

Characterisation and Technology from Studies of Clay Bodies of Local Islamic Production in Zaragoza (Spain)

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Abstract

The ceramic material considered in this study belongs to Islamic culture and was found inside a kiln dated from 11th to 12th century in Zaragoza (Spain). A selection of 100 samples of various types, function and morphology, have been analysed by petrography and X-ray diffraction. The mineralogical study has been aimed at the characterisation of the different clay bodies completed by textural analysis of their inclusions by means of the Image Digitalisation System applied on the representative microphotographs. Quantitative chemical analyses of major, minor and trace elements were carried out by Inductively Coupled Plasma–Atomic Emission Spectrometry (ICP–AES) in 56 of these sherds which were submitted to computer assisted multivariate statistical treatment. The analytical results indicate that a selection of raw material was used for each production variant. Significant differences in the technology have been detected for each group, above all by the estimation of firing temperature. © 1999 Elsevier Science Limited. All rights reserved

Keywords: islamic ceramics, petrography, X-ray methods, clays, traditional ceramics.

1 Introduction

The study of ancient ceramics from archaeological contexts contributes to our understanding of the society that manufactured and used them. Such investigation becomes more interesting when dealing with ceramics found in their own workshop, since this may include aspects such as the availability

and preparation of raw materials, techniques and traditions of fabrication and the methods of decoration together with an assessment of how these may have changed both spatially and temporally.

The studied samples come from excavations carried out recently by the Archaeological Service of the County Council of Zaragoza, in the NE of Spain. These archaeological remains are located on the Western side of the medieval city where the Muslim people exploited a flourishing potterywork dating from the 10th–11th century. Remains have been found of kilns, wasters, potter's tools and a large quantity of richly decorated ceramics. These kilns are of oriental type and very little known in Europe.^{1,2}

The ceramic material considered in this study was found inside one of these kilns dated from 11th to 12th century AD. Production variants show different forms and decorations ranging from: elite pottery finely made and elaborately decorated, crockery pots with different decoration, coarsewares or utilitarian pottery and pottery for cooking (Fig. 1).

Decoration techniques comprise a great variety of coatings applied to the bodies:

- *melado*, a honey-yellow transparent lead glaze,
- *verde y manganeso*, an opaque tin glaze decorated with green and black designs, used especially on plates (*ataifores*),
- *cuerda seca* consists of drawing the design with a black slip on the surface body and filling up the spaces between the lines, with coloured glazes, either total or partially,
- *almagra*, deep-red unglazed slip.

The aim of this study is to characterise the pottery production of this workshop by determining the mineralogical–chemical composition and the

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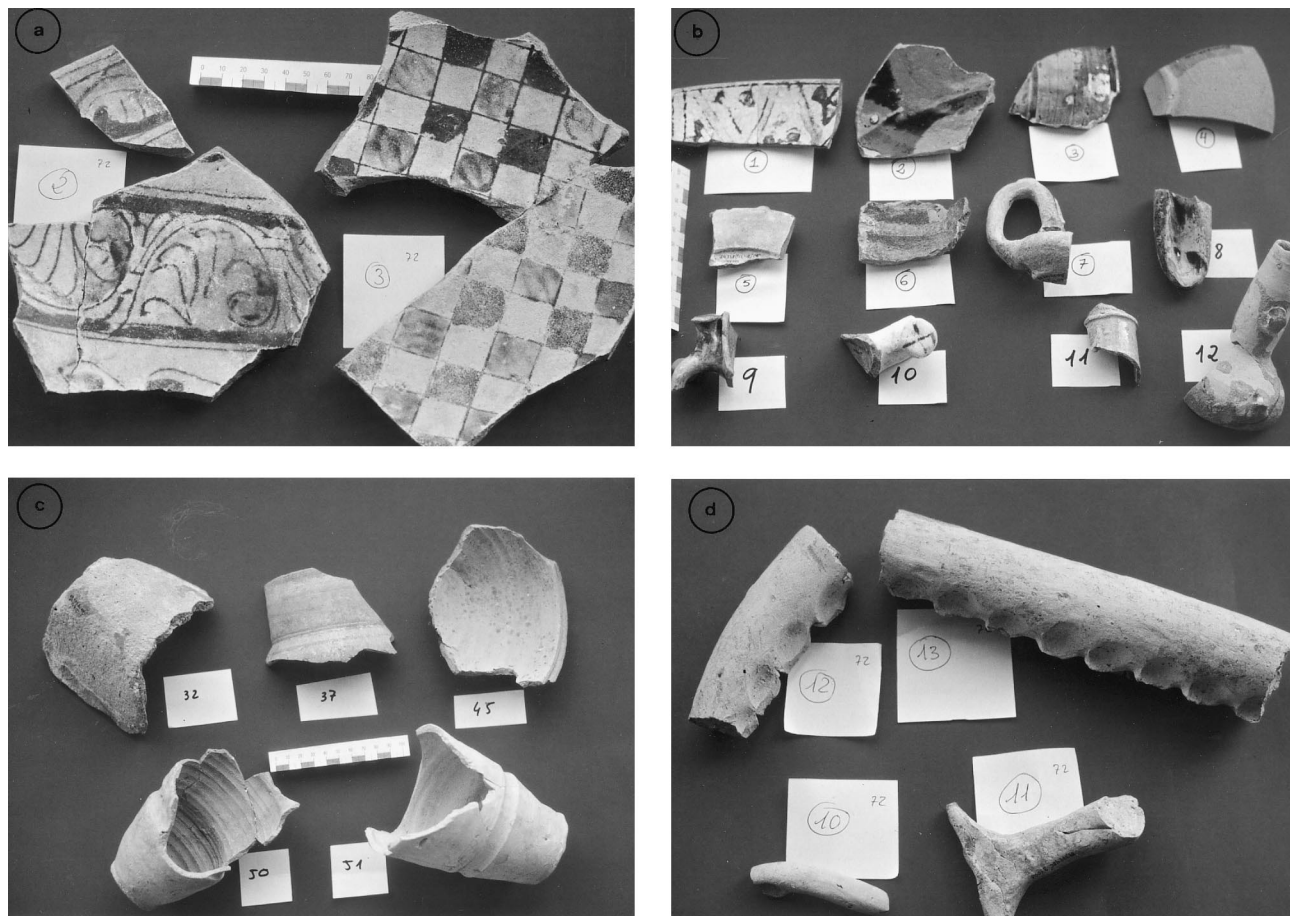


Fig. 1. Different ceramic material of the Islamic culture (11th to 12th century) found in archaeological excavations from Zaragoza (Spain): (a) two different plates (*ataifores*) with *verde y manganeso*; decoration; (b) small ceramic pieces with glazed decoration (*cuerda seca* and *melado*); (c) utilitarian coarsewares; (d) kiln tools.

texture of the ceramic bodies in order to know whether the variation of form and decoration implies the use of different raw materials. Finally, this study intends to create a ceramic reference group of the Islamic culture for further studies of material distribution in other parts of the *al-Andalus* (Islamic Spain). The technology for decorating these Islamic objects by using different coatings (transparent glazes, tin-glazes and slips) was also studied by scanning electron microscopy.³⁻⁷

2 Experimental Methodology

A selection of 100 samples of various types, function and morphology, of five different excavated levels in the same kiln have been analysed by the usual mineralogical techniques (XRD and optical microscopy). The diffractometric analysis was performed using the method of non-oriented powders with a Philips X-ray diffractometer under the following experimental conditions: K_{α} Cu radiation, graphite monochromator, 0.5 2θ step scan, 5 s/step counting time, 4 to 50° 2θ range, 20 mA and 40 kV tube conditions. Semiquantitative data were estimated following the Schultz method.⁸ Estimations

of body colours were made using the Munsell charts.

Petrographic analysis were carried out on adequate oriented thin sections of each sample, cut across the thickness of the sherds. Some of these were stained with Alizarin red S to distinguish dolomite from calcite. Particular attention was paid to the optical properties of the clay matrix (birefringence), the presence of some relict phases (such as carbonates in the matrix, or carbonate ground-mass containing free CaO derived from the dissociation of carbonate during firing), as well as reaction rims around carbonate grains produced by increasing the firing temperature, with the formation of prograde phases (diopside, gehlenite). Some secondary or retrograde phases such as gypsum or calcite can occasionally be found cementing the pores and/or fissures, even as a carbonatic matrix.

Image analysis of thin sections offers great scope to the petrologist for recording a wide range of quantitative data to support qualitative descriptions of ceramics. Its application to the textural analysis was undertaken in order to discriminate between groups of fabrics and clarify information about the possible manipulation of raw materials. Measurement of morphological properties and

grain size distribution were carried out using imaging processing from representative microphotographs. In most cases the system works well with crossed polarised light, but limitations sometimes occur in identifying the boundaries of composite inclusions. To solve this problem, a combination of both light conditions (crossed and plane polarized) were used. The software utilised has been developed by the Technical and Scientific Services of Zaragoza University.

Chemical analysis of major, minor and trace elements was carried out on 56 of these sherds by Inductively Coupled Plasma–Atomic Emission Spectrometry (ICP–AES). This analysis was undertaken at the Department of Geology, Royal Holloway College, University of London, on representative 500 mg sub-samples drilled from freshly fractured surfaces of the sherds. Fusion was used to prepare sample solutions. Ten major elements (Si, Al, Fe, Mg, Ca, Na, K, Ti, P, Mn) and some trace elements (Ba, Ce, Dy, Eu, La, Li, Nb, Nd, Sc, Sm, Sr, V, Y, Yb, Zr) were determined in every sample. Chemical data were submitted to a computer assisted multivariate statistical treatment by cluster analysis.

3 Analytical Results

3.1 Petrographic and mineralogical results

The results of Optical Microscopy and XRD analyses have led to distinguishing four main Groups (I, II, III, IV). The granulometry of the inclusions characterised by means of digitalised microphotographs has led to a further subdivision of Group I into two subgroups, a and b, respectively.

Table 1 resumes the characterisation of each group based on the application of the mentioned techniques. Note that Groups Ia and Ib correspond to elite pottery finely made and crockery pots with glazed decoration (*melado*, *verde y manganeso* and *cuerda seca* techniques). Groups II and III represent the utilitarian pottery (coarsewares and kiln tools) while the unglazed ceramics coated *a la almagra* belong to Group III. Finally, in Group IV are exclusively pottery for cooking.

The grain size distribution of each distinguished group is given by the average value in Table 2. Its representation, in accumulative percentage, is shown in Fig. 2.

Figures 3–6 illustrate the results of the image analyses of Groups Ia, Ib, II and III, respectively.

3.1.1 Group I

Group I consists of a paste with isotropic optical behaviour, indicating its amorphous vitreous character after firing, together with dispersed iron oxides,

Table 1. Resume of petrography, XRD, granulometry of the inclusions and statistical analysis of chemical results

Production variants:	Glazed decoration		Kiln Tools/ Pottery		Cooking
	Ia	Ib	II	III	
Groups				Almagra	IV
Matrix					
Anisotropic: Clay (Illite)				X	
Calcite			x	X	
Dolomite				X	
Isotropic:	X	X	X		X
Neoformation minerals: (Di, Gh, Ess, An)	X	X	x		x
Inclusions					
Qtz + Kfs	x				
Qtz + Kfs + Met. Rock + Chert		x	x	x	x
Carbonates			x	x	
Grogs					x
Granulometry					
< 7% (sand < 4%)	x				
7–12% (sand < 8%)				x	
7–12% (sand > 8%)			x		
10–15% (sand > 8%)		x			
15–25% (sand > 8%)					x
Chemical analysis	Cluster 1	Cluster 2	Cluster 3	Cluster 4	

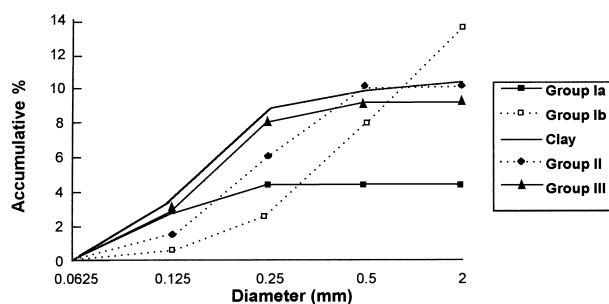
occasionally small laths of prograde minerals and yellow rims of reactions around lime nodules (Fig. 7). The presence of microvoids is common, which is confirmed by SEM observations. XRD analyses have corroborated the presence of prograde minerals from the thermal modifications such as diopside, esseneite, gehlenite, anortite, etc. No evidence of carbonates or clay minerals was reported from this technique, as a consequence of the loss of their crystal structures.

The ratio between clay matrix and aplastic inclusions allows the separation into subgroup Ia (with an abundant clay matrix, > 93% in volume) and Ib (with a variation of matrix between 85 and 90%). Both percentages correspond to the extreme values from all the distinguished groups. The lithology of inclusions is homogeneous in subgroup Ia, with subangular quartz and occasionally quartzite, while Ib presents quartz, K feldspar and rock fragments such as quartzite, graywacke, phyllite and chert. These components have a different grain size distribution in both subgroups, Ia presents silt and fine sand (< 0.25 mm of diameter) which corresponds to a volume less than 4%. However, 10–15% of inclusions in Ib consist of a very small quantity of silt and all the sand sizes (fine, medium and coarse) which correspond to more than 8%, with a median of 12%. On the other hand, from all the established groups, this latter distribution is the only one that presents coarse sand (0.5–2 mm of diameter) [Fig. 8(a)].

Group I corresponds to decorated pieces which were glazed using the *cuerda seca*, *melado* and *verde*

Table 2. Average values of the grain size distribution of each ceramic group and the average value of 25 samples of miocene regional clay

	%matrix	% silt (< 0.0625 mm)	% fine sand (< 0.25 mm)	% medium sand (< 0.5 mm)	% coarse sand (< 2 mm)
Group Ia	95.63	2.65	1.72	0	0
Group Ib	86.57	0.52	2.14	5.03	5.74
Group II	89.01	1.38	4.66	4.05	0
Group III	90.89	2.78	5.18	1.15	0
Clay	89.71	3.41	5.20	1.16	0.52

**Fig. 2.** Grain size distribution of the inclusions in each ceramic group. It is also represented the average distribution of 25 samples of miocene regional clay.

y manganese techniques. Their bodies have a pale yellow (2.5Y 8/3, 2.5Y 8/4) to pinkish white colour (7.5YR 8/2). The differences between subgroups Ia and Ib, seem to be related to the morphology of the ceramics. The larger vessels of coarser grained fabric generally belong to subgroup b.

3.1.2 Groups II and III

Group II is characterised by the presence of a semi-isotropic matrix with no evidence of clay minerals by XRD. The existence of a small quantity of calcite in the ground-mass gives its slightly birefringence together with the incipient prograde minerals [Fig. 8(b)]. The presence of phyllosilicate platelets in the matrix of Group III marks the anisotropic behaviour typical of their samples [Fig. 8(c)]. Poorly crystallised iron ores are also present. The XRD analyses confirmed the results of the thin section studies, with particular regard to the differences between both groups. Calcium silicates are not present in the matrix of Group III where, however, illite, calcite and dolomite were identified. These results indicate a relevant difference in the maximum temperature reached in both calcareous groups, also evidenced by comparing the micro-structural features of the ceramic bodies, observed under optical and electron microscopy.

The lithology of their inclusions is similar to that of Group I with the characteristic difference of containing subrounded limestone fragments and calcareous alga fragments in Group III and eventually in Group II. On the contrary, bodies of these

groups do not present lime nodules or evidence of an extensive melting of their matrix. While individual mineral particles are readily distinguishable in samples of Group III, the degree of interconnection of the particles increases and the porosity decreases from Group II to Group I.

Based on the textural analysis, samples from Groups II and III consist of a relatively abundant matrix (88–93% in volume) with fine-grained inclusions which correspond to silt and fine-medium sand size. Differences were found between the grain size distribution of both groups, which are related to the total quantity of sand (<8% in Group III, >8% in Group II whose distribution coincides in equal parts of fine and medium size).

The vessels corresponding to these groups show no decoration with the exception of the unglazed ceramics coated *a la almagra* belong to Group III. In the same way, the potter's tools correspond to both groups. Their bodies have a pink (7.5YR 7/4) to pale red colour (2.5YR 7/2).

3.1.3 Group IV

Samples of this group are exclusively cooking wares, a few of which are glazed.

This group is characterised by the high content of sand-sized inclusions (>8%), the total ranging between 15 and 25% of temper material. These coarse aplastic inclusions are made up of angular to subangular siliceous rock fragments, quartz, iron ores and grogs held within a matrix having a vitreous textural aspect produced by the high temperatures reached [Fig. 8(d)].

These characteristics allowed resistance to the thermal shock when the wares were placed directly on or in a fire. As the bodies were made porous and coarse, their open texture would allow whatever expansion that might occur to be accommodated by the pore spaces. On the other hand, grog is a chemically inert material which limits shrinkage, reduces drying time and eliminates cracking.⁹

3.2 Chemical results

Chemical analyses by ICP–AES are summarised in Tables 3 and 4. Chemical data were treated statistically by cluster analysis. This statistical treatment

showed samples grouped in four clusters (see dendrogram in Fig. 9), according to their different body compositions and the mineralogical characteristics of their inclusions. In one of these, the smallest cluster

(Fig. 9, cluster 4, at the bottom), samples from Group IV of the mineralogical study were included. They were fragments of non-glazed cooking-pots. The only glazed cooking-pot analysed was kept

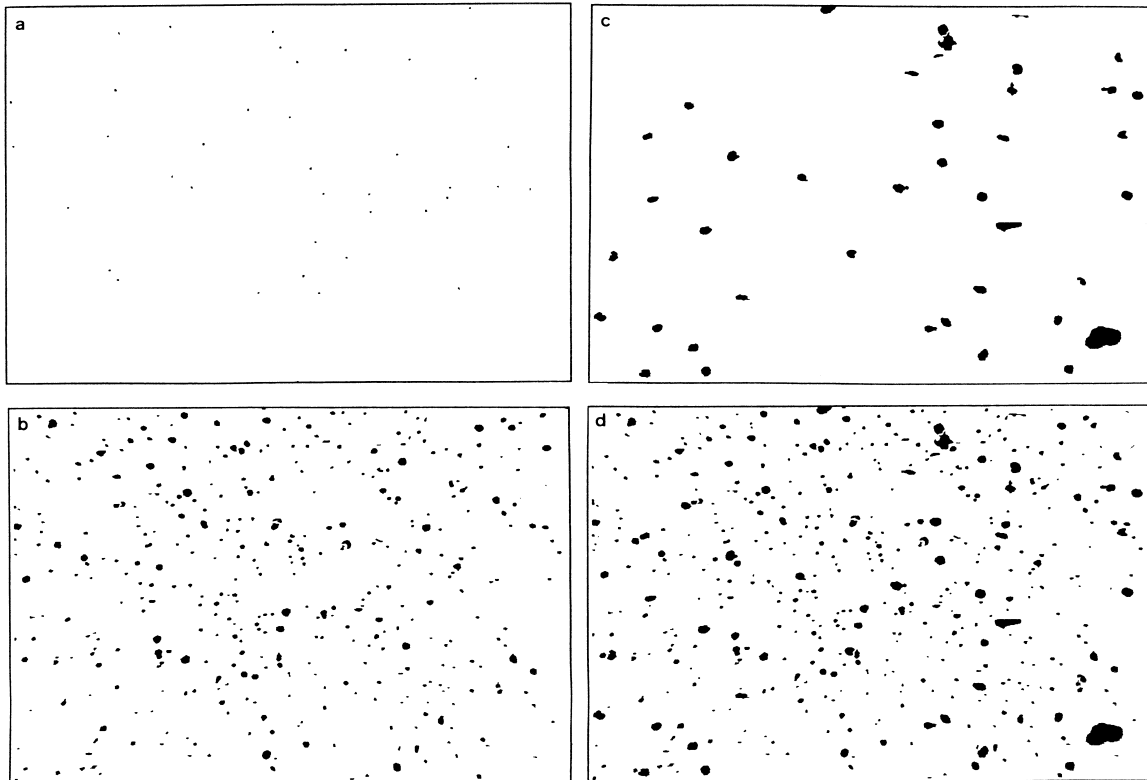


Fig. 3. Example of ceramic Group Ia from the digital image analysis. Image (a) fine silt (<0.0156 mm maximum diameter); (b) coarse silt (0.0156 – 0.0624 mm); (c) fine and medium sand (0.0625 – 0.499 mm); (d) sum of particle sizes. Long side is 3 mm.

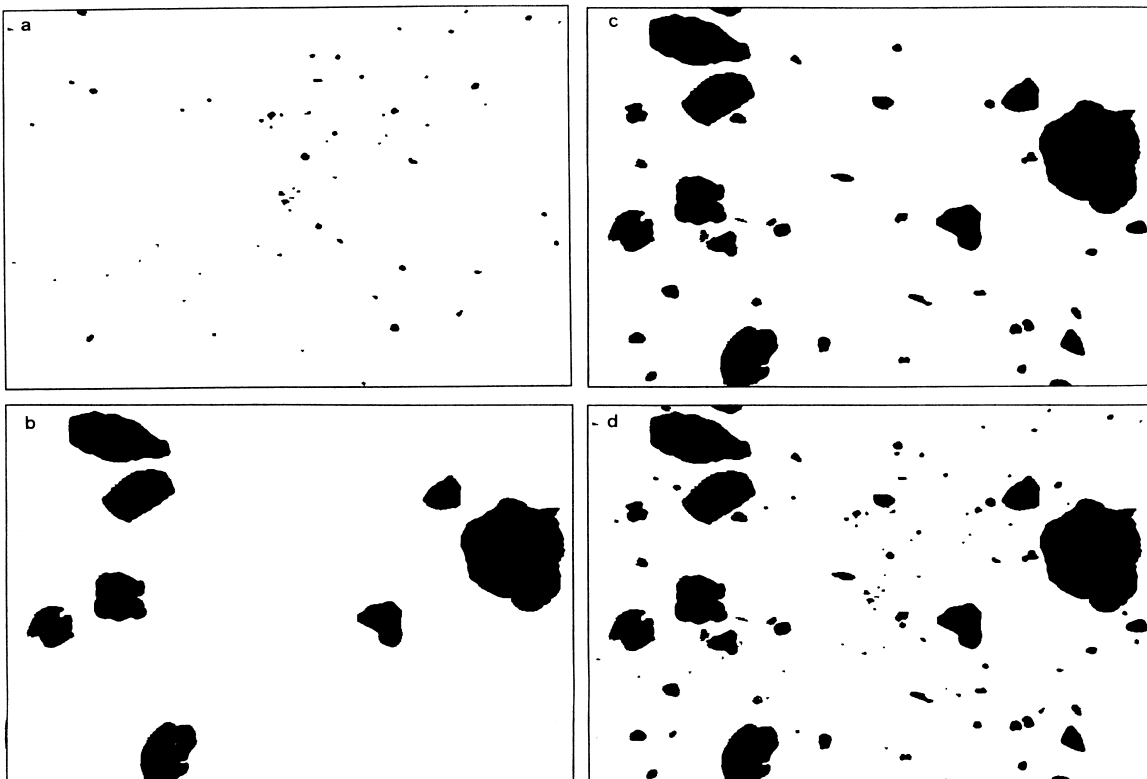


Fig. 4. Example of ceramic Group Ib from the digital image analysis. Image (a) fine and coarse silt (<0.0156 – 0.0624 mm, maximum diameter); (b) medium sand (0.25 – 0.499 mm); (c) fine and medium sand (0.062 – 0.499 mm); (d) sum of particle sizes. Long side is 3 mm.

separate from them. Another three more important clusters were also found: cluster 1 with samples from Groups Ia and Ib, cluster 2 of samples from Groups Ib and II, and cluster 3 with objects only from Group

III. Average compositions of these clusters were calculated and are shown in Tables 3 and 4, with the major and some minor components, quoted as wt% oxide, and trace elements, quoted as ppm.

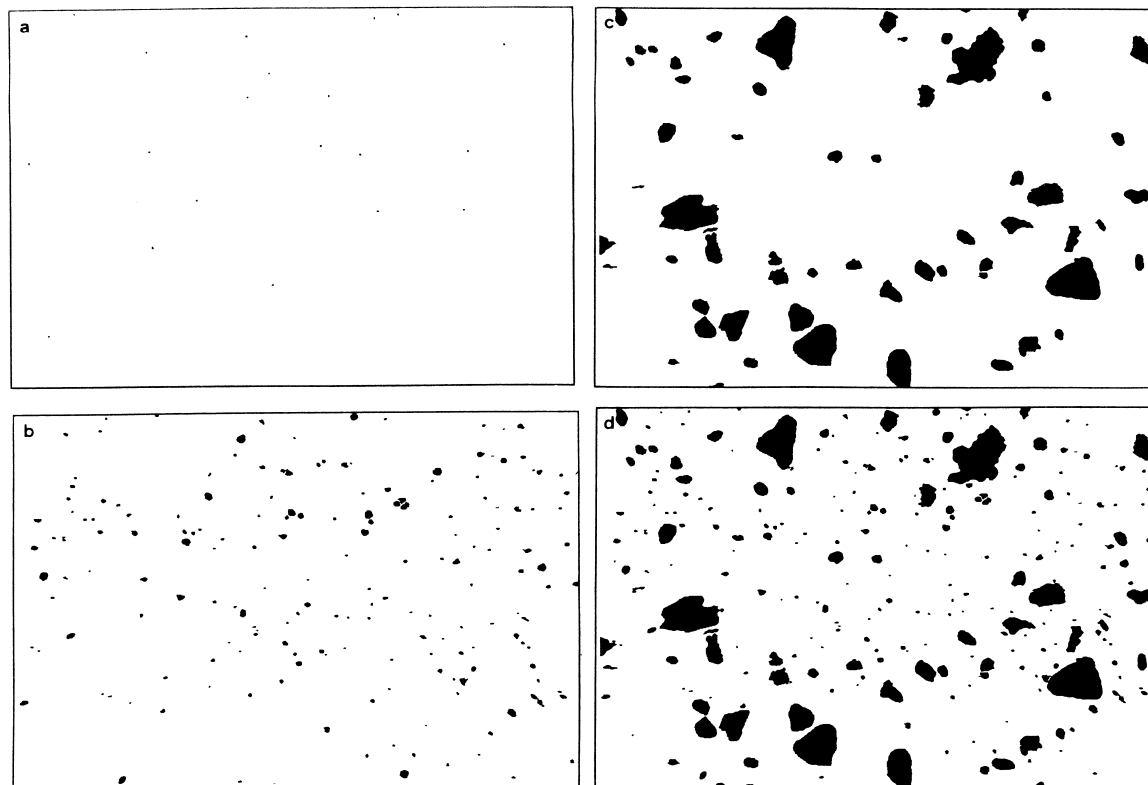


Fig. 5. Example of ceramic Group II from the digital image analysis. Image (a) fine silt (< 0.0156 mm maximum diameter); (b) coarse silt (0.015–0.00624 mm); (c) fine and medium sand (0.0625–0.499 mm); sum of particle sizes. Long side is 3 mm.

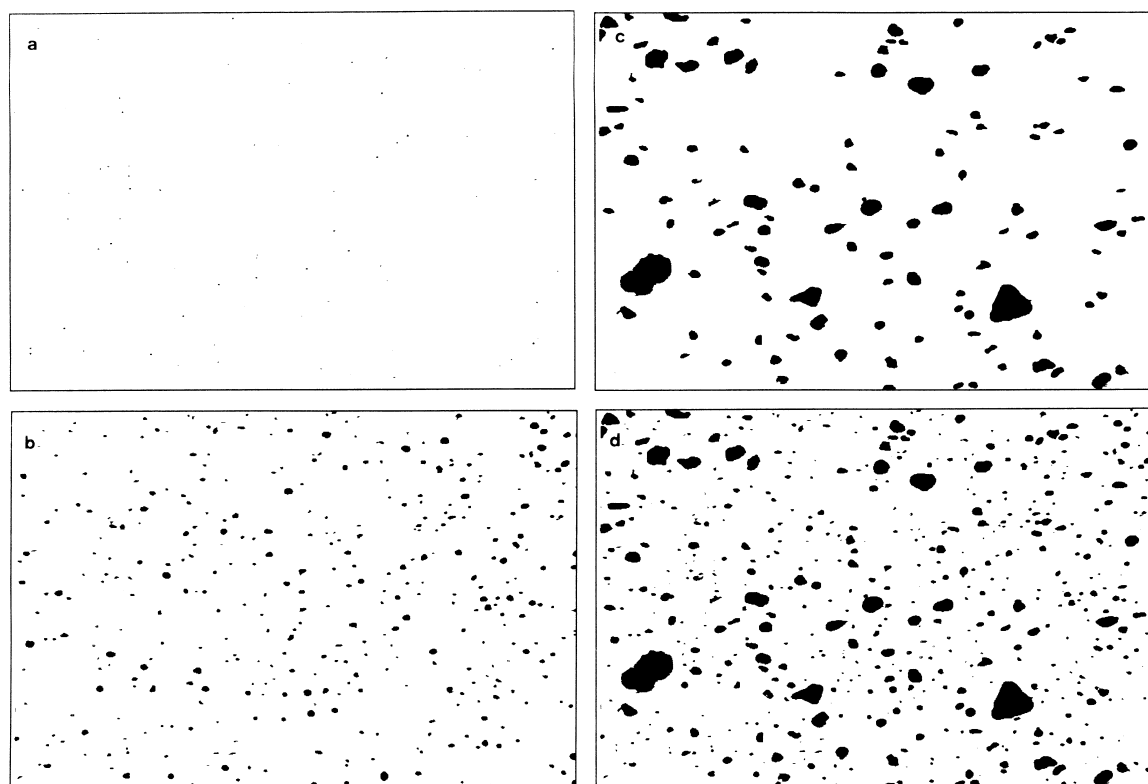


Fig. 6. Example of ceramic Group III from the digital image analysis. Image (a) fine silt (< 0.0156 mm, maximum diameter); (b) coarse silt (0.015–0.00624 mm); (c) fine and medium sand (0.0625–0.499 mm); (d) sum of particle sizes. Long side is 3 mm.

Table 3. Chemical composition of ceramic bodies by ICP–AES (major and minor elements), quoted as wt% of oxides

	<i>SiO₂</i>		<i>Al₂O₃</i>		<i>Fe₂O₃</i>		<i>MgO</i>		<i>CaO</i>		<i>Na₂O</i>		<i>K₂O</i>		<i>TiO₂</i>		<i>MnO</i>	
	<i>x^a</i>	<i>σ^b</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>
Cluster 4 (<i>n</i> = 3)	59.40	±2.26	29.23	±1.75	6.63	±1.35	0.42	±0.03	0.79	±0.18	0.33	±0.18	1.25	±0.33	1.37	±0.11	0.04	±0.05
Cluster 1 (<i>n</i> = 18)	48.55	±2.46	17.23	±0.88	5.90	±0.27	4.10	±0.24	16.89	±1.30	1.73	±0.36	1.95	±0.50	0.62	±0.03	0.08	±0.01
Cluster 2 (<i>n</i> = 18)	49.52	±2.13	16.03	±0.98	5.55	±0.31	3.60	±0.38	16.46	±0.89	0.81	±0.20	3.25	±0.47	0.58	±0.04	0.08	±0.01
Cluster 3 (<i>n</i> = 12)	42.17	±2.20	14.45	±1.39	5.01	±0.45	2.90	±0.38	17.46	±1.21	0.57	±0.14	3.77	±0.39	0.53	±0.04	0.07	±0.01

^a average value.^b standard deviation.**Table 4.** Chemical composition of ceramic bodies by ICP–AES (trace elements), quoted as ppm

	<i>Ba</i>		<i>Sr</i>		<i>Li</i>		<i>Nb</i>		<i>Sc</i>		<i>V</i>		<i>Y</i>		<i>Zr</i>		<i>La</i>		<i>Ce</i>		<i>Nd</i>		<i>Sm</i>		<i>Eu</i>		<i>Dy</i>		<i>Yb</i>	
	<i>x^a</i>	<i>σ^b</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>	<i>x</i>	<i>σ</i>		
Cluster 4 (<i>n</i> = 3)	266	±103	130	±13	113	±12	28	±2	26	±2	182	±48	35	±5	255	±12	55	±5	98	±4	40	±4	8.4	±1.6	1.2	±0.3	4.1	±0.9	3.0	±0.4
Cluster 1 (<i>n</i> = 18)	674	±127	570	±45	69	±11	12	±1	15	±1	105	±6	26	±2	132	±22	37	±3	70	±3	39	±2	7.2	±0.4	1.1	±0.1	3.6	±0.2	2.0	±0.1
Cluster 2 (<i>n</i> = 18)	576	±62	525	±45	84	±9	11	±1	14	±1	98	±9	23	±2	133	±30	35	±2	66	±4	38	±2	6.8	±0.4	1.0	±0.1	3.1	±0.2	1.7	±0.2
Cluster 3 (<i>n</i> = 12)	528	±59	553	±93	78	±10	9	±1	12	±1	87	±9	19	±1	119	±29	28	±3	56	±4	30	±3	5.6	±0.4	0.8	±0.1	2.6	±0.2	1.4	±0.1

^a average value.^b standard deviation.

The group of non-glazed cooking-pots (cluster 4) had low contents of calcium (0.79% CaO), as the main difference, they were made with a non-calcareous matrix, which showed a completely different chemical composition (see silicon, aluminium, iron, potassium, magnesium, etc., in Table 3). It had the highest silicon and aluminium content (59.40% SiO₂, 29.23% Al₂O₃), higher iron and titanium proportions (6.63% Fe₂O₃; 1.37% TiO₂) and the lowest percentages of sodium (0.33% Na₂O), potassium (1.25% K₂O) and magnesium (0.42% MgO). Considering trace elements (Table 4), this group had the highest concentration of most elements (Li, Nb, Sc, V, Y, Zr, La, Ce, Yb).

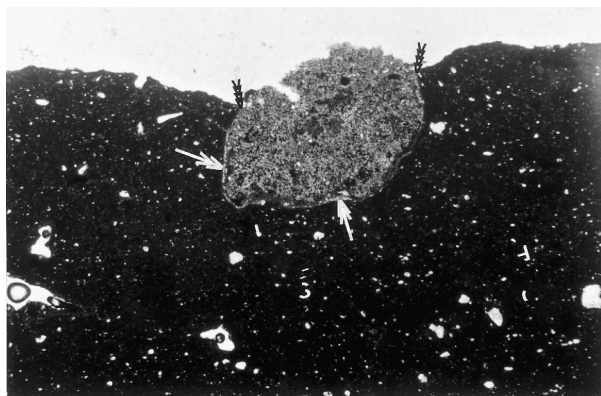


Fig. 7. Photomicrograph of Group Ia showing a yellow rim of reaction around a lime nodule (plane-polarized light). Long side is 3 mm.

Although all cooking-pots were included in the same mineralogical Group IV, chemical results revealed the first technological division between them, since the isolated glazed cooking-pot had a calcareous body (10.48% CaO). Therefore, non-glazed cooking-pots have non-calcareous bodies and on the contrary, glazed objects have calcareous bodies.

The other three clusters had a more similar composition (see Table 3), all with a calcareous matrix. Some differences however, can be observed especially regarding a higher or lower proportion of inclusions. Calcareous bodies had calcium contents between 14.6 and 19.8% CaO. This important quantity of calcium could originate from the use of marly clays for preparing the ceramic bodies. This is corroborated by the petrographic features of Group III where rounded limestone and calcareous algae were identified as natural inclusions. No correlation between magnesium and calcium contents was observed. Calcareous matrices however corresponded to higher proportions of barium and strontium. Concerning other elements, a total correlation between aluminium and iron contents could be observed (Regression analysis, $R^2 = 0.989$) in all calcareous bodies, which would prove that one sole source of raw material was used. Another good correlation exists between potassium and sodium, probably because they are associated to the presence of feldspar in the body inclusions.

Cluster 1 included samples from Group Ia, with the finest body, it had the highest contents of aluminium

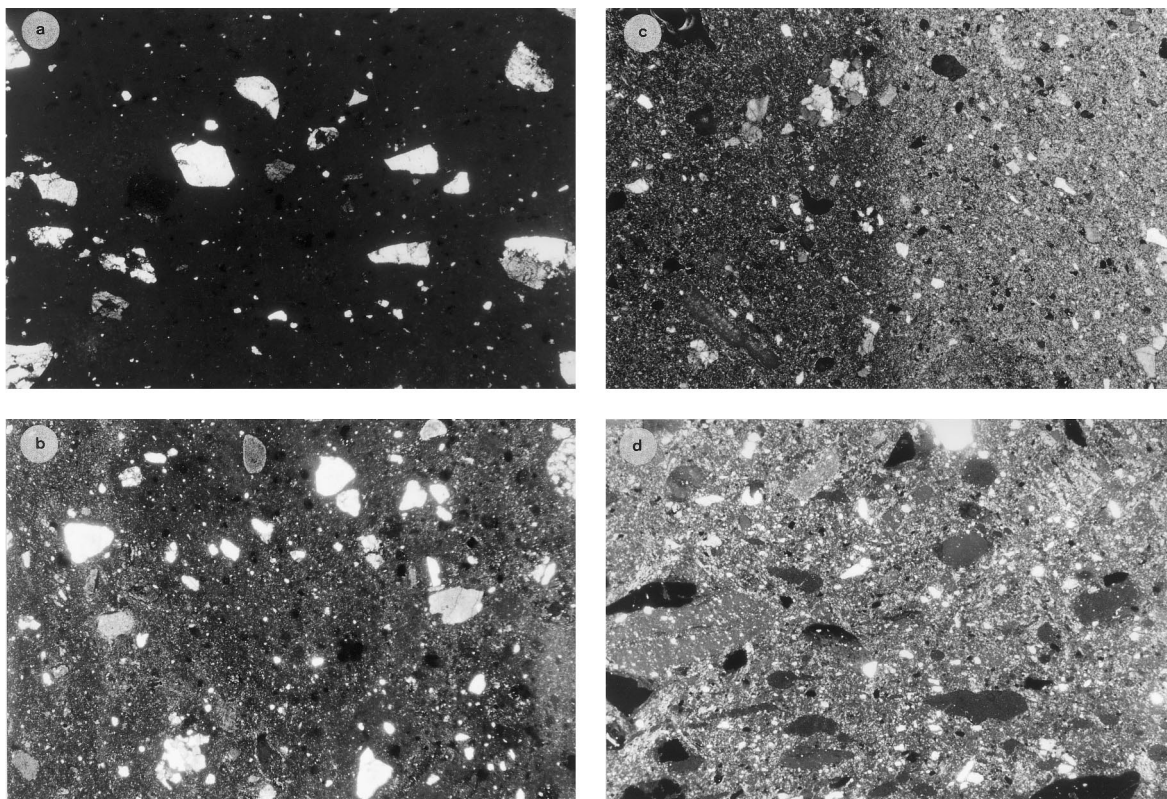


Fig. 8. Photomicrographs of ceramic groups: (a) Group Ib; (b) Group II; (c) Group III; (d) Group IV (crossed nicols). Long side is 3 mm.

(17.23% Al_2O_3), because of the high proportion of clay matrix, and the lowest percentage of potassium (1.95% K_2O), as a consequence of fewer inclusions. This alkaline decrease is discussed below in terms of a possible addition of salt.¹⁰ Cluster 2 included samples from Groups Ib and II, those with more inclusions but with a grain size distribution that indicated some material manipulation, that showed a higher percentage of SiO_2 (49.52%), which agreed with the presence of quartz inclusions in Group Ib. Cluster 3 contained the objects from Group III, the most similar to the clay raw material, with the lowest content of SiO_2 (42.17%) and Al_2O_3 (14.45%), and the highest proportion of K_2O (3.77%).

Considering trace elements (see Table 4), these repeat still further the differences established through major and minor elements, between the three clusters of the calcareous body. Normalised values for rare-earth element (REE) concentrations are represented in Fig. 10 with respect to a sedimentary standard rock as North American Shale Composite (NASC). Normalisation against NASC is usually chosen for sedimentary rocks as a measure of how typical a sediment is and may identify subtle enrichments and deficiencies in certain elements. REE present in a sediment are chiefly transported as particulate matter and reflect the chemistry of their source. REE patterns of the first three clusters (Fig. 10) show a similar behaviour, with a relative abundance of light REE analogous to NASC and a small deficiency in heavy REE (Dy, Yb). REE pattern of the source is most faithfully represented in the clay-sized fraction of the sediment, cluster 1 (Group Ia) in this case, and it can also show the diluting effect of quartz and other inclusions in clusters 2 and 3. Cluster 4 displays a different REE pattern, also according to the use of a different manipulated source.

Technologically, cluster 1 included glazed ceramics, in general small objects; in cluster 2 there were *verde y manganese* tin-glazed plates (*ataifores*) and objects with *cuerda seca* decoration, while the unglazed objects belonged to cluster 3.

4 Discussion

Two aspects are of interest in the discussion of ceramic manufacture: the selection of raw materials and the estimation of firing conditions, with particular emphasis on maximum temperatures of firing. The physical and chemical properties of a ceramic ware are, in fact, determined in the first place by the properties of the clay selected for its production and the possible manipulation (levigation, mixtures, or temper addition during the

manufacture). Once these have been defined, however, the physico-chemical features of the final product depend on the conditions of the firing process.

4.1 Selection of raw material

The application of the image digitalisation technique for characterising the textures of this Islamic pottery has been used successfully to discriminate between different fabrics.

Differences between forms and products with or without decoration were established by the textural analysis. It can be supposed that the same raw material consisting of calcareous illitic clays, or lacustrine marly clays, could have been used for the manufacture of these local Islamic ceramics, with the exception of some cooking-pots. This assumption is based on the chemical results and on the limited differences found in the mineralogical composition of the bodies, which are discussed below in terms of firing conditions.

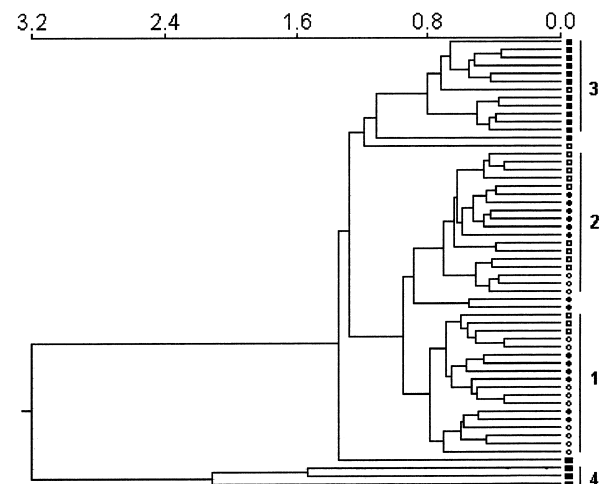


Fig. 9. Dendrogram from statistical analysis, showing the four clusters (○: group Ia; ●: group Ib; □: group II; ■: group III; ■: group IV).

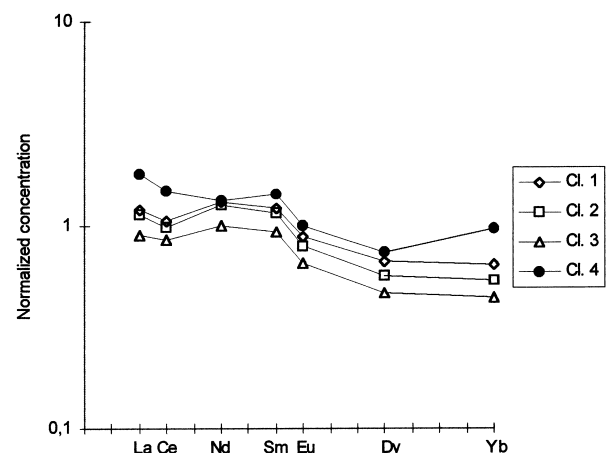


Fig. 10. Rare-earth element abundances in the four ceramic clusters, normalised to North American Shale Composite (NASC), plotted against atomic number.

From a geological point of view, the workshops were located in an area where not only miocene clay deposits have been exploited since ancient times, but also alluvial floods with fine sediments were relatively frequent along the Ebro valley. The lithofacies of the miocene deposits consist of sandy lutites and marly lutites with gypsiferous levels and were deposited in distal alluvial flats and lacustrine systems.¹¹ Their mineralogical composition agree with those determined in the clay bodies.^{11,12} However, comparing the grain size distribution of each group with those of the clay material,¹² no identical patterns were found, especially with regard to the content of coarse sand. Only Group III shows a similar distribution to samples from different deposits.

These miocene calcareous clays could have been used directly for making the samples of Group III, or modified by decanting or levigating to eliminate impurities and also the coarse material. This latter procedure would be required to make them suitable for elaborating fine bodies with elite decoration such as those of Group I. Moreover, the addition of sieved sand would be necessary for manufacturing coarse bodies for larger pieces, such as in the case of samples attributed to Groups Ib and II. The mechanical behaviour of the quartz–calcareous clay system is very sensitive to the proportion of quartz inclusions, which have significant effect in its strength and toughness and other physical properties. The inclusions seem to have an optimum concentration around 20% for vitrified calcareous matrix,^{13,14} which is not very different from the proportion of inclusions in Groups Ib (10–15%) and II (7–12%). Sand or quartz forms part of the raw material used in these Islamic workshops, for making glazes, and the same material was also taken for tempering the clay, which can be seen by the presence of lead-glaze surrounding many quartz inclusions in bodies from Group Ib (Fig. 11)

Moreover, the use of a calcareous clay would have the advantage of getting buff colour in an oxidising atmosphere, and its bodies tend to have higher thermal expansion coefficients than those made with non-calcareous clay, therefore with a reduced risk of glaze ‘crazing’.¹⁵

There is no clear evidence of the addition of lime, either in the excavation or deduced from the analytical results, which is commonly found in other traditional cultures.¹⁶

Another substance used to modify the properties of clays is salt (NaCl). This mineral appears to counteract the tendency of calcite to spoil the surfaces of low-fired pottery^{17–19} and also contributes to the change of the surface colour to what may be a more desirable light or white colour.²⁰ This phenomenon is similar to that which occurs when

firing calcareous clays without chloride, but appears at lower temperature.^{17,21} NaCl accelerates the formation of calcium aluminosilicates susceptible of Fe³⁺ trapping.¹⁰ This manipulation of adding salt, or salt water, to calcareous clays is referred to in ethnographic studies from Pakistan,²² Melanesia²³ and the Middle East.²⁴

In technical literature concerning this phenomenon^{9,25} cites that the addition of salt to the clay can sometimes be determined in petrographic thin sections because the salt reacts with calcareous fragments, leaving a yellow ‘reaction rim’. This effect has been registered in some samples of Group I (see Fig. 7). On the other hand, during the firing of the calcareous bodies, the presence of salt causes the partial loss of alkaline metals such as potassium, in the form of chlorides^{26,27} together with Li and Rb.¹⁰ Such an effect is also evidenced from the comparison of the chemical contents between Group I and the rest of calcareous bodies (cluster 1 in Tables 3 and 4).

This salt was readily available from the sources such as the mines in the Zaragoza area, which were among the most important suppliers of halite during this period, according to medieval al-‘Udrī texts.^{28,29}

4.2 Firing conditions

The effect of the firing temperature on the fabric of clay artefacts is important in order to understand their manufacturing processes and can be used to evaluate the level of sophistication of the crafts. The firing conditions may be estimated from the modifications of both, the mineral composition and the examination of micro-fabric.

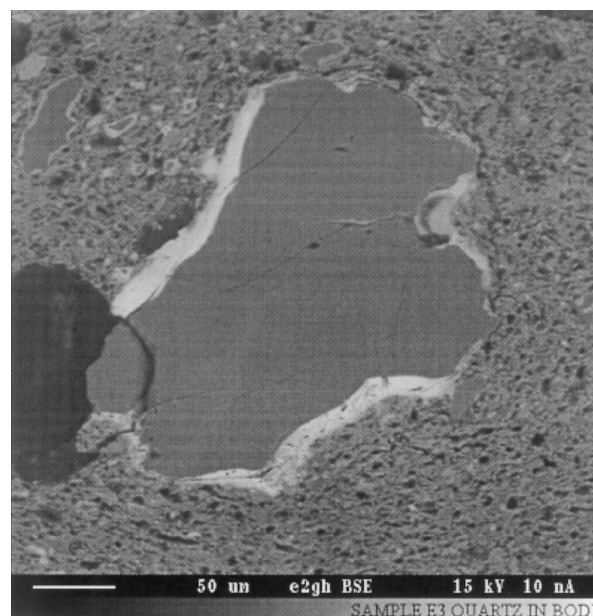


Fig. 11. Backscattered electron microphotograph of a quartz surrounded by lead-glaze in the ceramic body.

The petrographic and mineralogical study showed that there were significant differences in the maximum firing temperature for the different calcareous groups. The manufacture of decorated vessels from Group I would have required high temperature conditions, ranging from 800 to 950°C, as evidenced by the loss of the crystal structure of the clay minerals and also by the decomposition of the calcite and the neoformation of prograde minerals.³⁰ Samples from Groups II and III are of similar composition in their inclusions, differing only in the mineralogical phases found in their fabric which is due to the various firing temperatures reached. In Group II, the presence of calcite and the loss of the illite structure leads to suppose that these ceramics were fired at temperatures ranging around 850°C. The estimated firing temperature for Group III would not have been above 700°C evidenced by the presence of dolomite and also calcite and illite.

The calcareous clays were widely used in the Mediterranean area in antiquity. Pottery produced using calcareous clays possesses distinctive vitrification properties which facilitate firing at significantly low temperatures (extensive vitrification can be achieved at about 850°C) and have a particularly stable microstructure over the temperature range 850–1050°C, and therefore control of the temperature attained is not particularly critical. This is a considerable margin of error in firing and such clays would guarantee a consistently high quality ceramic product. On the contrary, the quality of pottery made with non-calcareous clays can vary quite considerably with changes in firing temperatures of about 50°C, increasing the extension of vitrification progressively.

5 Conclusions

The petrographical–mineralogical analyses completed by chemical data and textural analyses allowed not only the characterisation of these ceramics, but also the formulation of some hypotheses regarding the specific manufacturing techniques of their bodies.

Conclusions obtained regarding the technology of manufacture of the Islamic ceramic workshop located in Zaragoza (Spain) may be summarised as follows.

No mineralogical or chemical differences were found in pieces of extremely different chronology and consequently it can be inferred that no technological changes occurred during at least two centuries. The analytical results indicate that a selection of raw material was used for each production variant. Significant differences in the technology have been detected for each group.

Starting with the clay raw material, it can be said that a ceramic group (Group III) was made directly with a calcareous illitic clay, fired at low temperature and oxidising atmosphere to prepare non-glazed and slip-coated ceramics (for crockery or potter's tools). With a slight addition of temper and fired at medium temperature, another group of non-glazed ceramics and kiln-tools can be separated (Group II). But, if this raw material is refined and then fired at high temperature, the bodies of small glazed ceramics can be obtained (Group Ia). However, the body for a large glazed ceramic requires a refined clay but tempered with coarse sand (Group Ib). Glazed objects were always joined to calcareous bodies, as they offer the greatest adherence with the glaze.

There is petrographical and chemical evidence of the addition of salt to the calcareous clays, particularly in objects with decoration, whose bodies are, moreover, whiter in colour.

Nevertheless, for making non-glazed cooking-pots, the potters worked with a different raw material (non-calcareous) and added natural siliceous temper and grog in order to increase their refractory properties, firing the objects at higher temperature (Group IV).

Finally, in order to establish compositional reference groups for further provenance studies, it is worth mentioning that in this Islamic workshop, different fabrics have been used for producing different types of pottery. As a consequence, there is not only one geochemical group of reference and therefore, only the products of the same typology and function should be compared.

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