Article

Characterization of Islamic Ceramic Production Techniques in Northeast Iberian Peninsula: The Case of Medieval Albarracin (Spain)

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Abstract: Ceramic objects found in the Islamic *Taifa* of Albarracin (Spain), 12th century CE, were studied to ascertain the main characteristics and influences of its manufacture. Production centers even from small kingdoms can add new insights in medieval ceramic technology. Several types of decorated ceramics, such as tin-opacified glazed, monochrome glazed and *cuerda seca*, were investigated. Ceramic bodies were analyzed by ICP-Optical Emission Spectrometry, and glazes were studied by Scanning Electron Microscopy with Energy Dispersive X-ray Spectrometry. All the ceramic bodies showed the use of Ca-rich pastes, although three groups could be distinguished and related to their decoration. Lead and silicon were the main components of the glazes, as well as scattered tin oxide in the case of white or green opacified glazes. Some features, such as calcareous bodies, double firing for tin-opacified glazes, glaze components, and coloring oxides, were common in Albarracin samples and other Islamic production centers in the Iberian Peninsula. However, some differences were also highlighted in lead/silica proportions and *cuerda seca* decoration, and several influences from northern or southern pottery centers. Lead isotope ratios, measured by ICP-Quadrupole Mass Spectrometry, revealed two different sources or suppliers of lead raw materials according to the type of glaze to be prepared.

Keywords: characterization; ceramic; glaze; tin opacified; *cuerda seca*; lead isotopes; SEM; ICP-QMS; Islamic

1. Introduction

Islamic ceramic manufacture in northern al-Andalus (the Muslim part of the Iberian Peninsula) is less known; however, the study of production centers from some small, but splendorous, kingdoms in the Peninsula can improve general knowledge and add new insights into medieval ceramic technology. The Iberian Peninsula was part of the Islamic culture since the 8th century CE, but it was in the 10th century CE when it reached its political independence from the Damascus caliphate. At the beginning of the 11th century CE, al-Andalus was divided in small kingdoms (called *Taifas*), such as Toledo, Cordoba, Granada, or Zaragoza (Figure 1). This was the case of Albarracin *Taifa*. Nowadays, Albarracin is a town located in the NE of the Iberian Peninsula, in a mountain range in the current Aragon. Its good situation in an environment with lots of natural resources (water, vegetation and animals) has favored human habitation since ancient periods, as is shown by the rock art found in the surroundings that dates from the 6th millennium BCE [1]. Albarracin *Taifa* was governed by a Berber family, the Banu-Razin, and it had a flourishing cultural and economic development which reflected the influence of the Muslim society in different aspects of its way of life and also in ceramic production [2,3]. At the beginning of the 12th century CE, the kingdom was under the influence of the Almoravid

political power from Valencia and Murcia. At the end of the 12th century CE, in 1170, Albarracin was given to the Christian Azagra family, but at the same time it preserved its independence from Aragon and Castile during two centuries; this allowed it to continue economic relations not only with northern Iberian regions dominated by Christians but also with Muslim kingdoms [2,3]. All these features reveal a great interest on the study of ceramic productions in Albarracin *Taifa* in order to understand pottery characteristics, technology and actual exchanges between different workshops in the 11–12th centuries CE. Moreover, this area could be the origin and the connection with later important glazed ceramic manufacture centers, such as workshops from Teruel and Valencia.

Figure 1. Map of the Iberian Peninsula, showing the approximate border between Islamic and Christian areas at the beginning of the 11th century CE, with some of the main centers of ceramic production.

During the Islamic period, ceramics was one of the most important industries in the Iberian Peninsula due to great trade development, with the production, exportation and importation of ceramic objects. Pottery reflected the influence of Muslim culture, not only in decorative styles but also in decoration technology. As well as a continuity of the production of lead glazes developed in earlier periods, some new technologies were widespread in the *Taifa* kingdoms. The addition of tin oxide to the glazes to obtain opaque decorations was imported from the Middle East and northern Africa in the 10th century CE, or at the end of the 9th century CE. Initially, tin-opacified glazes decorated with green and brown designs on white surfaces were produced in the caliphal Cordoba [4], but the style was fast spread to all the other later kingdoms [4,5]. Another kind of decoration introduced in this period by Muslims was *cuerda seca*. This technique was an important style in al-Andalus, confirmed by the different productions found around the Iberian Peninsula [6,7]; it is characterized by a black line that delimits the drawing of the decoration, and the spaces between lines were filled totally or partly with colored glazes. The geometric and vegetable motifs were common in the techniques of both white tin-opacified glazes and *cuerda seca*.

The continuous Islamic influence in the ceramic production of the Iberian Peninsula was not only present in decoration technology, such as the introduction of tin-opacified glazes and the *cuerda-seca* technique, but also in the decorative styles. All these characteristics and techniques were applied in ceramic production from Albarracin *Taifa* dating from the 11–12th centuries CE, as the numerous objects found during several archaeological excavations have proved. This production was the subject of our study. The research was focused on the technology of production and decoration of the diverse ceramics found in Albarracin from the *Taifa* period.

The aim of this work was the characterization of the Islamic ceramic production of Albarracin in order to establish ceramic reference groups of this period in the *Taifa* and determinate the possible influences of the pottery manufactured in other Islamic kingdoms. To carry out these objectives, the study was focused on the chemical composition of the materials employed in the clay bodies, and on aspects of the decoration related to the composition of the materials and their contribution to the glaze characteristics and to the manufacture process. Finally, this study intended to establish relations between the variations of the raw materials depending on the type of decoration and compare the compositions used in their manufacture with other workshops of the same period.

2. Materials and Methods

Nearly one hundred ceramic fragments, dating to the 11th century CE from archaeological sites in Albarracin [3], all of them decorated, were chosen to carry out the study, although only the results of glazed pottery are discussed in this paper. Most of them belonged to the tableware group (bowls, beakers, and dishes). According to their decoration, the studied samples can be classified in four groups:

- *Cuerda seca* (Figure 2a): ten fragments were studied, decorated only on the main side of the ceramic;
- White tin-opacified glazes (Figure 2(b1,b2)): twenty-two samples with green and brown-black designs on white glazes (Figure 2(b1)) of the main object side were studied. The secondary sides were coated with yellow glazes (Figure 2(b2));
- Yellow/honey glazes (Figure 2(c1,c2)): almost all the ceramics covered with yellow glazes were coated on both sides with the same color. Sometimes these fragments were decorated with green (Figure 2(c1)) or brown lines (Figure 2(c2)) on the yellow glaze. This type of decoration was very frequent, with a great range of colors among the clay bodies. Twenty-two fragments were chosen for the study;
- Green glazes (Figure 2d): fragments decorated with green glazes included ceramics glazed on both sides, with the same green color in many cases, but some fragments had the main side colored in green and the secondary side of yellow color. Nineteen green glazed samples were selected.
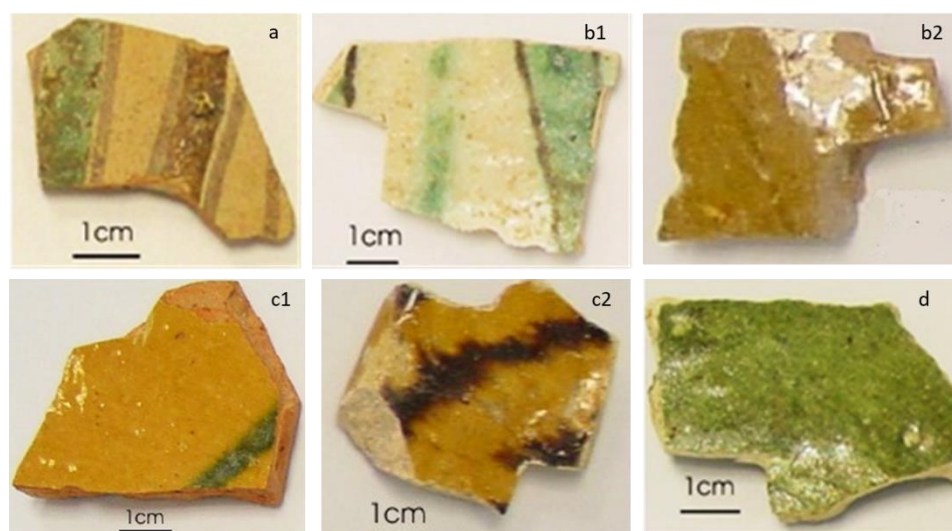

Figure 2. Ceramic fragments with different decorations found in Albarracin: (**a**) *Cuerda seca*; (**b1**) White tin-opacified glaze; (**b2**) Yellow glaze on secondary-side of white-tin opacified ceramic; (**c1**) Yellow/honey glaze with green line; (**c2**) Yellow/honey glaze with brown line;(**d**) Green glaze.

Chemical analysis of major and minor elements of the ceramic bodies was carried out in a Thermo Elemental Iris Intrepid Radial Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES) (Thermo, Bremen, Germany). Powdered samples were extracted

from ceramic bodies by drilling in freshly fractured surfaces of the sherds, using a diamond-tip drill. Samples (50 mg) were digested in open Teflon-vessels. The procedure consisted of successive additions of HNO_3 + HCl, and HF, and a final addition of HNO_3 + $HClO_4$, heating in every step [8]. After dissolution, every sample was diluted in a 50 mL volumetric flask. For ICP-OES analysis, an 1150 W RF power was used. The following components were determined: Na, Mg, Al, K, and Ca as major elements, and Ti, Mn, Fe, Ba, and Sr as minor elements. Results were submitted to a multivariate statistical treatment by hierarchical clustering analysis (using XLSTAT software, version 2021.2.2).

Small fragments of glazed ceramics were cut perpendicularly to the glaze-body interface to prepare polished cross-sections in order to examine them by Optical Microscopy (OM) and Scanning Electron Microscopy (SEM). Cross-sections of samples were observed by means of OM, using a Nikon Eclipse 50ipol (Nikon, Melville, NY, USA). For the determination of glaze composition, a JSM 6360 LV SEM (JEOL, Tokyo, Japan) equipped with an energy-dispersive X-ray analysis (EDS, Inca software, Oxford Instrument, Oxford, UK) with ZAF correction was used, applying an acceleration voltage of 20 kV (oxygen was not measured, oxides were calculated by stoichiometry). The cross-sections were previously carbon coated. X-ray analyses were carried out by scanning large areas of the glaze matrix (normally using ×2000 magnification, although sometimes ×5000 magnification was used where the decoration layer was very thin) and acquiring data from ten different points in every sample.

A Perkin Elmer SCIEX Elan 6000 Inductively Coupled Plasma-Quadrupole Mass Spectrometer (ICP-QMS) (Perkin Elmer, Ontario, Canada), equipped with a cross-flow nebulizer, was used to measure lead isotope ratios in glazes [9]. The measured masses were 204, 206, 207, 208, and 202 to avoid ^{204}Hg interferences.

3. Results and Discussion

3.1. Body Compositions

Chemical compositional data of ceramic bodies obtained by ICP-OES were treated statistically by hierarchical clustering analysis using eight variables (Na, Mg, Al, K, Ca, Ti, Mn, and Fe) and Euclidean distances between samples (Ward method) to group them. This multivariate statistical treatment (Figure 3) revealed several differentiated groups whose compositions are summarized in Table 1 (oxides were calculated by stoichiometry). Calcium was one of the elements whose content varied between groups and, together with other components, allowed the characterization of each group. The groups presented calcareous matrix whose composition ranged from 5 to 18% CaO. Group 3 presented lower average calcium proportion (7.5% CaO) than the other calcareous matrixes, but potassium, magnesium, and aluminum contents were higher. The other two groups with calcareous bodies (group 1 and 2) had similar calcium quantities, but different minor element contents. All studied decorated ceramics were produced with calcareous clay, only a less frequent group of slip-ware decorated with red/brown painted designs (not included in this paper) was made with non-calcareous clay (0.5–1% CaO) [10]. The use of calcareous clays for manufacturing glazed pottery was expected in order to obtain light-colored bodies [11].

The reference groups obtained by cluster analysis, according to the chemical composition of the bodies, could be related to the decoration types manufactured in Albarracín. Groups 1 and 2 corresponded to ceramics decorated with white or green glazes. Most of the *cuerda-seca* fragments were included inside group 1. Group 3 was mainly formed by honey-glazed objects, although some yellow fragments were also included in groups 1 and 2. This fact suggested that yellow-glazed ceramics could be manufactured with different raw materials or in different workshops.
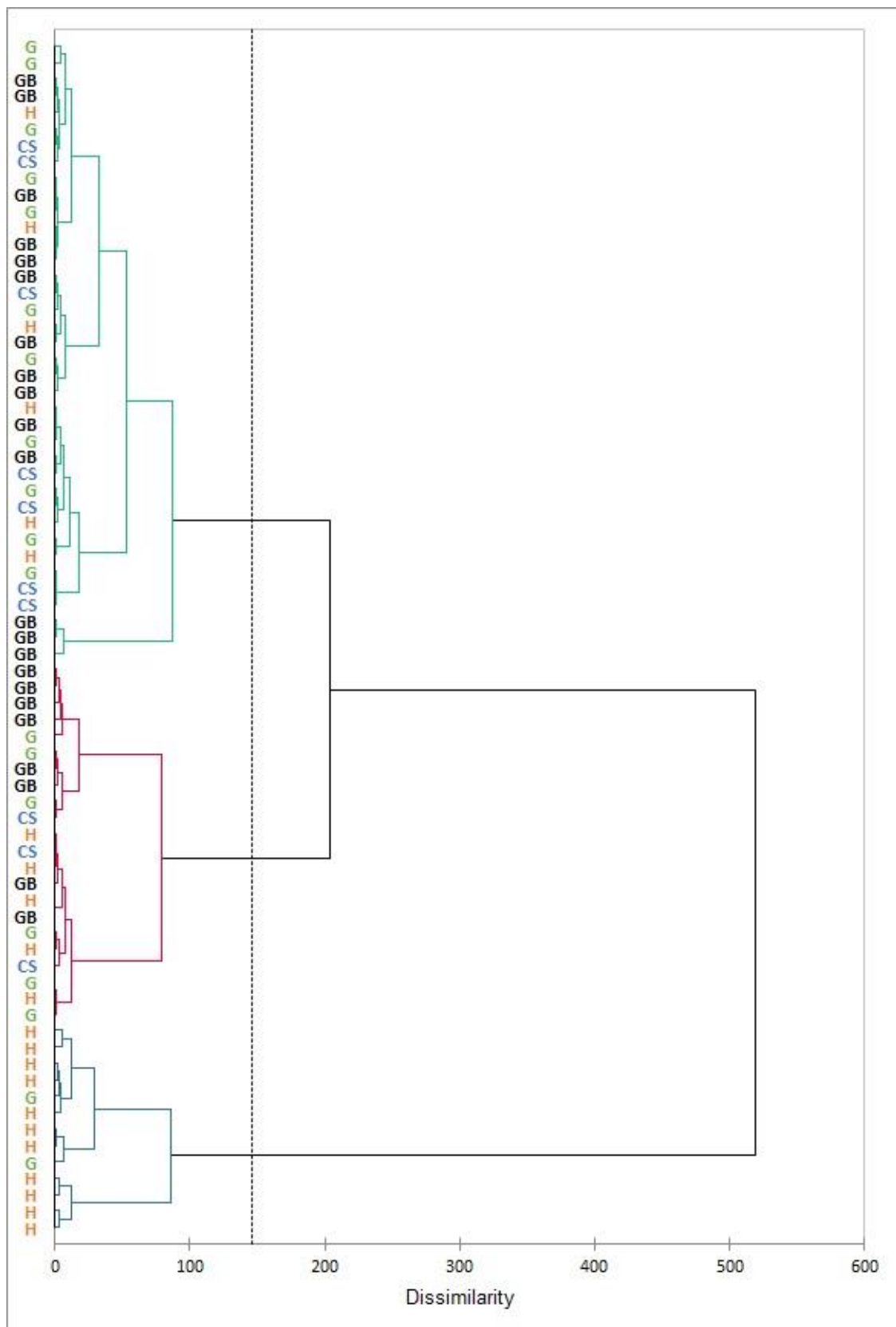
Figure 3. Dendrogram from hierarchical clustering analysis of the body composition data (CS: *cuerda seca*; GB: green and brown decoration on white glaze; G: green glaze; H: yellow/honey glaze).

Table 1. Chemical composition of ceramic bodies, determined by ICP-OES (data as %wt, except Ba and Sr results given as μg.g^{-1}, *s*: standard deviation; *n*: number of samples).

		Na$_2$O	MgO	Al$_2$O$_3$	K$_2$O	CaO	TiO$_2$	MnO	Fe$_2$O$_3$	Ba	Sr
Group 1	*mean*	0.845	2.91	14.8	3.31	12.6	0.587	0.0545	4.78	544	446
	s (n = 38)	0.453	0.94	1.9	0.88	1.4	0.087	0.0136	0.66	152	247
Group 2	*mean*	0.898	3.56	15.6	3.03	16.2	0.636	0.0643	5.65	597	509
	s (n = 22)	0.495	0.71	1.8	0.67	1.4	0.062	0.0150	0.92	105	148
Group 3	*mean*	0.734	5.28	16.20	4.70	7.54	0.593	0.0626	5.34	801	393
	s (n = 13)	0.466	0.97	2.39	0.73	2.25	0.107	0.0116	1.31	232	90

According to chemical results, it seems that selection of the raw materials was performed depending on the decoration. In groups 1 and 2, it can be assumed that the same type of illitic clay was employed for the manufacture of the pottery. However, the raw materials used in the ceramic production of group 3 differed in the chemical composition from those employed for groups 1 and 2. Therefore, ceramics included in this last group can belong to another production workshop or even another area.

Comparing the results obtained from Albarracin ceramics with analysis of clay bodies of Islamic productions from other areas of the Iberian Peninsula [4,12–16], it can be pointed out that all the clays used in decorated ceramics were also Ca-rich (8–22% CaO) and had similar contents of iron (4–6% Fe$_2$O$_3$) and fluxing agents (4–5% Na$_2$O+K$_2$O). The use of calcareous clays for decorated pottery justified the buff colors of the bodies due to the growth of calcium silicates, which incorporated some iron atoms to their structure and decreased the content of iron oxides, responsible for red color [11]. A great similarity was found when we compared these objects from Albarracin with other pottery productions from Muslim centers, such as Cordoba, Denia, Granada, Mallorca, Murcia, Pechina, or Zaragoza [4,13–16]. For instance, the Islamic production from Zaragoza during the 11–12th centuries CE was studied [13] and four reference groups were described, based on the type of body clays employed. In these workshops from Zaragoza, differences among the calcareous groups were also based on the amounts of alkalis and the groups were related to the final decoration and to the utility of the pottery. The selection of the raw materials implied great knowledge of their characteristics and properties.

In the south of al-Andalus, the production of ceramics was very popular with a great number of workshops. Cordoba, Denia, Granada, Murcia, and Pechina were important pottery centers during the 11th–13th centuries CE [4,14,16]. The body composition of these glazed productions from Islamic workshops was very similar to ceramics from Albarracin with high contents of calcium (groups 1 and 2), although ceramics manufactured in some of them were richer in calcium content than Albarracin. The study carried out in the workshop of San Nicolas [14], situated in Murcia and dating to the 10th century CE, revealed some differences with the pastes from Albarracin, but three different references groups can be also distinguished; these three groups were also related to the decoration to be applied. Again, this fact demonstrated the specialization of the potters on the selection of the material.

3.2. White Tin-Opacified Glazes

Different types of decoration on the Islamic pottery from Albarracin *Taifa*: white tin-opacified glazes, colored glazes, and *cuerda seca* were studied for their characterization. In the case of white tin-opacified glazed samples, the compositions of white glazes on the main side and the yellow-honey glazes on the secondary side of these fragments were studied. Further, brown and green decorations on the white glazes were analyzed in most ceramic samples. Tin-opacified glazes presented an average chemical composition that was very homogeneous in the main side, as can be seen in Table 2. The major elements found in the white glazes were silicon (SiO$_2$ 42.8%), lead (PbO 36.2%), and tin (SnO$_2$ 7.0%), mixed with contents below 5% of aluminum, iron, alkalis (potassium and sodium) and calcium oxides. Glazes produced during the medieval period in the Iberian Peninsula were characterized by lead-rich contents. The low percentage of aluminum (Al$_2$O$_3$ < 4.5%)

could be attributed to the use of small amounts of clay to produce the glaze, or also to the diffusion of aluminum from the paste to the glaze during firing [17]. Iron and calcium were not added to the recipes of glazes as main components. Their very low proportions (FeO < 1% and CaO 2.86%) were also due to the migration of iron and calcium into the glazes. Higher contents of iron would induce yellow or brown colors in the glaze. Therefore, raw materials employed in the production of tin-opacified glazes were carefully chosen in order to obtain a white color of glaze.

Table 2. Chemical composition (%wt) of white tin-opacified glazes and their colored decoration by EDS.

Glaze Color		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	MnO	FeO	CuO	SnO_2	PbO
white	*mean*	1.13	0.60	4.20	42.8	3.48	2.86	-	0.76	-	6.95	36.2
	s (n = 18)	0.27	0.18	0.64	2.2	0.77	0.65		0.29		1.72	2.8
green	*mean*	1.13	0.56	4.10	42.5	2.89	3.38	-	0.96	2.04	4.53	37.4
	s (n = 10)	0.21	0.17	0.68	2.6	0.56	1.11		0.31	0.66	2.10	4.0
brown	*mean*	1.10	0.60	3.98	41.1	2.81	3.64	2.83	1.00	1.15	5.06	36.6
	s (n = 18)	0.19	0.21	0.69	2.5	0.70	0.96	1.34	0.41	0.39	1.88	3.9

Green and brown decorations on tin-glazed samples from Albarracin *Taifa* did not differ in their compositions from the white ones, except for the presence of metallic oxides responsible for the colors (Table 2). Analyses carried out on green areas were rich in copper oxide, and brown zones had higher contents of manganese oxide. The content of CuO was in the range of 1.4–3.4% and MnO varied between 1.2 and 6.0%. The presence of copper in brown decorations can be explained due to the thin brown design that made it difficult to determinate only the composition of brown areas without including green zones.

Opacity was accomplished by the presence of small crystals of cassiterite (SnO_2) within the glaze that reflected and scattered the light [18]. These crystals were achieved by the addition of tin oxide to the raw materials of the glazes. During the heating (above 700 °C) of the mixture, the tin oxide was dissolved and began to recrystallize as cassiterite [19]. The amount of tin, the size of tin oxide crystals, their distribution in the glaze, and the thickness of the glaze were factors with great influence on the grade of opacification [12,18,19] and they were related to the process of manufacture. The average amount of tin oxide (7% SnO_2) employed in the studied samples was in good agreement to obtain enough opacity (5–10% SnO_2) without increasing too much the price of the product. The size of tin oxide crystals in Albarracin samples varied from 500 to 800 nm. These dimensions of the crystals indicated that high temperatures (above 800 °C) were reached during the firing process and the cooling occurred fast, as it was demonstrated in the literature [19]. The homogeneous distribution of the cassiterite crystals suggested that the tin glazes were prepared by fritting the raw materials before applying them to the ceramic body [15,20]. Fritting was an extended method used since ancient times to obtain glasses and glazes, which consisted in melting the raw materials (lead, tin, and silica) previously to use them as components of the glazes. Finally, these fired materials were ground to powder before preparing the glaze suspension that was applied on the surface of the body. Besides the homogeneity of the fritted material, frits presented some advantages with respect to raw materials: they reduced the risk of contraction, avoiding cracking the surface, improved the maturation of the glaze, avoided bubbles, and allowed better control of the process [19].

Only one tin-opacified glazed sample had also white glaze on the secondary side; the others had honey glazes on the secondary side, as it has been explained before. The composition of these secondary glazes revealed a higher heterogeneity between specimens (Table 3), but it was possibly to classify them in two different subgroups. The first one included samples with contents of SiO_2 and PbO about 37% and 46% respectively, and the other group was composed by glazes with higher SiO_2 (45%) and lower PbO (37%) proportions. In both cases, the elements added as fluxes were lead and some alkalis with contents around 3–4%. Tin oxide was not present and the proportions of aluminum and iron oxides were increased regarding main tin-opacified glazes, which evidenced the use of

clays and sand not as pure as those used to manufacture white glazes. The honey/yellowish color of the glazes was achieved by iron (>2% FeO) dissolved in the glaze.

Table 3. Chemical composition (%wt) of glazes coating the secondary side of white glazed objects by EDS.

Honey Glazes		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	FeO	PbO
Subgroup 1	*mean*	0.45	0.81	5.11	36.6	2.57	5.20	2.27	46.5
	s (n = 8)	0.10	0.18	0.52	0.8	0.45	0.74	0.33	2.0
Subgroup 2	*mean*	0.58	0.93	5.54	44.5	3.62	4.94	2.30	36.9
	s (n = 9)	0.20	0.43	1.74	5.6	0.80	2.09	0.99	3.6

Microstructure of glazes observed by SEM differed depending on the fragment side (Figure 4). Tin-opacified glazes on the main side of the objects (Figure 4a) had few inclusions, while on the secondary side (transparent glazes) were abundant inclusions with large sizes (often bigger than 60 μm) and well distributed through the glaze (Figure 4b,c). In both cases, the inclusion compositions were the same: potassium feldspars and quartz. Those microstructural differences reflected that the materials for the main decoration were carefully chosen and probably fritted for producing the main glazes [14].
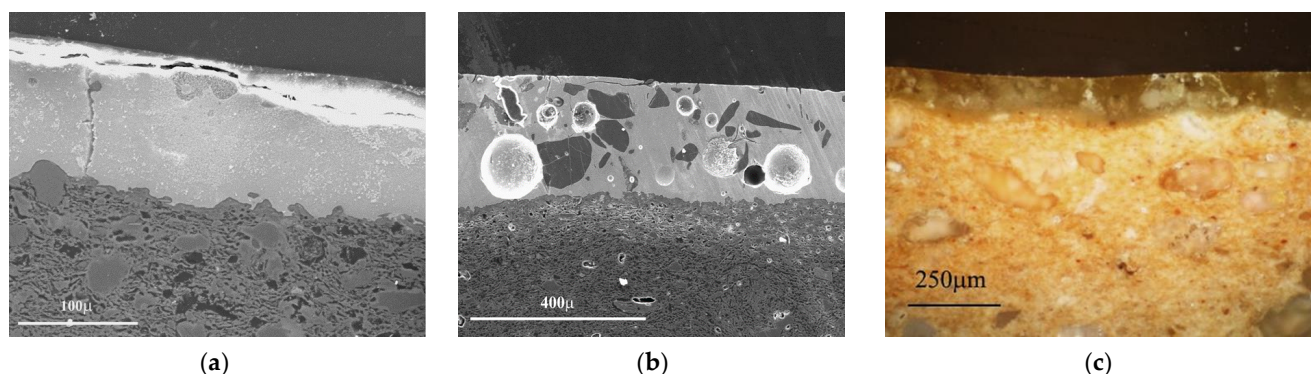

| (**a**) | (**b**) | (**c**) |

Figure 4. Tin-opacified glazed ceramics: (**a**) Secondary electron-SEM image of the main-side glaze with cassiterite crystals scattered in the glaze; (**b**) Secondary electron-SEM image of the secondary side with non-plastic inclusions; (**c**) Optical Microscopy image of the secondary-side glaze.

Concerning thickness, it is outstanding that glazes of the main side were thicker than the coatings applied on the secondary side in all studied samples. On average, the thickness of the layers ranged from 100 to 200 μm for tin-opacified glazes, and from 50 to 100 μm for transparent glazes.

In the contact zone between clay body and glaze, several reactions occurred during firing that favored the migration of atoms between both phases. Diffusion of the elements produced the growth of Pb-rich feldspars crystals. The development of these crystals in the interface and the reaction degree between clay body and glaze were related to the type of clay body and the firing/cooling process [17]. Not only in tin-opacified glazes from Albarracin but also in the transparent ones, the amount of crystals and the interaction between clay paste and glaze (interface thickness < 10 μm) was minimum. It was described that, when the glaze was applied on an already fired clay body, the diffusion between both phases was reduced, and consequently the number of crystals formed and the thickness of the interface were lower [17,21]. This feature indicated that tin-opacified glazes manufactured in the *Taifa* of Albarracin were applied on a previously fired paste (biscuited) and a second firing process was carried out to produce the decoration.

If the results obtained for tin-opacified glazes of Albarracin production are compared with other workshops of the Islamic period in the Iberian Peninsula [4,14], it can be pointed out that the technology (firing process and opacity) seemed to be very similar and the glaze recipes differed only slightly. In all Iberian production centers, white opaque

glazes were composed by a mixture of lead–silica with tin oxide contents from 5 to 10%. Each workshop had a characteristic lead/silica ratio that could help to differentiate the productions. Islamic ceramic centers in the south and the SE of the Peninsula had higher contents of lead (45–60% PbO and 30–42% SiO_2, in general; then, PbO/SiO_2 ratios from 2 to 1.1) than Albarracin (PbO/SiO_2 = 0.8) and Zaragoza samples (in the NE), except for some examples from the Vega of Granada [16]. The low lead contents of these northeastern groups were compensated by higher amounts of alkaline components (2–5% K_2O); this fact allows the definition of these tin-opacified glazes as potassium-lead glazes. These differences were also revealed in the results obtained with the same methodology in our laboratory in samples from Pechina (9th–10th centuries CE), given by the Laboratoire de Céramologie (Lyon, France) (unpublished data). Those characteristics proved that white tin-opacified glazes manufactured in Albarracin and Zaragoza *Taifas* could be differentiated by their chemical composition from other production areas.

3.3. Monochrome Colored Glazes

Colored glazes were used for coating always both ceramic sides. Yellow/honey glazed samples were decorated on both sides of the objects with the same color; however, green glazed fragments were decorated with either green or yellow glazes on the secondary side. Compositional analyses of green and yellow monochrome glazes (Table 4) revealed that they were rich in lead and silicon, with minor quantities of iron, aluminum, calcium, and alkali oxides. Similar chemical compositions were determined on the secondary sides. Only slight compositional differences were observed among green and yellow glazes. Some of these differences were due to elements responsible of the color of glazes. Although the final color of glazes was also influenced by the body color and the metallic oxide added into the glaze [12,14,17], the latter was the responsible of the compositional differences. Therefore, yellow/honey glazes had higher amounts of FeO (1.6–4% in yellow glazes, versus 0.6–1.7% in green glazes) and green coatings were richer in CuO proportions (1.2–4% in green glazes).

Table 4. Chemical composition (%wt) of monochrome glazes by EDS.

Glaze Color		Na$_2$O	MgO	Al$_2$O$_3$	SiO$_2$	K$_2$O	CaO	FeO	CuO	SnO$_2$	PbO
honey	*mean*	0.54	0.94	5.00	37.8	2.10	5.03	2.59	-	-	45.1
	s (n = 19)	0.24	0.29	0.72	1.4	0.46	1.44	0.74			3.7
green	*mean*	0.94	0.74	4.53	40.4	2.84	3.98	1.17	2.56	1.80	41.0
	s (n = 17)	0.36	0.21	0.79	3.5	0.78	1.05	0.35	0.85	1.71	4.5

An important feature of these monochrome glazes was the fact that all green glazes included tin oxide in their composition (Table 4). Tin was detected with EDS analysis and tin-oxide crystals were observed in the green glazes. Although the tin-oxide percentages were lower than white tin-opacified glazes (Table 3), the proportion was enough to give a certain opacity. This characteristic was also highlighted in green glazes from Zaragoza *Taifa* [22].

Inclusions were occasional in the main monochrome glazes (Figure 5), but they were always present in the secondary sides, which implied careful treatment of raw materials again, especially for the main sides. These inclusions were mainly quartz and feldspars. It is not easy to establish an average size of the inclusions because there was a lot of variation even inside the same specimen. The glaze thickness was quite constant in the same sample, but it varied significantly among different ceramic fragments. Additionally, not only the yellow/honey glazes but also the green glazes were thicker in the main side (100–150 μm for the yellow glazes, and 150–200 μm for the green glazes) than in the less important side (80–100 μm in the yellow glazes, and 100–150 μm in the green ones).
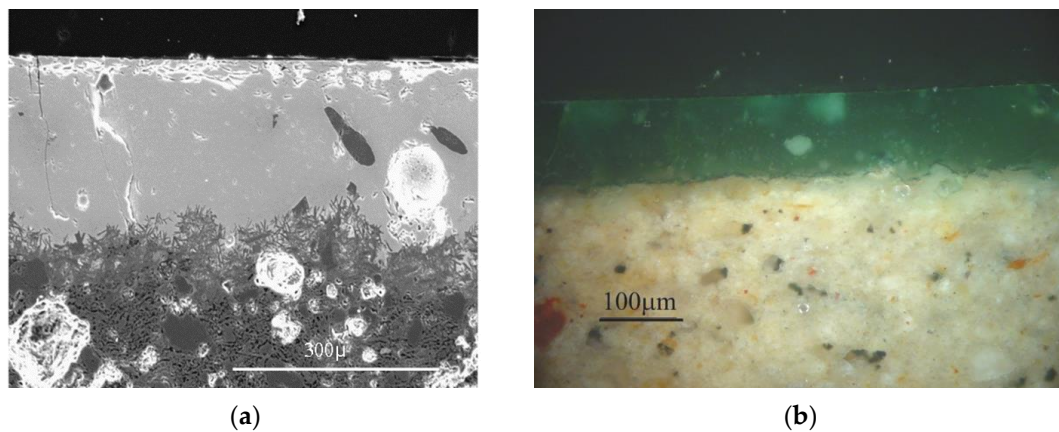
Figure 5. Monochrome colored glazes: (**a**) Secondary electron-SEM image of monochrome coating where a great interface between glaze and ceramic body can be observed; (**b**) Optical Microscopy image of a green glaze.

In these types of glazes, greater interactions between body and glaze were observed by SEM (Figure 5a). Since the reaction between ceramic paste and glaze was related with the type of developed firing process [12,17,21], it can be assumed that the thickness of the interface (35–40 μm) in monochrome glazes corresponded to a single firing process. This technique of manufacture justified the great amount of lead-potassium feldspar crystals that existed in the interface, and that favored the adhesion between both layers [23]. In addition, this type of firing process enhanced the diffusion of certain elements (such as aluminum and iron) from the body to the glaze. Aluminum and iron were incorporated into the glaze and helped the stabilization of PbO-SiO_2 glazes. Therefore, the presence of aluminum and iron in the glazes can be attributed, on the one hand, to the raw materials of the glazes and, on the other hand, to the diffusion of components from the clay to the glaze during the firing process [12,17].

It can be pointed out that the composition of transparent yellow/honey glazes (Table 4) was very similar to that of the glazes employed on the secondary side of white glazes, especially to subgroup 1 (Table 3). Inclusions were abundant in both secondary-side glazes of yellow-honey glazed and white glazed ceramics.

The comparison of chemical compositions of the honey/yellow glazes from Albarracin with productions from other Islamic workshops [4,12,16,22] confirmed differences in lead/silicon ratios. Almost all productions whose workshops were situated in the south and SE part of the Iberian Peninsula had lead oxide content between 45–60% PbO. Lead percentages of glazes produced in Albarracin were among the lowest compared with the rest of the workshops. However, productions from Albarracin and Zaragoza *Taifas* had in common that their ceramics were manufactured with a unique firing process, whereas the rest of the centers of production seemed to prepare monochrome glazes on biscuited ceramics.

3.4. Cuerda-Seca Decoration

Concerning *cuerda-seca* samples, we determined the composition of the black line which divided different areas of the drawing, and the glazes applied inside those areas.

The analyses of the black lines showed inhomogeneous compositions (Table 5) that made it not realistic to establish average proportions. However, the high content of SiO_2 as major element in all the samples is remarkable, mixed with lead, manganese, aluminum, and iron. Manganese was the responsible for the black color of the line, probably in the form of different oxides. Lead could be added to enhance the adherence of the pigments to the paste [24], because the black line was applied directly on the non-fired paste, as can be deduced by the thickness of the interface (Figure 6).

Table 5. Chemical composition (%wt) of the black line, yellow and green glazes in *cuerda-seca* decorated ceramics by EDS.

Glaze Color		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	MnO	FeO	CuO	SnO_2	PbO
black	*interval* (n = 8)	0.4–3.4	0.4–9.0	2.3–19	8.5–58	0.5–13	2.2–26	1.3–50	0.6–21	-	-	0.3–30
green	*mean*	1.00	0.68	3.89	46.0	2.65	3.14	-	0.96	2.29	1.67	37.5
	s (n = 5)	0.22	0.39	0.74	1.7	1.17	0.76		0.38	0.65	1.00	5.1
yellow	*mean*	0.80	1.22	6.26	38.9	2.04	7.11	-	2.70	-	-	37.9
	s (n = 3)	0.14	0.46	1.25	5.4	0.67	2.02		0.03			11.9
turquoise	*mean*	2.47	1.09	1.36	50.6	2.90	4.21	-	0.56	1.86	11.6	23.3
	s [1] *(n = 1)*	0.19	0.15	0.47	2.2	0.23	0.43		0.21	0.16	3.4	1.4

[1] In this case, standard deviation (*s*) corresponds to the number of analyzed points (9) in one sample.
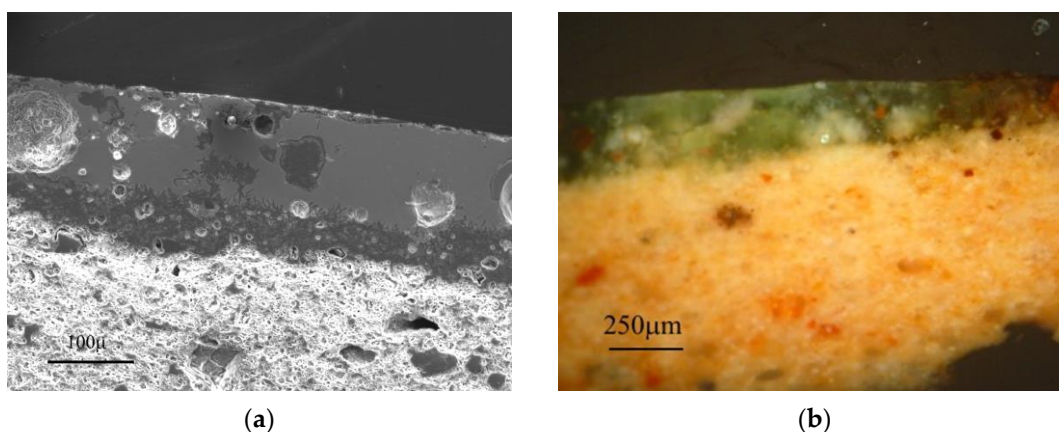

| (**a**) | (**b**) |

Figure 6. *Cuerda-seca* decorated glazes: (**a**) Secondary electron-SEM image of the glaze; (**b**) Optical Microscopy image of *cuerda-seca* decoration.

The glazes used to fill the spaces between black lines were rich in silicon and lead (Table 5). The green glazes had tin oxide in lower proportions than white tin-opacified glazes. Although there were not many *cuerda-seca* samples with yellow and green glazes among the studied samples, and the yellow ones showed great variation in lead content, the average composition of the PbO was similar to the secondary yellow/honey glazes (subgroup 2, Table 3) on green-and-brown decorated objects, and to green monochrome glazes (Table 4). However, the silicon proportion of *cuerda-seca* glazes differed more in both colored glazes.

Once again, the presence of copper and iron oxides with percentages around 2–3% contributed to the final color of the glazes. Only a sample showed *cuerda-seca* decoration with honey and green-turquoise colors. In this case, the color was due to the presence of copper, but its green hue was shifted to turquoise because the glaze had alkaline-lead composition with a significant contribution of sodium (2.5% Na_2O) and potassium (2.9% K_2O) and lower lead content (23.3% PbO). These differences in green hues were already highlighted in *cuerda-seca* ceramics of the 12th century CE produced in the Iberian Peninsula [25].

Studies of pottery with *cuerda-seca* glazes manufactured in the Iberian Peninsula during the 10th–12th centuries CE revealed the use of silicon and lead as main components [6,24,25]. The amounts of lead decreased from the 10th century (43–55% PbO) to 12th century CE (34–43% PbO), mainly in productions from the south of al-Andalus [25]. Nevertheless, samples manufactured during the 11th century CE in the NE workshop of Zaragoza were characterized by high lead proportions [24]. Contrary to other types of decoration, pottery decorated with *cuerda seca* produced in Albarracin in the 11th-12th centuries CE was more similar to the recipes employed in the south of the Peninsula with lead contents around 35–38% PbO.

Examinations by SEM of polished sections of *cuerda-seca* glazes confirmed the presence of abundant inclusions scattered homogeneously in the glaze (Figure 6). The inclusion sizes varied from 50 to 70 µm and their composition was mainly quartz. The use of such an amount of inclusions and their size, plus the tin-oxide content, could be responsible for the opacity. Opacification can be reached either with undissolved inclusions into the glazes or with tin-oxide crystals scattered in the glaze, and even with both of them [26,27]. *Cuerda-seca* ceramics manufactured in the same period in al-Andalus employed both techniques [6,24,25], although only tin oxide was employed as an opacifier in later centuries. Therefore, the inclusions found in the glazes played an important role in the opacity of the glazes. The thickness of the glazes reached values higher than 200 µm and the thickness of the clay-glaze interfaces was also higher (>45 µm), which implied that a single firing technique was carried out during the pottery's manufacture. The type of firing process (single or double) depended on the workshops; both techniques were used without a clear tendency. Single firing process was more frequent, especially in workshops such as Almeria and Zaragoza [24,25]. This characteristic was also followed in the studied production from Albarracin *Taifa*.

As it has been explained before, the technology of opacification also underwent an evolution from a double method (tin oxide and inclusions) employed in the 10th century CE to the opacity technique due exclusively to tin oxide used in the 12th century CE. However, in our case, the double methodology was followed, as in the case of the southern workshops.

It seems clear that the *cuerda-seca* technique underwent an evolution during the Islamic period with variations in recipes, firing processes, and methods of opacification. The evolution did not follow a regular extension to all the production centers, but we can highlight that Albarracin was influenced by the southern centers of al-Andalus. Although archaeological studies based on the decoration motifs of *cuerda seca* during the period of *Taifas* emphasized that the same type of vegetal designs was also employed in Zaragoza [6], both influences could be possible taking in mind the good relationships with northern and southern *Taifas*, and the wide commercial distribution of Islamic glazed ceramics [28,29].

3.5. Lead and Raw Materials for Glazes

Lead isotopes ratios were measured by ICP-QMS in a selection of representative glazed samples, because of the important information in lead sources that could be provided. Natural lead consists of four isotopes: three of them (^{208}Pb, ^{207}Pb and ^{206}Pb) come from the radioactive decay of U and Th, but the fourth one (^{204}Pb) has a non-radiogenic origin and natural lead changes its isotopic composition. The results of twenty-three samples from Albarracin, plus two samples from Islamic workshops of Zaragoza and one sample from Pechina are shown in Figure 7.

The lead-isotope ratios of the samples, plotted as ^{208}Pb/^{206}Pb vs. ^{207}Pb/^{206}Pb and ^{206}Pb/^{204}Pb vs. ^{207}Pb/^{206}Pb ratios (Figure 7), offered the possibility of establishing several groups and distinguishing different features of the used lead for the production of glazes. Samples from Albarracin can be clearly separated into two groups. One of the groups was related to the fragments with yellow/honey glazes (Figure 7a,b, on the right of both plots). The second group was formed by white tin-opacified and green glazes from Albarracin (Figure 7a,b, on the left of both plots). Moreover, these last two types of glazes showed slight differences between them, but both had in common being tin-opacified glazes. If we observed the results of the three samples from other areas (Zaragoza and Pechina), the white tin-opacified glaze from Pechina was grouped with the tin-opacified samples from Albarracin. However, the glazes from Zaragoza indicated differences in their source of lead, because both samples were closer to the honey glazes from Albarracin, and even the white tin-opacified glaze from Zaragoza (marked with a red circle in Figure 7a,b) was unexpectedly similar to the transparent honey glazes from Albarracin.
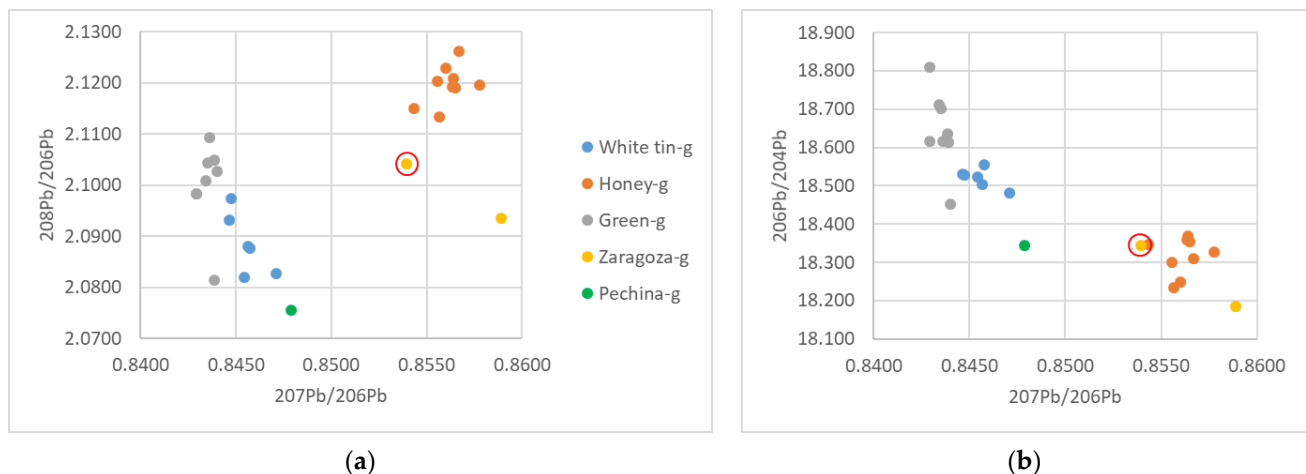
Figure 7. Lead-isotope ratios of glazes from different ceramics (white tin-opacified glazes, honey glazes, and green glazes from Albarracin; glazes from Zaragoza workshops (white tin-opacified glaze is marked with a red circle); and glaze in a sample from Pechina): (**a**) $^{208}Pb/^{206}Pb$ ratio vs. $^{207}Pb/^{206}Pb$ ratio; (**b**) $^{206}Pb/^{204}Pb$ ratio vs. $^{207}Pb/^{206}Pb$ ratio.

Therefore, all of these glazed ceramics, manufactured in Islamic Albarracin, had lead isotope abundances very well characterized in two groups related to the type of glaze (tin-opacified or transparent) applied on the ceramics. This suggested diverse lead sources for the glaze raw materials depending on the type and quality of glaze; then, it could be related to the use of a type of commercial lead for producing tin-opacified glazes and another different lead product for preparing transparent glazes. The first one could be linked to obtaining a best-quality glaze or to the preparation (fritting) of tin-lead glazes. In later periods (16th century CE), differences in the commercial lead supplied to potters were documented: it was explained that the lead used to produce glazed cooking pots was cheaper than the lead acquired to prepare tin-opacified glazes [30].

It is important to underline that this possible source or supplier of lead for tin-opacified glazes from Albarracin was quite similar to that for the white glaze from Pechina; as such it could perhaps come from one of the lead sources in the south/southeast of the Iberia Peninsula. However, lead used in Zaragoza workshops seemed to be different, or closer to that introduced in transparent glazes; slight differences could be highlighted between these two samples (honey and white) (Figure 7a,b), but there are not enough results to find significant dissimilarities.

4. Conclusions

The results of this study allowed the characterization of the pottery production from Albarracin *Taifa* in the 11–12th centuries CE, and establishing differences and similarities with Islamic productions manufactured in other *Taifa* kingdoms of the Iberian Peninsula during the same period. The analyses carried out on the ceramic bodies confirmed that the selection of raw materials depended on the decoration to be applied afterwards. All the samples had a Ca-rich body in order to obtain buff colors and to avoid cracking during firing process.

Firing process depended on the kind of decoration. Tin-opacified glazes were applied on pre-fired clay bodies, but *cuerda-seca* and transparent yellow/honey glazes were applied on the unfired clay body. Once again, this differentiation between the two techniques implies a specialization of the potters.

Concerning to the decoration, it can be pointed out some conclusions about Albarracin production. Glazes manufactured in Albarracin *Taifa* during 11th century CE were rich in lead and silicon oxides. Their composition ranged from 32% to 48% PbO and from 36 to 49% SiO_2, but differences could be established between white tin-opacified and colored glazes. Secondary-side glazes had similar compositions, although the number of inclusions and the size of them were in both cases bigger. However, the thickness of the coatings was

thinner on the less important side. Those features corroborated the idea of a specialization and knowledge of the techniques of production.

As well as silicon and lead as main components, tin-opacified glazes included cassiterite crystals that were responsible for opacity. The buffer color of the bodies required only ~7% SnO_2. Tin-oxide crystals were well distributed inside the glazes, and the size of them (<800 nm) was an important factor to achieve the opacification. Green monochrome glazes were always opacified with tin oxide. However, opacity in *cuerda-seca* samples was reached by the combination of tin oxide plus undissolved grains of quartz. This technique of opacification was also used in the south of Iberian Peninsula in the 10th century CE.

Copper, iron, and manganese oxides were added to the glazes to obtain, respectively, green, yellow, and black glazes. Manganese oxide was employed to draw the black lines that described the designs of *cuerda-seca* specimens.

Comparing the lead/silicon ratios used in the manufacture of ceramics with transparent and tin-opacified glazes in Albarracin *Taifa* with other production centers of the Iberian Peninsula during the Islamic period, we can conclude that PbO/SiO_2 ratios were lower in the northeastern workshops than in the southern ones. However, the potassium content was higher in the northeastern centers.

In all the studied ceramic workshops of the Iberian Peninsula, tin-opacified glazes were applied on a biscuited body, as in Albarracin *Taifa*. However, there existed differences in transparent glazes, because the southern workshops seemed to use a double firing for producing this type of glaze. In the case of *cuerda-seca* decoration, the production of Albarracin had more similarities in the method of opacity and in the lead/silicon ratios with southern workshops; however, for the firing method, Albarracin samples followed a single firing process, as with the Zaragoza and Almeria centers.

According to these results, it can be highlighted that Islamic ceramics with tin-opacified and transparent glazed decorations manufactured in the *Taifa* of Albarracin during the 11th century CE were influenced by the production of the north *Taifa* of Zaragoza. Nevertheless, ceramic decorated with the *cuerda-seca* technique was influenced in some aspects by southern workshops, and in other ones by centers of production from Zaragoza or Almeria.

These possible influences and the Islamic potters' knowledge and specialization were also revealed by the lead-isotope ratio results, where two different lead sources or suppliers were shown: one to prepare tin-opacified glazes, and another one for yellow/honey glazes.

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