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ARTISANAL EXPERIENCES IN ARPI: COLOURS AND RAW MATERIALS OF APULIAN RED FIGURE PRODUCTION

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Abstract

This work aims at studying the manufacturing and technology peculiarities of Apulian red figure (4th - 3rd century BCE) production from Arpi. In particular, the relationship between the red figure production from Arpi with coeval pottery of different classes retrieved in the same contexts in Arpi is investigated, as well as the connection between the red figure production from Arpi and same class vases from different Apulian sites. This goal has been achieved through widening the archaeometrical studies so far performed to red figures finds from two more tomb complexes in Arpi – i.e. tomb of the Niobids vase and 4/2004 tomb – and also organically reorganizing the so far obtained experimental data on Arpi findings of various ceramic classes. The systematic characterization of red figure vases was obtained by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray Spectroscopy (EDS), Powder X-ray diffraction (PXRD) and Raman Spectroscopy. Analytical data allow to define the elemental and mineralogical composition of ceramic pastes and identify pigments and coatings. The results allow to validate the archaeological hypotheses that Arpi red figure production is different from the one from other Apulian areas and that pictorial experiences were shared among tombs, *domus* and ceramic production in Arpi.

Keywords: Archaeometry, Apulian red figure pottery, Arpi, ceramic provenance, pigments, ceramic technology

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1. Introduction

Arpi (Mazzei 1995, Marin 1970) was located in northern Apulia, a few kilometers from Foggia, in the center of the Tavoliere, in a large alluvial plain bordered to the north by the Fortore river, to the south by the Ofanto, to the east by the Gargano massif, and to the west by the Daunian sub-Appennine mountains. It was crossed by several waterways that were navigable in ancient times (Figure 1). The Celone river, flowing into the Candelaro, allowed navigation to Lake Salso and then to the ports of Siponto and Salapia – a river road that was useful for trade and exchange with the coastal populations and those across the Adriatic. The other centres of Daunia were arranged in a crown around Arpi. Like many ancient centers in Apulia, it boasted about having been founded by Diomedes who, according to Stephen of Byzantium, walled it with stones brought from Troy (Notarangelo 2008). The form of the city was recently reinterpreted and clarified (Arpi Project).

Arpi and Canosa, from the end of the 4th to the end of the 2nd century B.C., were deemed the largest, richest and most populous cities in Daunia (Grelle 1995, Corrente 2012).

Written sources and archaeological evidence suggest that Arpi had remained an active center until the 2nd century AD, even after the Hannibalic period and the loss of the port of Siponto due to its defection from Rome. Its abandonment was likely due to the swamping of the river, which made the area unhealthy (Mazzei 1992). The ancient archaeological center has been the scene of original artistic and artisanal experiences which have accompanied the cultural, economic and social evolution of this Daunian population in its interaction with other Greek, Samnite and Roman communities.

This work, which aims to study Apulian red-figure ceramics from Tomb of the Niobids vase and Tomb 4/2004, is part of a wider project. Its purpose is to investigate the manufacturing peculiarities of Apulian red figure pottery (4th - 3rd century BCE) and the relationship between the Arpi red figure production with coeval pottery of different classes retrieved in the same contexts (Mangone et al. 2009c, Giannossa et al. 2019), as well as their connection with the red figure production from different Apulian sites. The ceramic collection from Arpi is extremely rich and varied, yet productive structures and other archaeological indicators of production are seldom found. Extremely few archaeometric data can identify local or imported production, outline the manufacturing process and thus identify the workshops; to date, the vases analyzed come from only two grave contexts (Giannossa et al. 2019, 2020).

The results of the present work are eventually combined with those previously obtained on the finds from Arpi (Giannossa et al. 2019, 2020) and compared with those related to red-figure vases from

two other coeval Daunian centres, i.e. Canosa (Bitetto et al. 2016) and Ruvo (Giannossa et al. 2016, 2017b). This may contribute to validate the hypotheses that Arpi red figure production differs from the one of other Apulian areas and that pictorial experiences were shared among tombs, domus and ceramic production in Arpi. This theory has been originally proposed by Patete et al. (Patete et al. 2021) on the basis of archaeological investigations on the ancient city of Arpi.

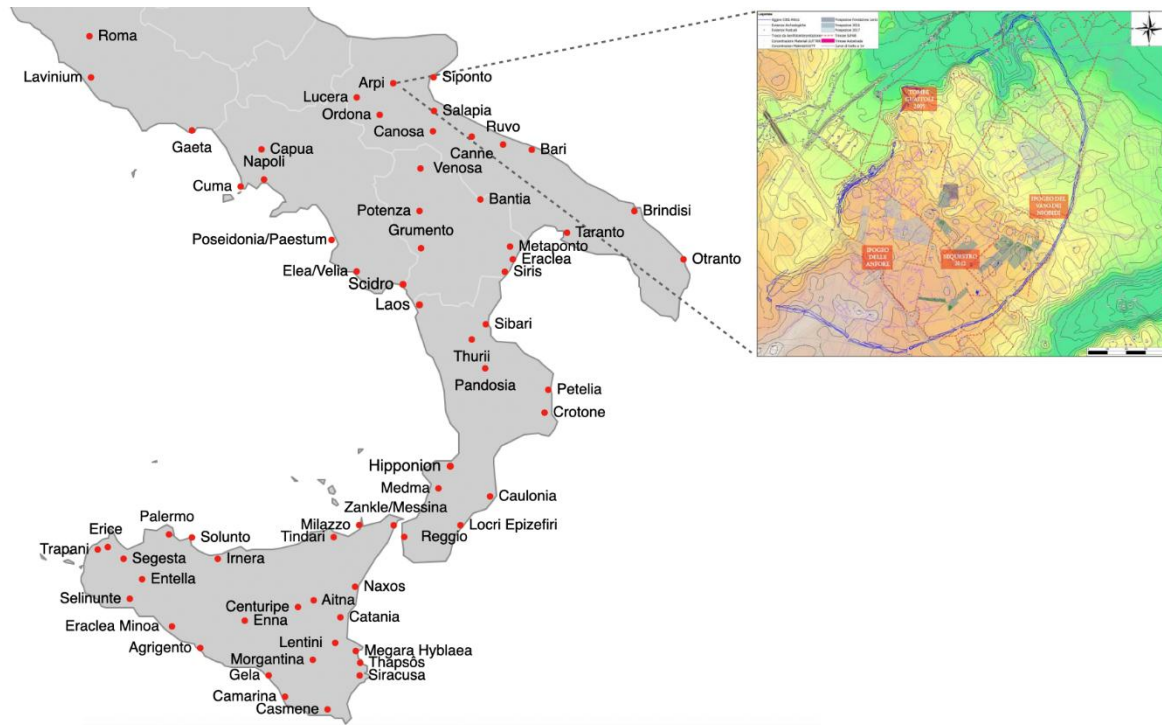


Figure 1: Magna Grecia. In the inset: excavated tombs in Arpi (L. Fornaciari, M. Leone, G. Stello and V. Soldani).

1.1. Tomb of the Niobids vase and Tomb 4/2004

The Tomb of the Niobids Vase was discovered in 1972 at Arpinova (De Juliis 1992). It is a chamber tomb with a rectangular room built of limestone blocks, measuring 6 m long and 3 m wide, with a maximum height of 2.5 m, with plastered walls and a double-pitched roof. It was accessed through a long corridor and a “portal”, consisting of three large blocks, two vertical and one horizontal resting on top, embellished with a carved architrave painted yellow, surmounted by a pediment painted red and decorated in the corners with blue spirals. The form of this monumental tomb is known in Arpi from the middle of the 4th to the first decades of the 3rd century. It belongs to a type of which fifteen examples have been attested (Type VIC corresponds to chamber tombs with a double-pitched roof (Leone and Munzi in press)). Although part of the finds was taken away by robbers, the intervention

of the Soprintendenza allowed the recovery of 159 vases, now stored in the Civic Museum of Foggia. The archaeological study dates the tombs within a range from 350 to 310 BC.

The Tomb 4/2004 is a pit tomb found in 2004 in the area of the hypogeum of the Amphorae in the south-western sector of the settlement not far from the hypogeum of the Medusa. Unpublished, it is currently being studied (Progetto Arpi). The pit was intercepted in the dromos of a grotto tomb (T4). Lined and covered with flat tiles, it belongs to type III attested in Arpi (Leone and Munzi in press). The inhumed person was in a lying position with a belt and the bell-shaped krater was deposited at his feet.

2. EXPERIMENTAL

2.1 Samples

13 finds -12 coming from the Niobids vase tomb and 1 find from 4/2004 tomb- were analyzed (Table 1).

Different forms and stylistic attribution to painters from two different workshops can be highlighted, the Painter of the Patera (Ar34) and the Painter of Baltimore (Ar27, Ar33, Ar29, Ar26, Ar24, Ar25, Ar35) (Trendall and Cambitoglou 1982). Both workshops can be dated to the last third of the 4th century BC. The productions linked to these painters are attested in the centers of Peucetia and Daunia.

The bell-shaped krater from the tomb 4/2004 (Ar43) is typical of a standardized production with a generic iconography (on one side a satyr follows a maenad, on the other two cloaked men are depicted) that can be related to workshops operating in the second half of the 4th century BC, in particular the Lucera Painter.

2.2 Instrumentation

Investigations were conducted using an integrated approach of analytical techniques. To obtain compositional data of ceramic pastes, about 40 mg of ceramic body were sampled and subjected to dissolution through acid attack, using a controlled microwave technique (Milestone Start D (FKV) microwave oven). The solutions were analyzed by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS Nexion 300X Perkin Elmer). The *brick clay* standard (679 National Bureau of Standards) was used as a reference to test the analytical procedure. Five replicate readings were performed on both the standard and each sample. Indium was used as an internal standard (25 ppb). The analytical details of this procedure have been described in Mangone et al. 2009a, 2009b and Genga et al. 2008.

Multivariate statistical treatment by Principal Component Analysis (PCA) was carried out on compositional data using the software package Minitab®.

To investigate the mineralogical composition of the paste, optical microscopy (OM, Axioscop 40 – Carl Zeiss microscope), scanning electron microscopy (SEM, EVO-50XVP LEO – Carl Zeiss) equipped with an energy dispersive X-ray spectrometer (X-max (80 mm²) Silicon drift Oxford detector supplied with a Super Atmosphere ThinWindow®) and X-ray diffraction analyses (Philips X'Pert Pro) were used. Working conditions for XRD were: CuK α Ni filtered radiation, 40 kV and 40 mA of power supply, divergence slit 1°, anti-scatter slit 0.5°, receiving slit 0.2 mm, scan speed of 0.5° (2 θ) per minute (Giannotta et al. 2006). The PANICSD database (High Score software package) was used to process the analytical data. The semi-quantitative estimate of the mineral phases was determined using Hillier method (Hillier 2003) on whole-rock random powders.

In order to identify the pigments used as overpainting, Raman investigations were carried out on the surface and on the polished cross sections of the micro-samples, after OM and before SEM observations (LabRAMHR Evolution® (Horiba®, Kyoto, Japan), He-Ne 633 nm laser).

3. RESULTS

3.1 Ceramic bodies

3.1.1 Chemical composition and statistical treatment of data

Data from Table 2 have been treated by PCA alongside both previously obtained ones regarding finds from Tomb ONC28 (Giannossa et al. 2020) and Tomb 5/2005 (Giannossa et al. 2019) in Arpi, to highlight similarities and differences between the composition of ceramic pastes of finds from different tombs in Arpi. The results are shown in Figure 2. Three clusters can be distinguished: A, B1 and B2. All the finds coming from the same tombs are efficiently grouped together, samples Ar36, Ar37 and Ar38 being the only ones left out. Cluster A groups the samples from the tomb of the Niobids vase, cluster B the ones from the ONC28 tomb and cluster C the ones from the 5/2005 tomb. The sample Ar38 is located inside cluster B, while Ar36 and Ar37 are located between clusters A and B. The position of sample Ar43, outside all three clusters, is justified by the fact that it is the only sample from tomb 4/2004.

Cluster A differs from B along PC1 principally due to the loadings relative to Fe, Mn, Al, Ti and Ca parameters. In particular, the A scores are characterized by positive values of PC1 due to higher amounts of Fe, Mn, Al and Ti, while the scores of the cluster B are spread along negative PC1 values due to the higher Ca contents. The scores of both clusters are characterized by positive PC2 values.

The negative PC2 values of the scores of the samples from cluster C, mainly due to the loadings relative to K and Sr parameters, justify the separation of this cluster from the other two.

The inclusion of the finds of other two coeval Daunian centres (Ruvo (Giannossa et al. 2016, 2017) and Canosa (Bitetto et al. 2016)) to the statistical treatment highlighted three different clusters, each grouping samples retrieved from the same site (Figure 3). This result upholds the supposition (Giannossa et al. 2021 and references therein) that Apulian production has its own distinctiveness shared by many manufacturing sites.

The Arpi cluster in Figure 3 is considerably homogeneous in chemical composition, hinting to a very similar raw material being used. However, different subclusters can be outlined, agreeing with their provenance context. The compositional difference detected among samples from different tombs (Figure 2) can therefore be attributed to different technological processes employed for their manufacturing, possibly performed in different workshops.

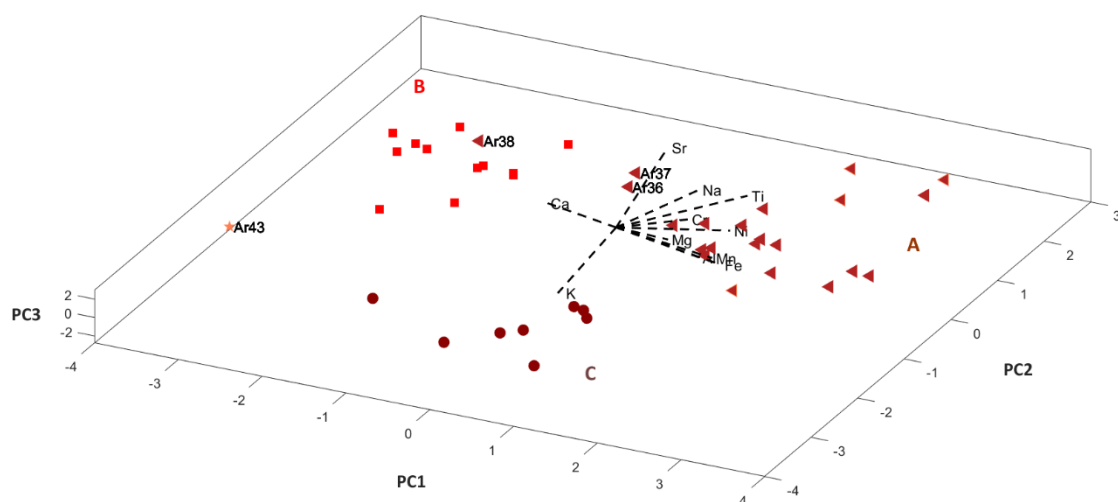


Figure 2: Scores and loadings plot of the ceramic bodies of finds coming from Arpi (circles: 5/2005 tomb, squares: ONC28 tomb, triangles: Niobids vase tomb, star: 4/2004 tomb). The accounted variance is 73% of the total one.

3.1.2 Mineralogical composition

The pastes of the finds from Niobids vase tomb are quite homogeneous in terms of mineral assemblage (Table 3), hinting to the use of a common raw material. Small variations have been found in quantitative abundance, especially for newly formed ones (pyroxenes, gehlenite and hematite). SEM analyses showed in some samples the presence of accessory minerals – i.e., rutile, ilmenite, zircon, Ba-feldspar (celsian), Ca- and rare earth-phosphates, - which cannot be detected via PXRD.

Their presence and distribution are not uniform, possibly due to their traces amount in the original rocks and to their current position in sedimentary eluvial deposits.

Calcite is present both as secondary recrystallisation within the porosity and, in some samples, as limestone clasts. In samples Ar26, Ar33 and Ar35, limestone clasts as small as fine sand can be also recognized by SEM analyses, probably due to an incomplete dissolution of too big starting clasts during firing. An indirect confirmation of the presence of calcite in the raw material can be obtained from the abundance, shape and size of the pores, as these features also depend on the size and shape of the original limestone clasts. However, it should be pointed out that microporosity is mainly linked to the degree of sintering of the clay during firing.

The porosity in some samples (Ar27 and Ar28) is rather homogeneous in shape, dimension and distribution. Mica minerals and pores are parallel oriented to the walls of the pottery wares, highlighting a very accurate manufacturing. On the same grounds, the same cannot be said for the other samples (Ar25, Ar26, Ar29, Ar33, Ar34 and Ar35).

On the other hand, the microporosity, related to clay matrix sintering, appears overall constant. This fact and the high quantity of pyroxenes, gehlenite and hematite suggests that firing occurred in the 1000 – 1100 °C range for a long enough time, except for Ar26, Ar33 and Ar35 samples.

According to the size of clasts (quartz and feldspar mainly) present in pastes, the samples can be divided in three groups: A) fine to medium silt (8-16 µm) (Ar28 and Ar31); B) medium to coarse silt (16-62 µm) only (Ar26, Ar27, Ar29, Ar32 and Ar35); C) both medium to coarse silt (16-62 µm) and very fine sand (62-125 µm) (Ar25, Ar33 and Ar34).

As for sample Ar43, clay matrix is more abundant than in the Niobids vase tomb samples, while the skeletal fraction is composed of a medium-fine silt (quartz, feldspar and micas), with a small amount of coarse silt and scarce amount of very fine sand. Zircons are present in the sample. Pores are heterogeneous in size and distribution and mostly parallel oriented to the walls of the pottery wares, as well as micas. Its sintering degree is high. It can be therefore assessed that the firing temperature was similar to the one for the vases in Niobids vase tomb, although kept for a shorter time, as suggested by the lower amount of newly formed minerals.

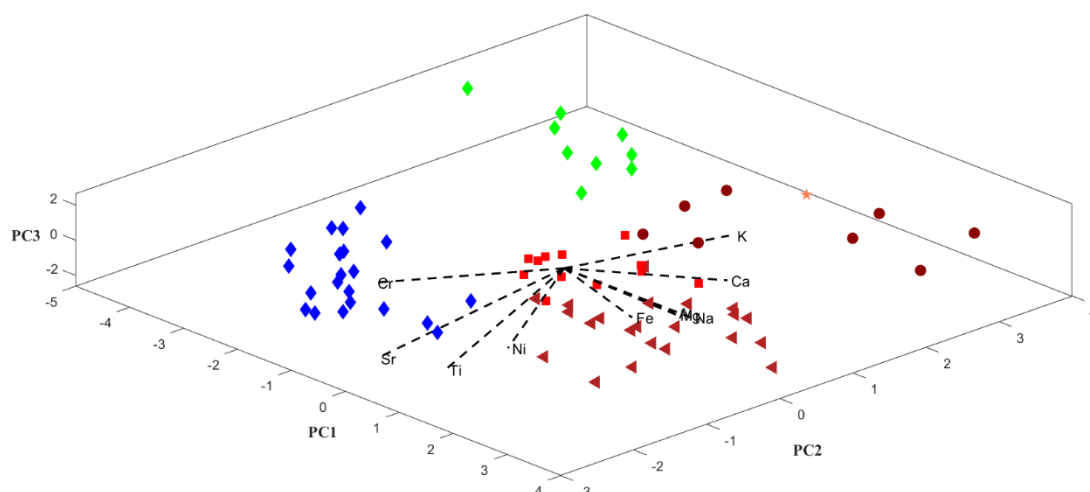


Figure 3: Scores and loadings diagram of the ceramic bodies of finds coming from Ruvo (blue diamonds), Canosa (green diamonds) and Arpi (red, circles 5/2005 tomb, triangles Niobidis vase tomb, squares ONC28 tomb, and star 4/2004 tomb). The accounted variance is 83% of the total one.

Comparing these results with the ones from previously studied tombs – ONC28 and 5/2005 (Giannossa et al. 2019, 2020) – fundamental homogeneities can be brought up, i.e., optimal sintering, matrices made of marly clay and skeletal parts mostly constituted by quartz, feldspar, micas and Fe- oxides and hydroxides clasts. However, minimal differences can be underlined among samples from different tombs, such as body texture and content of accessory minerals.

In samples from both 5/2005 and ONC28 clusters, the grain size distribution varies from fine to medium silt (not exceeding 32 μm). The size and the unimodal distribution of clasts suggest the use of a refined clay for paste manufacturing. On the other hand, a coarser texture, with grains of sizes ranging from medium silt (>32 μm) to very fine sand (62-125 μm), characterizes samples Ar36, Ar37 and Ar38, sample Ar43 and the Niobidis vase tomb cluster -except samples Ar28 and Ar31-.

It is interesting to point out that compositional similarities (chemical and mineralogical) only occur among finds retrieved in the same tomb and belonging to the same ceramic class. When the statistical treatment is indeed extended to coeval vases, coming from the same context but belonging to different ceramic classes, significant compositional differences can be emphasized (Giannossa et al. 2019). This is probably due to the use of different raw materials and/or manufacturing procedures. Apulian red figure pottery is characterized by a more careful selection of raw materials and more accurate manufacturing process (Giannossa et al. 2020).

3.2 Overpaintings

3.2.1 White

SEM-EDS analyses of the white surfaces of all vases showed the same structural and compositional features of all Apulian red figure vases studied so far (Figure 4). Kaolinite $[\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4]$, added with feldspars and albite, was the supposed raw material for white overpainting (Mangone et al. 2009d). The same features are reported in literature for Attic ones as well (Noble 1960).

Raman analyses, on the other hand, do not show clear signals which can be associated to any white pigment compatible with the historical period. Indeed, signals from anatase (i.e. $\beta\text{-TiO}_2$, 140, 195, 385, 512 and 625cm^{-1}), poorly crystalline quartz (463 cm^{-1}) (Wopenka et al. 2002), feldspar (153, 284, 407, 465 and 512 cm^{-1}), rarely gypsum (140, 414, 494, 615, 1008 and 1136 cm^{-1}) and calcite (150, 275, 708, 1083) were identified (Figure 4).

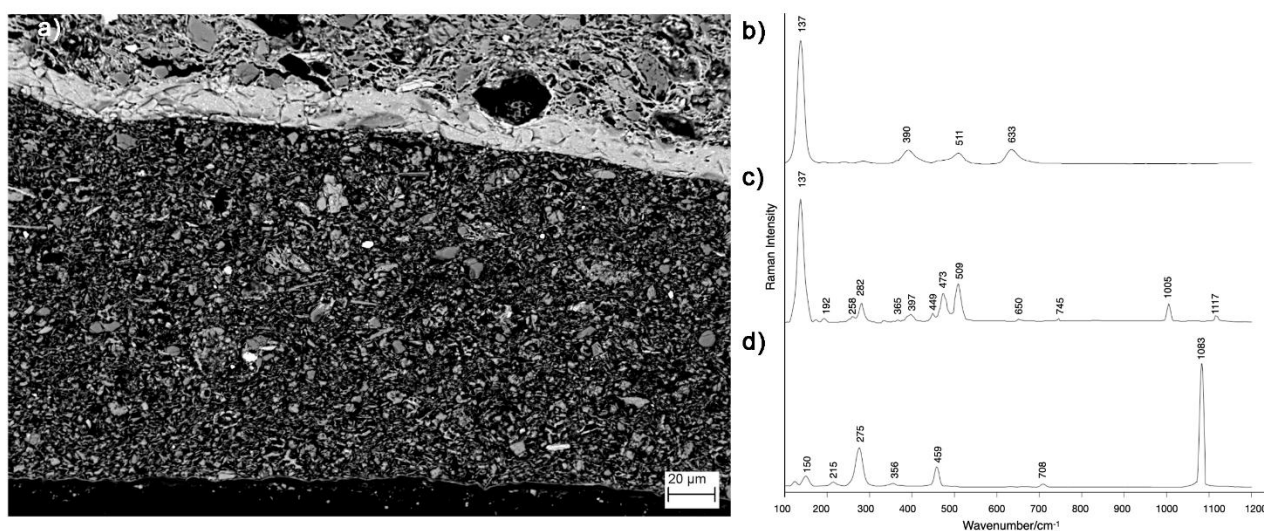


Figure 4: a) SEM-BSE photomicrograph of cross sections of fragment Ar31 showing the structure of white overpainting (lower layer). Right: Representative baseline-subtracted Raman spectra of b) anatase, c) feldspar, d) calcite on the white surface of AN6 vase. Acquisition parameters: 532 nm laser, 50X LWD objective, 0.6 mW, 3600 s.

The complete absence of kaolinite associated Raman signals is certainly to be ascribed to the manufacturing process, consisting of vase firing after its overpainting. Indeed kaolinite, a very weak Raman scatterer *per se* (Murad 1997, Clark 2007), loses its crystalline form upon firing, as it first (400 – 600 °C) dehydrates into meta-kaolinite and then (950 °C approx.) transforms into Al-Si spinel, a cubic phase with poor crystallinity degree. Anatase, a minority component of kaolin, is instead a strong Raman scatterer (Schroeder et al. 2003, Clark 2007) and its Raman fingerprint has been used (Zuo et al. 1999) as an indicator of kaolin. However, anatase cannot be associated with TiO_2 use as white pigment since no analysis of artefacts dated prior to the 20th century suggests the intentional use of native anatase for this purpose.

Moreover, kaolinite is abundant in Apulian red earths and in bauxites deposits. When no Al- and/or Si- amorphous compounds are present, the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio for kaolinite is 1.17. As can be seen from Table 4, its value is higher in the analyzed samples owing to the presence of quartz and other phyllosilicates (illite) in the sediments used as raw material for white overpainting. This is also compatible with the small amount of Ca retrieved, as it is commonly found in sediments derived from limestone leaching and weathering. The presence of calcite can be explained mostly as secondary crystallization due to circulating water, or raw material impurity. Gypsum is likely derived from sulphation phenomena.

3.2.2 Red

The structure of red overpainting appears coarse-grained and heterogeneous with aggregates of iron oxides, sometimes very large in size (Figure 5). Its average chemical composition is shown in Table 4.

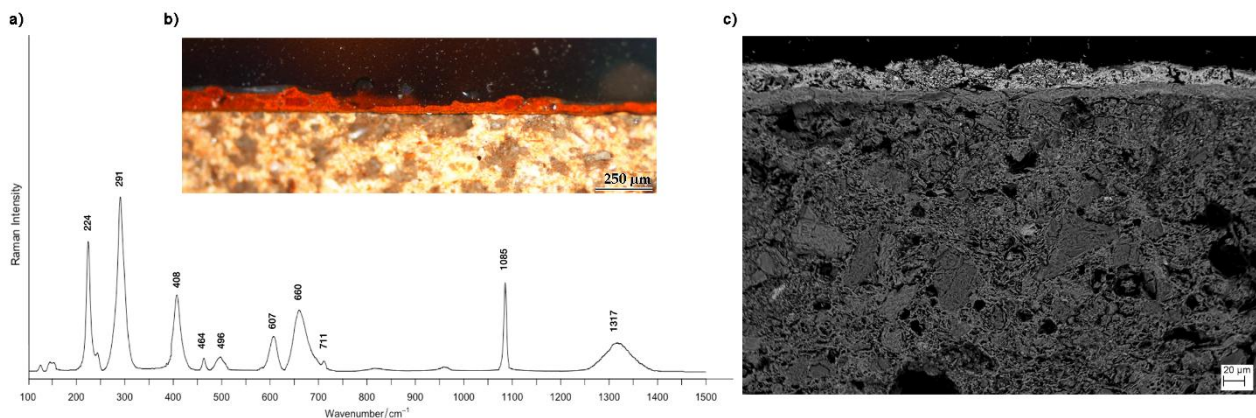


Figure 5: a) Representative baseline-subtracted Raman spectra of the red overpainting (acquisition parameters: 633 nm laser, 50X LWD objective, 0.8 mW, 2000 s); b) OM and c) SEM-BSE photomicrographs of cross sections of fragment Ar27, showing the structure of red overpainting.

In the measured Raman spectra, besides the dominant hematite peaks (226, 292, 410, 615 and 1332 cm^{-1}), peaks related to α -quartz and/or feldspar ($126, 160, 510\text{ cm}^{-1}$) and calcite (711 and 1085 cm^{-1}), were also observed (Figure 5). The broad band observed at about 660 cm^{-1} in the spectrum of hematite can be related to magnetite (Fe_3O_4) or maghemite ($\gamma\text{-Fe}_2\text{O}_3$) contamination or associated to the Raman forbidden, IR active longitudinal optical (LO) E_u mode of hematite (De Faria et al. 1997, Bersani et al. 1999, Forleo et al. 2022).

3.2.3 Yellow

Two different yellow shades as overpainting pigments were highlighted on Apulian red figure pottery from Arpi (Giannossa et al. 2020): a matt one used for the lightness of some painted fabrics and a bright one to outline the contours and to paint weapons. They differ in texture, sintering degree, chemical composition, covering power and brightness.

The matt one is poor both in vitrification degree and in compactness. It is similar in structure to the white overpaint, while differing for small Fe quantities. Its Raman spectra (Figure 6) show peaks related to hematite (225, 292, 410, 608 and 1325 cm^{-1}), magnetite (554, 669 cm^{-1}), quartz (464 cm^{-1}), gypsum (1008 and 1140 cm^{-1}), anatase ($\beta\text{-TiO}_2$) (144 cm^{-1}), alkali feldspar (511 cm^{-1}) and hercynite (749 cm^{-1}).

On the other hand, the bright one has a high vitrification degree and a great compactness. Its Raman analyses (Figure 6) mainly display magnetite and hematite, alongside weak peaks that can be attributed to quartz, anatase and alkali feldspar.

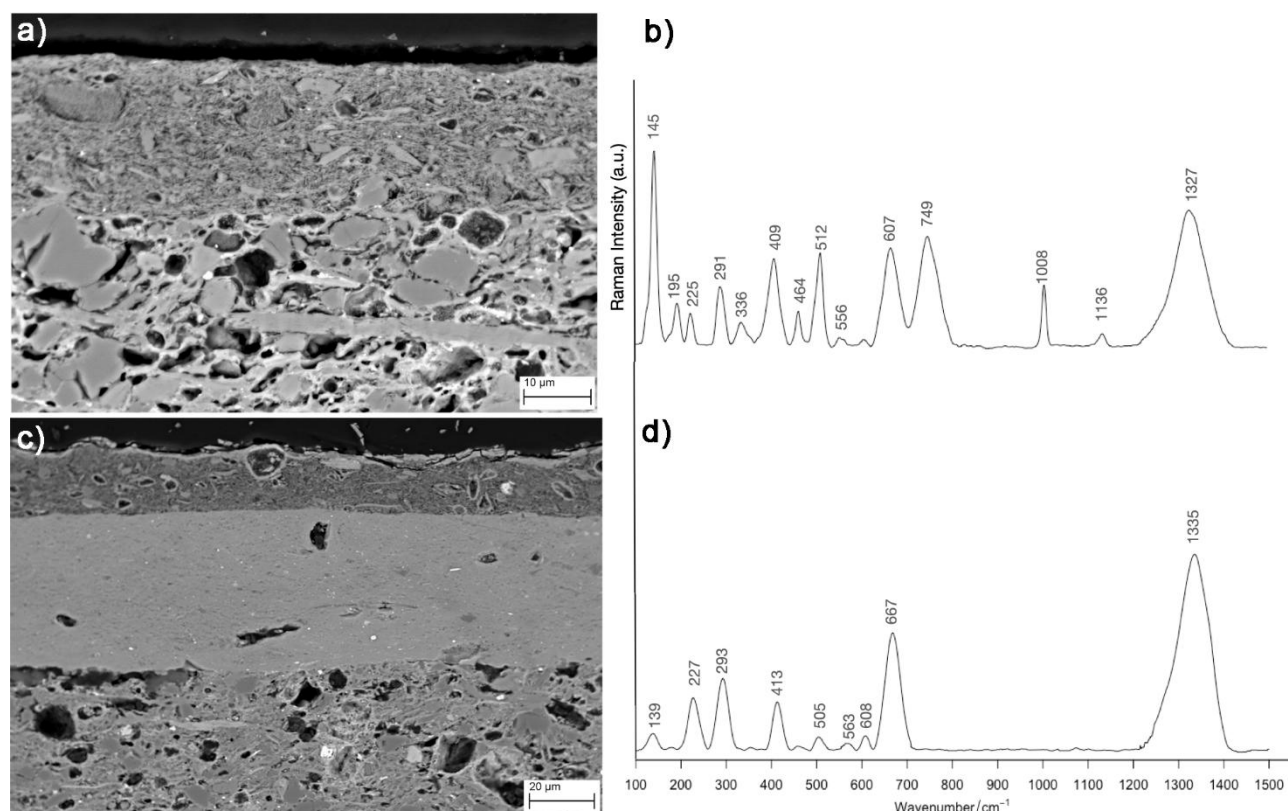


Figure 6: SEM-BSE photomicrographs of the cross section of fragment Ar28 showing the different structures of the matt (a) and the bright (c, intermediate layer) yellow. Representative baseline-subtracted Raman spectra of the matt (b) and bright (d) yellow overpaintings on Ar28 vase (acquisition parameters: 633 nm laser -50X LWD objective, 0.6 mW, 3600 s).

These data are consistent with both yellow pigments being prepared from black gloss mixed with white pigment in varying ratios, with greater quantity of the latter to yield the matt shade. This hypothesis has also been suggested by Noble (Noble 1960) for Attic red figure pottery, showing cases

of bright vitrified yellow overpaintings. No information has been found in literature about matt yellow in either pottery classes.

3.2.4 Light brown

Light brown overpainting is used for perspectival purposes of architectural structures on vases. SEM analyses shows a black gloss-like structure, yet thinner ($< 20 \text{ mm}$), richer in Ca and poorer in Fe (Figure 7 and Table 4).

The Raman spectra show mainly hematite ($224, 244, 291, 405, 608$ and 1327 cm^{-1}) and magnetite (675 cm^{-1}) (Figure 7).

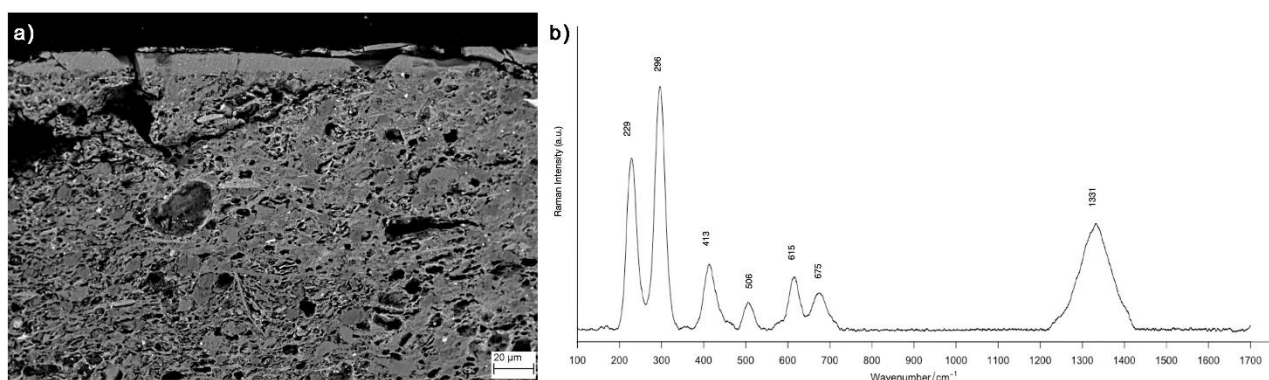


Figure 7: a) SEM-BSE photomicrographs of cross sections of Ar31 vase, showing the structure of the light brown overpaintings; b) representative baseline-subtracted Raman spectra of the light brown overpainting, showing the presence of hematite and magnetite. (Acquisition parameters: 633 nm laser, 50X LWD objective, 0.6 mW, 3600 s).

4. Conclusions

The archaeometric results concur in hypothesizing the use of local clayey raw materials, most likely retrieved from the area closest to the Arpi necropolis – i.e., Candelaro river valley – (Giannossa et al. 2019) for creating the ceramic bodies.

Small compositional differences – chemical and mineralogical – among finds of the same class belonging to different tombs and also the major ones among finds of different classes but belonging to the same context have been brought up. They could be explained supposing either supplies of raw materials from different local sources or the use of manufacturing processes with different extents of accuracy (possibly from different workshops).

Among all the ceramic classes of Arpi analyzed so far, the most refined pottery making is found in red figure pottery. The differences in manufacture accuracy indeed mirror the differences in the vases value and purpose.

A conscious use of locally available raw materials and an in-depth knowledge of manufacturing processes should also be assumed for the realization of overpainting. This hypothesis is supported by red overpaintings being made of *terra rossa* (Dell'Anna 1967, Dell'Anna and Laviano 1991, Mongelli et al. 2015), a raw material easily available, especially in the area immediately next to the site of Arpi. It is also reinforced by the finding of *terra rossa* inside a vase (Ar43) from tomb 4/2004. Likewise, the skillful mixing of black gloss and white pigment made it possible to obtain two yellow overpaintings with different covering power and brightness from the same raw materials. Furthermore, the mixing of black gloss clayey raw material with lime water to obtain light brown overpainting is another clear evidence.

All the outcomes suggest that Apulian potters operating in Arpi during the 4th century BC had an excellent expertise in pottery making. A well-thought-out selection of raw materials were made, also choosing the most suitable technological process in line with the market value, importance and purpose of the final product.

The comparison of data from Arpi with those relating to Apulian red figure vases from other sites (Canosa and Ruvo) provided further validations of the hypothesis of a local production for late Apulian red figure pottery.

All the results could allow to validate the hypotheses that the Arpi red figure production is different from the one of other Apulian areas (Bitetto et al 2016, Eramo and Mangone 2019, Giannossa et al 2016, 2017a, 2017b, 2021, Mangone et al 2008, 2013) and that pictorial experiences were shared among tombs, *domus* and ceramic production in Arpi, as originally proposed on the basis of archaeological investigations (Patete et al. 2021 in press).

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