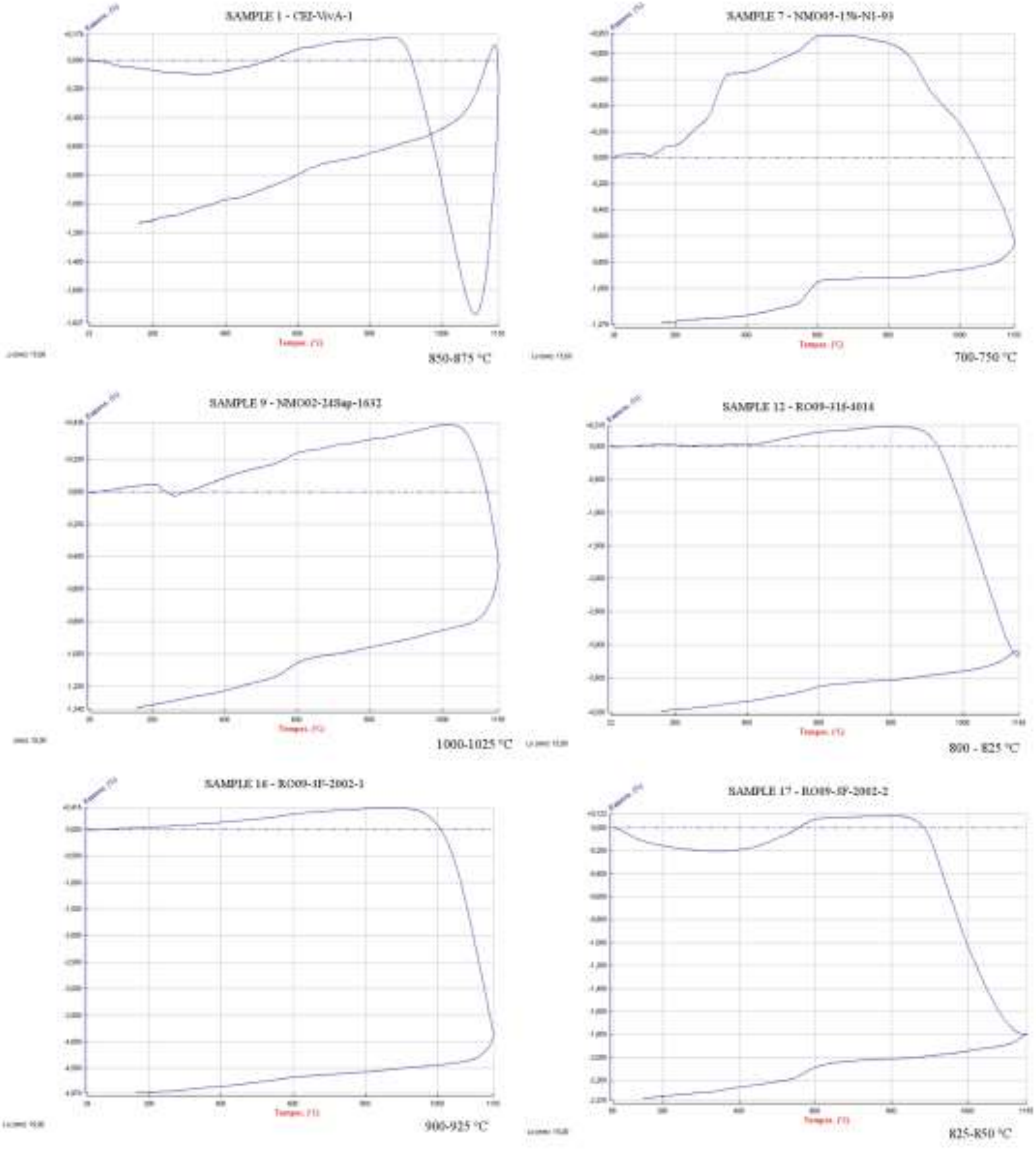

Figure S2. Pictures obtained by DLT. Samples from Los Rodiles hillfort.

Table S1. Mineralogical composition obtained by XRD (expressed in %).

Sample	Qz	Fsp	Calc	Dol	Hem	Phyllos	Ilt	Kln
1	8	7	traces	---	---	85	X	---
2	8	7	---	---	---	85	X	---
3	100	traces	---	---	---	traces	X	---
4	100	traces	---	---	---	traces	X	---
5	100	traces	---	---	---	traces	X	---
6	100	traces	---	---	---	traces	X	---
7	7	10	8	10	---	65	X	---
8	8	7	traces	---	---	80	X	---
9	75	---	25	---	---	traces	X	---
12	10	8	7	---	---	75	X	---
16	10	10	---	---	---	80	X	---
17	10	5	15	---	---	70	X	---
18	<5	<5	<5	traces	---	90	X	X
19	8	traces	15	22	traces	54	X	X
20/1	7	traces	35	50	traces	54	X	X
20/2	7	---	40	53	traces	traces	X	X
20/3	5	<5	18	17	6	50	X	X

Qz: quartz; Fsp: feldspar; Calc: calcite; Dol: dolomite; Hem: hematite; Phyllos: phyllosilicates; Ilt: illite; Kln: kaolinite.

Table S2. Chemical composition of mayor elements (expressed in % oxide).

Sample	1	3	4	5	6	9	16	17	18	19	20/1	20/2	20/3
Na_2O	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
MgO	2.8	0.83	2	0.7	0.8	0.8	1	1	4.7	2	3	3.2	3.3
Al_2O_3	13.6	25.3	17.5	23.3	27.3	26.3	21.1	21	16.6	13.5	5	11.1	3.7
SiO_2	50.7	53.6	45.2	63.1	62.1	60.4	62	52	48.8	65	13.3	28.8	6.7
P_2O_5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
SO_3	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	6.7
K_2O	5.4	2.4	3.1	2.2	2.7	3.2	4.6	2.2	4.5	4.2	0.7	1.1	0.4
CaO	2.9	0.8	10.9	2.1	0.7	0.7	1.7	2.3	4.5	3.8	34.1	22.8	39.1
TiO_2	0.8	0.9	0.9	1.2	0.9	1.1	0.8	0.7	0.8	0.5	< 0.5	< 0.5	< 0.5
Fe_2O_3	7.4	3.8	4.6	4.2	2.9	4.4	3.9	7.4	6.5	3.8	0.9	2.7	0.7
LOI	8.7	11.4	14.4	2.4	1.8	2.3	3.9	11.8	12.4	6	41.6	29.2	45.3

Table S3. Chemical composition of trace elements (expressed in ppm).

Sample	Error	1	3	4	5	6	9	16	17	18	19	20/1	20/2	20/3
Sc	± 0.00017	17.1	22.8	18	17.9	16.1	24.9	14.3	17.3	17.5	9.9	6.3	12.2	4.2
V	± 0.00086	115.6	90.9	106.2	87.9	86.6	96.1	84	134.8	113.3	59.7	20.7	43.4	15.1
Cr	± 0.00053	80.2	66.8	83.9	61.1	49.6	58.3	55.4	86	72.6	49.2	19.6	25.9	17.8
Co	± 0.00011	17.3	8.8	15	10.1	1.6	12.5	7.8	4.7	13.2	7.7	3.4	8.6	5
Ni	± 0.00164	48.4	18.6	54.5	21.1	7.9	31	17.5	32.6	46.6	16.8	6.6	13.4	4.8
Cu	± 0.00046	41.4	16.6	37.4	16.1	11.6	16.5	18.9	50.8	22.6	14.4	7.2	9.9	18
Zn	± 0.00086	87.3	34.5	101.7	43.3	29.5	37.8	42	44.8	54.1	29	28.8	23.5	20.6
Ga	± 0.00036	24.4	34.6	19.8	27.8	30	29.6	24.3	25	19.9	10.2	4.5	8.4	3.2
As	± 0.00037	28	14.1	18.7	12.7	11.5	9.5	16.2	31.7	19.9	18.4	8.6	11.3	9.7
Rb	± 0.00048	150.6	143.8	126.6	115.8	147.6	155.9	176.6	91.4	108.7	75.8	27.3	43.6	17.2
Sr	± 0.00246	314.3	100.1	127	127.4	94.6	60.7	136	105.6	525.5	166.5	119.3	82.8	128.8
Y	± 0.00021	29.5	27.6	31.2	47.2	26	65.8	28.2	18.4	25.9	15.7	8.8	16.7	5.4
Zr	± 0.00112	158.9	274	203.7	338.7	325.1	233.6	286.8	188.3	180.9	113.7	53.8	162.1	19.8
Nb	± 0.00055	18.6	26.3	18.7	26.7	23.4	24.4	21.1	17.1	17.5	10.5	7.1	10	5.6
Mo	± 0.00006	1	n.d.	0.9	0.5	n.d.	n.d.	0.3	1.8	1.1	0.6	0.5	0.4	1.1
Sn	± 0.00012	1.1	6.1	n.d.	5.3	7.5	4.6	2.6	0.4	n.d.	n.d.	n.d.	n.d.	n.d.
Cs	± 0.00049	18.9	15.6	10.2	11.8	15.2	21.2	12.1	13.3	6.2	15.2	n.d.	2.9	n.d.
Ba	± 0.00147	1043.7	587.5	1216.2	472.6	456.6	299.6	563.7	218.1	563.6	357.3	96.7	131.6	69.2
La	± 0.00127	52.7	51.8	44.2	65.9	91.3	117.3	50.7	42.6	45	31.3	8.8	26.8	8.1
Ce	± 0.00146	81.7	78.4	75.2	118.3	136.4	258.6	81.2	54	83.3	53	15.1	39	12.7
Hf	± 0.00011	4.5	8.4	6.2	9.5	11.2	7.1	8.7	5.4	4.5	4.3	1.9	5.5	1.3
Ta	± 0.00015	0.1	3.4	2.1	3.2	2.8	3.1	2.9	0.4	n.d.	n.d.	0.2	n.d.	0.3
W	±0.00037	2.9	5.5	2.6	6.6	6.2	7.9	4.1	2.2	2.5	1.5	2.5	1.5	0.4
Pb	± 1.65604	29.9	29.5	25.3	29.9	48.5	27.9	25.6	18.3	11.7	12.7	37.8	16.7	29.2
Th	± 0.00018	11.1	20.1	14.8	20.5	24.1	19.9	17.3	13.3	9.4	6.4	2.8	8.3	n.d.
U	± 0.00013	4.2	4	4.7	4.1	4.6	5.3	4.5	3.5	2.4	2.8	1.4	2.5	1
Nd	± 0.00072	36.2	26.2	32.8	62.3	37.4	140.2	33.4	19.5	37.2	27.2	9.7	19.1	10.3

n.d. Not detected