A comparison of the earliest faience tiles produced in Lisbon with earlier and later types

João Manuel Mimoso, Sílvia R. M. Pereira, Alexandre Pais, Maria Augusta Antunes, Ana Margarida Cardoso, Maria de Lurdes Esteves, António Candeias

ABSTRACT

The manufacture of majolica azulejos in Portugal started, as far as we know presently, during the 1550s by the workshop of Hans Goos, a Flemish potter established in Lisbon as João de Góis.

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 starting with his own productions before 1560 and lasting at least until the 1580s, possibly beyond.

To address the characteristics of those productions and what makes them distinguishable, we have to compare them with those that chronologically preceded or followed it. This article presents such a study concluding that the productions of the circle of João de Góis have defining characteristics that allow to identify azulejos of unknown origin as stemming from workshops using the same basic materials and technology.

RESUMO

A fabricação de azulejos de faiança em Portugal começou, tanto quanto sabemos atualmente, durante a década de 1550 pela oficina de Hans Goos, um oleiro flamengo estabelecido em Lisboa como João de Góis.

As produções do que podemos chamar de "círculo de João de Góis" (talvez apenas uma única oficina, ou talvez várias compartilhando a mesma tecnologia) abrangem um período que se inicia com as suas próprias produções antes de 1560 e que continuou pelo menos até à década de 1580, prolongando-se possivelmente até uma época mais tardia.

Para abordar as características dessas produções e o que as torna distinguíveis, temos que compará-las com aquelas que as precederam ou que lhes sucederam cronologicamente. Este artigo apresenta um estudo realizado nessa base concluindo-se que as produções do circulo de João de Góis apresentam características próprias que as tornam distinguíveis permitindo, em geral, identificar como portugueses azulejos cuja origem é desconhecida mas apresentam morfologias e composições que os ligam ao mesmo circulo tecnológico.

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KEYWORDS: Renaissance majolica / Hispano-Moresque tiles / Azulejos / João de Góis / / Museu Nacional do Azulejo

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1. INTRODUCTION

Architecturally integrated tiles are used in Portugal since at least the 16th century. The oldest are believed to have been glazed slabs of a single colour and Hispano-Moresque tiles. Despite the possibility of a local production in small numbers, many Hispano-Moresque tiles applied in Portugal depict a compositional proximity that suggests they were imported from a single place, namely Seville, where they were manufactured in the Islamic tradition [1, pp. 95-98]. Hispano-Moresque tiles are decorated according to a *cloisonné* technique: raw glass pigmented in several colours is used to fill partitions on the surface of the tile. Once fired, a colourful decorative design is obtained with the shape of the partitions.

In majolica (also known as faience) a raw lead-tin glass which can be fired to an opaque white glaze is used to cover the surface of the tile. The raw glass can be painted, as if it was a canvas, with colours based on pigments able to sustain temperatures above 1,000 °C. Some of the earliest faience tiles used in Portugal were imported from Antwerp, of which the tiles dated "1558" used in the Ducal Palace of Vila Viçosa are an example [2]. The manufacture of majolica azulejos in Portugal started, as far as we know presently, during the 1550s by the workshop of Hans Goos, a Flemish potter established in Lisbon as João de Góis [3].

The productions of what we may call *the technological circle of João de Góis* (maybe only a single workshop, or maybe several sharing the same technology) encompasses a period starting with his own work before 1560 [3] and lasting at least until the 1580s [4], and possibly beyond. To address the technical characteristics of this production and what makes it distinguishable, we have to compare it with those that chronologically preceded or followed it. This article attempts such a study, noting however that the notions of "antecedent" and "subsequent" do not imply, in this case, a technological connection in the form that the latter is rooted in the former.

2. EXPERIMENTAL

2.1. Samples

We considered four groups of azulejos. Besides the productions of the workshop of João de Góis or his circle, as chronological predecessors we considered Hispano-Moresque tiles and majolica tiles from Antwerp. As chronological successors we considered Portuguese productions of the 17th century. Except for the Antwerp tiles, of which only two clearly distinct productions were available, six tiles or panels from each group (Hispano-Moresque, circle of João de Góis and 17th century Portuguese) were selected. The samples used in this study were chosen to correspond to the largest chronological spread in each group. They were collected from:

- six Hispano-Moresque tiles of different origins, mostly unknown but presumed to be from the workshops of Seville, from the collections of the Museu Nacional do Azulejo (MNAz – National Museum of Azulejos) and tentatively dated from the early 16th century to the 1560s (Figure 1);
- two majolica tiles produced in Antwerp, one from the Flemish panels applied in the Ducal Palace of Vila Viçosa and one from a group excavated in Lisbon [5] and



Figure 1. From left to right and top to bottom: tiles and panels from which samples were collected: Hispano-Moresque Az317/00; Az304/00; Az017/00; Az301/00; Az065/00 and Az066/00; Antwerp Az030/A1 and Az031/A; Portuguese 17th century Az003/00; Az024/00; Az100/01; Az197/01; Az198/AR2 and Az052/00

presumed older (Figure 1);

- six tiles from the three panels ascribed to the circle of João de Góis: two samples from the earliest confirmed panel (Graça church in Lisbon Figure 2, presumed to date from the 1560s), two samples from the latest (São Roque chapel in Lisbon Figure 3, dated "1584") and two samples from the only other azulejo ensemble that may, at this time, be indisputably ascribed to a Portuguese production by the workshop of João de Góis or one of the same circle the panel Nossa Senhora da Vida (Figure 4) tentatively dated to ca. 1580 and belonging to the MNAz collection. More images of the panels and details of the tile units from which samples were taken can be found in the literature [6; 7; 8];
- six Portuguese panels or loose tiles ascribed to the 17th century from ca. 1620 to ca. 1690 (Figure 1).

Table 1 includes further relevant data on all samples.



Figure 2. The monogrammed panel of Graça church from which samples Az013/L1 and Az013/ L2 were collected



Figure 3. The signed and dated panel at São Roque chapel from which samples Az068/03 and Az068/13 were collected



Figure 4. The lower part of the panel Nossa Senhora da Vida from which samples Az032/00 and Az032/01 were collected

| Reference | Group* | Date | Origin/ Location | Glaze Colour | Notes | |
|-----------|--------|--|----------------------------------|----------------------------|--|--|
| Az317/00 | HM | 1500-1510 | Palace of Vila Viçosa / MNAz | white | Seville (?) | |
| Az304/00 | HM | early 16 th century | Royal Palace of Sintra / MNAz | white | Seville (?) | |
| Az017/00 | HM | 2 nd quarter 16 th century | collection MNAz, Lisbon | blue | Seville (?) | |
| Az301/00 | HM | 1 st quarter16 th century | collection MNAz, Lisbon | white | probably Seville, excavated (?) | |
| Az065/00 | HM | ca. 1520-1560 (?) | collection MNAz, Lisbon | white | probably Seville | |
| Az066/00 | HM | ca. 1520-1560 (?) | collection MNAz, Lisbon | white | probably Seville | |
| Az030/A1 | Antw | before 1558 (?) | collection MNAz, Lisbon | grey | archaeological find | |
| Az031/A | Antw | 1558 | Palace of Vila Viçosa / MNAz | white and blue | fragment | |
| Az013/L1 | JGc | 1560s | Graça church, Lisbon | green | from tile bearing monogram | |
| Az013/L2 | JGc | 1560s | Graça church, Lisbon | white | from different tile, as above | |
| Az032/00 | JGc | ca. 1580 | Igreja de Santo André / MNAz | white | child Jesus elbow/arm | |
| Az032/01 | JGc | ca. 1580 | Igreja de Santo André / MNAz | yellow-brown | child Jesus crib | |
| Az068/03 | JGc | 1584 | São Roque chapel, Lisbon | blue | signed panel of the dog | |
| Az068/13 | JGc | 1584 | São Roque chapel, Lisbon | dark blue | signed panel of the dog | |
| Az003/00 | PT17 | 1600-1620 | collection MNAz, Lisbon | white with orange paint | fragment | |
| Az024/00 | PT17 | before 1640 (?) | collection MNAz, Lisbon | white and blue | fragment | |
| Az100/01 | PT17 | 2 nd quarter of the 17 th century | collection MNAz, Lisbon | white | panel decorated with <i>grotteschi</i> | |
| Az197/01 | PT17 | 1660-1680 | Graça church, Lisbon | white and blue | azulejo panel still in situ | |
| Az198/AR2 | PT17 | 1660-1690 | Graça church, Lisbon | white and blue | azulejo arch still in situ | |
| Az052/00 | PT17 | 1670-1690 | collection MNAz, Lisbon | white | fragment | |

 Table 1.
 Samples references and characteristics

* HM - Hispano-Moresque; Antw – Antwerp; JGc – 16th century, circle of João de Góis; PT17 – 17th century Portuguese

2.2. Analytical methodology

Small fragments were detached from the azulejos, stabilized in epoxy resin, lapped and polished to obtain a flat surface for observation and analysis by scanning electron microscopy coupled with an X-ray energy-dispersive spectrometer (SEM-EDS). SEM-EDS observations and analyses were made at the HERCULES Laboratory in Évora using a HITACHI 3700N SEM coupled to a BRUKER XFlash 5010 EDS. The specimens were uncoated and the observations were made in backscattered electrons mode (BSE) with a chamber pressure of 40 Pa and at an accelerating voltage of 20 kV. The acquisition of X-ray spectra was done with the detector set at ca. 8 mm working distance.

The selection of areas for EDS analysis avoided inclusions in the glaze or biscuit representing more than ca. 5 % of the full area analysed. The area sizes were ca. 200 x 200 μ m² for glazes and 500 x 500 μ m² for biscuits but acceptable repeatability was verified in areas four times smaller. For comparison purposes, only the elements usually representing the major contents were considered, excluding tin (Sn) in the glaze and lead (Pb) in the biscuit due to their variability with the area chosen (in the case of Sn because of local aggregations of SnO₂ crystals; in the case of Pb because its content in the biscuit increases with proximity to the interface with the glaze). The results of the EDS analyses are given in weight % of each element identified.

Principal component analysis (PCA) was made of EDS results using the SPSS[®] software platform by IBM Analytics.

3. RESULTS

3.1. Glaze morphology

Figures 5 to 8 depict, at the same scale for comparison purposes, SEM images of the samples showing the main micro-morphologic characteristics generally associated with the glazes and their interfaces.

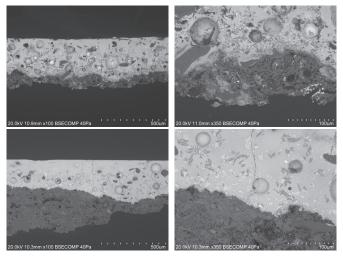


Figure 5. SEM-BSE images showing the main micro-morphologic characteristics of Antwerp majolica tiles glazes (left side) and biscuit-glaze interfaces (right side). From top to bottom: samples Az030/A1 and Az031/A

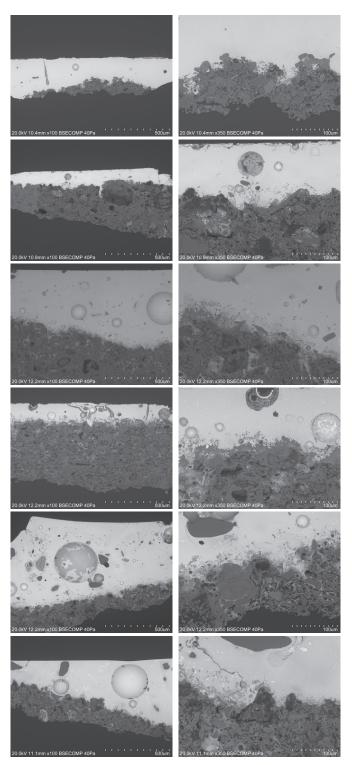


Figure 6. SEM-BSE images showing the main micro-morphologic characteristics of Hispano-Moresque tiles glazes (left side) and biscuit-glaze interfaces (right side). From top to bottom: samples Az317/00; Az304/00; Az017/00; Az301/00; Az065/00 and Az066/00

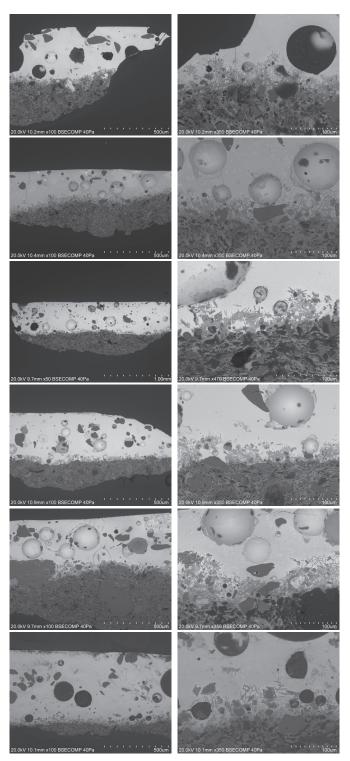


Figure 7. SEM-BSE images showing the main micro-morphologic characteristics of glazes (left side) and biscuit-glaze interfaces (right side) ascribed to glazed tiles of the circle of João de Góis. From top to bottom: samples Az013/L1; Az013/L2; Az032/00; Az032/01; Az068/02 and Az068/13

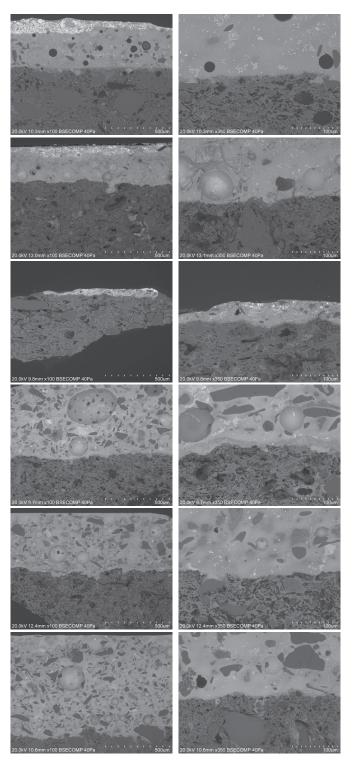


Figure 8. SEM-BSE images showing the main micro-morphologic characteristics of 17th century Portuguese tiles glazes (left side) and biscuit-glaze interfaces (right side). From top to bottom: samples Az003/00; Az024/00; Az100/01; Az197/01; Az198/AR2 and Az052/00. The darker colour of the glaze as compared with e.g. the previous figure results from a lower content in Pb

3.2. Glaze composition

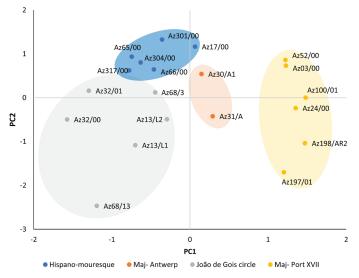
Table 2 includes the semi-quantitative results of analyses of the glazes by EDS in weight %. Sn was excluded for the reasons pointed out in section 2.2. The amount of oxygen was calculated through the remaining elements stoichiometry considering their most commonly considered oxides (Na₂O, MgO, Al₂O₃, SiO₂, K₂O, Fe₂O₃ and PbO). The results were normalized to 100 % and the table also indicates the ratios Si/Pb.

| Sample | Group* | Na | Mg | Al | Si | К | Fe | Pb | 0 | Si/Pb |
|-----------|--------|-----|-----|-----|------|-----|-----|------|------|-------|
| Az317/00 | HM | 2.3 | 0.7 | 1.3 | 17.9 | 2.5 | 0.4 | 47.8 | 27.1 | 0.4 |
| Az304/00 | HM | 2.0 | 0.6 | 1.4 | 17.1 | 2.9 | 1.0 | 48.6 | 26.5 | 0.4 |
| Az017/00 | HM | 3.5 | 0.4 | 2.1 | 20.4 | 1.8 | 2.3 | 38.5 | 30.9 | 0.5 |
| Az301/00 | HM | 2.7 | 0.8 | 1.8 | 19.3 | 1.6 | 1.1 | 43.6 | 29.2 | 0.4 |
| Az065/00 | HM | 2.5 | 0.7 | 1.6 | 19.1 | 1.1 | 0.7 | 45.8 | 28.6 | 0.4 |
| Az066/00 | HM | 2.6 | 0.6 | 1.6 | 19.8 | 2.0 | 0.7 | 43.5 | 29.3 | 0.5 |
| Az030/A1 | Antw | 2.2 | 0.7 | 1.2 | 23.6 | 4.4 | 0.7 | 34.0 | 33.1 | 0.7 |
| Az031/A | Antw | 1.1 | 0.5 | 1.5 | 24.2 | 6.9 | 0.9 | 31.0 | 33.8 | 0.8 |
| Az013/L1 | JGc | 1.2 | 0.4 | 2.9 | 19.5 | 1.7 | 0.5 | 44.2 | 29.5 | 0.4 |
| Az013/L2 | JGc | 1.1 | 0.5 | 3.3 | 20.2 | 1.9 | 1.1 | 41.2 | 30.7 | 0.5 |
| Az032/00 | JGc | 0.9 | 0.5 | 2.4 | 14.5 | 0.9 | 0.6 | 56.1 | 24.1 | 0.3 |
| Az032/01 | JGc | 1.4 | 0.7 | 2.6 | 15.3 | 0.8 | 0.6 | 53.3 | 25.3 | 0.3 |
| Az068/3 | JGc | 1.3 | 0.8 | 3.5 | 17.6 | 1.9 | 0.9 | 45.6 | 28.4 | 0.4 |
| Az068/13 | JGc | 0.7 | 0.1 | 2.3 | 19.4 | 1.4 | 0.8 | 46.6 | 28.7 | 0.4 |
| Az003/00 | PT17 | 1.3 | 1.1 | 2.9 | 25.9 | 6.0 | 2.4 | 23.1 | 37.3 | 1.1 |
| Az024/00 | PT17 | 1.7 | 0.8 | 4.7 | 26.5 | 5.8 | 1.2 | 20.5 | 38.8 | 1.3 |
| Az100/01 | PT17 | 2.4 | 0.9 | 4.3 | 28.7 | 4.4 | 0.9 | 17.7 | 40.6 | 1.6 |
| Az197/01 | PT17 | 2.1 | 0.3 | 3.7 | 30.8 | 4.6 | 0.5 | 16.1 | 41.8 | 1.9 |
| Az198/AR2 | PT17 | 2.5 | 0.4 | 4.2 | 29.0 | 6.3 | 0.7 | 16.2 | 40.7 | 1.8 |
| Az052/00 | PT17 | 4.1 | 0.9 | 4.3 | 24.3 | 4.1 | 1.3 | 24.4 | 36.7 | 1.0 |

Table 2.Semi-quantitative composition (% w/w) of the glazes determined by EDS (weight of
the elements normalized to 100 %) and Si/Pb ratio

* HM - Hispano-Moresque; Antw – Antwerp; JGc – 16th century, circle of João de Góis; PT17 – 17th century Portuguese.

Figure 9 shows the results of a log-based principal component analysis (PCA) of the glazes of all samples, considering the analytical results in Table 2, through a plot in the plane of the two first principal components (PC1 and PC2). PC1 explains 47 % of the variation and is controlled in the positive sense mostly by the contents in Si and K; and in the opposite sense singly by the content in Pb, as can be seen from the loadings plot of Figure 10 in which the projections of the vectors on an axis show the contribution of each element to the respective principal component. PC2 explains 21 % and is controlled in the positive sense mostly by the contents in Mg, Na and Fe; and in the opposite sense mostly by the contents in Mg, Na and Fe; and in the score plot of Figure 9, and these positions together with the provenance of the samples, when known, are the bases for a graphical clustering that is much clearer than the perception obtained directly from the tables of results.





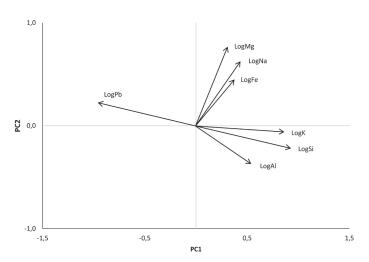


Figure 10. Loadings plot of the PCA analysis of the glazes

3.3. Biscuit composition

Table 3 includes the semi-quantitative results of analyses of the biscuits by EDS in weight %. Pb was excluded for the reasons pointed out in section 2.2. The amount of oxygen was calculated through the remaining elements stoichiometry considering their most commonly used oxides (Na₂O, MgO, Al₂O₃, SiO₂, K₂O, CaO and Fe₂O₃). The results were normalized to 100% and the table also indicates the ratios Ca/Si.

| Sample | Group* | Na | Mg | Al | Si | К | Ca | Fe | 0 | Ca/Si |
|-----------|--------|-----|-----|------|------|-----|------|-----|------|-------|
| Az317/00 | HM | 3.5 | 2.9 | 7.1 | 22.9 | 1.9 | 15.7 | 2.8 | 43.3 | 0.7 |
| Az304/00 | HM | 1.6 | 4.2 | 7.2 | 19.7 | 1.3 | 19.8 | 3.9 | 42.1 | 1.0 |
| Az017/00 | HM | 1.1 | 7.8 | 7.9 | 21.3 | 1.7 | 12.3 | 4.0 | 43.8 | 0.6 |
| Az301/00 | HM | 1.9 | 2.5 | 7.0 | 21.2 | 2.1 | 18.3 | 4.5 | 42.4 | 0.9 |
| Az065/00 | HM | 1.7 | 3.3 | 7.5 | 20.2 | 1.8 | 19.1 | 4.2 | 42.2 | 0.9 |
| Az066/00 | HM | 2.2 | 2.1 | 7.3 | 25.3 | 1.5 | 13.9 | 3.1 | 44.6 | 0.5 |
| Az030/01 | Antw | 1.8 | 1.8 | 8.5 | 21.5 | 1.6 | 18.3 | 3.6 | 43.0 | 0.9 |
| Az031/A | Antw | 1.0 | 2.7 | 6.8 | 21.9 | 1.2 | 20.0 | 3.5 | 42.9 | 0.9 |
| Az013/L1 | JGc | 1.3 | 1.7 | 8.4 | 26.5 | 2.6 | 10.6 | 3.3 | 45.5 | 0.4 |
| Az013/L2 | JGc | 1.2 | 1.4 | 8.3 | 26.8 | 3.2 | 9.2 | 4.4 | 45.5 | 0.3 |
| Az032/00 | JGc | 1.5 | 1.7 | 10.5 | 25.3 | 3.8 | 6.4 | 5.3 | 45.4 | 0.3 |
| Az032/01 | JGc | 1.2 | 2.2 | 9.1 | 24.6 | 2.6 | 9.5 | 5.9 | 44.9 | 0.4 |
| Az068/03 | JGc | 1.3 | 1.3 | 7.7 | 28.9 | 2.5 | 8.7 | 3.2 | 46.4 | 0.3 |
| Az068/13 | JGc | 0.7 | 0.9 | 7.9 | 28.5 | 4.0 | 8.7 | 3.2 | 46.0 | 0.3 |
| Az003/00 | PT17 | 1.4 | 2.3 | 5.9 | 17.8 | 1.0 | 28.9 | 2.5 | 40.3 | 1.6 |
| Az024/00 | PT17 | 1.4 | 1.5 | 6.7 | 16.8 | 1.6 | 28.5 | 3.6 | 39.9 | 1.7 |
| Az100/01 | PT17 | 1.7 | 2.0 | 7.5 | 20.1 | 2.1 | 20.9 | 3.9 | 41.9 | 1.0 |
| Az197/01 | PT17 | 1.6 | 2.0 | 6.2 | 19.0 | 1.3 | 25.9 | 3.0 | 41.0 | 1.4 |
| Az198/AR2 | PT17 | 1.4 | 1.9 | 6.8 | 17.8 | 1.1 | 26.7 | 3.8 | 40.5 | 1.5 |
| Az052/00 | PT17 | 1.6 | 1.6 | 6.8 | 16.3 | 1.6 | 28.4 | 4.0 | 39.7 | 1.7 |

Table 3.Semi-quantitative composition (% w/w) of the biscuits determined by EDS (weight of
the elements normalized to 100 %) and Ca/Si ratio

* HM - Hispano-Moresque; Antw – Antwerp; JGc – 16th century, circle of João de Gois; PT17 – 17th century Portuguese

Figure 11 shows the results of a log-based principal component analysis (PCA) of the biscuits of all samples, considering the analytical results in Table 3, through a plot in the plane of the two first principal components (PC1 and PC2). PC1 explains 46 % of the variation and is controlled in the positive sense mostly by the contents in Al, Si and K; and in the opposite sense mostly by the content in Ca, as can be seen from the loadings plot of Figure 12. PC2 explains 18 % and is controlled in the positive sense mostly by the content in Si (Figure 12).

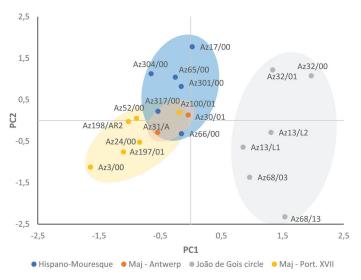


Figure 11. Score plot of the PCA analysis of the biscuits with tentative clustering of the groups

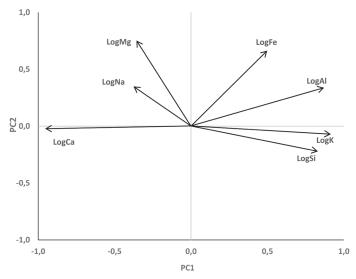


Figure 12. Loadings plot of the PCA analysis of the biscuits

4. DISCUSSION

4.1. Morphology of the glazes

A comparison of the images in Figures 5 to 8 shows that the morphology of the glazes of the productions within the technological circle of João de Góis is different from all others. The most revealing difference lies in the crystalline outgrowth from the biscuit into the glaze. This is determined by both the composition and the firing cycle, but particularly by the period the tiles are kept at a temperature high enough for the glaze to be molten and by a long cooling time [9; 10]. Antwerp and later Portuguese productions that we analysed are completely different in this respect, depicting minimal crystalline outgrowths in the interface. Only Hispano-Moresque tiles depict, at times, a similar interface – Figure 6 and [1, white glazes¹ in pp. 161-196] – but the size of the crystals is usually smaller and the glaze is often devoid of any large inclusions, contrarily to tiles from the circle of João de Góis.

4.2. Composition of the glazes

As pertains the glazes, the PCA score plot of Figure 9 allows the graphical separation of the productions of the circle of João de Góis from all the other groups studied. They separate from the coeval Antwerp majolica and later Portuguese tiles by their lower Si/Pb ratio that allowed firing at a lesser temperature, and from Hispano-Moresque productions, which share with them a similarly low Si/Pb ratio, by their higher content in Al and much lower content in Na. It is interesting to note that the same plot also clearly allows a tentative separation of the other groups but particularly of the 17th century Portuguese productions because their Si/Pb ratio is very different.

Except for the tiles from Antwerp, of which we had only two distinct samples available, Table 4 summarizes the results of Table 2, depicting the estimates of the averages of the elemental contents of the glazes, as well as of the Si/Pb ratio. To each value is associated a ca. 90 % confidence interval on the reasonable assumption that Student's t-distribution with 5 degrees of freedom is approximately applicable. Important similarities with the results for the circle of João de Góis are displayed on a green ground while remarkable contrasts are highlighted on a red ground.

| Averages with ca. 90 % confidence intervals | Na | Mg | Al | Si | K | Fe | Pb | Si/Pb |
|---|---------|---------|---------|----------|---------|---------|----------|---------|
| Circle of João de Gois | 1.1±0.2 | 0.5±0.2 | 2.8±0.4 | 17.8±2 | 1.4±0.4 | 0.7±0.2 | 47.8±4.7 | 0.4±0.1 |
| Hispano-moresque | 2.6±0.5 | 0.6±0.1 | 1.6±0.3 | 18.9±1.2 | 2±0.6 | 1±0.6 | 44.6±3.7 | 0.4±0.1 |
| Portugal 17th century | 2.3±0.9 | 0.7±0.3 | 4±0.7 | 27.5±2.4 | 5.2±0.9 | 1.2±0.7 | 19.7±3.6 | 1.5±0.4 |

Table 4.Averages of glaze elemental contents (% w/w) and Si/Pb ratios estimated with ca.90 % confidence intervals

¹ Faience glazes are white and for a strict comparison of Hispano-Moresque to them, the white glazes should be preferred.

4.3. Composition of the biscuits

As pertains the biscuits, the PCA score plot of Figure 11 also allows the separation of the productions of the circle of João de Góis from all the other groups studied. The separation is based on their much lower Ca/Si ratio as well as on the higher content in K.

The Hispano-Moresque and later 17th century Portugal majolica azulejos groups are partially entangled. The later Portuguese productions have on average higher Ca/Si ratios (1.5 vs 0.8) compared to the Hispano-Moresque tiles. However, the separation is not as clear as it is for the glazes. Antwerp tiles cannot be separated from the previous groups based on the two first principal components depicted in Figure 11.

Table 5 summarizes the results in Table 3, depicting the estimates of the averages of the elemental contents of the biscuits, as well as of the Ca/Si ratio. To each value is associated a ca. 90% confidence interval as before. The tiles from Antwerp where not considered since we had only two distinct samples available.

In this case, similarities do not seem particularly relevant as they pertain mostly to lowcontent elements present in clays. However, contrasts are revealing and possibly valuable as rough indicators of provenance and are therefore highlighted on a red ground for comparison with the values for the circle of João de Góis, shown on a green ground. They refer to the low Ca content and related Ca/Si ratio of the productions of João de Góis as compared to the remaining groups studied. The high content in K is also important since it does not have a counterpart in either Hispano-Moresque or later Portuguese productions.

| Averages with ca. 90 % confidence intervals | Na | Mg | Al | Si | К | Ca | Fe | Ca/Si |
|---|---------|---------|---------|----------|---------|---------|---------|---------|
| Circle of João de Gois | 1.2±0.2 | 1.5±0.4 | 8.6±0.9 | 26.8±1.4 | 3.1±0.5 | 8.9±1.2 | 4.2±1.0 | 0.3±0.0 |
| Hispano-moresque | 2.0±0.8 | 3.8±2.1 | 7.3±0.3 | 21.8±2.0 | 1.7±0.3 | 16.5±3 | 3.7±0.7 | 0.8±0.2 |
| Portugal 17th century | 1.5±0.1 | 1.9±0.3 | 6.6±0.6 | 18±1.4 | 1.4±0.4 | 26.6±3 | 3.5±0.6 | 1.5±0.3 |

 Table 5.
 Averages of biscuit elemental contents (% w/w) and Ca/Si ratios estimated with 90 % confidence intervals

4.4. Origin of the earliest azulejo technology in Portugal

Given that the earliest Portuguese azulejo productions are different from those that preceded them in the Peninsula, a question can be put forward: from where was the technology acquired? To hypothesize about a source based on the characteristics discussed in this paper, three potentially indicative aspects may be considered: the composition of the biscuits; the composition of the glazes; and the morphologic features.

For the production of the biscuits, potters had to rely on clays and marl available locally and therefore the compositions could result more from the geological context than from technical options. The glaze, on the other side, is a product of the technology of the workshop, made from selected raw materials according to a prescription presumably obtained elsewhere and eventually incorporating local specificities or improvements. The morphology of the glaze and of its interface with the biscuit depends, in a great measure, of the kiln and associated firing technique [9; 10] – therefore it may reflect conditions previously established for the firing of glazed ceramics other than azulejos, using an already extant kiln, rather than a workshop option. Accordingly, the glaze composition is the most promising discriminant characteristic as pertains the identification of a technological source. However, for the purposes of this section, the Si/Pb is not particularly relevant because it, too, may well derive from an imposition of the firing conditions.

On dealing with the preparation of the glazes in his 16th century treatise *Li tre libri dell'arte del vasaio* (*The three books of the potter's art*), Cipriano Piccolpasso describes the raw glaze as a mixture of *marzacotto* with lead and tin compounds. The marzacotto follows several recipes according to the region of Italy but those more often mentioned are: i) a mixture of sand with calcined lees or tartar (potassium carbonate); or ii) the same with a third component- sea salt [11, pp. 62-81].

For a visual support to the argument that will follow, Figure 13 depicts the relevant parts of eight EDS glaze spectra: two for the circle of João de Góis (Az013/L2 and Az068/03); two for Hispano-Moresque tiles (Az017 and Az065); two for both Antwerp tiles (Az030 and Az031); and two for 17th century Portuguese tiles (Az100 and Az198). The peaks considered more important for comparative purposes are identified in all spectra and on considering them it will be noticed that in the glazes produced within the circle of João de Góis the content in sodium is low, sometimes only residual. A study of the glazes of Graça church, the earliest group of panels from that circle identified so far [6], demonstrated, through the correlation matrix of the elemental composition of the glaze, that the contents in Si and K were highly correlated, but Si was uncorrelated with Na. From all this we may deduce that the workshop of João de Góis, at least in an early phase, prepared a *marzacotto* from sand and a source of potassium but did not add sea salt. Reviewing the spectra in Figure 13, the only sample where we recognize a similar and unusually low content in Na is Az031/A – one of the azulejos supplied in 1558 by an Antwerp workshop for the Ducal Palace of Vila Viçosa [2] but this is not enough to define a correlation.

The fact that no obvious similarity was observed with the Hispano-Moresque glaze composition and that by the time João de Góis may have been in Seville there seemingly was not a local majolica technology capable of quality results [12] is relevant and leaves as possible technological sources of his craftsmanship Antwerp and Italy. Two Flemish majolica masters are known to have been in the Iberian Peninsula in the 1550s [13] and João de Góis employed a Venetian [3].

The remark that dissimilarities are more noteworthy than similarities leaves the question of the technological ancestry of the technique used open until more results are available but we should also advance the possibility that the technology was perfected locally from several sources and therefore a definite origin may be impossible to pinpoint.

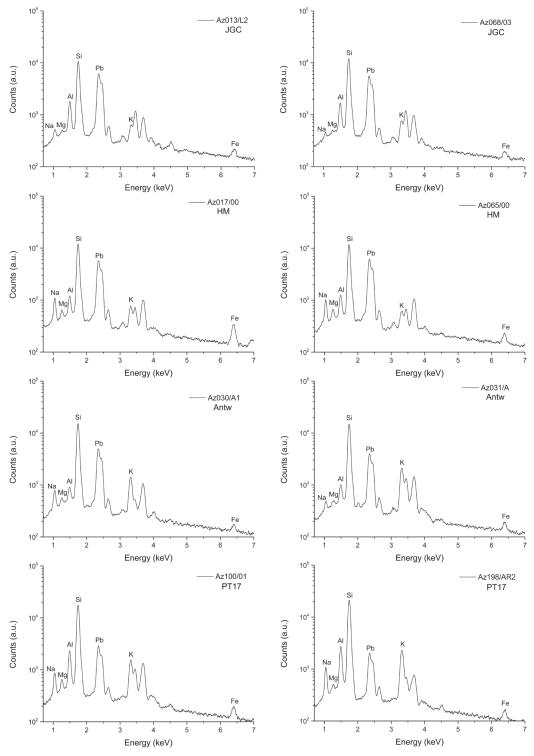


Figure 13. EDS spectra of the glaze analyses of (from left to right and from top to bottom): Az013/ L2 and Az068/03 (João de Góis circle); Az017/00 and Az065/00 (Hispano-Moresque); Az030/A1 and Az031/A (Antwerp); Az100/01 and Az198/AR2 (17th century Portugal)

5. CONCLUSION

The azulejo productions of the circle of João de Góis, as represented by the monogrammed panels of Graça church in Lisbon, the panel Nossa Senhora da Vida conserved at the Museu Nacional do Azulejo, and the signed and dated panels lining the São Roque chapel in Lisbon, constitute a cluster distinct from previous productions (Hispano-Moresque tiles presumed of Seville manufacture and majolica tiles from Antwerp). They are also very different from later Portuguese productions of the 17th century. The differences pertain mostly to the morphology and composition of the glazes. The very distinct morphology that endured through the period of ca. 20 years covered by the three panels considered is related, not only with the composition, but also and particularly with a possibly long firing cycle.

This result is capital because the composition and morphology of the glazes of the circle of João de Góis should now be sufficient, in most cases, to separate them from other nearly contemporary productions imported to Portugal, as well as from later Portuguese productions. The biscuits, though more variable, also depict distinguishing characteristics that may help sustain doubtful attributions to the circle of João de Góis and to a Portuguese production spanning from the 1560s (maybe even before) to, at least, the 1580s.

It is certainly fortunate for the research of the early production of majolica azulejos in Portugal that such clear differences were found. The composition of the glazes as pertains the Si/Pb ratio was, in a large measure, connected to the firing method, which depended on the kiln technology available. The constancy of the morphologic characteristics suggests that the firing conditions remained essentially unchanged during this period and maybe the kiln used was always the same. When a new kiln or improved technology became available, allowing a quicker firing at probably a higher temperature, the ratio Si/ Pb was increased to save on the cost of lead and the duration of the cycle was decreased to save on both time and fuel, leading to a sharp diminution of the interfacial crystalline outgrowths.

The focus on the kiln technology and the constancy of the firings brings to memory that on both the Holly Inquisition process against João de Góis and the denunciation against his brother Filipe, a kiln is mentioned ("the kiln where glazed ceramics are fired" [3]), maybe precisely the kiln whose firing cycle made these productions so very recognisable.

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