Studies in Heritage Glazed Ceramics

The majolica azulejo heritage of *Quinta da Bacalhôa*

3

December 2021

ISBN 978-972-49-2317-8

Special Series

Volume I

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This number is published within the scope of the national infrastructure E-RIHS.pt funded by FCT, the Portuguese Foundation for Science and Technology.

Site: http://glazedceramics.lnec.pt/

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Graphic design: Helder David

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www.lnec.pt

Publisher: LNEC
Collection: Studies in Heritage Glazed Ceramics
Series: SHGC 3
1st. edition: 2021
Printing: 200 copies

Descriptors: Azulejo / Historic heritage / Glazed ceramics / Manufacture / Physical-chemical characterization / PT

Descritores: Azulejo / Património histórico / Cerâmica vidrada / Fabrico / Caracterização físico-química / PT

CDU 691.833"16"(469)
691.434"16"(469)

ISBN 978-972-49-2317-8

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PREFACE

Studies in Heritage Glazed Ceramics was forcefully interrupted for almost two years due to the COVID-19 crisis, but returns with its third number, the first of a special series of four volumes dedicated to the renaissance majolica azulejo heritage of *Palácio e Quinta da Bacalhôa* in Azeitão, Portugal.

The azulejos of Bacalhôa have a legendary status in the studies of renaissance majolica in the Iberian Peninsula in general, because of their extraordinary variety and quality and the fact that its most mythical panel, representing the biblical episode of *Susanna and the Elders*, is dated "1565" – a chronology hardly compatible with the then-recent production of azulejos in Portugal. Several hypotheses were advanced over the years to cope with this seemingly impossibility, almost always involving Flemish potters immigrated to the Peninsula.

Another problem stems from the assortment of patterned tiles, often depicting variations in the design or the quality of the workmanship only explainable by the involvement of, not one, but several workshops… But what could those workshops be? Who were the potters and painters behind such artistic achievement? What is the approximate chronology of the different (non-dated) panels and linings with patterned tiles? And most of all: how does the unrivalled treasure of renaissance tiles of Bacalhôa fit within the history of the diffusion of the majolica technology and its firm establishment in Spain and Portugal, where azulejos developed to become a cultural trait still flourishing today?

Following the studies in the early production of majolica azulejos in Portugal, published in the first two numbers of this journal, a multidisciplinary research team was formed to try and reply (within the bounds of the possible) to those questions, as well as to shed light on a number of other perplexing details related to the surviving panels. The research lasted for two years and the results will be published in a dedicated special series. This first volume includes four articles which deal with the basic issues and establish the basis for the detailed study of the panels and patterned tiles that will follow. Four more articles will be published in the second volume, in January 2022, and the last eight articles will be published in the third and fourth volumes within the following 12 months.

The scientific production stands on several pillars, one of them the peer-reviewers of the authors' papers, whose names are often unknown but whose importance in the final output is singular. The editors wish to heartily thank the reviewers for this number: Professor Nuno Senos of *Instituto de História da Arte* of *Universidade Nova de Lisboa*, Doctor Alexandre Nobre Pais, Director of *Museu Nacional do Azulejo* and Doctor António dos Santos Silva of *Laboratório Nacional de Engenharia Civil* (LNEC) who have graciously accepted the hardship of the revisions.

LNEC thus presents this third number of its journal dedicated to azulejos and other glazed ceramics with a set of articles resulting from the cooperation of the tools of Humanities and Natural Sciences aiming to support in solid foundations the study and understanding of one of today's most prized cultural heritages of Portugal.

The Editors

EDITORS

SCOPE

Studies in Heritage Glazed Ceramics is dedicated to the results of scientific studies in the field with a particular emphasis on analytical results, conservation issues and historical studies and very specially to multidisciplinary research in the domain.

The contents will include:

- Archaeometry studies, namely the application of analytic methods to the identification of materials and the establishment of technologies, provenance or the setting of chronologies;
- The artistic and historical context of productions, materials and evolving technologies, as well as the origin, preparation and trade routes of pigments and other raw materials;
- Decay of glazed ceramics, techniques and materials for conservation;
- Other innovative research results in the field.

A technical overview of 16th century majolica panels and patterned tiles from *Palácio e Quinta da Bacalhôa* in Portugal

João Manuel Mimoso, Alfonso Pleguezuelo, Maria Augusta Antunes, Dória Costa, Sílvia Pereira, Álvaro Silva

ABSTRACT

The manufacture of azulejos in Portugal started, as far as we know presently, around 1560s by the workshop of Hans Goos, a Flemish potter established in Lisbon as João de Góis. His younger brother, of whom we only know the adopted Portuguese name Filipe de Góis, was referred to in 1575 as a "master of glazed ceramics" and so he may have also produced azulejos around that time. The productions of what we may call the "circle of João de Góis" (maybe only a single workshop or maybe several sharing the same technology) encompasses a period thus starting around 1560 and lasting at least until the 1590s. Practically all the potentially locally-produced azulejos from this period that we have studied (fifteen different panels) could be fitted into the productions of that rather consistent circle, while confirmed imports from Antwerp and Seville are characterized by different technological traits.

We had now the opportunity to study all the azulejo panels and patterned linings at the Palace, annexes and gardens of Bacalhôa, amounting to more than 50 different types, a research made all the more difficult by the mixing of tiles of like patterns but certainly different provenances and by the use of clays, glaze formulations, or firing schedules that could not be safely included within the bounds of the circle of João de Góis. Since it is not viable to encompass all the types in a single paper, the most representative azulejo panels still extant at Bacalhôa were chosen, as well as the patterned types used at the Pleasure House by the lake and in the Palace itself. They were sampled and their morphological and chemical characteristics were compared between them and with previously published results, following which the panels and patterned tiles were organized in groups with a view to their future individual study.

RESUMO

Tivemos a oportunidade de estudar todos os painéis, azulejos de padrão e de emolduramento do Palácio, anexos e jardins da Bacalhôa, perfazendo mais de 50 tipos diferentes. A complexidade do estudo foi aumentada pela mistura de azulejos do mesmo padrão, mas de proveniências certamente diferentes e pelo uso de argilas, formulações de vidrados, ou ciclos de cozedura que não permitiam uma inclusão segura nas produções do círculo de João de Góis. Não sendo viável abordar conjuntamente tantos tipos diferentes num único artigo, escolhemos os painéis mais representativos ainda existentes na Bacalhôa, bem como os padrões de azulejos de faiança utilizados na chamada Casa de Prazer (também referida como Casa do Lago ou Casa de Fresco) e no próprio Palácio. Estes foram amostrados e caracterizados morfológica e composicionalmente e os resultados comparados entre si e com os de produções já conhecidas, conduzindo a uma proposta de agrupamentos que irá enquadrar o seu subsequente estudo individual.

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KEYWORDS: Renaissance majolica; Palace of Bacalhôa; Portuguese azulejos; João de Góis

Acknowledgements

We are thankful to Mr. Joe Berardo and to Mr. Renato Berardo, who authorized the sampling of the unique azulejo heritage of *Palácio e Quinta da Bacalhôa*; and to *Associação de Colecções | The Berardo Collection* and *Bacalhôa Vinhos de Portugal* for support to this project.

We thank *Museu Nacional do Azulejo* in Lisbon for authorization to use samples from their collections (identified here as Bac167 (MNAz, Inv. 53), Bac168 (MNAz, Inv. 54), and NSVida (MNAz, Inv. 138)) as well as *Igreja da Graça* in Lisbon (samples Graça I and Graça II, code Az013).

Instrumental research developed at *Laboratório Nacional de Engenharia Civil* (LNEC) within the scope of the national infrastructure E-RIHS.pt funded by FCT, the Portuguese Foundation for Science and Technology. LNEC Research Project 0202/1101/19014.

How to cite

MIMOSO, J.M. et al. – *A technical overview of 16th century majolica panels and patterned tiles from Palácio e Quinta da Bacalhôa in Portugal* in Studies in Heritage Glazed Ceramics, No. 3, LNEC, Lisbon, Portugal, 2021, pp. 109-142.

1. INTRODUCTION

The manufacture of azulejos in Portugal started, as far as we know presently, around 1560 by the workshop of Hans Goos, a Flemish potter established in Lisbon as *João de Góis* [1]. His younger brother, of whom we only know the adopted Portuguese name *Filipe de Góis*, was already active in 1565 [2] and was referred to in 1575 as a "master of glazed pottery" [1, p. 18] and so he may have also manufactured azulejos around that time. The productions of what we may call the "circle of João de Góis" (maybe only a single workshop, or maybe several sharing the same technology) encompasses a period thus starting around 1560 and lasting at least until the 1590s. Tiles imported to Portugal at around the same time from Antwerp and Seville, are characterized by different technological traits [3] and practically all the others we studied up to now could be fitted into the productions of this rather consistent circle. In a few instances, however, we found azulejos that, albeit similar, did not match closely the characteristics of the circle of João de Góis. Such cases were hypothesized to represent productions of others, maybe foreign potters temporarily in the country, or local potters whose production was still unknow. It should be kept in mind that the tally of professionals made in Lisbon in 1565 [2] mentions at least three other potters working with tin-glazed ceramics, presumably tableware but maybe also azulejos: João Fernandes, Pero Fernandes and Francisco Jácome (Flemish), the two last ones living near João de Góis.

We had now the opportunity to study all the panels, patterned linings and frame tiles at the palace and gardens of *Bacalhôa*, amounting to more than 50 different types. The domain now known as *Bacalhôa* in Vila Fresca de Azeitão (35 km SE of Lisbon) was acquired by Brás [Afonso] de Albuquerque in 1528 (all unreferenced historical mentions to Bacalhôa or Albuquerque stem from the study by Pleguezuelo et al. [4] that updated the original research by Joaquim Rasteiro published in 1895 [5]). Brás [Afonso] de Albuquerque modernized the palace and added a number of annexes. The year "1554", inscribed over the entrance gate, dates the completion of the work at the palace, while the Pleasure House by the lake was likely built soon afterwards. The palace, annexes and gardens are colourfully decorated with azulejos, initially of the Hispano-Moresque type and later with a profusion of majolica tiles. Patterned tiles of attractive and often unique designs line the walls of the Pleasure House, interrupted here and there by figurative panels of mythological or biblical inspiration, one of which - *Susanna and the Elders* - has the date "1565" inscribed over a painted portal, as if to date the tiling of the whole chamber where it is located (the central and most lavishly decorated room of the Pleasure House).

Brás de Albuquerque (1500-1581) was the only son of Afonso de Albuquerque (1452-1515), the military commander and governor of Portuguese India who gained the appellation *O Grande* (The Great). After the death of his father, King Manuel I (1469-1521) instructed that Brás should take his father's name, thus becoming *Afonso de Albuquerque*. After Brás' death, a judicial dispute for the property ensued leading to an unstable situation until 1609 when the rights of ownership were definitively settled in favour of Maria de Mendonça e Albuquerque, married to a Jerónimo Manuel from whose nickname, *Bacalhau*, stems the now prestigious name of the property itself. Given the troubled situation after Brás [Afonso] de Albuquerque's death, the year 1581 likely sets a chronological limit to the 16th century azulejo linings which art historians have singled amidst the *chefs-d'oeuvre* of renaissance majolica tile decorations. Their provenance, foreign or domestic, and authorships have, however, remained disputed.

Based on the letters "TOS" painted on a fragment of a baseboard slab [6, Estampa XXXVII] Joaquim Rasteiro attributed, rather rashly, all the figurative panels to a Francisco de Matos [5, pp. 34-35] who, in 1584, signed one of the panels at *Igreja de São Roque* in Lisbon [7, p. 95]. Santos Simões [8] gave proof of his proverbial insight by systematizing the linings and panels and attributing them to Flemish potters working in Lisbon, with collaboration of a local painter. On his attempted systematization, Santos Simões considered firstly a division between patterned linings and figurative panels and slabs, and then he discussed further separations based on style and the quality of the drawing and painting skills. However, as Reynaldo dos Santos admitted in 1957, no significant advances seemed possible without supporting documental evidence [9].

Resourcing to modern analytical means, we studied the panels and patterned tiles of Bacalhôa based on their manufacturing technology and accordingly attempted an objective systematization such as was not feasible when Reynaldo dos Santos and Santos Simões wrote their pioneering books. Besides the sheer number of types identified, the study was made particularly difficult by the mixing of tiles of the same pattern having certainly different provenances or chronologies and by the use of clays, glaze formulations or firing schedules unknown to us, that did neither match the familiar parameters of the circle of João de Góis, nor the known Flemish or Spanish productions. Since it is not viable, in this first approach, to encompass all the types in a single paper, the study concentrates on the azulejo panels still extant, complete or incomplete, still *in situ* or surviving in fragments but whose original setting is known, as well as the majolica baseboard slabs decorated with *grotteschi* or playful children. Also included were the patterned tiles used in the palace and the Pleasure House, of which, whenever several versions of the same pattern were clearly present, only the presumably older one was considered. The decision in such cases, albeit subjective, was based on the assumption that the originals would be superior to the copies and therefore the version considered best in terms of the quality of the pigments, the skill of the painter and the absence of remarkable defects caused by the manufacturing technique was selected.

The panels, slabs and patterned tiles studied for the purpose of a systematization of types are identified and illustrated in Figure 1 and referenced by the codes of the local museum, the *Museu do Palácio da Bacalhôa*: **301-***nn* (in the case of panels) and **301-P***nn* (in the case of patterned tiles), in which "nn" represent the itemization numerals. It will be noted that two flowerbed panels with fauns (coded 301-23 and 301-27 in Figure 1), once decorating the Secret Garden, ante-chamber to the Pleasure House, and now removed for restoration, were considered independently. We had the possibility to examine closely the remaining panels of this set and at least two different types were seemingly apparent, each represented by one of the panels considered here. Even neglecting the obviously distinct skills of the painters, the biscuits were different and so were the backside markings. On the other side, two tile fragments (coded 301-96 in Figure 1) were considered together as being remains of a coat of arms. This was recognized to be that of Albuquerque's first wife Maria de Noronha [4] and the fragments are therefore referred to as the "Noronha Coat of Arms panel".

Samples were obtained and, once prepared, they were observed by scanning electron microscopy to define their characteristic morphologies, and the chemical composition of the biscuit and glaze of each was determined by energy dispersive X-ray spectroscopy. The panels, slabs and azulejos were then tentatively clustered based on morphology and compositional results, following the methodology previously used on the singling of 16th century azulejo productions by the workshops of Lisbon [3]. Once defined, the clusters were examined for consistency and compared between them and with similar results from panels of known Portuguese origin and similar chronology [10].

A comprehensive study of panels and patterned tiles, including those in the garden, India House and the frame tiles will be published at later times in dedicated articles.

***Susanna and the Elders* (301-8)**

***Abduction of Hippodamia* (301-9)**

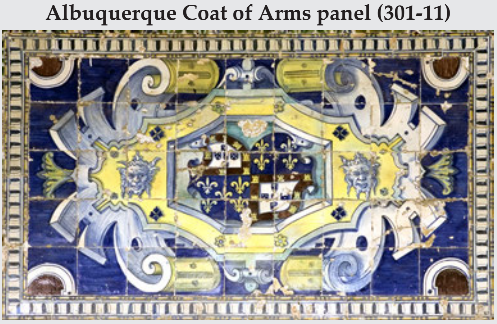
Tagus river god (301-10)

Loggia river gods panels (example: Douro 301-5)

***Rape of Europa* (301-6)**

Mascarons (301-7)

Albuquerque Coat of Arms panel (301-11)



Noronha Coat of Arms panel (301-96)

Flowerbed panel with fauns (301-23)

Flowerbed panel with fauns (301-27)

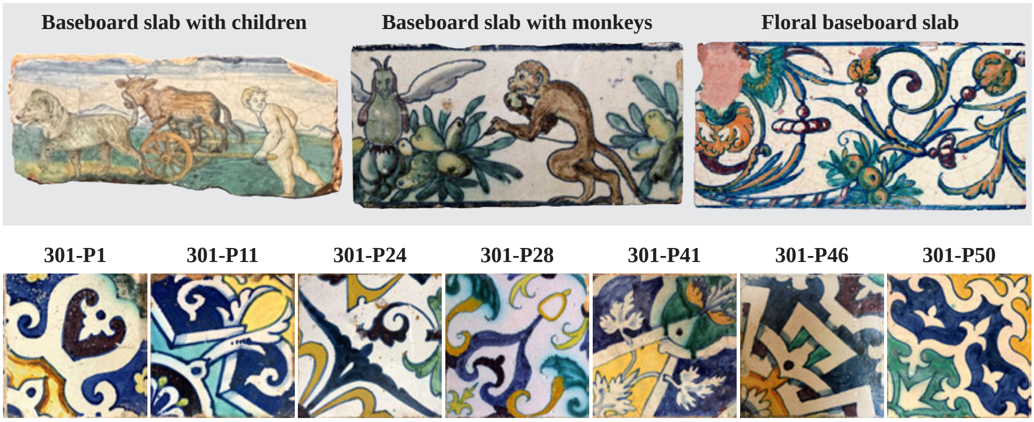


Figure 1. Bacalhôa panels, baseboard slabs and patterned tiles addressed by this paper (images: slab with monkeys and floral slab - Museu Nacional do Azulejo; all others © Associação de Colecções | The Berardo Collection).

2. EXPERIMENTAL

2.1. Samples

Very small samples were taken from the panels, the patterned tiles and the baseboard slabs. Whenever possible, and to improve the accuracy of the analytical results, larger samples were taken from fragments in storage in the reserves of the local museum. All samples were identified by a technical reference **Bac***nnn* to which was added an additional numeral whenever the sample was later split.

Table 1 includes references and data on each case studied. The first column (*Identification*) repeats the codes in Figure 1 and includes a short name for panels and baseboard slabs that will be used in the diagrams and tables for their easier identification. The second column (*Description/Location*) presents the name of the panels and the location of the patterned tiles. The third column (*Sample Ref.*) includes the technical reference of the samples collected and prepared for observations and analyses. In the case of patterned tiles, the underlined reference is always of the main sample that was collected directly from the walls. The remaining columns include the tile dimensions (when measurable) and indicate how many samples and measurements were averaged in the semi-quantification of the chemical composition of each. In Tables 2 and 3 each value represents the average compositions of all samples and measurements of a single panel or pattern, except in the case of the five panels in the Loggia of the River Gods which, because of their presumed common provenance and chronology, confirmed by the morphological and compositional similarities (to be addressed in a future paper) were, for the sake of legibility of the results, averaged as a single panel composed of five parts.

The panels in *Igreja da Graça* (Graça Church in Lisbon), tentatively dated from 1565 to 1570 [11], were used for comparative purposes. Their study, published in 2019, identified two variants readily separable by the clays used: one includes the tiles bearing the monogram of João de Góis and named "Graça I"; the other includes the 16th century frame tiles with the Greek pattern and the remainder of the panel composed by tiles other than of the first type, named "Graça II" [11, fig.13; 12, p 32]. Besides both these types, the other panel presumably signed by João de Góis, the superb *Nossa Senhora da Vida* (NSVida - Our Lady of Life) conserved at *Museu Nacional do Azulejo* in Lisbon, probably dating from the second half of the 1570s [13] was also included as a reference for comparison.

Table 1. Short name of items, locations, sample references, dimensions of tiles, and number of results averaged

Identification / short name	Description/Location	Sample Ref.	Approximate tile dimensions (cm)	Nr. of samples	Total nr. of results
301-8 / Susanna	*Susanna and the Elders*	Bac001	13 x 13 x ?	3	5 (g) 3 (b)
301-9 / Hippodamia	*Abduction of Hippodamia*	Bac024	13 x 13 x 2.5	2	3 (g) 2 (b)
301-10 / Tagus	Tagus river god	Bac006, 017, 083	13 x 13 x 2.5	4	9 (g) 9 (b)
301-1; -2;-3;-4;-5 / River gods	Palace Loggia of the River Gods [4]	Bac004, 010, 011, 012, 013	12.5 x 12.5 x ?	10	17 (g) 13 (b)
301-6 / Europa	*Rape of Europa*	Bac015	13 x 13 x ?	2	6 (g) 2 (b)
301-7 / Mascarons	Mascarons	Bac016	13 x 13 x ?	2	2 (g) 2 (b)
301-11 / Shield 1	Albuquerque coat of arms	Bac018	13 x 13 x ?	2	3 (g) 1 (b)
301-96 / Shield 2	Noronha coat of arms	Bac007, 009, 104	13 x 13 x 2.0	4	8 (g) 4 (b)
301-23 / Fauns 1	PH Secret Garden flowerbed	Bac107	18 x 18 x 2.5	1	2 (g) 1 (b)
301-27 / Fauns 2	PH Secret Garden flowerbed	Bac008, 023	18 x 18 x 2.5	2	2 (g) 2 (b)
Children slab	PH (?) baseboard slab with children	Bac005, 022	26 x 13 x 2.5	2	2 (g) 2 (b)
Monkey slab	Skirting PH Room 5	Bac167	26 x 13 x 2.5	1	3 (g) 1 (b)
Floral slab	Skirting PH Room 1	Bac168	26.5 x 13 x 2.5	1	2 (g) 1 (b)
301-P1 / P1	Pattern used in the Palace Coat-of-Arms Room	Bac109, 120	13 x 13 x 1.5	3	3 (g) 3 (b)
301-P11 / P11	Pattern used in front court yard and PH exhibition panels	Bac082, 113, 123	13 x 13 x 2.0	4	11 (g) 6 (b)
301-P24 / P24	Pattern used in Palace	Bac144	14.5 x 14.5 x 1.5	1	3 (g) 3(b)
301-P28 / P28	Pattern used in Palace	Bac014, 127, 141	15.5 x 15.5 x 1.5	4	3 (g) 4 (b)
301-P41 / P41	Pattern used in PH Room1	Bac100, 125, 138, 143	12 x 12 x 2.0	3	3 (g) 3 (b)
301-P46 / P46	Pattern used in PH Room 3	Bac003, 040, 078; 142	12 x 12 x 1.5 (Bac078 & Bac142)	6	9 (g) 6 (b)
301-P50 / P50	Pattern used in PH Room 5	Bac042, 085, 103	13 x 13 x 1.5	4	4 (g) 4 (b)

Notes: PH = Pleasure House:
- underlined samples were collected from the walls;
- dimensions given are rounded estimates of the average. There are variations of ca. ± 0.5 cm in face dimensions and ca. ± 0.3 cm in thickness;
- g = glaze; b= biscuit.

2.2. Methods, instrumental means and software

The azulejo samples were stabilized in epoxy resin, lapped and polished to obtain a flat cross-section for observation and analysis by scanning-electron microscopy coupled with an X-ray energy-dispersive spectrometer (SEM-EDS).

SEM observations and EDS analyses were made at LNEC using a TESCAN MIRA 3 field emission microscope combined with a BRUKER XFlash 6|30 EDS system. The samples were uncoated and the observations were made in backscattered electrons mode (BSE), with a chamber pressure of typically 10 Pa, at an accelerating voltage of 20 kV with the samples sections at a distance of 14 ± 1 mm from the detector. SEM images were typically acquired at magnifications of 350x and 700x for the glaze and 3,000 to 6,000x for the biscuit.

The selection of areas for EDS quantification avoided large inclusions in the glaze or biscuit representing more than ca. 5% of the full selected area. From our previous experience, the adequate minimum measurement areas are 200 x 200 μm for glazes and 500 x 500 μm for biscuits. In general, multiple measurements were made and in such case the results are averages and smaller non-overlapping areas may be used to the same effect. Whenever possible, the analyses were performed on white glazes to avoid interference from elements diffused from the blue, green or violet pigments which, when present, were neglected. The yellow pigments remain at the surface and therefore do not present the same problem. Still, in the case of zinc-bearing yellow pigments, the analyses must be performed at a safe distance from the colour.

Having in view comparisons through Principal Component Analysis (PCA), ancillary elements usually representing less than 1% of the compositions, such as magnesium (Mg) and iron (Fe) in the glazes, were not included in the tables of results. Calcium (Ca) was also excluded from the glazes because, as will be seen further on, in cases such as one of the flowerbed panels with fauns and the baseboard slabs, the addition of a powdery Ca-rich substance (perceived as clouds of minute dark inclusions in some samples of Figure 2) results in high Ca contents that might overshadow the variability of other elements in the graphs.

The quantification of tin (Sn) in the glazes may be problematic because the aggregation of crystals often results in a large variance. That problem was dealt with by using larger areas whenever aggregation was visually detected in the SEM images or, when that was not possible, averaging the results of multiple analyses on different areas.

The amount of oxygen (O) was calculated through the remaining elements stoichiometry of their most commonly considered oxides (Na_2O, MgO, Al_2O_3, SiO_2, K_2O, CaO, Fe_2O_3, SnO_2, PbO) and the result was normalized to 100 %.

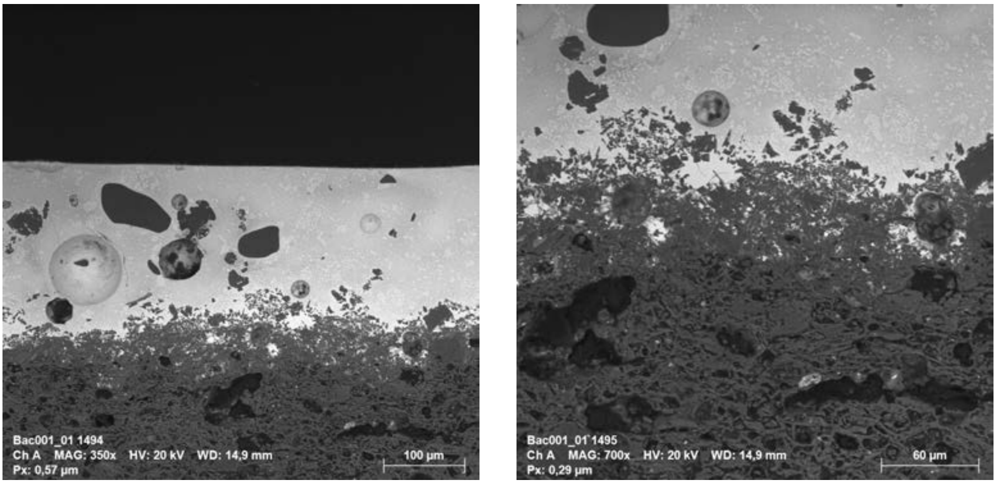
PCAs of EDS results were made using the SPSS® software platform by IBM Analytics.

3. RESULTS

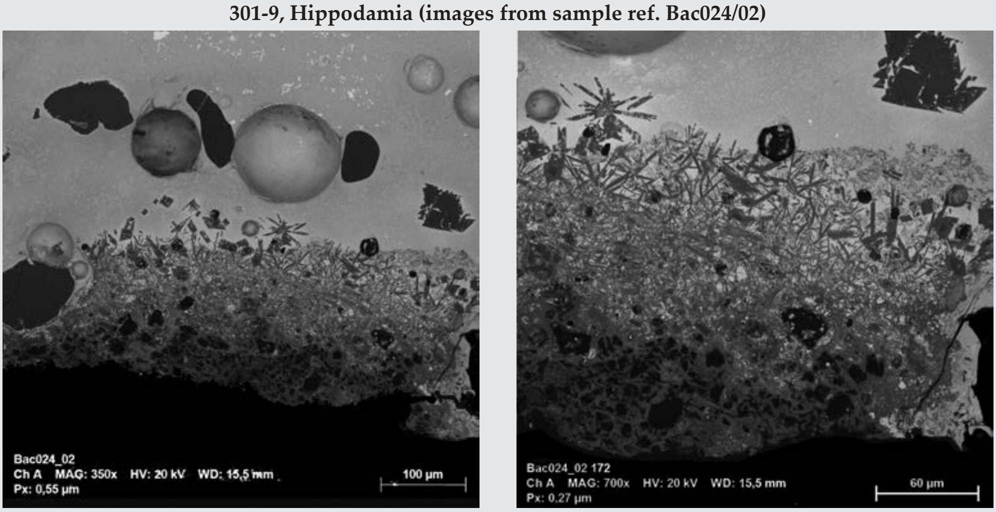
3.1. Morphology of the glazes

Figure 2 depicts, at the same magnifications for comparison purposes, SEM images of the samples showing the main micro-morphological characteristics generally associated with the glazes and their interfaces. The light grey area on top is the glaze, while the dark grey area corresponds to the biscuit. Because of its colour, the inclusions in the glaze are conspicuous: gas bubbles retained in the glass, grains of sand (larger compact dark inclusions, usually with rounded edges) and bits of feldspars, often in disaggregation. The white spots in the midst of the glaze are crystals of the opacifier (tin oxide), while a continuity of similar white spots near the surface may correspond to the lead-rich yellow pigments.

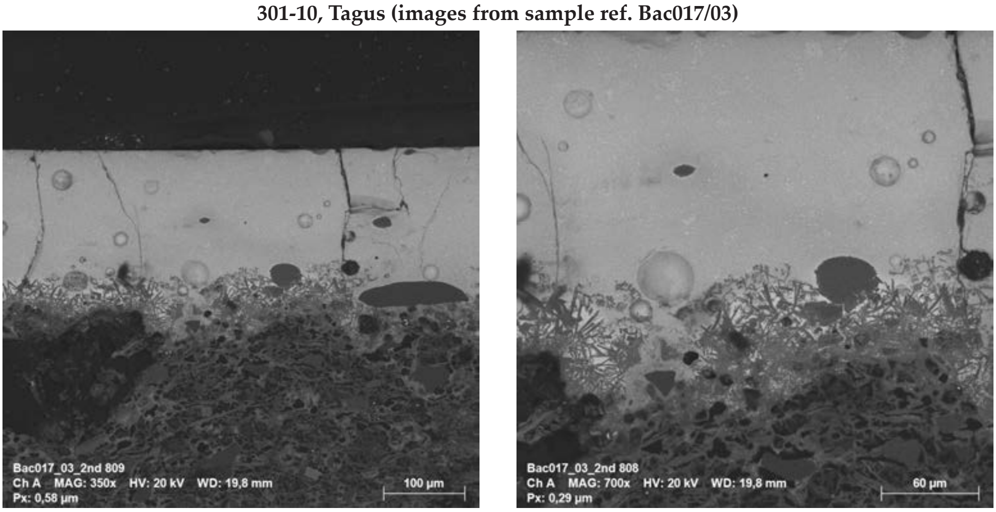
301-8, Susanna (images below from sample ref. Bac001/01)



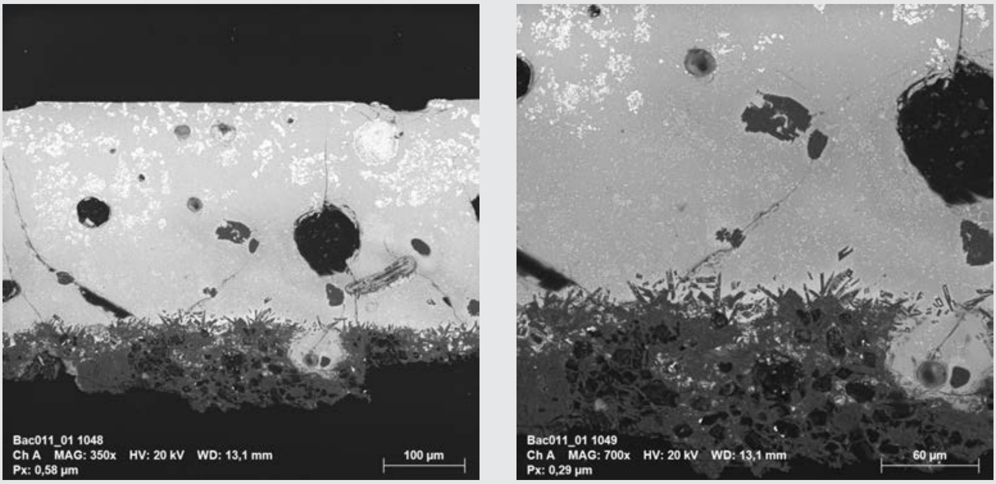
301-9, Hippodamia (images from sample ref. Bac024/02)



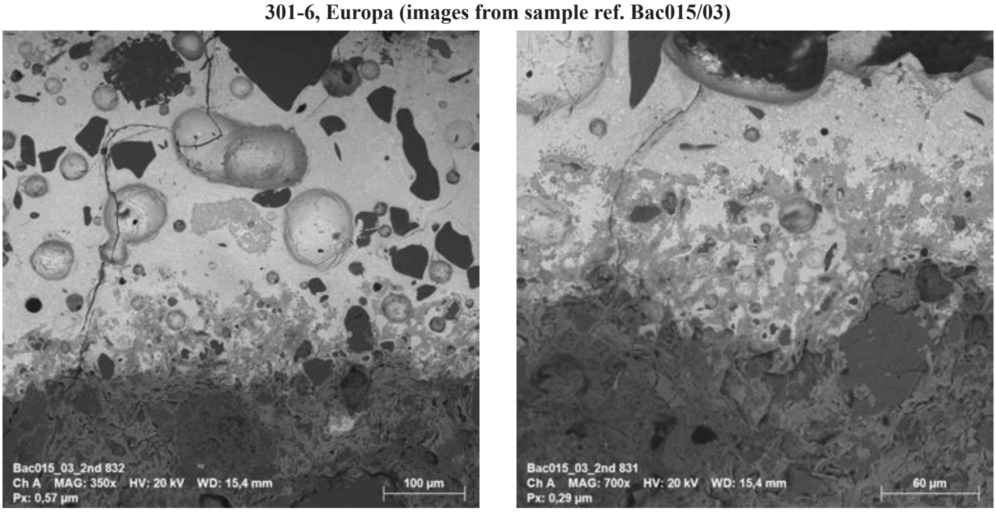
301-10, Tagus (images from sample ref. Bac017/03)



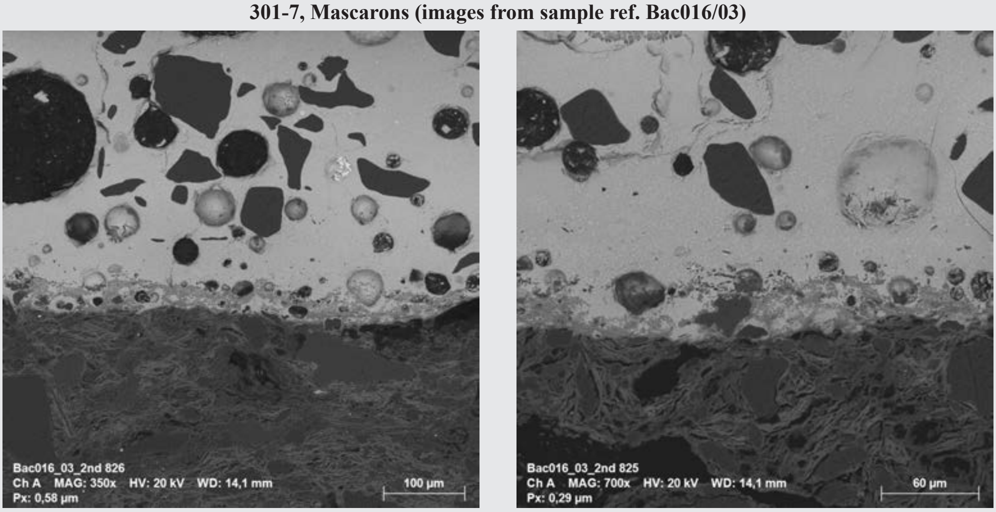
River gods (in the images 301-2, Mondego - sample ref. Bac011/01)



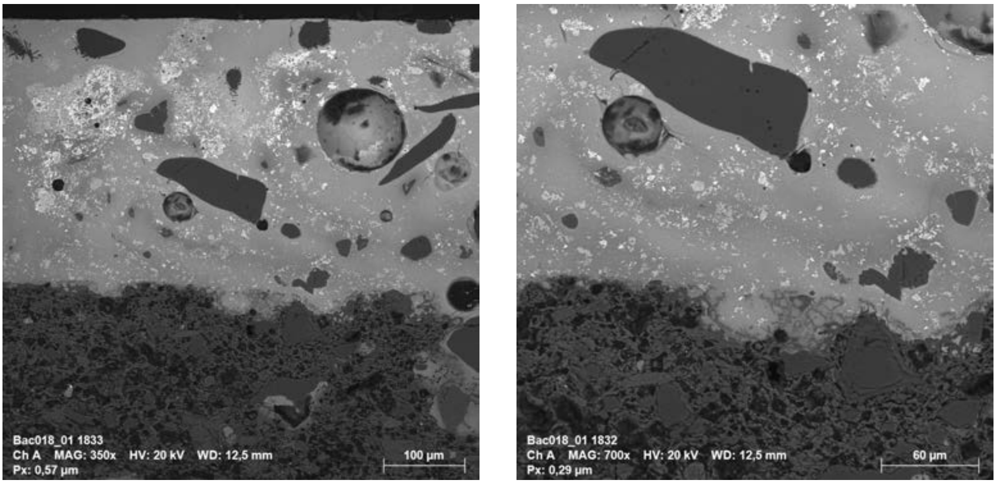
301-6, Europa (images from sample ref. Bac015/03)



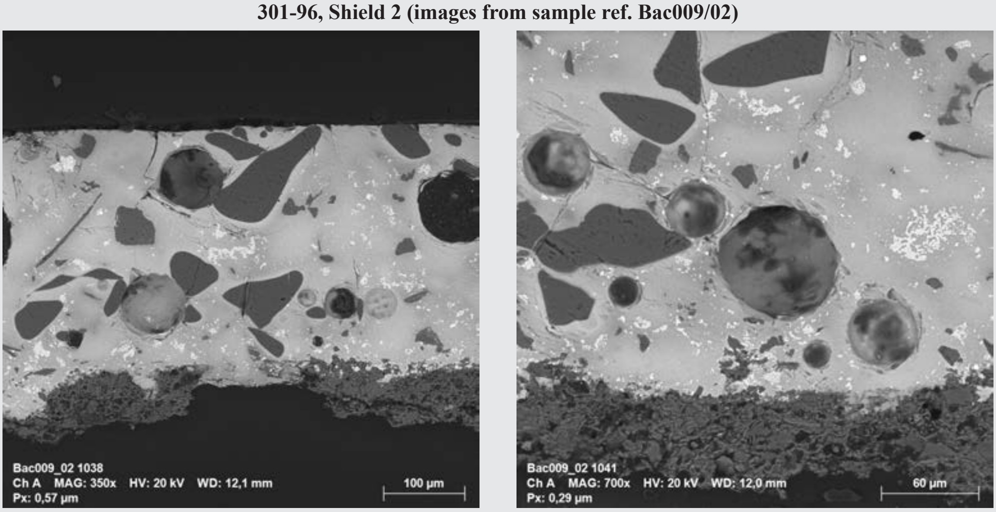
301-7, Mascarons (images from sample ref. Bac016/03)



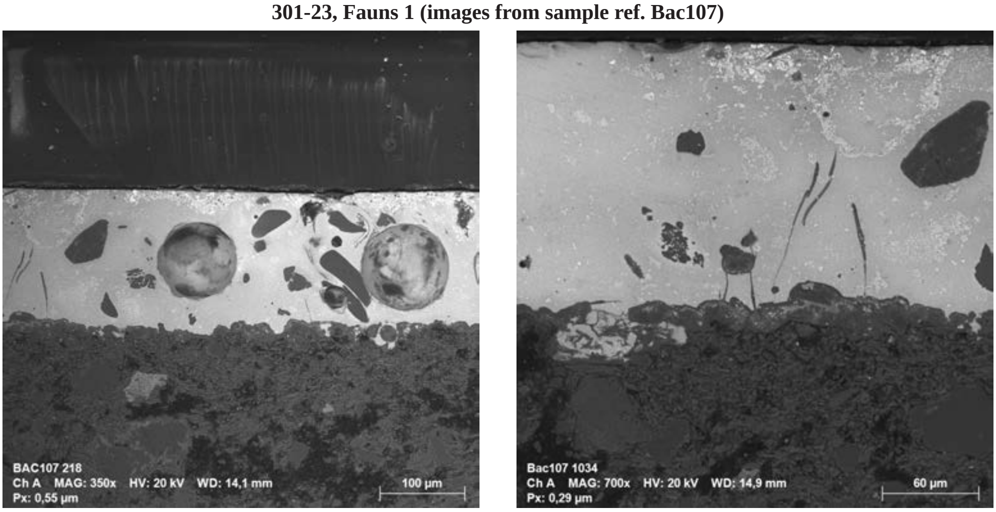
301-11, Shield 1 (images from sample ref. Bac018/01)



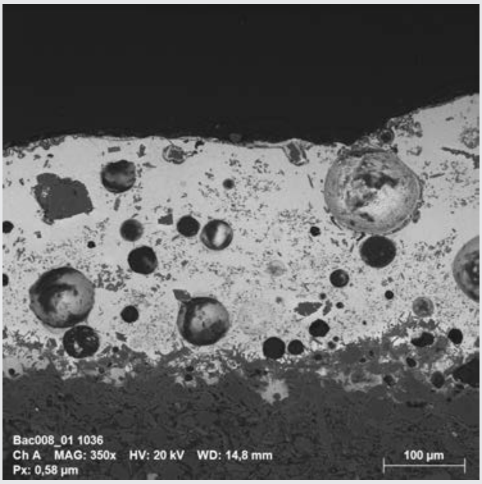
301-96, Shield 2 (images from sample ref. Bac009/02)

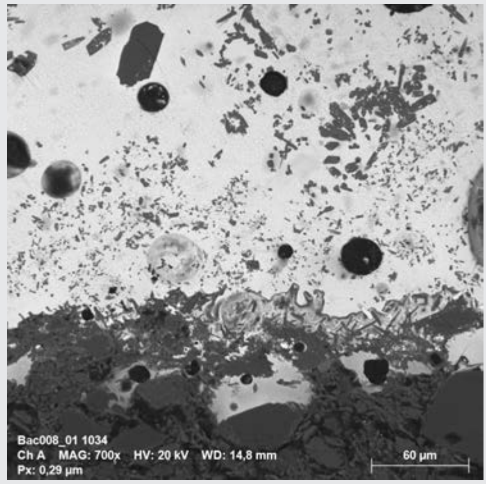


301-23, Fauns 1 (images from sample ref. Bac107)

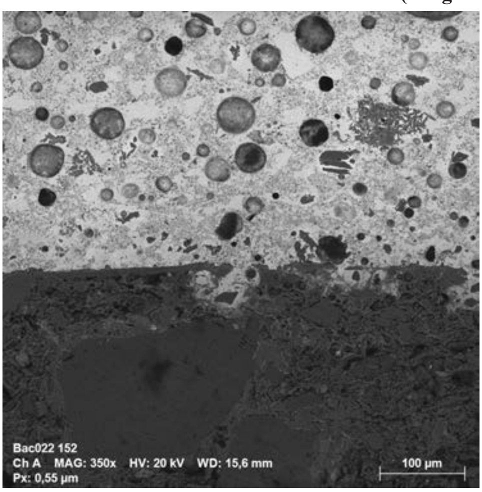


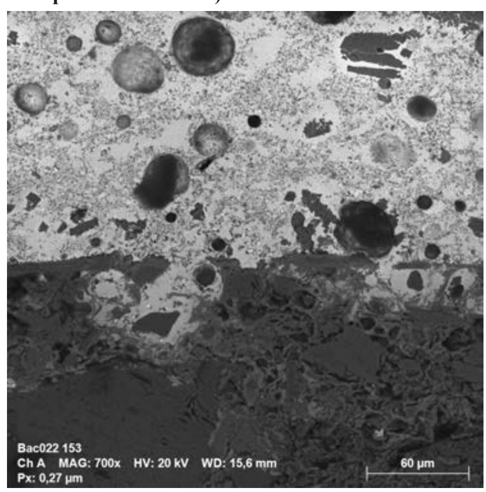
301-27, Fauns 2 (images from sample ref. Bac008/01)



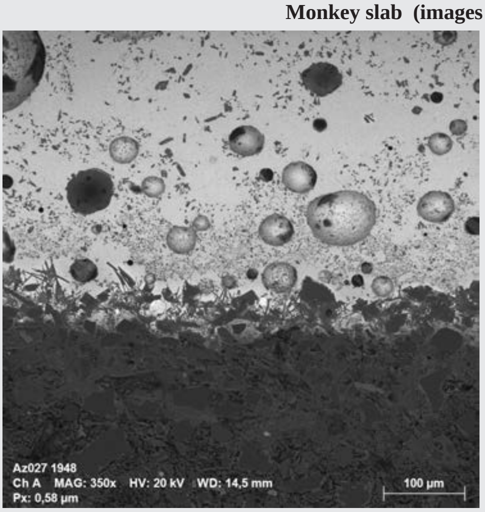


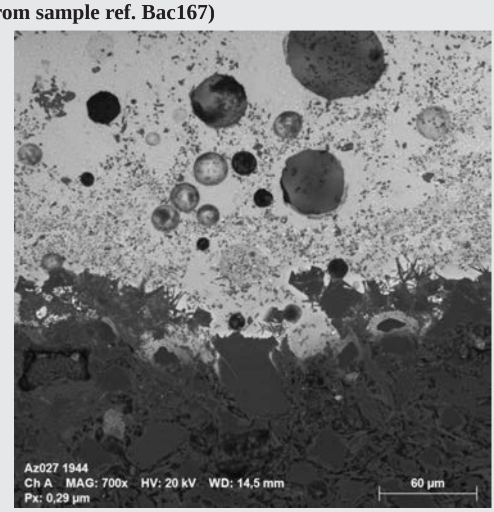
Children slab (images from sample ref. Bac022/01)



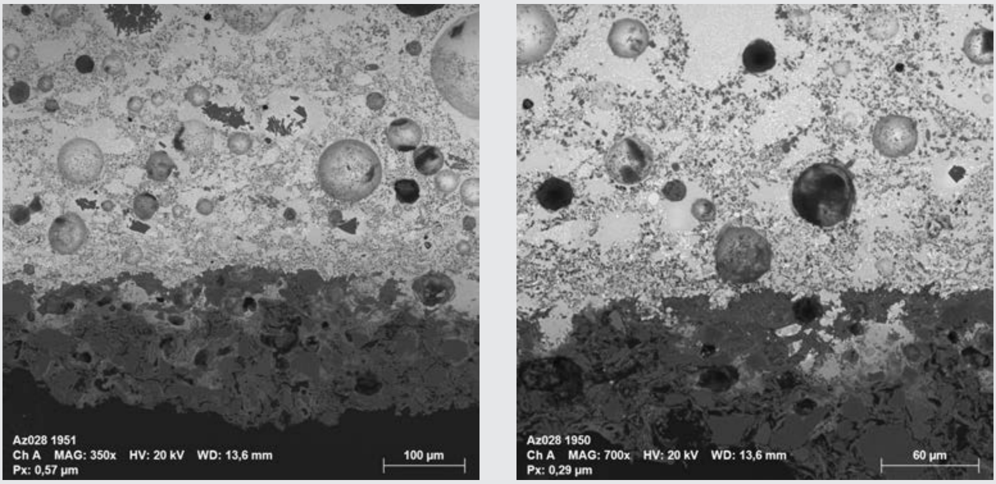


Monkey slab (images from sample ref. Bac167)

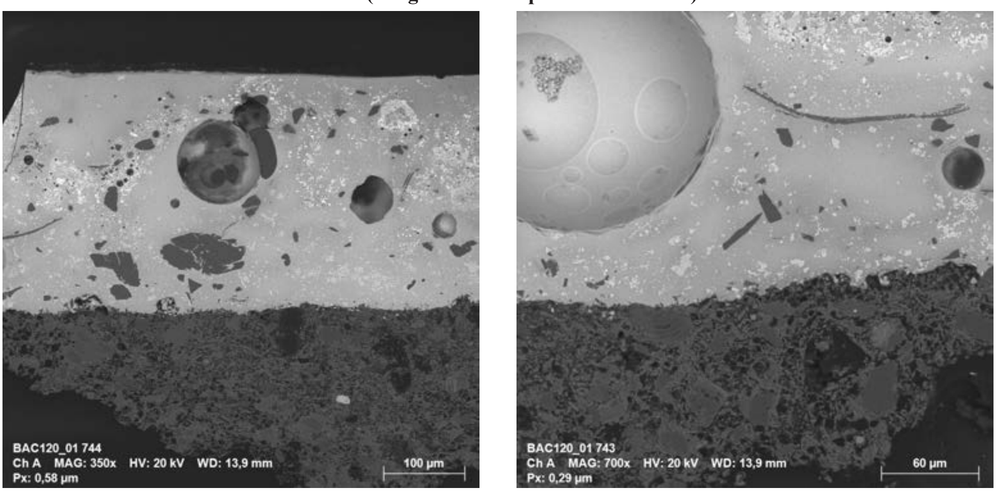




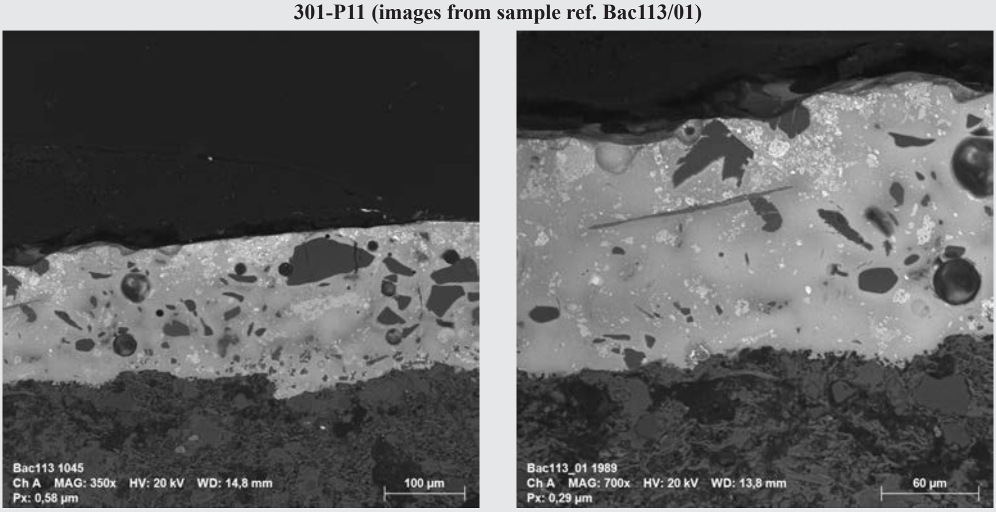
Floral slab (images from sample ref. Bac168)



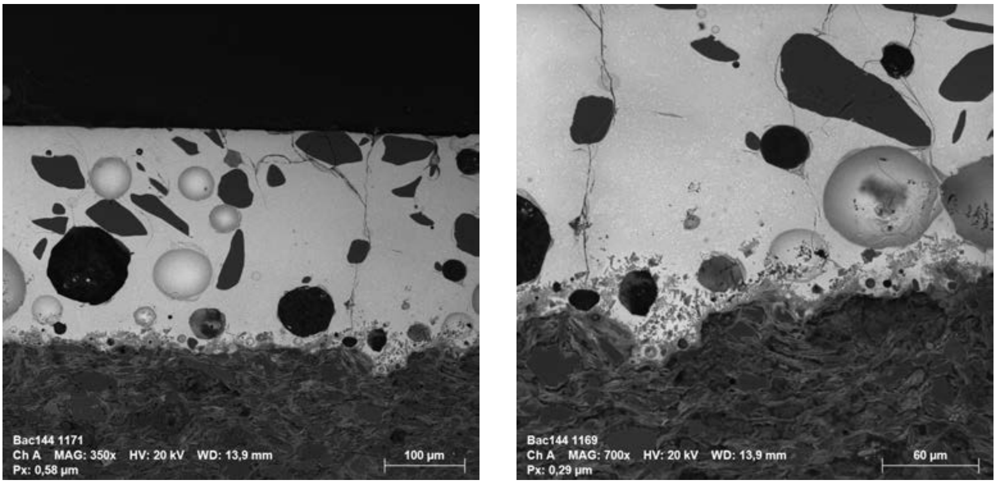
301-P1 (images from sample ref. Bac120/01)



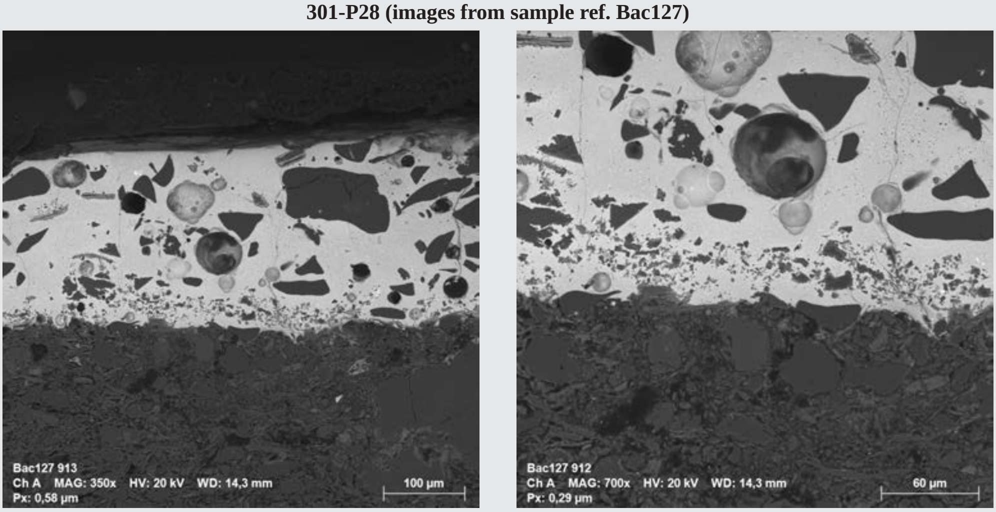
301-P11 (images from sample ref. Bac113/01)



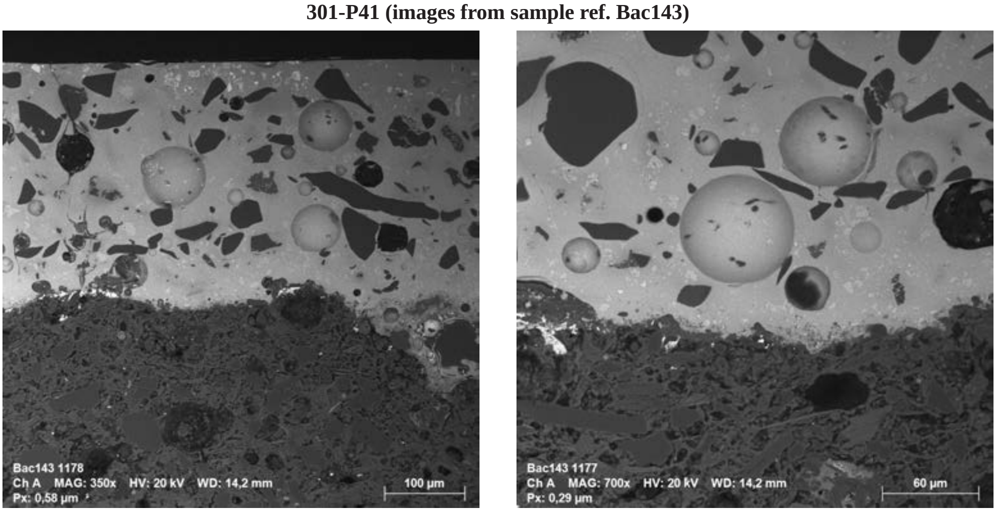
301-P24 (images from sample ref. Bac144)



301-P28 (images from sample ref. Bac127)



301-P41 (images from sample ref. Bac143)



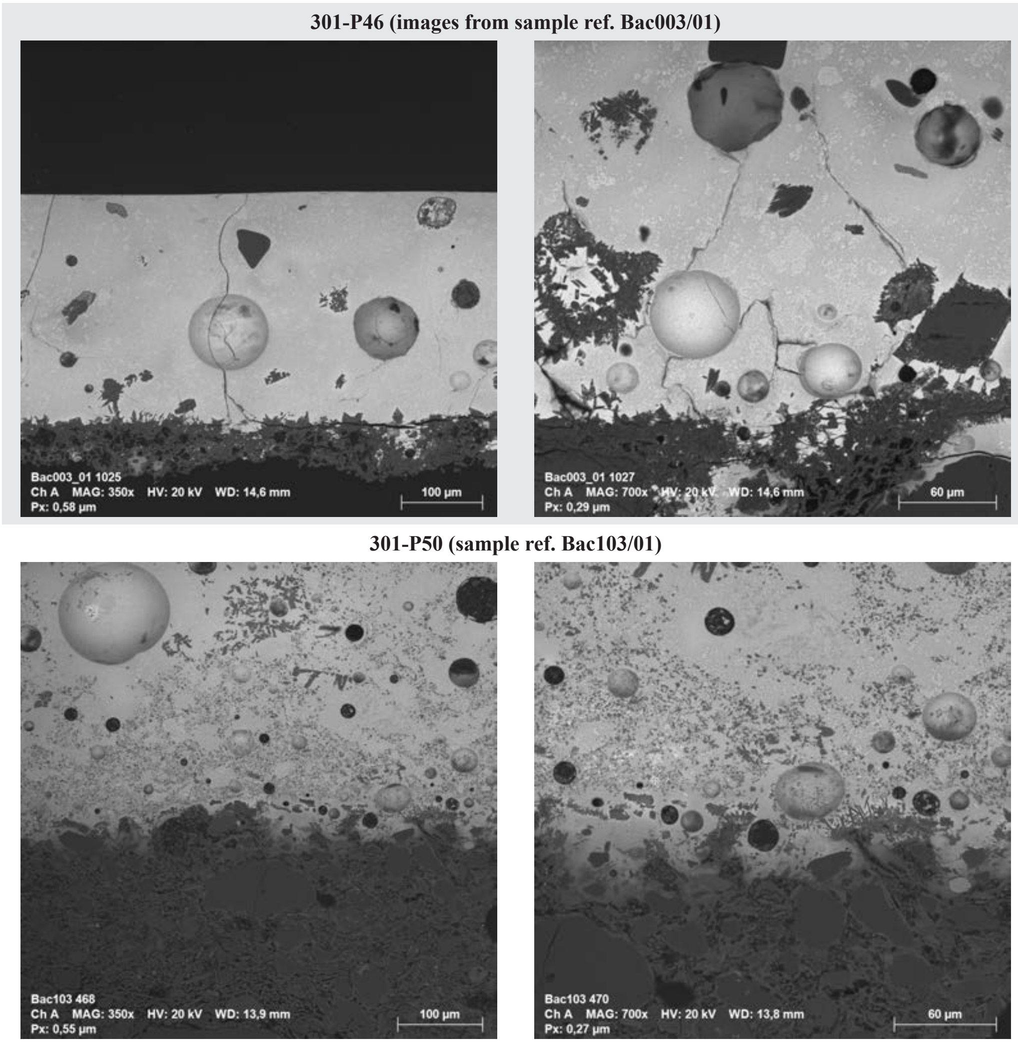


Figure 2. SEM-BSE images showing the main micro-morphological characteristics of the Bacalhôa azulejo glazes (left side: 350 x) and biscuit-glaze interfaces (right side: 700 x) (images: LNEC).
Note: samples were chosen that offered clear and representative images of each case.

3.2. COMPOSITION OF THE GLAZES

Table 2 includes the semi-quantitative results of analyses of the glazes by EDS in weight %. Each result is the average of all determinations of the element in the panel, slab or frame tiles, given with the standard deviation. The important silicon to lead ratios (Si/Pb), related with the minimum temperature at which the glazes may be properly fired, have been determined and are also included in the table, which is sorted by their descending values.

Table 2. Average values (with standard deviations in parentheses) for the semi-quantitative composition of the glazes of the Bacalhôa samples determined by EDS (values in wt.% with oxygen obtained by stoichiometry and elements normalized to 100%) with high Si/Pb and low Si/Pb ratios shaded in blue and in red

PANEL/TILES	O	Na	Al	Si	K	Sn	Pb	Si/Pb
301-96 Shield 2	38.13	2.63 (0.46)	3.08 (0.51)	26.52 (0.99)	6.89 (0.42)	5.69 (1.37)	17.05 (2.42)	**1.56**
301-11 Shield 1	36.76	2.87 (0.75)	3.21 (0.24)	25.12 (2.24)	5.64 (0.50)	5.66 (0.86)	20.74 (4.15)	**1.21**
301-P11	35.63	0.93 (0.10)	2.66 (0.29)	24.78 (1.57)	6.40 (0.70)	5.79 (1.96)	23.82 (4.09)	**1.04**
301-P1	34.81	1.93 (0.31)	2.55 (0.74)	23.27 (0.91)	5.77 (1.19)	9.02 (1.07)	22.66 (3.13)	**1.03**
301-23 Fauns 1	35.30	1.95 (0.13)	2.52 (0.25)	24.27 (1.96)	5.32 (0.00)	6.62 (1.37)	24.02 (2.01)	**1.01**
301-P41	35.25	2.59 (0.57)	1.38 (0.13)	25.44 (2.00)	3.79 (0.76)	4.80 (0.60)	26.75 (4.22)	**0.95**
301-P46	30.74	1.64 (0.50)	2.41 (0.74)	19.16 (1.62)	2.76 (0.66)	11.86 (2.57)	31.43 (5.88)	**0.61**
301-8 Susanna	30.50	1.19 (0.16)	2.26 (0.36)	19.41 (2.00)	3.14 (0.32)	10.18 (1.82)	33.32 (4.02)	**0.58**
301-P50	28.65	2.56 (0.26)	1.78 (0.37)	16.41 (1.33)	1.50 (0.29)	17.55 (3.50)	31.54 (1.76)	**0.52**
301-10 Tagus	29.23	1.21 (0.53)	2.10 (0.24)	18.27 (1.61)	2.70 (0.74)	10.28 (3.16)	36.21 (4.02)	**0.50**
301-9 Hippodamia	29.58	1.42 (0.10)	3.14 (0.27)	17.95 (0.91)	2.46 (0.35)	9.54 (0.93)	35.91 (2.84)	**0.50**
Floral slab	27.60	2.05 (0.08)	1.59 (0.02)	15.66 (0.67)	1.95 (0.09)	17.08 (2.91)	34.06 (4.81)	**0.46**
Loggia river gods	28.10	0.71 (0.39)	1.92 (0.42)	17.76 (1.69)	2.12 (0.68)	8.66 (1.65)	40.72 (5.02)	**0.44**
301-P28	27.80	0.79 (0.40)	2.02 (0.11)	17.91 (1.31)	2.19 (0.25)	5.62 (1.01)	43.68 (2.72)	**0.41**
Children slab	26.65	2.17 (0.54)	1.77 (0.34)	14.85 (1.28)	1.23 (0.16)	15.74 (4.84)	37.58 (1.71)	**0.40**
301-27 Fauns 2	26.98	1.76 (0.17)	2.01 (0.10)	15.76 (1.54)	1.62 (0.57)	11.88 (2.56)	39.99 (0.62)	**0.39**
301-P24	26,42	0.25 (0.04)	3.09 (0.40)	14.65 (1.42)	1.01 (0.13)	16.26 (2.81)	37.71 (6.10)	**0.39**
301-6 Europa	27.03	0.26 (0.07)	2.74 (0.13)	15.59 (0.54)	1.11 (0.06)	9.07 (0.85)	44.81 (1.60)	**0.39**
301-7 Mascarons	25.50	0.16 (0.07)	2.60 (0.39)	14.16 (0.26)	0.67 (0.08)	12.84 (0.19)	44.07 (0.99)	**0.32**
Monkey slab	24.84	1.59 (0.09)	1.65 (0.07)	14.06 (0.44)	1.17 (0.03)	11.37 (1.58)	45.32 (0.77)	**0.31**

3.3. Composition of the biscuits

Table 3 includes the semi-quantitative results of analyses of the biscuits by EDS in weight %. Lead (Pb) occurs in most cases deriving from diffusion into the biscuit when the raw glaze is applied. Its content was determined but not considered because it is not part of the natural composition of the biscuit and depends on the proximity to the interface. The presence of lead renders the quantification of sulphur (S) doubtful because of a superposition of X-ray elemental peaks and therefore it too was not considered. In each case the ratio of the calcium to silicon contents (Ca/Si), that in part defines the suitability of the clay to glazing, has been determined and the table has been sorted by its descending value.

Table 3. Average values (with standard deviations in parentheses) of the semi-quantitative composition of the biscuits of the Bacalhôa samples determined by EDS (values in wt.% with oxygen obtained by stoichiometry and elements normalized to 100%) with high and low Ca/Si ratios shaded in blue and in red

PANEL/TILES	O	Na	Mg	Al	Si	K	Ca	Fe	Ca/Si
301-P11	41.66	0.98 (0.27)	4.12 (0.60)	6.96 (0.58)	19.02 (0.73)	1.24 (0.28)	22.43 (2.17)	3.61 (0.23)	**1.18**
301-P1	42.13	1.03 (0.12)	3.64 (0.29)	7.54 (0.60)	19.74 (1.33)	1.58 (0.24)	19.86 (2.41)	4.48 (0.12)	**1.01**
301-96 Shield 2	42.74	0.96 (0.20)	4.47 (1.01)	7.23 (0.31)	20.83 (2.96)	1.55 (0.73)	18.33 (3.99)	3.89 (0.29)	**0.88**
301-P41	43.27	1.29 (0.16)	2.83 (1.13)	8.02 (0.03)	21.89 (2.17)	1.57 (0.43)	17.06 (3.48)	4.05 (0.22)	**0.78**
301-9 Hippodamia	43.26	1.09 (0.04)	4.73 (1.93)	9.40 (0.53)	20.28 (0.38)	1.64 (0.03)	15.16 (2.19)	4.45 (0.23)	**0.75**
301-11 Shield 1	43.69	0.86	4.44	7.69	22.23	1.22	15.91	3.95	**0.72**
301-P46	43.69	1.03 (0.30)	4.24 (0.81)	9.13 (1.53)	21.38 (1.79)	1.37 (0.36)	14.97 (1.24)	4.21 (0.44)	**0.70**
Loggia river gods	43.49	0.87 (0.25)	4.75 (0.77)	8.31 (0.82)	21.59 (1.38)	2.27 (0.50)	14.55 (2.69)	4.18 (0.38)	**0.67**
301-8 Susanna	43.89	1.83 (0.67)	4.71 (0.10)	8.02 (0.49)	22.37 (0.57)	1.23 (0.34)	13.88 (0.60)	4.07 (0.50)	**0.62**
301-10 Tagus	43.99	1.14 (0.39)	4.75 (1.40)	8.89 (0.75)	22.18 (1.61)	2.30 (0.43)	12.60 (1.89)	4.15 (0.43)	**0.57**
301-23 Fauns 1	44.64	0.81	4.88	9.90	22.52	1.59	12.10	3.56	**0.54**
301-P24	45.28	0.85 (0.08)	1.20 (0.06)	9.45 (0.35)	25.67 (0.08)	3.14 (0.16)	9.61 (0.67)	4.80 (0.12)	**0.37**
301-27 Fauns 2	45.99	1.38 (0.29)	0.99 (0.22)	8.86 (0.03)	27.55 (0.64)	3.06 (0.35)	8.74 (0.31)	3.44 (0.22)	**0.32**
Floral slab	45.80	1.86	1.33	7.19	28.43	3.79	8.64	2.96	**0.30**
301-P50	46.40	1.76 (0.68)	1.11 (0.35)	8.58 (0.22)	28.41 (0.53)	2.62 (0.17)	8.28 (0.91)	2.83 (0.37)	**0.29**
Monkey slab	46.45	1.35	0.75	7.67	29.25	2.92	8.41	3.19	**0.29**
301-P28	46.63	1.06 (0.23)	0.89 (0.29)	7.72 (0.57)	29.46 (2.24)	2.71 (0.73)	8.45 (2.17)	3.08 (0.56)	**0.29**
Children slab	46.18	1.45 (0.44)	1.13 (0.05)	8.91 (0.23)	27.87 (0.54)	3.24 (0.35)	7.75 (1.20)	3.48 (0.10)	**0.28**
301-7 Mascarons	46.39	0.97 (0.09)	1.22 (0.11)	9.81 (0.14)	27.41 (0.14)	2.65 (0.35)	6.86 (0.11)	4.69 (0.43)	**0.25**
301-6 Europa	46.56	1.17 (0.23)	1.29 (0.60)	10.52 (0.09)	27.54 (1.43)	3.79 (1.05)	4.26 (2.28)	4.87 (0.00)	**0.15**

4. DISCUSSION

4.1. Technology

Considering the morphology of the glazes in Figure 2, one group, including all the baseboard slabs, one of the flowerbed panels with fauns (301-27, "Fauns 2") and the pattern 301-P50 from Room 5 of the Pleasure House, is immediately individualized because the whole glaze is speckled with dusty inclusions and that characteristic constitutes a key-marker, enough to discriminate a class (Table 4). The same type of inclusions has previously been found in the *Nossa Senhora da Vida* panel [13, pp 83-84] and the 1584 lining of *Capela de São Roque* (St. Roch's Chapel in the church of the same dedication in Lisbon) [7, pp 103-104] but in those two cases the inclusions were present only in the blue or black smalts used to sketch the outlines. EDS analysis of those inclusions revealed a composition rich in calcium (Ca) and silicon (Si) which was confirmed also in the present case. But here we have a considerably earlier case in which the inclusions were added to the glaze itself.

The composition and small granulometry of the inclusions suggested that they might result from an addition of powdered calcium oxide to the glaze frit, resulting in the genesis of a calcium silicate over firing. This hypothesis was confirmed through reproductions following a procedure already tested in the past [14] and indeed the same result as seen in the glazes was obtained and the compositions of the inclusions in both cases were confirmed to be practically identical (Figure 3 and results to be published).

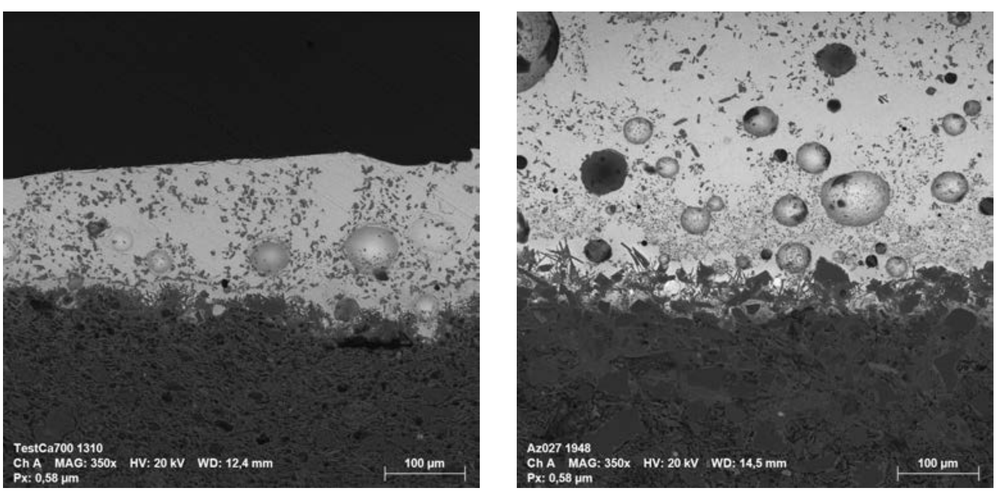


Figure 3. Left side: reproduction tile ref. "TestCa700"; right side: baseboard slab with monkeys, sample Bac167 (images: LNEC).

The purpose of the addition of powdered calcium oxide to the glaze frit is arguable but it was only used on biscuits made of local clay that often fired to a dark reddish colour. In one case (to be published) only a layer of Ca-rich inclusions can be seen near the glaze-biscuit interface, suggesting a white engobe applied over the biscuit for the purpose of hiding its dark colour, making the glaze whiter. That single case may represent a trial after which the powdered calcium oxide was simply added to the whole glaze. The use of a calcium-rich engobe, possibly lime-based, was also found in majolica tiles from

Antwerp[1] but its bulk use dispersed in the glaze is unknown from there,[1] suggesting that it may represent a solution specifically developed for Bacalhôa by a foreign potter who left the country after completing his commission there.

Table 4. Systematization of samples solely based on the addition of powdered calcium oxide to the glaze frit

Group A - glaze speckled with minute Ca-rich inclusions	Group B - glaze clear of Ca-rich specks	
301-27 (Fauns 2)	301-11 + 301-96 (Shields 1 & 2)	301-8 (Susanna)
Floral slab	301-23 (Fauns 1)	301-9 (Hippodamia)
Monkey slab	301-6 (Europa)	301-10 (Tagus)
Children slab	301-7 (Mascarons)	Loggia river gods
301-P50	301-P1 + 301-P11 + 301-P24	301-P28 + 301-P41 + 301-P46

4.2. Composition of the glazes

In the composition of a glaze, the ratio silicon/lead (Si/Pb) is important because it represents a capital technological trait: the mixture proportions used by the workshop that contribute mostly to define the minimum temperature at which the glaze can be properly fired. Lead is a fusing agent and therefore a low Si/Pb ratio means that the glaze can be properly fired at a lower temperature, eventually making use of a less sophisticated kiln than would be necessary if the ratio was much higher, or else completing firing cycles at a faster pace than would be possible with a high Si/Pb ratio.

Considering the Si/Pb ratios (Table 2), it will be seen that they decrease from 1.56 to 0.31 with a divide between samples with ratios of around 1.0 or higher, and samples with a ratio of around 0.6 or lower. This result is highlighted by the use of two colours in the table and it can be noted that in the tiles corresponding to the blue area of higher Si/Pb ratios, the contents in potassium (K) are in average higher and the contents in tin (Sn) are in average lower than in the red area. These remarks are particularly relevant because most of the silicon (Si) and potassium (K), and all of the tin and the lead (Pb) are introduced by the potter in raw materials that may be accurately weighed and therefore their contents are established by the glaze recipe.

Figure 4 shows the results of a log-based PCA of the glazes of all samples, considering the analytical results in Table 2, together with the reference samples Graça I, Graça II and NSVida. Graça I and very likely NSVida are known to be by the workshop of João de Góis, while Graça II is maybe by a different workshop within the same technological circle [11; 12; 13]. The result of the analyses is depicted through a plot in the plane of the two first principal components (PC1 and PC2). PC1 explains 63 % of the variance and is controlled in the positive sense by the contents in silicon (Si) and potassium (K), and in the opposite sense by the contents in tin (Sn) and lead (Pb). PC2 explains 22 % of the

1 Personal communications by Bauvois, Stefanie of the ARCHES Research Group, University of Antwerpen.

variation and is controlled in the positive sense mostly by the content in sodium (Na) and in the opposite sense mostly by the content in aluminium (Al), as seen in Figure 5, where the loadings plot is represented as a vector graph.

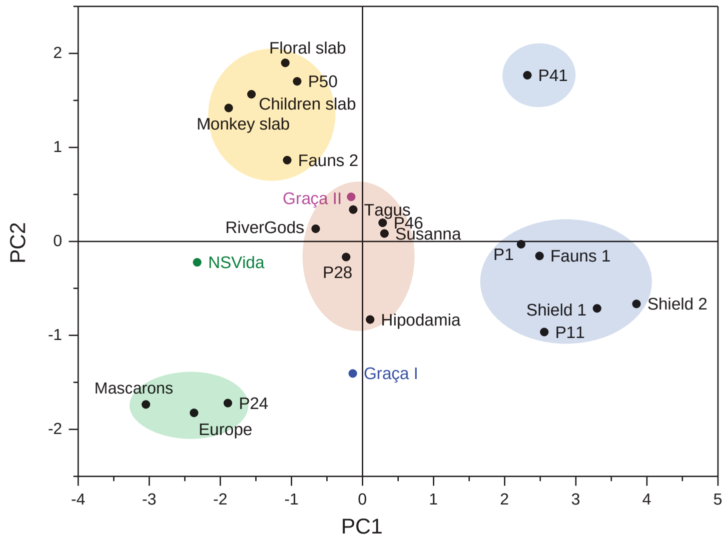


Figure 4. Score plot of the PCA of the glazes.

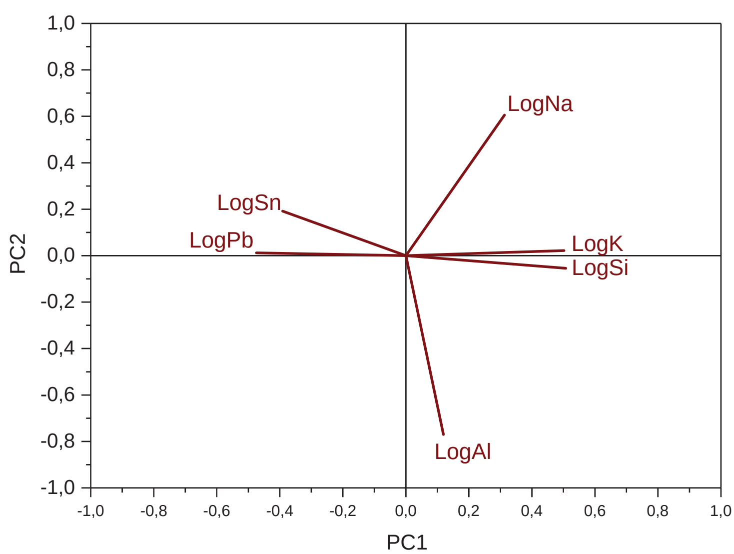


Figure 5. Loadings plot of the PCA of the glazes.

All the high Si/Pb ratio samples in the blue region of Table 2 are naturally on the right side of the PCA plot of Figure 4. All are close together except the pattern 301-P41, but consulting Table 2 it will be seen that the deviation upwards results, in this case, mostly from the low content in aluminium (Al). The presence of Al in 16th century glazes does not derive from a precisely weighted raw material called for by the recipe, but rather from an uptake from clay minerals in the biscuit during the second firing and also from additions to the glaze, such as feldspar, mica or ground clay. Some of these, together with grains of sand, are not fully digested during the firing cycle and result in the dark inclusions seen in the sectional images of Figure 2. Therefore, the recipe of the glaze of 301-P41 may well be similar to e.g. 301-P1 and 301-P11 and the different contents in Al may be related to the ancillary procedures of a specific workshop, or to the nature of the additions used at a specific time.

The fact that the Si/Pb ratio bundles together six panels and patterned tiles within the larger blue area, suggests that they should indeed be grouped. Reviewing Figure 2 it will be seen that all these, as well as 301-P41, share underdeveloped or mostly absent interface outgrowths, suggesting that the firing schedules used were similar. The tentative integration of 301-P41, that was clearly separated by the PCA, in this group can only be made with a note attached that it may stem from a different workshop sharing the same basic technology or, at least, represent the practice of a different potter in the same workshop.

Concerning the low Si/Pb ratio cases (red region of Table 2), the samples morphologically separated by the Ca-rich dusty inclusions (Group A in Table 4) are all within a small region marked in yellow in Figure 4, and glaze-wise these constitute a cluster whose members have likely a similar chronology. They are not too far apart from the cluster in the region marked with a red colour in Figure 4, a cluster that may also be considered to include, from the three reference samples, at least Graça II.

The only three other low Si/Pb samples that cannot be immediately clustered with the rest represent the Rape of Europa and the Mascarons panels, together with patterned tiles 301-P24 (region marked in green in Figure 4). What sets them apart are the lowest contents in sodium (Na) and potassium (K) as seen from Table 2, that push them to the far corner of the third quadrant of the PCA graph.

In a previous publication it was shown that the single characteristic that may distinguish the productions of the workshop of João de Góis or his near circle from all others, including the Hispano-Moresque from Seville which share with them a low Si/Pb ratio, is the low content in sodium [3, pp. 42-43]. It may prove relevant in the future to note from Table 3 that the panels and tiles with the lowest contents in sodium are, besides those in the green region of Figure 4, also the river god panels together with the patterned tiles 301-P28, precisely those that make up the decorative lining of the Loggia of the River Gods [4].

From our previous work, the Si/Pb ratio expected from the productions of the workshops of Lisbon at the time when the Bacalhôa azulejos and panels were in all likelihood manufactured (1560s-1570s) was 0.4 ± 0.1 for a 90 % confidence interval [3] or 0.4 ± 0.2 for a ca. 95% confidence interval. To attain this ratio the local potters had to use an excess of expensive lead, probably because of some technological limitation of the kiln or kilns available, or else to obtain a good match with the biscuit and a proper adhesion of the glaze to the distinctive low-calcium clay used at the time. The composition and firing conditions resulted in a characteristically well-developed interface with crystalline outgrowths [3, p. 40]. The glazes in the red area of Table 2 could fit into that interval, but the glazes in the blue area could not. Both this fact and the underdevelopment or absence of the interfacial outgrowths suggest for the glazes in the blue area a source other than the workshops of Lisbon.

4.3. Composition of the biscuits

Each clay or mixture of clays has a range of calcium/silicon ratios (Ca/Si) that characterize it in some measure, but ratios of around 1:1 in elemental weight are frequent and cannot, by themselves, identify a clay. However, our previous experience in the field shows that clays or mixtures with Ca/Si ratios of ca. 0.4 or below are rarely used in majolica azulejos at this time [3]. Kate van Lookeren Campagne, in her research of historical sources on early Dutch azulejo manufacture, states that "the importance of using calcium-rich clays in the production of tin-glazed wares has been mentioned in all of the earliest sources" [15]. Considering Table 3, the ratios vary from 1.18 down to 0.15, a very unusual low value. Also, there is a rather continuous decrease from 1.18 to 0.54 and then a discontinuity to the next value (0.37) suggesting that at least two different clay pastes may be present. This divide is highlighted by the use of two colours in Table 3 and it will be noted that in the tiles corresponding to the blue region of higher Ca/Si ratio, the contents in magnesium are noticeably higher than in the red region of calcium-poor biscuits, which is not unusual in itself, since the carbonates of magnesium (Mg) and calcium (Ca) are often associated, but in this case will be consequential.

Figure 6 shows the results of a log-based PCA of the biscuits of all samples, considering the analytical results in Table 3, together again with Graça I, Graça II and NSVida. The results are depicted through a plot in the plane of the two first principal components (PC1 and PC2). PC1 explains 55% of the variance and is controlled in the positive sense mostly by the contents in silicon (Si) and potassium (K), and in the opposite sense mostly by the contents in magnesium (Mg) and calcium (Ca). PC2 explains 25% of the variation and is controlled in the positive sense mostly by the contents in aluminium (Al) and iron (Fe) and in the opposite sense mostly by the contents in sodium (Na) and Ca, as seen in Figure 7, where the loadings plot is represented as a vector graph.

All the high Ca/Si ratio samples in the blue region of Table 3 are naturally on the left side of the PCA graph of Figure 6, forming a single cluster that also includes Graça II but not Graça I. However, this cluster includes, both above and below of PC2=0, glazes with high and with low Si/Pb ratios that were separated by the PCA of Figure 4. This is a rather perplexing situation that will be specifically addressed further on.

Regarding the low Ca/Si ratio region of Table 3 (red area), the samples morphologically separated by the Ca-rich dusty inclusions in the glaze (Group A in Table 4) are all within a small region marked in yellow in Figure 6, also including the patterned tiles 301-P28 from the Loggia of the River Gods, suggesting that they too use the same or a very similar clay. The *Rape of Europa* and the Mascarons panels, whose Ca/Si ratio is the lowest, are pushed to the far corner of the first quadrant by their lower sodium (Na) and calcium (Ca), and higher aluminium (Al) contents, where they group together with NSVida and patterned tiles 301-P24.

As a graphic counterpart of the biscuit compositions, Figure 8 depicts the relevant parts of four EDS biscuit spectra representing all the possible cases from crossing the clusters of Figure 4 with those of Figure 6, plus Graça I and Graça II used as references. The peaks of magnesium (Mg), potassium (K) and calcium (Ca) are identified and straight lines are used to accentuate graphically their relative sizes.

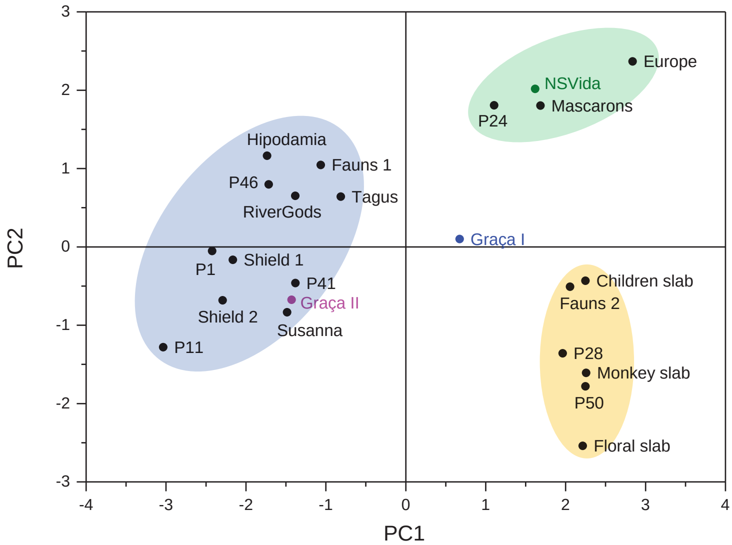


Figure 6. Score plot of the PCA of the biscuits.

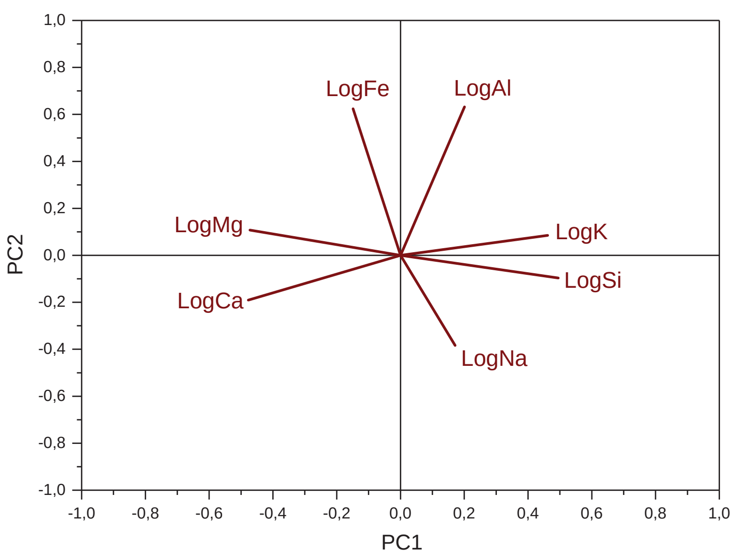


Figure 7. Loadings plot of the PCA of the biscuits.

As could be expected from the PCA result, all the spectra on the left side of Figure 8 look similar. They all depict a relatively high Mg peak, together with a high Ca peak. This is not an unusual feature in azulejo clays, including those used in Portugal from the 1590s onwards [12]. In comparison, the spectra on the right side present lower Mg and Ca peaks, which characterize the clay normally used by the workshops of Lisbon in the 16th century (exemplified by the spectrum of the biscuit of *Nossa Senhora da Vida*) until, at least, after the lining of the *Capela de São Roque* dated "1584" [7]. This type of clay is very different and quite unique amidst the clays we know from 16th century productions in other countries, being poor in Ca, with Ca/Si ratios around 0.3 and also relatively poor in Mg [3]. These features are a key-marker of this type of clay and suggest that the clay used on all the tiles and panels on the low Ca/Si red area of Table 3 (corresponding to the yellow and green clusters of Figure 6) may have been extracted from Miocene layers cropping out in Lisbon, as well as on the south bank of the Tagus. One of such layers, used by the local potters for many centuries, is the so-called *Forno do Tijolo* clay, cropping in the hill of the Lisbon castle and then northwards under the *Bairro das Olarias* (Potters Neighbourhood) [16, p. 136] towards Alvalade (near the airport of Lisbon) where it was cored in 2009. We had access to the cores and sampled and analysed the full 21 m thickness of the layer at that area, finding variable contents in calcium that may be used to define sublayers (to be published). The Ca-poor biscuits closely fit the thickest sublayer in which the Ca/Si ratios vary around 0.25. When fired in an oxidation atmosphere, the biscuits have a characteristic orange/reddish colour derived from the formation of red iron oxide that is not counterbalanced by light-coloured calcium-rich minerals, as seen also in several productions of the João de Góis circle (see, for instance [13, p. 82]). The clay used for both the *Rape of Europa* and the Mascarons is similar and the biscuits are red because they were formed from Ca-poor clays and fired in an oxidation atmosphere. However, when the same clay is fired in reducing atmospheres, eventually at a location inside the kiln where fumes prevail, the resulting biscuits are buff to a darker brownish (see e.g. [11, right side of fig.7]) and the tell-tale orange/reddish colour on 16th century biscuits that immediately suggests an attribution to the workshops of Lisbon, is lost.

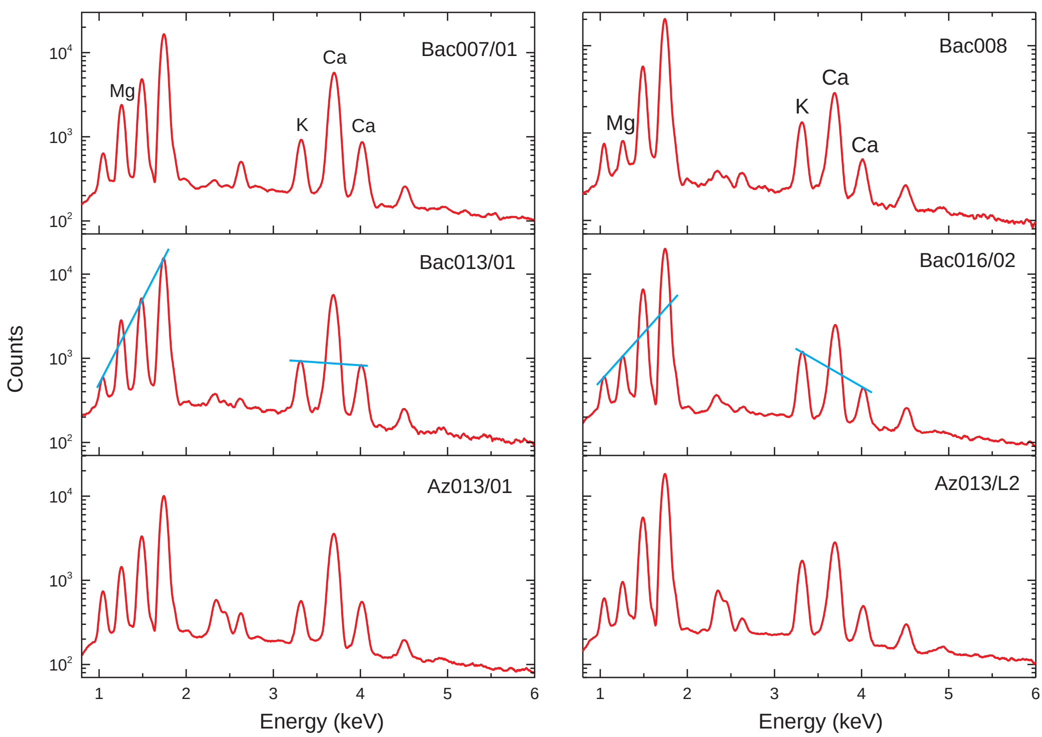


Figure 8. Biscuit EDS spectra. Left side (top to bottom): 301-96 (Shield 2); 301-5 Douro river god; Graça II (sample Az013/01). Right side (top to bottom): 301-27 (Fauns 2); 301-7 (Mascarons); NSVida (sample Az032/08).

Given their oddly low Ca/Si ratios, the biscuits making up the yellow and green clusters of Figure 6 allow a sound conjecture as to the origin of the clay and could even be combined to form a single cluster also with Graça I. However, the blue cluster of Figure 6 presents a major problem because it bundles together two very different glaze compositions, clustered in the red and in the blue areas of Figure 4, the second with characteristics unknown from the workshops of Lisbon at the time, and therefore suggesting a foreign production. So, either the blue cluster of Figure 6 mingles two different clays, casually with similar compositions, or else the same clay was used for two very different productions of which one is compatible with a known local provenance, and the other is not.

Starting with the supposition that two different clays are clustered together in the blue area of Figure 6, all biscuits were observed by SEM in BSE mode for any significant morphological evidence. A conspicuous detail immediately noticed was the occurrence of rhomboid hollows (Figure 9) left by rhombohedral crystals of dolomite or dolomitized limestone that were consumed by de-carbonization in the course of the firings, a process researched by Maria José Trindade et al. [17]. Sometimes a blob of the resulting neo-formed minerals could be perceived inside a hollow and the EDS analysis confirmed an excess of Mg, besides the elements characteristic of clays that make up the matrix (Figure 9).

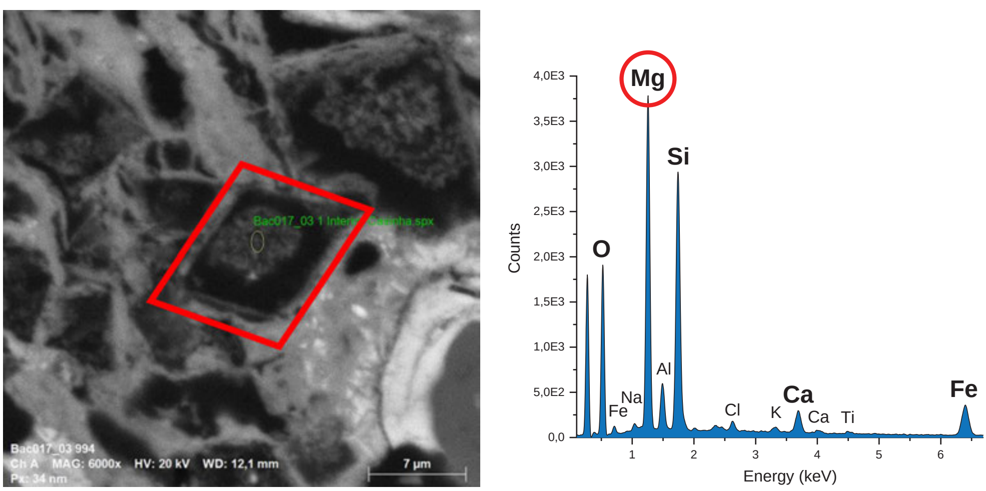


Figure 9. Rhomboid hollow left by the de-carbonation of a crystal of dolomite and analysis of the blob inside (301-10 Tagus, sample Bac017/03) (images: LNEC).

Figure 10 depicts sectional images of the biscuits of three samples from the red cluster of Figure 4, together with three samples from the blue cluster of the same figure. The widespread occurrence of the rhomboid hollows will be recognized in all cases. The epigenetic formation of these euhedral crystals of dolomite was discussed and even replicated by several authors who concluded that they could only be formed under conditions that favoured particular kinetics [18;19], while anhedral or subhedral forms were much more common. In our studies, we have only found the widespread occurrence of the shapes left by euhedral dolomite crystals in azulejo biscuits, in two other instances: one in samples from ca. 1600 attributed to the workshops of Talavera in Spain; and another from a ca. 1650 production attributable to the workshops of Lisbon, but in this last case the residue left by the consumption of the dolomite was also rich in manganese

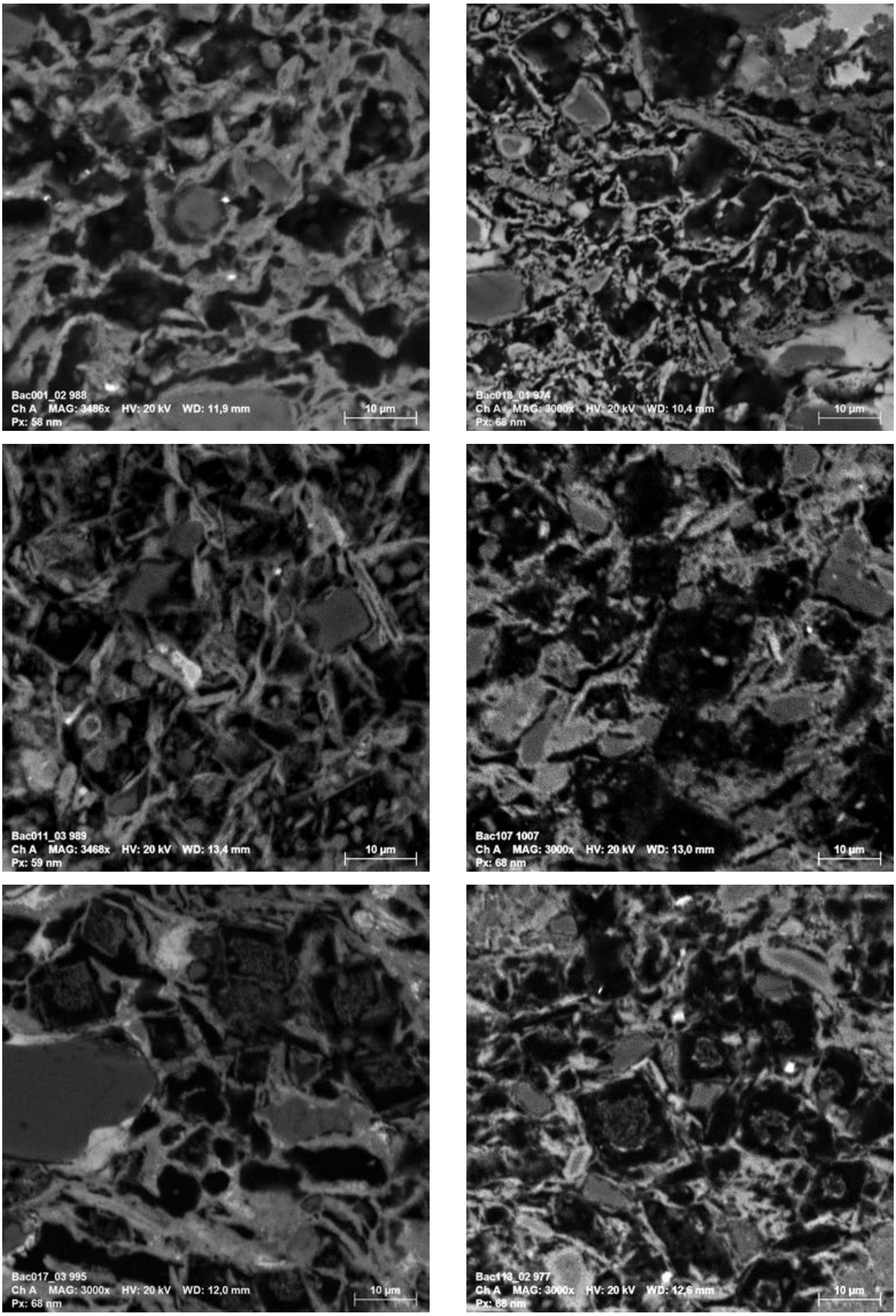


Figure 10. Left side (top to bottom): 301-8, Susanna (sample Bac001/02); 301-2, Mondego river god (sample Bac011/03); 301-10, Tagus (sample Bac017/03). Right side (top to bottom): 301-11, Shield 1 (sample Bac018/01); 301-23, Fauns 1 (sample Bac107); 301-P11 (sample Bac113/02) (images: LNEC).

(Mn), which is vestigial or non-extant in the Bacalhôa samples. The occurrence of the same unusual rhomboid morphology of the hollows in all the biscuits in the blue cluster of Figure 6 suggests that the same Mg- and Ca-rich clay was used in each and every one of them.

In the images of Figure 10, white specks can be perceived in some biscuits. They are inclusions of minerals containing elements of high atomic number (Z) that are seen in a conspicuous white colour because of the proportionally higher number of electrons backscattered by the nuclei of their atoms. Those inclusions range from common zircon and cerium-monazite, to rare minerals of tantalum or indium, which we have found only once. No inclusion is by itself determinant, but the occurrence of a set of uncommon or rare inclusions in a clay is expectably connected to a specific provenance [20].

The biscuits on the blue cluster of Figure 6 were again divided in two sets corresponding to the red and to the blue glaze clusters of Figure 4, and the nature of the high-Z inclusions was determined. Minerals with vanadium (likely vanadinite $Pb_5(VO_4)_3Cl$ because of the association with lead and chlorine that was clearly identified in the EDS results), copper, zinc, strontium (strontianite?), tin, barium (likely barite $BaSO_4$ identified because of the association with sulphur) and gold were found in both sets. Albeit uncommon, vanadinite, copper, tin, barite and gold are known from verified Portuguese biscuits made from local clay. Minerals of zinc and strontium, moderately common in the samples analysed, have not, yet, been found in 16th century Portuguese biscuits that we studied in the past. The clays could not be shown to be different in this manner either, and the results add to the presence of the rhomboid hollows to compellingly suggest that the same clay was used for all the biscuits aggregated in the blue cluster of Figure 6.

5. CONCLUSIONS

The analytical results of the biscuits suggest the use of two very different clays:

– One, poor in calcium and magnesium, firing to a shade of orange or dark red colour in an oxidation atmosphere, was very likely extracted from a Miocene layer of the region of Lisbon and may be the same clay already used in the 16th century by local potters to produce green-glazed red clay pottery. Two sorts of azulejos were made with this clay but all have glazes whose compositions are compatible with the productions of the workshops of Lisbon;

– The other clay is much richer in calcium and magnesium and we have never found biscuits made of a similar clay in tiles of this chronology manufactured by the workshops of Lisbon.

Two different sorts of azulejos were made with biscuits of this last clay:

– The first type includes, from the panels and patterned tiles studied, the Albuquerque coat of arms, the remains of the Noronha coat of arms, one of the faun panels that is also decorated with birds (301-23), and three different patterned tiles. The composition of the glazes used in these tiles, with Si/Pb ratios of ca. 1.0 or higher, is unknown to us from Portugal at this early chronology. Therefore, we conclude that they were likely manufactured abroad;

– The second type has a glaze whose composition is similar to those of the circle of João de Góis (as seen by e.g. the low Si/Pb ratio, low content in potassium and high in tin). These tiles were fired in conditions also compatible with a production by the workshops of

Lisbon, as seen by the generally high degree of development of the interfacial outgrowths. Therefore, the results suggest that, either the clay, or the biscuits, were imported, but they were glazed and fired in Portugal.

The importation of clays was not unknown in the 16th century and throughout the 17th century in Holland, even though appropriate clays were available locally, as Kate van Lookeren Campagne showed based on contemporary documental evidence [15]. And the reason was that the conditions under which a good fit between glaze and biscuit was obtainable could only be set, at that time, through an often lengthy trial and error process. Bastenaire-Daudenart, who worked in the late 18th century and had first-hand experience as shop-master, states that many would-be producers of faience were out of business before they could start a manufacture, because they were unsuccessful in their quest for a workable glaze-biscuit combination [21]. For that reason, once a fit was found and the parameters of a suitable firing schedule established, potters tended to continue using the same materials and technology, even though better clays might be available locally and less expensive production parameters would have been possible. In this case, an indication that the already formed and fired biscuits (and not the clay) were imported, is their thickness: Ca-rich biscuits used in panels where they were accessible to be measured and that were, according to our conclusions, glazed in Lisbon (the *Abduction of Hippodamia* and the Tagus river god) had dimensions of ca. 13 x 13 x 2.5 cm, while square tiles with side dimensions up to 15.5 cm made of local clay (such as 301-P24, 301-P28, 301-P50 and many others not included in this study) had a thickness of ca 1.5cm. If the biscuits used for the *Abduction of Hippodamia* and the Tagus river god had been made locally from imported clay, they would likely be formed with the lowest possible thickness to save on the raw material.[2]

Yet, a number of questions can be raised… Why import only the biscuits and not the finished tiles? Why would biscuits be brought to the country, when they could have been made from local clay? Accepting biscuits were imported, why not use the same glaze recipe used abroad whose compatibility was assured? In the absence of documental evidence, these and similar questions can only be replied hypothetically and will be addressed in future papers dealing with the historical context of the linings of Bacalhôa. Still, two of the perplexities can be replied on purely technical grounds- the importation of biscuits and their glazing with a different glaze recipe with its implicit risks.

The fact that the clay available in Lisbon fired to a dark colour implied a technical challenge to the whiteness of the glazes that foreign potters would certainly prefer to avoid by using their own biscuits already fired to a cream colour. And, in fact, the panels and patterned tiles for the Pleasure House manufactured in Lisbon were largely fired on imported biscuits. Local clay was used mostly for slabs and tiles whose dimensions excluded the use of imported biscuits and seemingly a specific correction was judged necessary by the potter: the adding of powdered calcium oxide to the glazes. On the other side, any glazes fired in Lisbon would have to be compatible with the kiln or kilns locally available for the purpose, and the firing schedule normally used to which the kiln owners would likely want to abide – if for no other reason, then at least because local glazed pottery had to be fired at the same time. Very likely the firing parameters

2 Research on tiles not addressed by this paper suggest that one or more small batches of clay were indeed imported and their utility extended by reducing the thickness of the tiles, or by mixing it with Lisbon clay, for use in the biscuits for Bacalhôa. Those interesting findings will be reported in future papers.

(maximum temperature and duration) called for a lower Si/Pb ratio than if they had been fired in the kiln used e.g. for the Albuquerque Coat of Arms panel. Therefore, the recipe of the glaze had to be altered so that, while maintaining its compatibility with the biscuit, it was suitable to be fired in conditions different from those that were standard at the origin. The starting point would be try-outs with the recipe used in Lisbon and if that worked, possibly with minor adjustments to the proportions, then it would be applied to the production. And while the tiles addressed by this article had relatively steady compositions (as seen by the standard deviations in Table 2) a process of testing the proportions of the glaze components can be recognized in lesser frame tiles of simple design in which the Si/Pb ratio may vary from 0.6 to more than 1.7, along with the density and nature of the inclusions, such as grains of sand, added to the frit, presumably to adjust the contraction of the glaze over cooling to that of the biscuit.

It is interesting to note, towards a future dedicated research, that the glazes of both Graça I (corresponding to the panels signed with the monogram of João de Góis) and Graça II are similar to those glazes that make up the red cluster in the PCA of Figure 4, but only the biscuit of Graça II is compatible with the blue cluster in the PCA of Figure 6 and it was confirmed that it too depicts the characteristic rhomboid hollows as in Figure 10. This leads to the important conclusion that Graça II was probably also made with the imported biscuits and is, then, very likely contemporary with the panels of Bacalhôa, one of them, *Susanna and the Elders*, dated “1565” as mentioned before. It also reinforces the connection of the disperse linings of *Igreja da Graça* with Brás [Afonso] de Albuquerque and possibly with the burial of Afonso de Albuquerque in that church on May 1566, as proposed in a previous paper [11, pp. 64-65].

The slabs, oversized tiles of some faun panels, and the patterned tiles from Room 5 of the Pleasure House, all using a local clay, have glazes depicting a technological trait (resulting from the addition of powdered calcium oxide to the frit) that was never found in any other tiles elsewhere and therefore suggests the intervention of a foreign potter that was in Lisbon specifically for this job.

The *Rape of Europa*, the Mascarons, and patterned tiles 301-P24, albeit standing separated from the other Bacalhôa panels in the PCAs, are clustered together with *Nossa Senhora da Vida* (presumably datable to the 1570s) as pertains to the biscuit, suggesting the possibility of a different, maybe later, chronology.

Figure 11 concludes the present study with a pictorial of the panels and main linings of Bacalhôa, grouped according to their presumed provenance and technology used. We must point that there are several other majolica patterned tiles, used elsewhere in the palace annexes and gardens, or removed and re-applied at an unknown time, or else exhibited at the local museum, which, for the sake of clarity, were not dealt with in this first study. They do, in general, fit into the groups of Figure 11 and will be addressed in future papers.

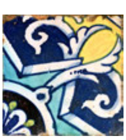
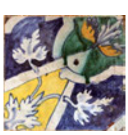
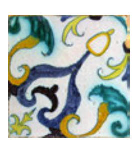
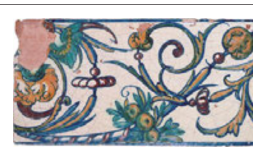
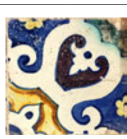
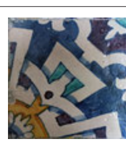
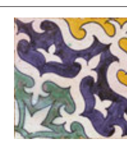
Group 1 Biscuit of unknown clay; composition of glaze unknown from Portugal at this chronology. Firing schedule seemingly different from the schedule used in Lisbon. **Likely imported from abroad as finished products.**	**Group 2** Biscuit as in Group 1; glaze similar to glazes from the circle of João de Góis. Firing schedule similar to workshops of Lisbon. **Likely manufactured in Lisbon using imported biscuits.**	**Group 3** Biscuits of Lisbon clays; glaze matches circle of João de Góis. Firing schedule seemingly same as used by workshops of Lisbon. **Attributable to the circle of João de Góis.**	**Group 4** Biscuit as in Group 3; glaze dusted with Ca-rich inclusions. Firing schedule similar to workshops of Lisbon. **Manufactured in the region of Lisbon, probably by a foreign potter.**
301-11, Albuquerque Coat of Arms panel	**301-8, *Susanna & the Elders***	**301-6, *Rape of Europe***	**301-27, Fauns 2**
			
301-96, Noronha Coat of Arms panel	**301-9, *Abduction of Hippodamia***	**301-7, Mascarons**	**Children slab**
			
301-23, Fauns 1	**301-10, Tagus River god**	**301-P24**	**Monkey slab**
			
301-P11 | **301-P41**	**301-1 to -5, Loggia River Gods panels**	**301-P28**	**Floral slab**
 			
301-P1	**301-P46**		**301-P50**
		—	

Figure 11. Conclusive grouping of the panels and patterns studied in this paper.

Notes to Figure 11.

- **Group 1** includes tiles (three panels and three different patterned tile types) with: i) a glaze formulation unknown to us from 16th century Portugal; ii) generally lacking the well-developed glaze-biscuit interfacial growth that characterizes Portuguese productions of the time; iii) using biscuits of a clay unknown to us in Portuguese azulejos of the 16th century, often (but not always) with a thickness of 20 mm or more;

- **Group 2** includes, from the set selected for this study, eight panels (of which five are at the Loggia of the River Gods) and one patterned tile type: i) with a glaze recipe similar to that used by the technological circle of João de Góis (albeit often with a higher content in sodium) and therefore known to us from the 16th century workshops of Lisbon; ii) fired in a manner that generally results in a well-developed interfacial outgrowth akin to the productions of the workshop of João de Góis in the 1560s-70s; and iii) that uses biscuits seemingly made from the same clays as used in Group 1.

- **Group 3** includes, from the set chosen, two panels and two patterned tiles: i) with a glaze fully characteristic of the technological circle of João de Góis, with a very low content in sodium; ii) fired according to a schedule that imparts an interface also known from the same circle; iii) on biscuits made of a local clay, seemingly from a Miocene layer quite similar to a clay known to be used by potters at this time - presumably the same used by the circle of João de Góis, with calcium contents variable but always low. Although Groups 3 and 4 are not separated by the clays used, it is relevant to note that 301-P28 of this group uses a clay somewhat different from the rest and similar to that of Group 4.

- **Group 4** includes one of the two panels with fauns studied, a large group of baseboard slabs and one patterned tile type: i) with a glaze formulation very similar to that of Groups 2 and 3 but with a slightly higher content in sodium than Group 3; ii) to the glaze of which was added a powdery Ca-rich substance (reproduction tests strongly suggest that a finely milled calcium oxide was used) resulting in a cloud of dusty inclusions spread throughout the glaze, representing a technological trait of likely foreign origin; iii) whose firing schedule is similar to that of Groups 2 and 3, but seemingly not exactly the same because, although developed, the interface is morphologically somewhat different; and iv) that use a local clay, maybe from the same Miocene layer as that of most of Group 3 but from a different sublayer.

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ISBN 978-972-49-2317-8

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