PERIODICO di MINERALOGIA established in 1930

An International Journal of
MINERALOGY, CRYSTALLOGRAPHY, GEOCHEMISTRY,
ORE DEPOSITS, PETROLOGY, VOLCANOLOGY
and applied topics on Environment, Archaeometry and Cultural Heritage

DOI: 10.2451/2012PM0018

Archaeometric reconstruction of Nuragic ceramics from Sant'Imbenia (Sardinia, Italy). Technological evolution of production process

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Abstract

The Nuragic village of Sant'Imbenia in Alghero in north-western Sardinia (Italy) was inhabited between approximately the 14th and the 7th century B.C. Foreigners including Eastern Phoenicians and perhaps Greeks settled in the village during the last stages of its existence in the Early Iron Age, importing their own culture and technology. Some of the pottery artifacts produced during this period do not seem to belong to the Nuragic tradition due to their particular shape and type, which suggests that local craftsmen were influenced by these contacts and exchanges with foreign cultures.

The objective of this work was to characterize the artifacts and analyze the evolution in production techniques during the Middle-Late Bronze Age and the Early Iron Age. The main changes seem to have occurred during the transition from the Bronze to the Iron Age, and there was also a clear distinction between cooking and serving wares. We observed that the decoration on the surface of the ceramics changed from the smooth surfaces of the Middle-Late Bronze Age to the partially vitrified, normally red slip wares of the Early Iron Age. Fragments of volcanic rocks were detected in the Early Iron Age cooking wares instead of calcite, the temper typically used during the Middle-Late Bronze Age. Firing temperatures were slightly higher in the Early Iron Age, as inferred by the presence of new mineral phases and the vitrification of the matrix. The surfaces of the ceramics were red, especially during the Early Iron Age, while the matrix often had a black heart.

Key words: Nuragic ceramics; Sardinia; mineralogy; texture; technology.

Introduction

The Nuragic civilization emerged and developed in Sardinia between the height of the Bronze Age (17th Century B.C.) and the Iron Age (7th Century B.C.), although several centres of population remained inhabited up until the 2nd Century B.C., when the Romans ruled the island (Lilliu, 1982).

The Nuragic village of Sant'Imbenia is in the north-west of Sardinia (Italy) in the Bay of Porto Conte (Figure 1), and is bounded to the west by the promontory of Capo Caccia and to the east by Punta Giglio. It has been a rich source of archaeological remains spanning a vast period from Prehistory to the Middle Ages (Bafico, 1986; 1998).

In geological terms the area consists mainly of Mesozoic layers, which lie on quaternary deposits related to alluvial fans and/or flat braided channels, and wind dune fields from the Würm Glaciation Stage (Delfrati et al., 2000). The Triassic consists mainly of dolostones, dolomitic marls, chalk marls and clay with foraminifers. Only a couple of miles north of the Nuragic village of Sant'Imbenia, the Triassic is characterized by continental facies, consisting of the typical red Buntsandstein sandstone. In particular, in the area around the settlement, there are outcrops of Triassic dolostones and aeolian sandstones with carbonatic cement. The Jurassic and the Cretaceous deposits are made up of platform carbonates: mainly dolostones, limestones and marl. On the top, there are volcano-sedimentary successions and lacustrine deposits from the early Oligocene-Miocene (Delfrati et al., 2000). The colour of these sediments varies from orange to ochre, due to the erosion of carbonatic rocks and volcanic deposits. The latter consist mainly of dacites and of rhyolitic ignimbrites (welded and unwelded) with subordinate basalts and andesites (Guarino et al., 2011).

Between the end of the Bronze Age and the

beginning of the Iron Age, a community of foreigners moved to the village of Sant'Imbenia, attracted by the wealth of the area, the accessibility of the site and its plentiful resources, in which they could trade. They did not interfere or clash with the local communities. as in other areas of the island (Rendeli, 2010). The absence of foreign constructions in this area suggests that these incomers found strong, wellestablished indigenous communities with whom they integrated (Bernardini, 2009). Several types of goods were exchanged in Sant'Imbenia, along with experiences, ideas and news. In short, a trade in culture was established (Ridgway, 2004; Rendeli, 2010; Bernardini et al., 2011). In fact, local ceramics, imported red-slip Phoenician wares, local imitations of Phoenician products and Greek, Euboean, Corinthian and Pithecusian drinking pots have all been found in this area (Oggiano, 2000; Ridgway, 2000; Rendeli, 2010; Rendeli and De Palmas, 2011).

The ceramics produced by the Nuragic culture have a well-defined chronology, typology and characteristic style (Campus and Leonelli, 2000). In this article we analyse Nuragic ceramics uncovered in Sant'Imbenia in order to learn more about the production techniques they used and their chronological development and to clarify a number of questions raised by archaeological investigation such as production areas, the choice of specific raw materials and additives and the technological level attained.

Materials and methods

Division of the archaeological samples

Prior to the selection of test samples in the laboratory, an archaeological survey was carried out on around 100 finds from the Nuragic village of Sant'Imbenia (Figure 1). 46 of these from the Middle-Late Bronze Age to the Early Iron Age were selected by visual inspection according to their archaeological significance, function, material and surface composition (Table 1).

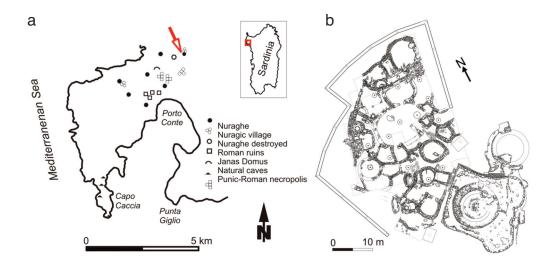


Figure 1. a) detailed representation of the Porto Conte archaeological area (from Bafico, 1998) in which the red arrow indicates the location of the Nuragic village of Sant'Imbenia; b) general plan of the Sant'Imbenia archaeological site (from Garau and Rendeli, 2011).

These 46 ceramics had previously been studied and dated by the archaeologists Bafico (1998) and Oggiano (2000). We extended their research with a detailed observation of the ceramics which allowed us to distinguish ceramics that were intended to be used in contact with fire (cooking wares) from those used for storing food products or for eating and drinking (serving wares) (Table 1 and Figure 2). Considering that some of the 46 ceramics can be used for both cooking and serving, the distinction was based on the quality of the product and the possible presence of burn-marks, i.e. the technological scope of the artifacts, and does not seek to suggest any symbolic, social or ritual role (Steele et al., 2010). We identified pans and jars from the Middle to the Late Bronze Age that were probably used for cooking, as they have visible burn-marks. They are quite porous and show a low degree of cohesion and large amounts of grains. Holes produced during the burning of organic matter are frequently observed.

The serving wares were mainly cups and

askoi, but there were also jugs and jars which could possibly have been used for cooking. However, they are all finer ceramics than the cooking wares, and have no burn-marks. They are from the Late Bronze Age and the Early Iron Age and have treated, decorated surfaces. Like the cooking wares, these ceramics also have voids produced during the burning of organic matter but in general they are more compact.

A lot of the ceramics (from both groups) have manufacturing imperfections: in some cases the thickness of the walls is irregular, with differences visible even to the naked eye, especially between the middle section of the body, which is thicker, and the final part, which is much thinner. Irregularities are often observed on the top of jars, right below the neck and finger marks can often be seen on the inner walls.

Analytical techniques

We began by observing the samples under a stereomicroscope (Leica ZOOM 2000) in order to identify the different treatments used to

Table 1. List of the studied samples with a brief description of the age, typology, presence of black core and surface treatment.

Age	San	nple Ceramic type	Black core	Class	Surface treatment	Analytical Techniques
Middle Bronze	59 60	scraped-comb truncated conical pan engraved-circlet truncated conical pan		CW CW	Smoothing Smoothing	Stm, POM, XRD, XRF Stm, POM, XRD,
B	61 68	engraved-circlet truncated conical pan engraved-circlet truncated conical pan		CW CW	Smoothing Smoothing	Stm, POM, XRD, XRF Stm, POM, XRD
ddle	69	engraved comb truncated conical pan	Yes	CW	Polishing	Stm, POM, XRD, XRF
Mi	70	scraped-comb truncated conical pan	103	CW	Polishing	Stm, POM, XRD, XRF
	7	dolium		SW	Smoothing	Stm, Spp, POM, XRD
	18	egg-shaped, bellied, small-brim, wide-embouchured jar	Yes	SW	Polishing	Stm, Spp, POM, XRD
	25	cylindrical jar with straight walls	Yes	SW	Polishing	Stm, Spp, POM, XRD
	28	cylindrical jar with straight walls		CW	Polishing XRD, XRF	Stm, Spp, POM,
	30	big rounded jar with neck inclined to the outside; its rim is rounded and slightly flattened	Yes	SW	Polishing	Stm, POM, XRD
	31	bellied jar	Yes	CW	Polishing	Stm, Spp, POM, XRD
	32	egg-shaped jar, convex walls, thinner rim	Yes	CW	Smoothing	Stm, POM, XRD
0	33	bowl with upper concave walls, very inclined to the outside, basin with slight-average depth	Yes	SW	Smoothing	Stm, Spp, POM, XRD
nze	37	cup		sw	Patina	Stm, Spp, POM, XRD
Late Bronze	39	truncated conical pan	Yes	CW	Smoothing XRD, XRF	Stm, Spp, POM,
La	42	bellied jar, developed rim with straight internal and external profiles, slight internal edge, inclined convex walls under the angle		CW	Polishing	Stm, Spp, POM, XRD, XRF
	58	tuyere		CW	Smoothing	Stm, POM, XRD
	65	carinated rounded bowl, rounded rim, slightly tapered and flared, straight wall, slightly thicker inside in the middle, strip loop starting from the bottom half of the wall	Yes	SW	Smoothing	Stm, POM, XRD
	66	egg-shaped jar, convex walls, thinner rim not separated from the wall and handles under it	Yes	SW	Smoothing	Stm, POM, XRD
	67	egg-shaped jar, convex walls, thinner rim		SW	Smoothing	Stm, POM, XRD

CW = cooking wares; SW = serving wares; Stm = stereomicroscope; Spp = spectrophotometer; POM = polarized optical microscopy; XRD = X ray diffraction; XRF = X ray fluorescence.

Table 1. Continued ...

Age	Sam	ple Ceramic type	Black core	Class	Surface treatment	Analytical Techniques
	6	bowl with concave walls, very inclined to the outside, shallow basin	Yes	SW	Patina	Stm, Spp, POM, XRD
	8	bowl with concave walls, very inclined to the outside, shallow basin	Yes	SW	Patina	Stm, POM, XRD
	9	bowl with concave walls, very inclined to the outside, shallow basin	Yes	SW	Patina	Stm, Spp, POM, XRD
	13	untrimmed rod-shaped vertical handle	Yes	SW	Patina	Stm, Spp, POM, XRD
	13bi	suntrimmed vertical handle	Yes	SW	Patina	Stm, Spp, POM, XRD
	14	small jar rim separated from the wall, flared		CW	Patina	Stm, POM, XRD
	15	bellied jar		CW	Patina	Stm, Spp, POM, XRD
	19	small egg-shaped jar rim, separate from the wall		SW	Patina	Stm, Spp, POM, XRD, XRF
	24	dolium		SW	Smoothing	Stm, POM, XRD
on	27	bowl with concave walls, very inclined to the outside, shallow basin	Yes	SW	Smoothing	Stm, Spp, POM, XRD
Early Iron	38	bowl with concave walls, very inclined to the outside, shallow basin	Yes	SW	Polishing	Stm, Spp, POM, XRD
斑	40	bellied jar	Yes	CW	Smoothing	Stm, Spp, POM, XRD
	41	bowl with concave walls, very inclined to the outside, shallow basin	Yes	SW	Smoothing	Stm, Spp, POM, XRD, XRF
	44	bellied jar		CW	Patina	Stm, POM, XRD
	45	bellied jar		CW	Patina	Stm, Spp, POM, XRD
	46	bellied jar	Yes	CW	Patina	Stm, POM, XRD
	47	bellied jar	Yes	CW	Burnished	Stm, POM, XRD
	48	compressed egg-shaped mug	Yes	CW	Smoothing	Stm, Spp
	49	small bellied jar		CW	Smoothing	Stm, Spp, POM, XRD
	51	bellied jar		CW	burnished	Stm, POM, XRD
	53	dolium		SW	Smoothing	Stm, Spp, POM, XRD
	56	egg-shaped jar, rod-shaped handles in the upper section of its body	Yes	CW	Smoothing	Stm, POM, XRD
	62	askos decorated with engraved parallel lines, starting from an embossed circlet	Yes	SW	Burnished	Stm, POM, XRD
	63	vertical handle, arch-like, burnished, decorated with engraved circlets and geometric patterns		SW	Burnished	Stm, POM, XRD
	64	askoid jug		SW	Smoothing	Stm, POM, XRD

CW = cooking wares; SW = serving wares; Stm = stereomicroscope; Spp = spectrophotometer; POM = polarized optical microscopy; XRD = X ray diffraction; XRF = X ray fluorescence.

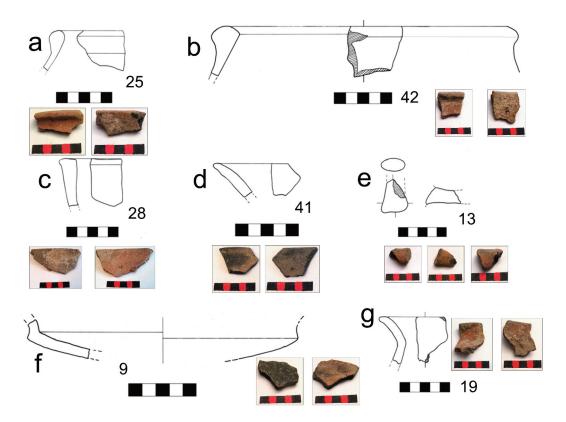


Figure 2. Representative samples of all the materials we studied; a, b, and c, date back to the Middle and Late Bronze Age; d, e, f, and g, date back to the Early Iron Age. The numbers refer to the samples listed in Tables 1. Samples 13, 28, and 42, are cooking wares. The others are serving wares.

smooth the surfaces. We used a portable Minolta CM700d spectrophotometer to analyse the colour of these pieces, both on the surface and in the matrix, and to calculate the L*, a* and b* coordinates. Due to the small size and curved surface of some samples, an illuminated area of just 3 mm in diameter was used. The measurements were performed by selecting CIE illuminant D65, which simulates daylight with a temperature colour of 6504 K.

We identified the mineral phases and ceramic texture by observing thin sections of ceramic fragments under an Olympus BX60 polarized optical microscope (POM) equipped with a

digital microphotography camera (Olympus DP10). Non-plastic component and porosity contents were estimated visually using comparative charts (Munsell® Soil Color Charts, 1994).

The mineralogical composition of the archaeological ceramics was determined using X-ray diffraction (XRD) analysis. A Philips PW 1710 diffractometer with automatic collimator was used. Working conditions were as follows: CuK α radiation emission (1 = 1.5405Å), 40kV voltage, 40 mA current, explored area 3° to 60° 2 θ , goniometric speed 0.05° 2 θ s⁻¹ The disoriented crystalline powder method was used. For this

purpose, samples were previously reduced to powder in an agate mortar and sieved to obtain grains with a diameter of less than 0.053 mm. Data interpretation was performed using the XPowder software (Martín, 2004).

Bulk chemical analyses were performed by means of the wavelength dispersive X-ray fluorescence (XRF) technique, using a S4 Pioneer (Bruker AXS) spectrometer with a Rh anode X-ray tube. 5 g per sample were finely ground and well mixed in an agate mortar before being pressed into an Al holder for disk preparation. Ten major and minor elements and eight trace elements were measured. The major and minor element contents are reported as wt.% oxide normalized to 100%, while trace elements are expressed in ppm, LOI-free. The analytical detection limits were: $SiO_2 = 0.08\%$; $Al_2O_3 =$ 0.1%; $TiO_2 = 0.01\%$; CaO = 0.01%; MgO =0.07%; MnO = 0.01%; Na₂O = 0.01%; K₂O = 0.02%; Fe₂O₃ = 0.01%; P₂O₅ = 0.01%; Cr = 11 ppm; Cu = 10 ppm; Zn = 15 ppm; Rb = 18 ppm; Sr = 20 ppm; Y = 16 ppm; Zr = 15 ppm; Ba =240 ppm. The relative standard deviation was < 1% except for Y, Ba and Mn which present values ranging from 3% to 4.8% (Niembro Bueno, 2009). ZAF correction was performed systematically (Scott and Love, 1983). The NCS DC 74301 (GSMS-1) standard (Chen and Wang, 1998) was applied.

It is important to remember that we were unable to perform all the analytical techniques mentioned here on all the samples (see Table 1), due to the fact that in some cases we had only very small amounts of sample at our disposal.

Results and discussion

Surface treatment and colour of ceramics

Stereomicroscope observations permitted a detailed study of the different surface treatments applied: smoothing, polishing, burnishing or patinating (Table 1). Observation was then extended to consider the different types of tools used to smooth the surfaces (hard or soft tools), the depth, the width, and the distribution of the traces (Cuomo di Caprio, 2007; Levi and Recchia, 1995; Levi, 2010).

We observed a number of differences between the Middle-Late Bronze Age samples and those from the Early Iron Age. During the Middle-Late Bronze Age, the most common treatment was

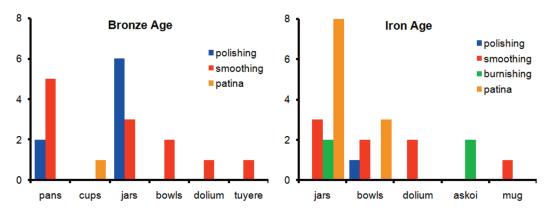


Figure 3. Description of surface treatments used during the Bronze Age and the Early Iron Age. The major differences are in the polishing process, which was much more common during the Middle and Late Bronze Age, and burnishing and patinas, which were usually more common during the Early Iron Age.

smoothing, followed by polishing (Figure 3). Often, this was not done on the whole surface, which was partially porous, irregular and contained grain fragments.

During the Early Iron Age, most samples show properly burnished patinas with well-covered, smooth, clean, impermeable surfaces.

More specifically, among the Bronze Age samples, the pans have smoothed (albeit still quite rough), polished surfaces. They are rich in grains and the pores are visible. The traces left by the smoothing processes are usually deep, with a random pattern and horizontal direction. In some cases, decorative geometrical figures can be seen, almost always engraved.

Jars were either smoothed or polished. The smoothed jars have the same characteristics as the pans, while the polished jars received more refined, more elaborate treatment, which covered more of the surface and the porosity is low. Polishing traces are shallow and run parallel to each other in horizontal and vertical directions. The best treated surfaces are the inner surfaces, which have very low porosity.

Bowls were smoothed and the treatment seems excellent, both inside and outside, covering the

whole surface and with medium porosity.

The dolium and the tuyere were smoothed perhaps after moulding only to reduce imperfections on the surfaces. A large number of grains, imprints of plant remains and high porosity are visible.

As far as the Early Iron Age samples are concerned, the jars were polished, smoothed, burnished or patinated; in all cases, the treatment was excellently applied, both on the inner and outer surfaces. It covers the whole surface, medium-low porosity is observed and surface traces tend to run in parallel usually in a horizontal direction.

The most elaborately treated ceramics are once again the bowls. Some have patinas that are totally impermeable, smooth and with no traces of treatment defects. Others are smoothed but with an extended treatment over a large part of the surface with shallow grooves tending to run parallel in a mainly horizontal direction and with very low porosity; others are polished. What does change, in comparison with the previous period, is the high percentage of samples with partially vitrified, compacted, non-porous, smooth, fully covered, red patina. This suggests

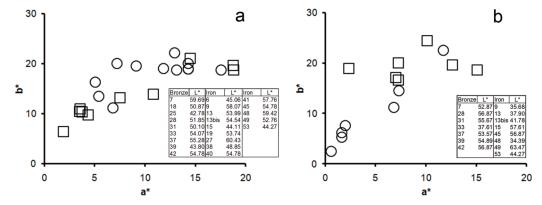


Figure 4. Chromatic coordinates (a* and b*) of ceramic fragments. a) shows the chromatism of the surfaces; b) shows the chromatism of the matrix. In both diagrams squares represent samples from the Middle-Late Bronze Age, while circles represent samples from the Early Iron Age. The lightness (L*) of the surfaces and of the matrix are included as tables in the two diagrams.

a possible imitation of the Phoenician red-slip wares brought to Sant'Imbenia by foreign incomers.

Askoi are very well burnished and their surfaces are shiny, smooth, very compact and impermeable and with no visible traces of treatments.

The dolium sample is smoothed and is similar to those from the Bronze Age, while the mug sample has smoothed surfaces.

As regards the hue (a* and b*) of the ceramics, with the exception of three red samples (numbers 7, 42 and 58), the surface of ceramics from the Middle-Late Bronze Age show the lowest a* and b* values (Figure 4a). By contrast, samples from the Early Iron Age show a quite constant b* value, while a* varies from 5 to 20, depending on the intensity of the red colour on the surface.

When we consider the colour of the matrix (Figure 4b) we observe a smaller range with a low a* value, while b* varies from 3 to 25 with values at the top of this range being observed more frequently in the Middle-Late Bronze Age samples. Only one sample (number 48) is located on the right of the diagram, denoting its high a* content.

The lightness (L*) of ceramics is generally lower in the matrix of the samples than on the surface because of the presence of a black (or at least grey) heart (Table 1), whose origin is described in the following section. The lowest L* value measured on the surfaces was for sample 25 (42.78) and six samples had a lightness value of less than 50 on the surface. As regards the lightness measured in the matrix, the lowest L value was measured in sample 48 (34.39) and half the samples had values of under 50.

Mineralogy and texture

Nuragic ceramics from Sant'Imbenia are quite homogeneous from the mineralogical and textural points of view. The main differences lie in the presence/absence of certain tempers and their grain size.

More specifically, samples from the Middle and Late Bronze Age are characterized by relatively low birefringence and by a 15-20% grains content with a bimodal distribution, in which the coarser fraction is more abundant (Table 2). Porosity values range between 20-30%, with two main families of pores: the smaller ones have a mainly sub-rounded shape, while most of the larger ones are elongated. As far as minerals are concerned, we detected quartz, calcite, muscovite, rare feldspars and metamorphic and sedimentary rock fragments (Table 2). The quartz is often fractured, with individual pieces that are angular and subangular in shape, and is the main component of both the coarse and the finest fraction of the temper (Fig 5a-d). The presence of calcite in the matrix often micritic - suggests that it was part of the natural sediment from the eroded Triassic carbonates that outcrop near the village, but this does not apply to the calcite grains, which are part of the temper. They are angular and subangular in shape and large and irregular in size, which suggest that they may have been crushed and then added to the mixture. Rock fragments consist of phyllites and litharenites, which may come from the nearby, very large Mores Formation from the Burdigalian Age (Carta Geologica d'Italia, 2012). Litharenites are the main component of the coarse fraction of the temper. Secondary calcite was observed on the surfaces and in the larger pores. Imprints of straw and other plant remains, which accounted for between 10 and 20% of the volume, were also detected. Presumably these were added to improve the plasticity and the rheological characteristics of the raw material (Maritan et al., 2006). voids resulting from The decomposition of plant remains may have created a reducing atmosphere inside the firing furnace (Cerdeño del Castillo et al., 2000), as the darkening of the matrix in certain specific areas suggests (Figure 5b).

From the Late Bronze age there was a

Table 2. POM analyses of ceramic samples from the Middle-Late Bronze Age and the Early Iron Age.

	Sample	Age	p	gs	Qz	Cal	Phy	Fsp	Ox	SRF	MRF	VRF	Chamotte	Organic matter
	59	0	M	20	++	++	+		+	++			++	+++
	60	onz(M	20	++	++	+	+	+	+			++	+++
	61	Br	M	20	++	++	+		+	++			++	+++
	68	Middle Bronze	M	20	++	++	+		+	++				+++
	69		M	20	++	++	+	+	+	+			++	+++
	70		M	20	++	++	+			++				+++
	28		M	15	+++	+		+	+			++		++
	31	e e	M	15	+++	++	+	+	+	++	++			++
nics	32	Late Bronze	Н	20	+++	++		+	+					++
ran	39	e Br	Н	20	+++	+		+	+	+		++		++
9 5	42	Lat	M	15	+++	+		+	+	+		++		++
Cooking ceramics	58		Н	20	+++	+	++		+	+	+			++
Co	14		M	10	+++		+		+	+		+++	+	
	15		M	10	+++		+	+	+	+		+++		
	40		M	10	+++			+	+	+		+++		
	44	_	M	15	+++			+	+	+	+	+++		+
	45	Early Iron	M	15	+++			+	+	+	+	+++		+
	46	ırly	M	15	+++			+	+	+	+	+++		+
	47	E	M	15	+++			+	+	+	+	++		+
	49		M	15	+++			+	+	+	+	++	+	+
	51		M	10	+++			+	+	+	+	++	+	
	7		Н	20	+++		+	+	+	+		++		+
	18		M	15	+++	++	+	+	+	+		++		
	25		Н	20	+++			+			++			
	30	ze	M	15	+++	++	+	+	+	+	+			+
nics	33	sron	M	15	+++		+	+	+	++	+			++
Serving ceramics	37	Late Bronze	M	15	+++	+		+	+	++	++			+
၁ ရွ	65	La	M	15	+++		+	+	+	++	++			+
ıvir	66		M	15	+++	+	+	+	+		+			
Se	67		M	15	+++	+	+	+	+	++		++		

High (H), medium (M), and low (L) grade of porosity (p); average grain size (gs, in %) of non-plastic component (temper plus skeleton); mineralogy of non-plastic component: Qz: quartz; Cal: calcite; Phy: phyllosilicates (in general); Fsp: feldspars (in general); Ox: metallic oxides; SRF: sedimentary rocks fragments; MRF: metamorphic rocks fragments; VRF: volcanic rock fragments; +: scarce; ++: abundant; +++: very abundant.

Table 2. Continued ...

	Sample	Age	p	gs	Qz	Cal	Phy	Fsp	Ox	SRF	MRF	VRF	Chamotte	Organic matter
	6		M	15	+++		+	+		++		++		+
	8		M	15	+++	+	+	+		+		++		+
	9		L	10	+++			+		+		+++		
	13		M	15	+++		+	+		+	+	+++		+
	13bis		Н	15	+++		+	+						++
cs	19		M	15	+++		+	+		+		+++		+
ami	24	Ţ,	M	15	+++			+		+	+	++		+
cer	27	Early Iron	L	15	+++		+	+		+		++		+
Serving ceramics	38		L	10	+++			+		+		+		
Ser	41	щ	M	15	+++			+	+	+		++		+
	53		M	15	+++	+		+	+	+	+	++	+	+
	56		M	10	+++	+		+	+	++	+	++		
	62		M	15	+++		+	+						+
	63		L	15	+++		+		+	+				+
	64		M	15	+++	+	+	+	+					+

High (H), medium (M), and low (L) grade of porosity (p); average grain size (gs, in %) of non-plastic component (temper plus skeleton); mineralogy of non-plastic component: Qz: quartz; Cal: calcite; Phy: phyllosilicates (in general); Fsp: feldspars (in general); Ox: metallic oxides; SRF: sedimentary rocks fragments; MRF: metamorphic rocks fragments; VRF: volcanic rock fragments; +: scarce; ++: abundant; +++: very abundant.

substantial change in the temper used in the pottery (Figure 5e-h) with fragments of volcanic rocks (pumices and ignimbrites) being detected in relatively constant proportions and sizes (Figure 5g). They are totally aphiric even if pyroxene, feldspar and oxide microcrystals can be also found in the pumices and ignimbrites from the Alghero area (Guarino et al., 2011). These ceramics did not show concentrations of calcite, the temper used in earlier ceramics. Fragments of metamorphic (mainly phyllites) and sedimentary rocks (litharenites) (Figure 5h) may also be found. The pumice and ignimbrites may come from the Tottubella area, about 15 km east of the Nuragic village, where these rocks outcrop in the "Piroclastiti di Olmedo", a formation from the Burdigalian Age (Ginesu, 1984; Carta Geologica d'Italia, 2012). Quartz, feldspars and plagioclases form the majority of the coarse fraction and are mainly angular to elongated in shape. Feldspars are mainly composed of sanidine (Figure 5e) denoting their volcanic origin and their provenance (together with the pumices and ignimbrites) from differentiated volcanic rocks. The matrices of these ceramics typically show a sandwich-like distribution of chromatic layers (black heart, Figure 5f), possibly due to the presence of reducing substances in the raw material (i.e. organic matter) or to a fast firing (Buxeda i Garrigós et al., 2003; Mirti and Davit, 2004; Maritan, 2004; Cultrone et al., 2011). Other changes noted in Early Iron Age ceramics

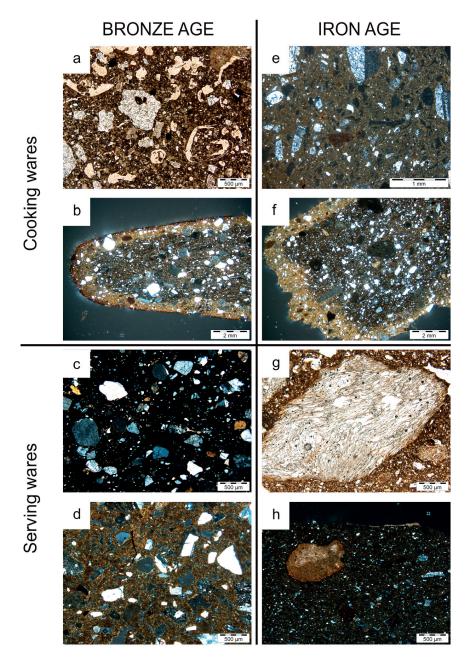


Figure 5. Optical microscopy micrograph of cooking and serving wares from the Bronze and Iron Ages: a) aspect of fractured quartz grains in sample 59; b) presence of a black core in sample 65; c) general view of sample 67; d) morphology of temper in sample 25; e) twinned sanidine crystals in sample 47; f) general view of sample 56 with black core; g) fragment of pumice in sample 39; h) litharenite fragment 1 mm wide in sample 63.

included the fact that less temper was used and the degree of porosity was lower. These samples show medium birefringence, a 10-15% temper content once again with bimodal distribution where both fractions (fine and coarse) are equally represented, and porosity values of between 5 and 10% (Table 2). Two families of pores can be distinguished: the larger, more common ones are elongated in shape and almost all share the same orientation; the smaller pores have rounded shapes. The muscovite has the same orientation as the large pores, i.e. perpendicular to the pressure that was exerted on the clay matrix during the preparation of the ceramics.

Microscope observation showed that the amount of temper is generally higher in cooking ceramics than in serving ones. This seems to have been a deliberate technological choice on the part of the potters, as the cooking wares had to withstand high thermal shocks. Other research have shown that high temper concentrations minimize the risk of crack propagation (Tite et al., 2001), although the strength of the finished ceramic is reduced substantially (Kilikoglou et al., 1998).

Based on the level of birefringence of the matrix and of some minerals, such as muscovite, as well as the presence of calcite, samples dating back to the Middle-Late Bronze Age could have been fired at temperatures of around 750 °C in an oxidizing atmosphere because of the risk of "lime blowing" (small spalls pushed out of the walls of the vessel) (Laird and Worcester, 1956). Generally, we observed that samples containing 5% or more of calcite, to which 10% of organic matter had been added (Cuomo di Caprio, 2007), were fired in a reducing atmosphere. Under such conditions, Maggetti et al. (2011) demonstrated that calcite can extend its stability up to 800-850 °C. Since the choice of crushed calcite grains was deliberate, in order to reduce or eliminate the risk of spalling if these temperatures were surpassed, ceramics may have been quenched after firing (Cultrone et al., 2004). It is also possible that seawater (the village is about 1 km from the coast) was used to mould the clayey material (Rye, 1976), as sodium chloride acts as a melting agent and helps to produce more resistant ceramics compared to other ceramics fired at the same temperature but made without NaCl (Cultrone et al., 2005). In the Early Iron Age wares in which they are present, the calcite grains are generally darker and reaction rims may sometimes be observed, while muscovite, when observed, shows lower interference colours. Another aspect worth noting is the presence of partially vitrified matrices. These data suggest a firing temperature of around 900 °C.

The fact that different kinds of temper were used may have been due to potters trying to improve functional aspects of their products (Martineau et al., 2007; Barone, 2010). Interestingly, this change in production techniques coincided with the influx of foreigners into the village of Sant'Imbenia.

Mineralogical changes due to firing temperatures

X-ray diffraction (XRD) analyses show that all samples are rich in quartz and have varying phyllosilicate (muscovite) and feldspar content (Table 3). Samples of cooking ceramics dating back to the period between the end of the Middle Bronze Age and the beginning of the Late Bronze Age are rich in calcite. From the Late Bronze Age onwards, the increase in the background noise of the diffractograms suggests the presence of an amorphous fraction (maybe, vitrification of the matrix as indicated above). Obviously, pumices ignimbrites observed under microscopy (see Table 2) cannot be detected by XRD, but they must have contributed to the increase in the amorphous fraction content. Storage and serving wares produced during the Middle-Late Bronze Age have a similar composition to the cooking wares, although some new silicate phases, gehlenite (Ca₂Al₂SiO₇) and diopside (CaMgSi₂O₆), start to appear. During the

Table 3. XRD analysis of ceramic samples from the Middle-Late Bronze Age and the Early Iron Age. The PDF2 database card of each mineral is indicated into brackets.

	Sample	Age	Qz	Phy	Fsp	Cal	Pl	Hem	Gh	Di	amorphous
	59	o		+++	++	+	++				
	60	zuc	+++	++	+	++					
	61	Bro	+++	++	+	++					
	68	Middle Bronze	+++	++	+	++					
	69	ido	+++	++	+	++					
	70	\geq	+++	++	+	++					
	28			+++	++	+	+				+
CS	31	IZe	+++	++	+	++					
Ξ.	32	ron	+++	++	+	++					
era	39	B	+++	++	+	+					+
<u>ത</u>	42	Late Bronze	+++	++	+	+					+
Cooking ceramics	58	Ι	+++	++	+	++					
90	14			+++	++	+	+				++
\cup	15		+++	++	+	tr					++
	40		+++	++	+	tr					++
	44	on	+++	++	+	++					
	45	Early Iron	+++	++	+	+					
	46	arly	+++	++	+	+					++
	47	Щ	+++	++	+	tr					++
	49		+++	++	+	++					
	51		+++	++	+	+					
	7			+++	++	+				+	+
	18		+++	++	+	++					
	25		+++	++	+				tr		
S	30	0	+++	++	+	++					
im.	33	nz(+++	++	+	++					
era	37	Late Bronze	+++	++	+	++					
် င	65	te I	+++	++	+					tr	+
Vin	66	La	+++	++	+	++					
Serving ceramics	67		+++	++	+				tr		+

Qz: quartz (33-1161); Phy: phyllosilicates (7-25); Fsp: feldspars (19-1227); Cal: calcite (5-586); Pl: plagioclases (20-528); Hem: Hematite (33-664); Gh: gehlenite (35-755); Di: diopside (19-239); tr: traces; +: scarce; ++: abundant; +++: very abundant (mineral symbols after Whytney and Evans, 2010).

Early Iron Age the number of samples in which gehlenite and/or diopside develop increased slightly, as did the content of the amorphous fraction (possibly an incipient vitrification of the matrix), while calcite content decreased. Using XRD we also detected plagioclase and hematite, which were not identified in the Middle-Late Bronze Age samples.

In general, the use of calcite in ceramics production decreased significantly between the Middle-Late Bronze Age and the Early Iron Age. The opposite occurred with the amorphous fraction, which increased from very low percentages in the Middle Bronze Age samples to higher percentages in the Early Iron Age. This indicates a clear change in the preparation of raw

Table 3. Continued ...

	Sample	Age	Qz	Phy	Fsp	Cal	Pl	Hem	Gh	Di	amorphous
	6			+++	+	+		+			++
	8		+++	+	+		+				++
	9		+++		+		+	+	tr		++
	13		+++	++	+	+					
ics	13bis	_	+++	++	+	+					
ceramics	19	Early Iron	+++		+		+	+			++
cera	24	<u>></u>	+++		+		+	+		tr	++
1g (27	ar	+++		+		+	+	tr		++
Serving	38	_	+++	+	+	tr					+
Se	41		+++		+		+	+			++
	53		+++		+		+	+	tr	tr	++
	56		+++	++	+	+					++
	62		+++		+		+	+	tr		++
	63		+++		+		+	+	tr		++
	64		+++	++	+	+					++

Qz: quartz (33-1161); Phy: phyllosilicates (7-25); Fsp: feldspars (19-1227); Cal: calcite (5-586); Pl: plagioclases (20-528); Hem: Hematite (33-664); Gh: gehlenite (35-755); Di: diopside (19-239); tr: traces; +: scarce; ++: abundant; +++: very abundant (mineral symbols after Whytney and Evans, 2010).

materials during the Bronze and Iron Ages. Moreover, the presence of mineral phases such as hematite and above all plagioclase in the serving ceramics dating back to the final phase of the Bronze Age and the Early Iron Age, indicates that potters may have used a clayey material that was different at least in part from the one used during the Middle Bronze Age.

XRD results confirm expected firing temperatures suggested by optical microscopy observations. Temperatures were estimated on the basis of the mineralogical transformation that takes place in the ceramic when it is fired, and which is manifested in the presence of calcite and/or muscovite (around 750-850 °C) or new silicate phases (gehlenite and diopside) (more than 800-900 °C) (Sterba et al., 2009). However, this temperature range may be wider because Nuragic ceramics were not fired in kilns with an oxidizing atmosphere but were open-fired in holes in the ground (De Palmas and Di Gennaro, 2004) making it impossible for us to control their

firing parameters (maximum temperature reached, heating rate, soaking time and firing atmosphere) (Maggetti et al., 2011). Both during the Bronze Age and the Iron Age, the cooking wares were fired at around 750-800 °C (or up to 850 °C under reducing conditions), as the presence of calcite and muscovite and the absence of new silicate phases suggest. By contrast, about half of the non-cooking wares were fired at higher temperatures, around or above 900 °C, as attested by the absence of carbonates and muscovite and the presence of gehlenite and diopside, albeit in small concentrations

Chemical composition

Despite the small number of samples available to be studied with this technique, we did observe a change in the chemical composition of the ceramics from the Middle-Late Bronze Age to the Early Iron Age (Table 4). The element that most varies with age is Ca. According to POM

Table 4. Major (wt.%), minor (wt.%) and trace elements (ppm) of ceramic samples from the Middle Bronze, Late Bronze and the Early Iron Ages.

		N	Iiddle Bro	onze		I	Late Bronz	ze	Early Iron		
Sample	59	60	61	69	70	28	39	42	19	41	
SiO ₂	61.98	65.80	59.87	64.30	60.97	69.41	65.90	69.46	65.70	65.78	
Al_2O_3	16.59	15.05	17.92	15.79	17.41	15.48	16.65	14.94	17.74	19.94	
TiO_2	0.95	0.40	0.48	0.55	0.60	0.39	0.51	0.78	0.73	0.62	
CaO	8.98	6.46	9.42	6.90	8.22	3.01	4.43	3.23	2.62	2.09	
MgO	1.27	1.44	1.46	1.42	1.62	2.15	1.64	1.82	1.53	1.41	
MnO	0.08	0.02	0.04	0.11	0.18	0.07	0.13	0.12	0.11	0.15	
Na_2O	1.83	1.83	1.84	1.91	2.00	1.32	1.60	1.41	1.87	1.21	
K_2O	4.21	4.27	3.79	4.18	4.17	3.86	3.89	3.83	4.01	3.16	
Fe_2O_3	3.77	4.55	4.84	4.63	4.67	4.13	4.89	4.25	5.55	5.48	
P_2O_5	0.32	0.18	0.34	0.21	0.16	0.17	0.35	0.15	0.15	0.16	
LOI	11.91	14.54	15.12	12.65	15.14	5.76	10.30	6.64	2.49	9.26	
Cr	137	196	147	n.d.	n.d.	n.d.	n.d.	103	108	n.d.	
Cu	78	76	81	53	99	53	85	60	n.d.	51	
Zn	131	172	147	82	88	105	116	91	90	59	
Rb	192	77	68	154	68	64	168	137	156	107	
Sr	199	670	746	190	217	468	282	217	201	232	
Y	22	30	n.d.	33	51	20	29	36	37	38	
Zr	192	188	207	315	377	230	232	299	346	280	
Ba	996	3608	653	584	803	1242	686	591	697	869	

Data are normalized to 100% (LOI-free). n.d. = not detected.

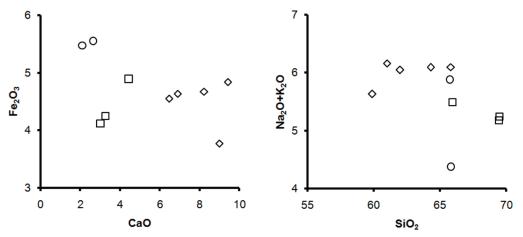


Figure 6. Chemical diagrams of CaO vs. Fe_2O_3 (on the left) and SiO_2 vs. $Na_2O + K_2O$ (on the right). The rhombi stand for Middle Bronze Age samples, the squares for Late Bronze Age samples and the circles for Early Iron Age samples.

observations, the High Ca content in ceramics from the Middle Bronze Age is due to the presence of micritic calcite in the raw material and the use of crushed calcite fragments as temper. From the Late Bronze Age the Ca content fell drastically, due mainly to the absence of calcite grains, which confirms a change in the temper used in making the ceramics. The two binary diagrams of Figure 6 show the variations in CaO vs. Fe_2O_3 and SiO_2 vs. $Na_2O + K_2O$: ceramics became richer in Si and Fe elements, while their Ca and alkalis content fell. Late Bronze Age samples are chemically closest to those from the Early Iron Age (see the low CaO content compared to that of Middle Bronze Age). Interestingly, Late Bronze Age samples were also similar in mineralogical terms (poor in calcite and with fragments of volcanic rock, see Table 3). As regards trace elements, sample 60 stands out for its high Ba content when compared to the other samples and it is also the richest in Cr and Zn elements. There is no clear trend in the distribution of trace elements except. perhaps, for lower Zn and higher Zr contents for ceramics from the Early Iron Age.

Conclusions

The mineralogical, textural, chemical and technological analyses conducted on Nuragic ceramics from Sant'Imbenia revealed a number of differences between the products from the Middle Bronze Age and the Early Iron Age. Firstly, in the use of temper: in the earliest ceramics a raw material rich in phyllites, quartz, calcite and feldspars was used almost exclusively, while at the end of the Bronze Age fragments of volcanic rocks began to be added, often in constant quantities and sizes. The use of carbonatic rock fragments (Hoard et al., 1995; Martineau et al., 2007; Cultrone et al., 2011) and volcanic rocks (Dickinson, 2007; Belfiore et al., 2010; Morra et al., 2012) was common in ceramic production areas near to where these rocks

outcrop. In our study, the presence of fragments of volcanic origin may help to date these ceramics, which are difficult to locate on the timeline.

The most ancient samples, those dating back to the Middle Bronze Age, are characterized by rather friable mixtures due to the high amount of temper and a porosity level of 20-30%, resulting from the use of organic matter and calcite. These samples were fired at temperatures of around 750-800 °C under oxidizing conditions or up to 850 °C under reducing conditions. Their surfaces were smoothed or polished and some decorations and engravings can be observed. From the Late Bronze Age onwards, the presence of volcanic observed fragments can be (especially ignimbrites and pumice). These materials had refractoriness levels that provided greater durability against changes in temperature (Hein et al., 2007), so avoiding the risk of fracture and ensuring that heat was distributed more evenly, so confirming the potters' expertise. These ceramics were more resistant than the older ones. in which a smaller amount of temper was used. and had a slightly lower porosity level (15-20%). The reduced birefringence of the matrix and the presence of new silicate phases (diopside, gehlenite) indicate firing temperatures of around or above 900°C. The surfaces were smooth, almost all of them had a patina, and they were slightly porous, glossy and bright. On the basis of the mineralogical and chemical analysis, the three samples which had originally been dated back to the Middle-Late Bronze Age also seem to belong to the Early Iron Age.

We also observed a change in the technology used to produce the ceramics, which varied according to their intended purpose. One example was the choice of clay raw materials that were capable of withstanding continuous contact with fire. Another was the treatment of the surface, which reduced surface permeability to minimum levels. The proficiency reached by potters in fire and kiln management was also observed; the presence of partially vitrified, non-porous and

very compact surfaces suggests that the potters of Sant'Imbenia were able to raise the temperature inside the kiln above 900 °C and keep it there long enough to cause the (at least partial) vitrification of the surface of the ceramics. The study of the surfaces also produced interesting results. The artifacts made between the end of the Bronze Age and the beginning of the Iron Age are almost all red on the surface, some of them have patinas and others are simply polished. This feature was not observed in earlier products, which may have been imitations of Phoenician red-slip products imported into Sant'Imbenia. In some cases, decorations were made with a high degree of specialization, and there are examples of very thin patinas, perfectly bonded with the ceramic body that are bright, non-porous and partially vitrified.

The results of this study shed more light on the lives and activities of the Nuragic villagers and provide the first mineralogical, textural and chemical characterization of the ceramics they produced. The technological evolution is confirmed by the changes in composition of the raw materials according to the intended function of the wares. There is also evidence of the important role played by contacts, trade and peaceful relations between culturally and ideologically different groups, aspects that were fundamental for the growth and development of individual centres of population or communities.

Acknowledgements

This study was financially supported by Research Group RNM179 of the Junta de Andalucía and by Research Project P09-RNM-4905. We are grateful to Nigel Walkington for his assistance in translating the original text. Celestino Grifa and an anonymous referee are thanked for their careful reading and their helpful comments.

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Submitted, July 2012 - Accepted, November 2012