Identification of microstructure and chemical composition of Celadon wares from the Harireh ancient cityof Kish Island, Iran

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ABSTRACT

The Celadon samples of the ancient port of Harireh in Kish Island from 11th-14th centuries were studied. The crystallite and chemical compositions of samples were investigated using quantitative X-ray diffraction (QXRD), X-ray fluorescence (XRF), and simultaneous thermos-analytic (STA). Moreover, thin section petrography and environmental scanning electron microscopy (ESEM-EDX) were applied to explore microstructural characteristics. The bulk chemical analysis of the investigated samples shows not only the high amount of K_2O and CaO as flux but also SiO_2 and Al_2O_3 as the major oxides. According to the STA and XRD results, the firing temperature of the samples is similar to the Chinese ones. The presence of mullite and moganite phases indicates a firing temperature up to 1050 °C for the samples. The formation of mullite derived from kaolinite clay and is more similar to the Chinese productions rather than the Islamic. The colorant element in Celadon samples is mainly Fe^{+2}. Chemical compositions of these Celadons and Chines wares also show strong similarities in raw materials and manufacturing processes.

Keywords: Celadon; Archaeometry; Composition; Microstructure; Kish Island; Harireh-Ancient city.

INTRODUCTION

Stonewares with Celadon glazes are firstly made by Chinese during the early Bronze Age in Shang Dynasty (Feng et al., 2008; Zhu et al., 2011; Tite et al., 2012). Chinese ancient Celadon began to be fired early in the Late Eastern Han Dynasty (AD 25-220) in South China (Li et al., 2012). Firing temperature of Celadon wares has been estimated around 1200 ºC (Zhu et al., 2011; Hao et al., 2013; Wood and Li, 2015). The color of Celadon develops through the formation of glassy and crystalline phases, generated by chemical reactions between the glaze and the clay-based body at high temperature under reduction atmosphere (Kim et al., 2011). The similarities and differences between the chemical compositions and microstructures of Celadon wares from China,

South Korea, Vietnam, and Japan have been previously analyzed using various analytical methods (Colomban et al., 2003; Kerr et al., 2004; Hidaka et al., 2011; Hao et al., 2013). The investigations on Chinese ceramics (Shanglinhu- in the late Tang Dynasty) have shed light on the provenance and manufacturing techniques of the body as well as the glaze (Wood et al., 2005). However, Vietnamese ceramics show enrichment of Ca (+K), which lead to better understanding the firing conditions. The presence of calc rich inclusions in a ceramic body chiefly provide the estimation about the firing temperature, due to their grain size since calcium carbonate decomposed very rapidly to calcium oxide (ca. 750°-850 °C) to form porous calcined calcium oxide (Tite, 1969). The grain size of carbonate has a high impact on the extent structural

disturbance in the ceramic body, at the temperature in which the carbonate being decomposed during the firing processes (Leicht, 1977).

Mineralogical and chemical investigations on the raw materials from the south and north China clay source have been essentially carried out based on the characterization of Mullite's habit and SiO_2/Al_2O_3 ratios in the body of the ceramics (Tite et al., 2012). The major chemical compositions of the body of Chines ware are characterized by SiO_2 (80.7-65.4 w%), Al_2O_3 (32.7-16.0 w%), and K_2O (6.1-1.1 w%) (Tite et al., 2012). The investigation of Li et al., (2012) indicates the high contents of K_2O, CaO, TiO_2 and Fe_2O_3 in Chines Celadon wares. Therefore, many systems are suggested, to provide a valid comparison of the chemical compositions such as SiO_2-Al_2O_3-($CaO+K_2O+Na_2O$) and CaO-K_2O-Na_2O (Colomban et al., 2003; Colomban et al., 2004). Chinese Celadons with very smooth green-grey surface reached this coastal line of Iran in around 1100-1500 AD (Pierson, 2012).

The production of Celadon reached its peak in the 14th century when they were exported widely from East coast of China to the Middle East through the Indian Ocean (Hobson, 1915; Cho, 2008). These kinds of ceramics had a very large distance of migration between the China Sea to the Persian Gulf through an extensive maritime trade. South Persian coastline has been a very important region for many centuries based on marine export/import of goods (McConkey and McErlean, 2007). Celadon wares have been reported in many provinces in Iran such as Kerman, Shiraz, Port of Siraf, Kish and Gheshm Island in the Persian Gulf (Pierson, 2012). This paper presents the chemical composition and firing temperatures of 7 pieces of Kish Celadon wares which were produced during the 11th-14th centuries to determine the raw materials and manufacturing process. The Celadon wares were sampled from the Port of Harireh in Kish Island, in the Persian Gulf (Figure 1). It also comparatively analyzes the composition and microstructure of the Celadons from China. The identification of the compositions and microstructures would lead the results to get suggestions about the origin of these Celadon wares (Colomban et al., 2003).

ARCHAEOLOGICAL SETTING

Kish is a ca. 95-square-kilometre Island in the Persian Gulf which is a part of the Hormozgān Province in the south of Iran (Figure 2). In the Middle Ages, Kish became an important trading center under its capital; Port of Harireh. The archaeological research of Whitehouse (1976), in Harireh port also provided a great collection of imported ceramics, such as Martaban stoneware, Celadon, porcelain and Ting ware (Whitehouse, 1976).

Figure 1. The location of Kish Island in the Persian Gulf.

MATERIAL AND METHODS

Seven Celadon wares were collected from the excavation at the ancient port of Harireh and subjected to mineralogical and chemical analyses. The characteristic shape and surface of these samples show macroscopically a very compact and homogeneous body with greenish-gray glaze on the external and the internal surface (Figure 3).

All analytical method carried out at the department of building material chemistry, University Siegen in Germany. XRF analysis has been done with an instrument Bruker® SPECTRA[plus] 2008, in order to have the bulk chemical composition of Celadons' body. Rh-anode end-window X-ray tube is used for analytical conditions. Long count times were used at both peak and background positions. It is essential that the whole sample has the same grain size; therefore the sample should be sieved through a 125 μm mesh. Because altered minerals within the ancient ceramic's matrices, they have a different hardness. In this case, 8g powder samples and 1ml PVP (polyvinyl alcohol) solution mixed together for making the pellet.

Quantitative XRD analysis was used for determining the crystalline phase occurrences in the body of ceramics by a PANanalytical instrument (analyses were carried out using an X-ray generator operated with a copper target at 45 kV and 40 mA. The powder patterns of ceramics were measured between 2θ=5° and 2θ=60°, with a scanning rate of 2θ=0.01° per minute. This large scanning area was required in order to gain a complete diffraction pattern

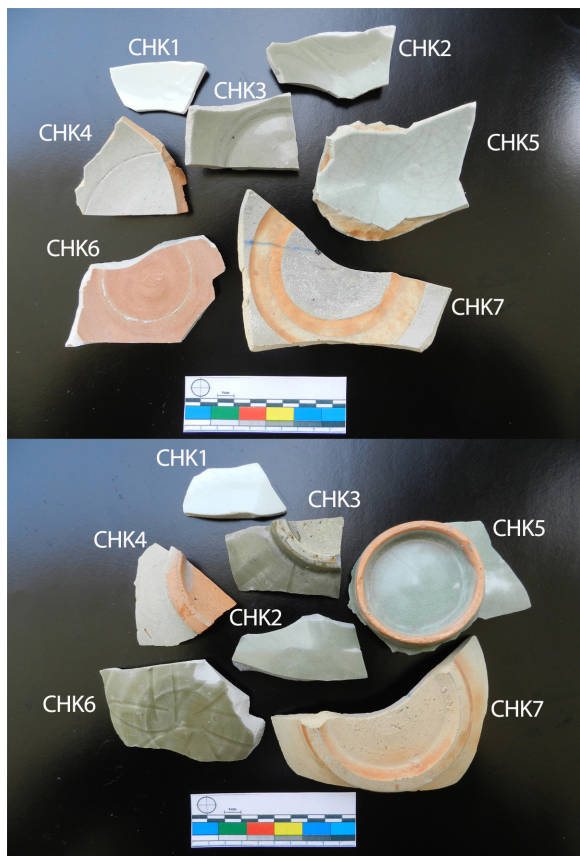Figure 2. Archaeological excavation at Port of Harireh in Kish Island.

Figure 3. Investigated Celadons from external and internal surfaces.

for minerals with low symmetry (Emami et al., 2016). The measured peaks refined with the Rietveld method for getting the best-preferred orientation of the phases. Clustering of analysis was carried out for distinguishing the ceramics according to the crystalline phase constituents (Emami and Trettin, 2010; Emami, 2012). Microscopic investigations are performed on thin sections. These samples were also examined by a polarizing microscope (an instrument from Olympus, XB 51 with Analysis Software) and high-resolution ESEM (ESEM Quanta FEG 250 FEI with Apollo XL30 EDX) fitted with an energy-dispersive X-ray (EDX) spectrometer. Finally, simultaneous thermal analysis (STA) was carried out with using a NETZSCH STA 449C instrument with a Jupiter & Proteus Analysis Software Version 4.8 in order to determine the firing process and crystalline phase decompositions in Celadons.

RESULTS
Bulk chemical composition of the body and Celadon glaze

Bulk chemical compositions of the body of the samples investigated samples are presented in Table 1. The Celadons are classified as high silica ware after Noll classification in the system SiO_2-Al_2O_3-CaO+MgO (Noll, 1991). SiO_2 and Al_2O_3 are mentioned as the major compositions in the investigated samples and show a negative correlation to each other which is probably associated to the clay source. CaO and K_2O are the major

Table 1. Chemical composition of oxides in the investigated Celadon shards.

Sample	SiO$_2$%	Al$_2$O$_3$%	Fe$_2$O$_3$%	CaO%	Na$_2$O%	K$_2$O%	MgO%	TiO$_2$%	MnO%	P$_2$O$_5$%	S%	LOI%
CHK1	66.96	21.61	1.96	3.89	0.87	4.86	0.19	0.140	0.057	0.014	0.015	1.79
CHK2	72.53	17.79	2.00	3.24	0.16	5.02	0.45	0.212	0.092	0.045	0.008	0.07
CHK3	74.26	15.86	1.45	4.05	0.27	4.12	0.20	0.103	0.113	0.025	0.026	2.21
CHK4	70.63	20.32	2.27	3.39	0.03	2.69	0.17	0.147	0.054	0.011	0.011	1.50
CHK5	66.35	20.71	2.01	2.41	0.29	3.35	0.24	0.261	0.033	0.002	0.023	6.04
CHK6	71.45	19.96	1.77	3.51	0.29	5.42	0.17	0.106	0.088	0.002	0.008	0.03
CHK7	74.21	17.06	1.45	4.40	0.43	2.92	0.39	0.242	0.053	0.043	0.019	1.59

oxides occurred within the body and discussed with two essentially points; firstly the origin of the clay or burial conditions and secondly the manufacturing process. CaO is a major oxide which varied between 2.41-4.05 w%. K$_2$O varied also from 2.69-5.42 w% which would be described as secondary contamination via alteration of potash bearing minerals. K bearing feldspar is also used as a fluxing material to form the shiny character to the glaze (Kim et al., 2011). However, due to the high fluxing effect of K$_2$O, even though via increasing the K$_2$O, decrease the shiny effect of the glaze and the glaze surface became matt. The variation of MgO usually associated with CaO due to the usage of dolomitic additives in the body (Daghmehchi et al., 2016). The amounts of FeO devise from different raw clayey materials and are furthermore responsible for the color of the glaze via a reduction atmosphere.

According to the chemical analysis, the amount of Iron is mostly constant which would be a reason for constant green-blue color for the glazes (Kim et al., 2011; Jeon et al., 2014). The chemical composition of the glazes is measured in some areas by SEM-EDX illustrated in Figure. 4. In this case, the glazes have definite chemistry with a major amount of SiO$_2$ and Al$_2$O$_3$ (zone A and B in both samples) and the most varieties are shown by the amount of K$_2$O and sometimes CaO. The glazes are classified as

high Potash glaze. They are mostly homogeneous and the bubbles which are visible within the sample CHK1 proved the high-temperature manufacturing process.

X ray Diffraction of the samples for determining the crystalline phases

Figure 5 shows the X-ray diffraction patterns refined via Rietveld methods of the investigated samples from Harireh (Table 2). The diffraction patterns are not largely different and the crystalline phase occurrences have not essentially changed in compared to each other. In the measured diffraction pattern, "Qz" is denoted for the reflection of α-quartz (quartz low) with its best intensity peaks in diffractogram at 2Θ=26.32 on (011) plane. The quartz peaks are in similar positions in the diffraction pattern and illustrated similar intensity. The XRD patterns illustrate the best crystallographic preferred orientation of quartz at 2Θ=21.12 on (100) and 26.32 on (011) crystallographic plane. The appearance of mullite as mineral composition of SiO$_2$+Al$_2$O$_3$ is also mentioned in this ceramics as it reported within the texture of Izumiyama ceramics (Hidaka et al., 2011). Mullite (in general Al$_6$Si$_2$O$_{13}$) have been used and reported in a very varied range of applications in high-temperature ceramic-based materials as well as technical ceramics (crucibles).
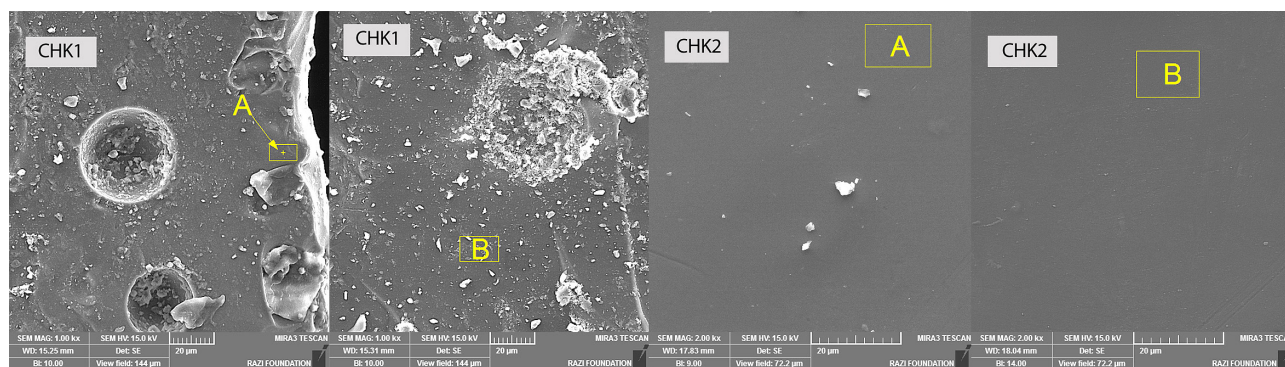

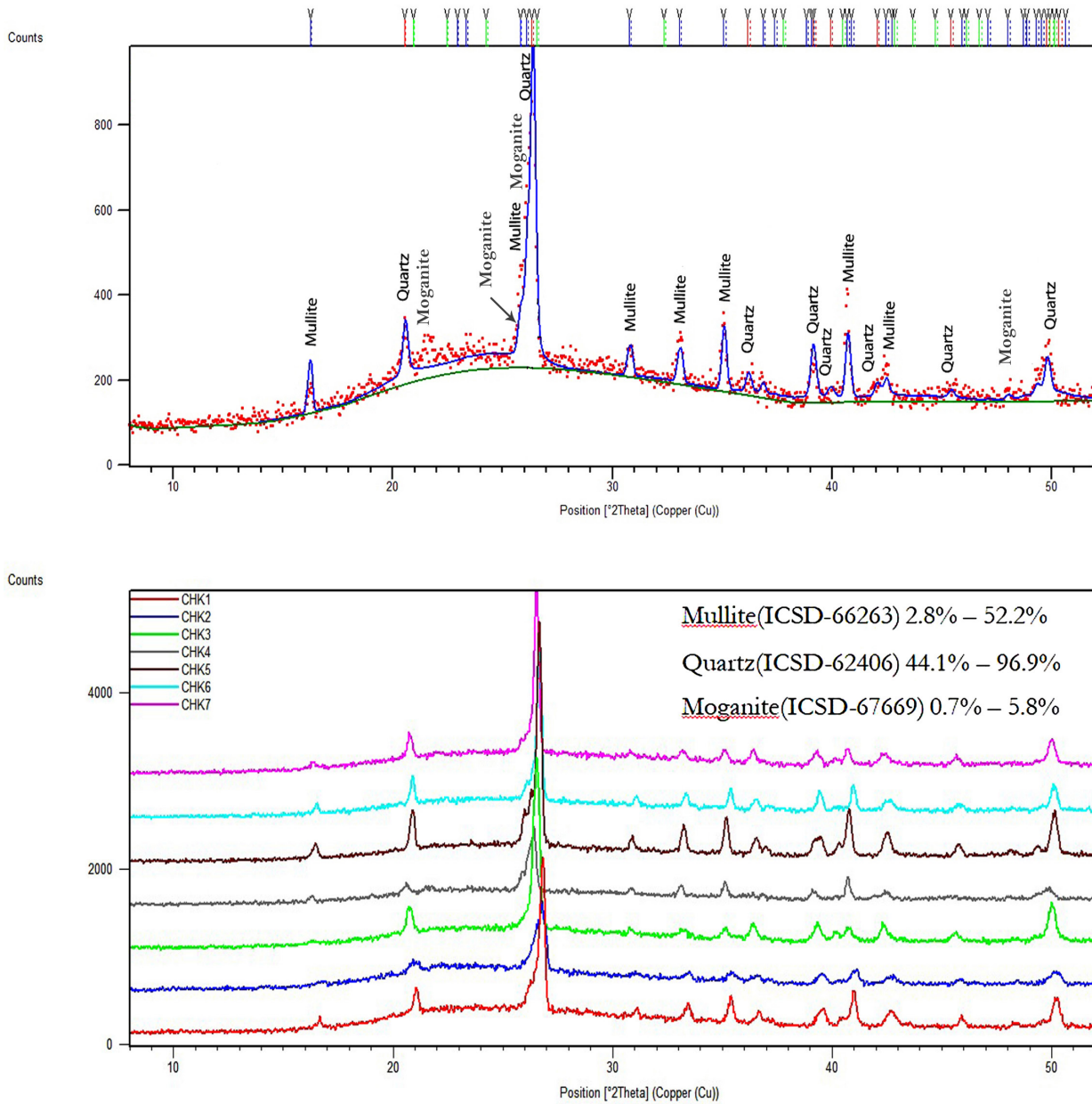Figure 4. Investigated areas on the glaze using ESEM for samples CHK1 and CHK2.

Figure 5. Diffraction patterns of all measured Celadons and crystalline phase composition due to their best preferred orientation.

Table 2. Quantitative Rietveld refining methods of the investigated Celadons.

	Quartz low-ICSD 62406 in %	Mullite-ICSD 66263 in %	Moganite-ICSD 67669 in %
CHK1	47.2	52.9	4.2
CHK2	91.4	2.8	5.8
CHK3	96.9	2.4	0.7
CHK4	90.0	4.9	4.2
CHK5	44.1	53.1	2.8
CHK6	51.2	47.2	1.6
CHK7	87.9	9.4	2.7

Mullite is dominated with its low thermal expansion, high temperatures formation, and its outstanding stability in aggressive chemical environments (Martinon-Torres et al., 2006).

The high intensity of mullite has been detected at $2\Theta=26.07$ on (210) plane and $2\Theta=25.79$ on (120) plane and the quality of mullite is increased by increasing the temperature (1000°-1200 °C) (Ntah et al., 2017). The other phase is Moganite which is appeared on $2\Theta=26.70$ on ($\bar{1}12$) crystallographic plane and $2\Theta=26.27$ on ($\bar{2}02$) crystallographic plane. This phase is a quartz polymorph structure with optically length-slow character. Moganite has a monoclinic symmetry with space group I2/a (Götze et al., 1998). In high-temperature ceramics, they could be found and contain varying percent ratios of moganite to quartz depending upon their source (Pretola, 2001).

Microscopic investigations of the Celadon bodies and glazes

Thin sections of Celadons were observed under a polarizing microscope in order to identify the fabrication as well as the types of additives in the matrix. The body of the wares has a fine-grained fabric (Figure 6A). The porosity of the Celadons is too low due to the firing process. Quartz aggregates are the common additives within the ceramic matrices. The appearance of quartz as additives in the antiquity is mostly highlighted due to its durability effect on the clayey matrix (Kilikoglou et al., 1998). As a matter of fact, to consider the quartz as a justified temper in ceramic ecology two points must be considered, accordingly. Quartz is naturally altered non-plastic materials have been accumulated in many sedimentation basins (raw material availability) and best functionally experiences to work with this material not accidentally, but indeed experimentally (Giurgiu et al., 2017).

Quartz aggregates are identified by their low birefringence and typical undulatory extinction effect. The grain size of quartz aggregates is estimated around 0.1-1 mm. Compact matrix with secondary calcite is observed due to the burial condition after the manufacturing process (Emami and Trettin, 2012) (Figure 6B). The CaO content
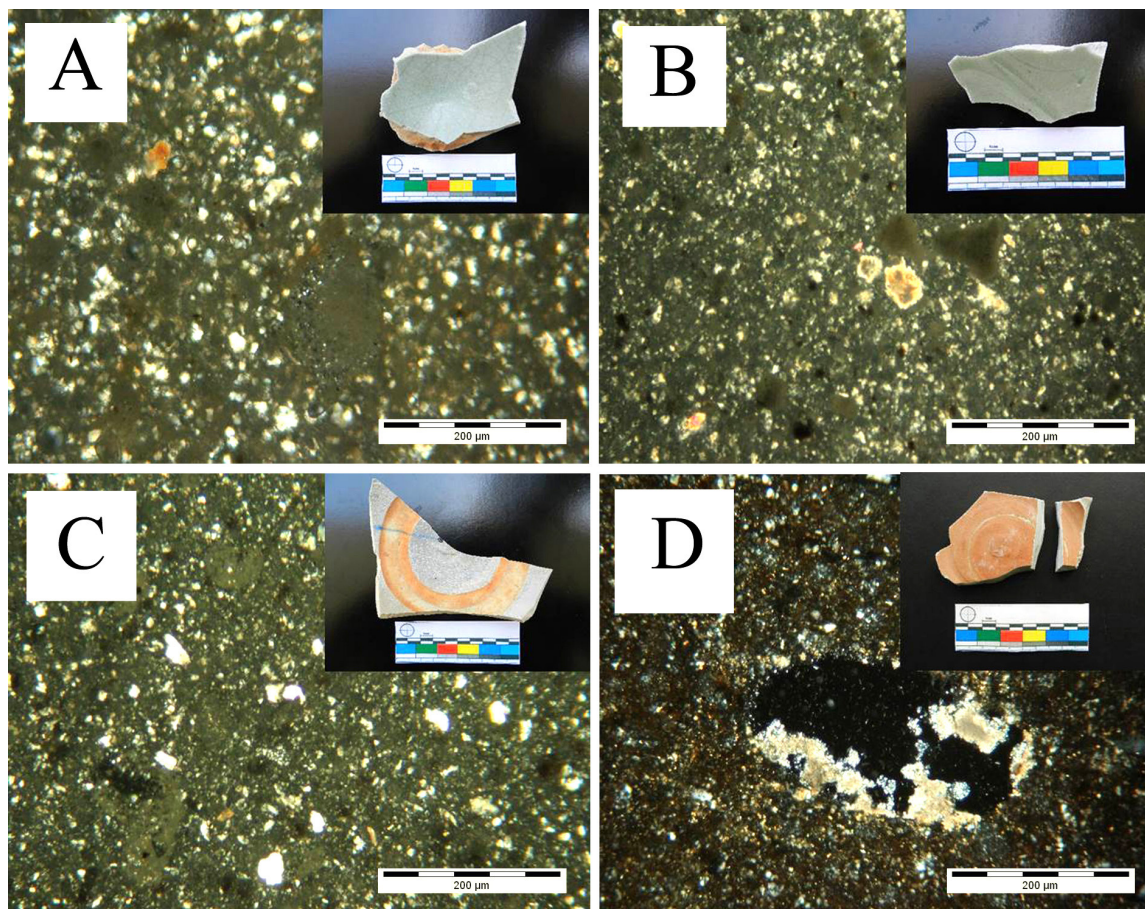

Figure 6. A) The body of the CHK1 is classified as a clayey matrix with a very compact fabric. B) Dense matrix with fine secondary calcite. C) Quartz grain with its typical undulatory extinction structure. D) Recrystallization of calcite as secondary formation around the pores.

is observed in a very fine aggregate. Quartz grain with its typical undulatory extinction structure is being visible only with great magnification and is most difficult to identify with magnification by 40x or 100x (Figure 6C). In sample CHK7 the recrystallization of calcite as new formed secondary calcite around the pores is noticeable (Figure 6D). Mullite needles exist within the fabric in all samples with low birefringence and colorless but only can be detected in high magnification (unsharp crystals under a microscope).

Thermal analysis of the Celadon samples

Firing temperature of 7 ceramics has been estimated by STA (simultaneous thermo-analysis). The mineralogical decomposition and crystallization occur in four important parts; dehydration, dihydroxylation, carbonization, recrystallization (new formation) and sintering during the heating process (Emami and Trettin, 2010, 2012). The DSC curves indicate the same firing temperature till 1050 °C for these Celadon wares (Figure 7).

Data on the thermal behavior of seven Celadons supported the conclusion in the DSC curves, that the Celadons can be roughly classified into a single category. Celadon samples show a very similar thermal behavior (Figure 7). Such thermal behavior is characteristic of ceramics sintered in one time at a high temperature (Colomban et al., 2004). By increasing the temperature, an immediate one-step exothermic behavior is seen in the DSC curve of the sample. However, from this, it may conclude that the phase composition of Celadons fired at a high temperature is quite comparable to each other.

DISCUSSION

Seven pieces of Celadon wares from Kish Island have been investigated in order to explain they're possible trends. They have mostly similar colors on the body as well as in the matrix of ceramics. The gray color of the body indicates the usage of ferruginous red clay, fired under reducing conditions (Pollard and Hatche, 1986; Kerr et al., 2004). The bulk chemical composition of the Celadons showed a low amount of CaO versus enrichment of $SiO_2+Al_2O_3$ within them. As a matter of fact, concentrations of CaO (in a very low amount) can be associated with secondary calcium-rich phases (new formed calcium-rich phases) via burial conditions. The high amount of Al_2O_3 (>19%) is probably related to the high quality of kaolinite clayey raw material which have been used (Zhu et al., 2011). CaO and K_2O are the major oxides with the ability of fluxing agents in the kiln. However, the more ratios of K_2O contents lead to a greater increase of firing temperature. The variation of K_2O depends also on the source of clayey raw materials which had been used in the manufacturing processes (Kerr et al., 2004; De Rosa et al., 2012).

The iron constituents varied from 1.45 to 2.27 w%. The investigation of Pollard and Hatcher (1986) on green-wares indicated similar compositions except for the lime contents (less than about 0.5%). Potassium constituents vary between 2.69-5.42 w%. High SiO_2, Al_2O_3, CaO, and K_2O contents and low concentrations of MgO, MnO, Na_2O, and Fe_xO_y (as Flux) are well documented in the chinse proto-Celadon glaze and body in antiquity (Li et al., 2012; Hao et al., 2013). The amount of TiO_2 (0.15%-0.22%) and Fe_2O_3 (1.63%-2.10%) in the Middle Ming dynasty (Zhu et
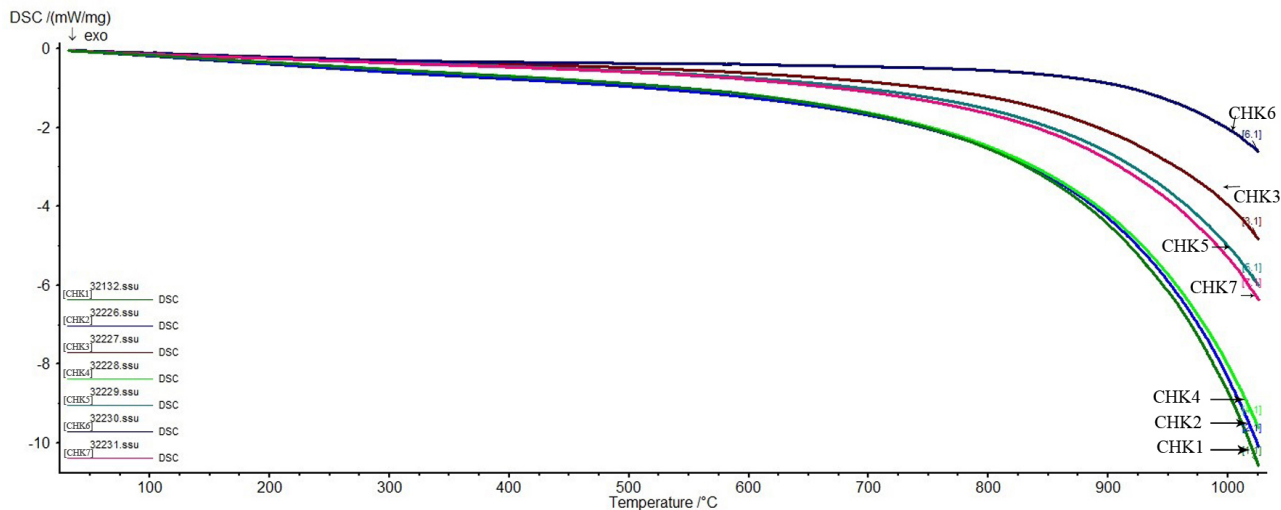

Figure 7. STA diagram of the analyzed Celadons from Harirah, Kish Island.

al., 2011; Li et al., 2012) is similar and considerable to the samples from Harireh ($0.14 < TiO_2 < 0.26$ and for Iron oxide $1.45 < Fe_2O_3 < 2.27$). Indeed, the K_2O contents are also similar to the previous studies on Middle Ming dynasty. According to Freestone (1982) and Li et al., (2012) alkaline oxides such as MgO, K_2O and CaO could be used as flux in the body of Celadon wares, but their amount in the glazes are particularly closed to a refractory material (Freestone, 1982; Li et al., 2012). The comparative analysis of the Celadon wares from Vietnam and China and the port show strong similarities in the amount of K_2O, CaO, and P_2O_5 in the glazes (Liem et al., 2000). The fluxing ability of K_2O is stronger than that of CaO.

Models of partial sintering process resulted in the collapse of the structures below the elastic instability pressures for both α-quartz and moganite. Certainly, moganite overlapped quartz on $\bar{1}12$ and $\bar{2}02$ crystalline plane and appears as a tiny hump on the left shouter of quartz peaks (at $2\Theta = 21.12$ and 26.32) in all diffractogram (Figure 5).

The chemical analysis of Celadons from Yuan Dynasty and early Ming Dynasty indicated that amount of K_2O is beneficial for reducing the firing temperature and developing shininess of porcelain glaze (Li et al., 2012). High variation of phosphate in the glaze can be related to its occurrences as amorphous and/or very fine crystal phase within the silica glaze (Freestone et al., 1985). Indeed, the presence of phosphate could be related also to post-burial conditions. The boundaries and face interface areas between the Celadons glaze and body are characterized by scanning electron microscope with integrated EDX system. The glaze on the surface is created very homogeneously and proved to have a well uniform layer above the body. The presence of the visible thicker layers and considerably more bubbles (Figure 8A) could also particularly be used as a main and continuous manufacturing process of the Chines ware which is also similar to those from Kish Island (Zhu et al., 2011). Under such circumstances the distinguishing between these materials would focus on local raw materials, as well as production technology employed there.

The typical occurrence of mullite is often called fibrolite which is a tiny aggregate of extremely fine whiskers. The characteristic structures and the temperature-time dependence of the structure of mullite were commonly similar to those of quartz. The possible source of this mineral can be formed primarily from the pure clay deposits as lithoclasts which are mostly associated with quartz, K-feldspar, and sillimanite (Havancsáka et al., 2014). It comes secondary via alteration of clay-feldspars-quartz originated from the acidic rocks (Tite et al., 2012).

The presence of mullite indicates a firing temperature above 980 ºC (Gonzalez-Garcia et al., 1990). Mullite formed from a non-calcareous kaolinitic clay reservoir based on the dehydration reaction of kaolinitic clay which has been followed by decomposition to metakaolinite (± Al-Si spinel) and at least the formation of mullite above 980°C (Noll and Heimann, 2016). Mullite and moganite are the phases which could be the thermobarometer minerals in this study. Mullite appears in a firing temperature > 980°C (Figure 8B and C) (Heimann and Maggetti, 1981; Gonzalez-Garcia et al., 1990; Rathossi and Pontikes, 2010). Presence of mullite ($3Al_2O_3.2SiO_2$) (also in 15th-century Vietnamese porcelain) via high temperature and moganite (which has not yet reported in Asian literature) via high temperature as well as, is the considerable points for adjusting the raw materials for developing such wares.

The crystallization of high-temperature phases such as mullite and sillimanite and their disappearance up to 1100°C indicate a firing temperature up to 1050 °C for the samples (Daghmehchi et al., 2016). The compact structure of the bodies also corroborates a firing temperature above 1000 °C (Hao et al., 2013). In the investigated Celadons vitrification has procced about 1000 ºC via secondary mullite formation out of liquid phase (Tite et al., 2012; Singh and Kumar, 2017). The pre-sintering occurred at 977ºC probably due to the stability of mullite till 1100 ºC.

CONCLUSION

The crystallite and chemical compositions of the 7 pieces of Celadons body and glaze were comparatively analyzed by XRD, XRF, STA, SEM-EDX and thin section petrography. The Celadon wares of Harirah Island suggest similar external appearance and chemical composition. The body of Celadon has lower Fe_2O_3 and higher K_2O. However, the existence of thicker glaze and the presence of knowingly more bubbles might be used as secondary structures allowing the similarity of Harirah Celadon to those reported from China. The ceramics proved to have high siliceous contents. The fabric of all investigated samples contains the same chemical composition. The microstructure of the material is very dense and well vitrified. The crystallite and chemical constituents in the body of Celadon are not compatible with the geological setting of the Island. The XRF analyses of the Celadons show a little amount of CaO as a flux. Anorthite is detected in the glaze and considered by CaO, Al_2O_3 and SiO_2 constituents. Moganite and mullite are also characteristic phases in these Celadons. DSC and XRD analyses indicate a firing temperature around 1000-1050 °C due to the crystallization of high-temperature phases. The glaze on the both internal as well as the external surface is very constant and seems to be covered with the same raw materials during the manufacturing process. The result shows that a lithoclastic source enriched in fibrolite were used as raw materials.
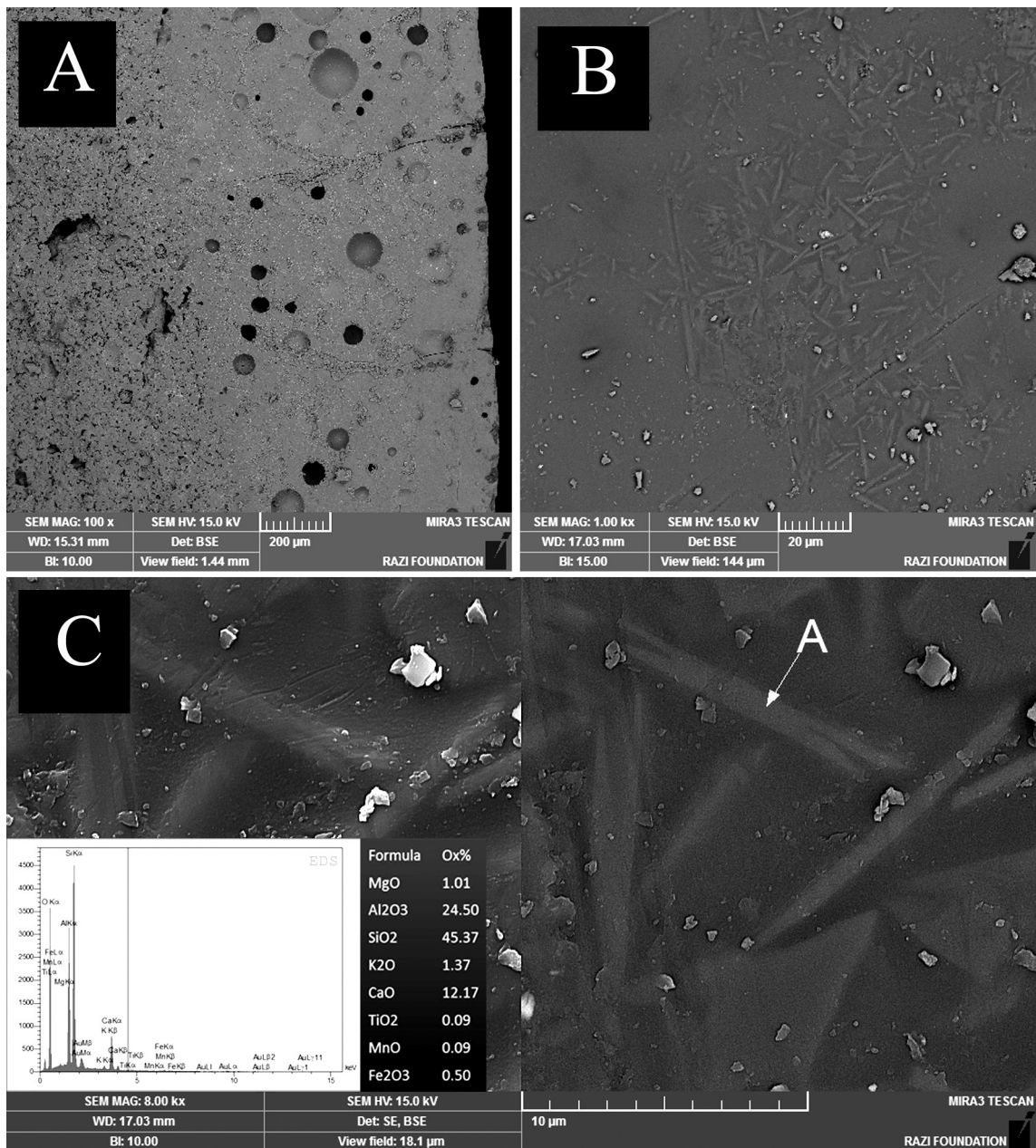
Figure 8. A) The boundaries and face interface areas between the glaze and the body of Celadons in sample CHK7, characterized by scanning electron microscope. Very homogeneously glaze surface is created and proved to have a well uniform layer above the body. B) the structure and distribution of mullite crystals in the samples as tiny prismatic crystals. C) Chemical composition of the mullite crystals by EDX. All pictures are from the sample CHK5.

This is also responsible for the different color of the body under different firing conditions and temperature. The presence of Moganite could be either due to its deliberate addition as part of the manufacturing process or as traces of raw material employed via technological features. These argumentations are mainly related to the suggestion of a local material, as well as production technology somewhere beyond the Iranian border.

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REFERENCES

Cho M.Y., 2008. How land came into the picture: Rendering history in the fourteenth-century "Jami al-Tawarikh", University of Michigan.

Colomban P., Liem N.Q., Sagon G., Tinh H.X., Hoành T.B., 2003. Microstructure, composition, and processing of 15th century Vietnamese porcelains and Celadons. Journal of Cultural Heritage 4, 187-197.

Colomban P., Sagon G., Huy L.Q., Liem N.Q., Mazerolles L., 2004. Vietnamese (15th Century) Blue-And-White, Tam Thai and Lustre Porcelains/Stonewares: Glaze Composition and Decoration Techniques. Archaeometry 46, 125-136.

Colomban P., Khoi D.N., Liem N.Q., Roche C., Sagon G., 2004. Sa Huynh and Cham potteries: microstructure and likely processing, Journal of Cultural Heritage, 5(2), 149-155.

Daghmehchi M., Omrani H., Emami M., Nokandeh J., 2016. Mineralogical and thermo-chemical characteristics of the Hellenistic ceramics and raw clay from Qizlar Qal'eh (northeastern Iran). Materials Characterization 120, 143-151.

De Rosa B., Cultrone G., Rendeli M., 2012. Archaeometric reconstruction of Nuragic ceramics from Sant'Imbenia (Sardinia, Italy). Technological evolution of production process. Periodico di Mineralogia 81, 313-332.

Emami M., 2012. QXRD, XRF and optical microscopy applied to characterization and provenance of ancient ceramics from Haft Teppeh (1500-1150 BC), southwest Iran. in: Hamdan, N., El-Khatib, S. (Eds.), IOP Conference Series: Materials Science and Engineering, IOP Publishing 37, 1-8.

Emami M., Kowald T., Trettin R., 2016. Preliminary archaeo-metallurgical studies on copper extraction from polymetallic ore sources in Meymand, south central Iranian desert. Archaeological and Anthropological Sciences 9, 1515-1528.

Emami M. and Trettin R., 2012. Mineralogical and chemical investigations on the ceramic technology in Čoġā Zanbil, (Iran, 1250 BC). Periodico di Mineralogia 81, 3, 359-377.

Emami M. and Trettin R., 2010. Phase Generating Processes in Ancient Ceramic Matrices Through Microstructure Investigation with High Resolution Microscopy Methods. Journal of Advanced Microscopy Research 5, 181-189.

Feng S. L., Feng X.Q., Zhu J.H., Xie G.X., Yan L.T., Li, L., Li G., Shen Q.H., 2008. Nondestructive analysis on ancient porcelain of Longquan Kiln in Zhejiang Province by WDXRF. Chinese Physics C 32, 284-288.

Freestone I.C., 1982. Applications and Potential of Electron Probe Micro-Analysis in Technological and Provenance Investigations of Ancient Ceramics. Archaeometry 24, 99-116.

Freestone I.C., Meeks N.D., Middleton A.P., 1985. Retention of Phosphate in Buried Ceramics: An Electron Microbeam Approach. Archaeometry 27, 161-177.

Giurgiu A., Ionescu C., Hoeck V., Tămaş T., Roman C., Crandell O.N., 2017. Insights into the raw materials and technology used to produce Copper Age ceramics in the Southern Carpathians (Romania). Archaeological and Anthropological Sciences 9, 1259-1273.

Götze J., Nasdala L., Kleeberg R., Wenzel M., 1998. Occurrence and distribution of "moganite" in agate/chalcedony: a combined micro-Raman, Rietveld, and cathodoluminescence study. Contributions to Mineralogy and Petrology 133, 96-105.

Gonzalez-Garcia F., Romero-Acosta V., Garcia-Ramos G., Gonzalez-Rodriguez M., 1990. Firing transformations of mixtures of clays containing illite, kaolinite and calcium carbonate used by ornamental tile industries, Applied Clay Science 5, 361-375.

Havancsák I., Bajnóczi B., Tóth M., Kreiter A., 2014. Archaeometric investigation of Celtic graphitic pottery from two archaeological sites in Hungary. Craft and Science, 191.

Hao W., Zheng T., Li Y., Zheng J., Zhu J., Glascock M.D., Wang C., 2013. Provenance study of Chinese proto-Celadon in Western Han Dynasty. Ceramics International 39, 6325-6332.

Heimann R.B. and Maggetti M., 1981. Experiments on simulated burial of calcareous Terra Sigillata (mineralogical change). Preliminary results, Scientific studies in ancient ceramics. British museum research laboratory, 163-177.

Hidaka M., Ohashi K., Wijesundera R., Kumara L., Watanabe M., Koga K., Choi J.-Y., Sung N., Park Y., 2011. Correlation between glaze-colors and structural properties of the HIZEN Celadons produced in the Edo period of Japan, by means of X-ray diffraction (I), Cerâmica 57, 106-114.

Hobson R.L., 1915. Chinese pottery and porcelain: an account of the potter's art in China from primitive times to the present day, Dover Publications.

Jeon A.Y., No H.G., Kim U.S., Cho W.S., Kim K.J., Kim J.Y., Kim C.M., Kim C.S., Kang G.I., 2014. Mössbauer Spectroscopic and Chromaticity Analysis on the Colourative Mechanism of Ancient Goryeo Celadon from GangJin and Buan. Archaeometry 56, 392-405.

Kerr R., Needham J., Wood N., 2004. Science and Civilisation in China: Volume 5, Chemistry and Chemical Technology, Part 12, Ceramic Technology, Cambridge University Press.

Kilikoglou V., Vekinis G., Maniatis Y., Day P., 1998. Mechanical Performance of Quartz-Tempered Ceramics; Part I, Streight and Toughness. Archaeometry 40, 261-279.

Kim J.-Y., No H., Jeon A.Y., Kim U., Pee J.-H., Cho W.-S., Kim K.J., Kim C.M., Kim C.S., 2011. Mössbauer spectroscopic and chromaticity analysis on colorative mechanism of

Celadon glaze. Ceramics International 37, 3389-3395.

Leicht R., 1977. Calcium Carbonate Inclusions In Pottery: Some Cautions For The Archaeologist. Nebraska Academy of Sciences Transactions of the Nebraska Academy of Sciences and Affiliated Societies 428, 1-3.

Li L., Feng S.L., Feng X.Q., Xu Q., Yan L.T., Ma B., Liu L., 2012. Study on elemental features of Longquan Celadon at Fengdongyan kiln site in Yuan and Ming Dynasties by EDXRF. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 292, 25-29.

Liem N.Q., Sagon G., Quang V.X., Tan H.V., Colomban P., 2000. Raman study of the microstructure, composition and processing of ancient Vietnamese (proto) porcelains and Celadons (13-16th centuries). Journal of Raman Spectroscopy 31, 933-942.

Martinon-Torres M., Rehren T., Freestone I.C., 2006. Mullite and the mystery of Hessian wares. Nature 444, 437-438.

McConkey R. and McErlean T., 2007. Mombasa Island: a maritime perspective, International Journal of Historical Archaeology 11, 99-121.

Noll W., 1991. Alte Keramiken und ihre Pigmente, Studien zu Material und Technologie, Schweizerbart'sche Verlagsbuchhandlung, Stuttgart.

Noll W. and Heimann R.B., 2016. Ancient Old World Pottery. Materials, Technology, and Decoration, Schweizerbart'sche Verlagsbuchhandlung, Stuttgart, 98-99.

Ntah Z., Sobott R., Fabbri B., Bente K., 2017. Characterization of some archaeological ceramics and clay samples from Zamala-Far-northern part of Cameroon (West Central Africa). Cerâmica 63, 413-422.

Pierson S., 2012. The Movement of Chinese Ceramics: Appropriation in Global History. Journal of World History 23, 9-39.

Pollard A. and Hatcher H., 1986. The chemical analysis of oriental ceramic body compositions: part 2-greenwares. Journal of Archaeological Science 13, 261-287.

Pretola J.P., 2001. A Feasibility Study Using Silica Polymorph Ratios for Sourcing Chert and Chalcedony Lithic Materials. Journal of Archaeological Science 28, 721-739.

Rathossi C. and Pontikes Y., 2010. Effect of firing temperature and atmosphere on ceramics made of NW Peloponnese clay sediments: Part II. Chemistry of pyrometamorphic minerals and comparison with ancient ceramics. Journal of the European Ceramic Society 30, 1853-1866.

Singh M. and Kumar S.V., 2017. Multi-analytical characterization of XVII century Mughal glaze tiles from northern India. International Journal of Conservation Science 8, 389-400.

Tite M., 1969. Determination of the firing temperature of ancient ceramics by measurement of thermal expansion: a reassessment. Archaeometry 11, 131-143.

Tite M.S., Freestone I.C., Wood N., 2012. An Investigation into the Relationship between the Raw Materials used in the Production of Chinese Porcelain and Stoneware Bodies and the Resulting Microstructures. Archaeometry 54, 37-55.

Whitehouse D., 1976. Kīsh, Iran 14, 146-147.

Wood N., Doherty C., Ratelli S., 2005. Some aspects of Yue ware production at Shanglinhu in the late Tang dynasty, in Science and technology of ancient ceramics 6: Proceedings of the International Symposium (ISAC'05) (Ed. Guo Jingkun), Shanghai Research Society of Science and Technology of Ancient Ceramics, Shanghai, 185-98.

Wood N. and Li H., 2015. A Study of the Techniques Used to Make Laohudong Guan Ware in China in the Southern Song Dynasty. Archaeometry 57, 617-635.

Zhu T., Huang H., Wang H., Hu L., Yi X., 2011. Comparison of Celadon from the Yaozhou and Xicun kilns in the Northern Song Dynasty of China by X-ray fluorescence and microscopy. Journal of Archaeological Science 38, 3134-3140.