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Prototype Doucai porcelain – A special form of ancient Honglvcai in Cizhou kiln, Jin Dynasty (1115–1234 AD), China

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ABSTRACT

Honglvcai (also named red and green porcelain) was prosperous in Jin Dynasty (1115–1234 AD) and had played a pivotal role in ancient ceramic development history in China, connecting with former glazed pottery Tang Sancai and later polychrome porcelain like Wucai and Doucai, but little scientific analysis has been reported up until the present day. In this research, one precious sherd unearthed from Linshui (one major site of Cizhou kiln system) in Hebei province was adopted as specimen for compositional and micro-structural analysis. Optical microscope (OM), X-ray fluorescence spectrometry (XRF) and energy dispersive spectrometry (EDS) in association with scanning electron microscope (SEM) were employed for investigation of the multi-colour decors and black outline. The extensive data reveal that although high lead content has been detected by surface analysis and the iron-rich pigment particles do locate in the middle of the whole vitrified region, the black outline in fact belongs to high temperature fired under-glaze painted decoration, rather than the deceptive low temperature fired lead based over-glaze decor or the generally believed 'sandwich' (glazed twice) structure. Furthermore, a corresponding criterion then has been established for differentiation among three main decorating methods, especially for the puzzling 'sandwich' effect scenario. Moreover, once this ongoing vexed debate over the nature of the black decoration in Cizhou kiln system has been clarified, it is then reasonable to deduce that, from idealistic and technical point of view, there is no difference between producing prototype like this sherd and real Doucai in Ming dynasty (1365–1644 AD) except for the types of under-glaze pigment. Overall, all these results together firstly provide realising scientific proofs for the origin of Doucai.

1. Introduction

DouCai is exquisite polychrome porcelain which boomed in Chenghua reign (1465–1487 AD) in Ming dynasty (1365–1644 AD). The term 'Dou' refers to a kind of complimentary patterns consisting of under-glaze and over-glaze decoration for a competitive aesthetic effect. Technically speaking, it is well acknowledged that the typical Doucai ware should include both high temperature fired under-glaze painted cobalt blue outlines and low temperature fired infilling polychrome decors. For instance, Fig. 1 presents a celebrated Doucai artefact, Jigang cup, which held the world record, about 280 million HKD in Sotheby's auction in 2014, of Chinese ancient porcelain with its recognised cultural and economic value.

Due to its unique artistic design and distinguished status, the origin and development of Doucai attract lots of attention in ancient ceramic researches. Jiang [1] considered that the technique of combining low temperature fired lead based over-glaze decors and the high temperature fired decoration firstly appeared on Honglvcai, which is well-known for low temperature fired over-glaze red and green decoration and flourished suddenly in Jin Dynasty (1115–1234 AD). As for the relationship of Doucai and Honglvcai, it is suggested that one kind of Honglvcai also consists of high temperature fired black under-glaze and low temperature fired over-glaze red/green decoration and thus should be the prototype of Doucai, because it is believed the black under-glaze decoration on this special type of Honglvcai stems from black and white porcelain produced in contemporary Cizhou kiln system, which is

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Fig. 1. The DouCai artefact, Jigang cup in which the blue outline was drawn by under-glaze decoration fired on high temperature whilst the red, yellow and green patterns over-glaze decoration fired on low temperature. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

famous for black under-glaze decoration. [2] However, according to optical examination, both over-glaze and under-glaze black decoration techniques had been observed in Dongshan kiln, located in Changzhi, Shanxi province but belongs to Cizhou kiln system during Jin Dynasty. [3] Given that little scientific analysis for the black decoration on Honglvcai has been reported up to our knowledge, the nature of black decoration on Honglvcai is actually unclear, i.e. whether it is high or low temperature fired over-glaze or under-glaze, so it is hard to judge – whether the special Honglvcai is the prototype of Doucai.

More importantly, it is reported that for the black decoration on black and white porcelain, the black pigment particles locate in the middle of glaze under SEM observation [3–6]. Such interesting scenario generate three explanations: 1) high temperature fired over-glaze [7]; 2) sandwich glaze, which means that the black decoration is initially applied on the glaze and then glazed again, thus the black pigment locates in the middle of glaze, i.e. glazing procedure was carried out twice [5]; 3) high temperature fired under-glaze. [3] The first opinion claims that the pigments sink down from glaze surface during the sintering process. The second one is questioned because this sandwich technique should not be feasible technically as the underlying glaze is too thin to apply the black decoration, and would increase the cost which is not suitable for commercial orientated folk kilns. Then Zhang [3] raises his opinion that it is actually under-glaze with explanation that the pigments would be pushed up by the crystal growth and bubbles produced during firing. However, Meng [8] challenged such perspective in terms of the fact that the pigments are mainly distributed around the bubbles in glaze when they are pushed up during sintering process of blue and white porcelain, instead of forming a thin, straight horizontal belt. Obviously, the previous optical and SEM observation is not eligible to solve such disputes, thus more scientific analysis is needed since the identification of the nature of black decoration is crucial for tracing the origin of Doucai, considering that the black decoration on Honglvcai is identical to that on black and white porcelain.

In this study, multiple analyses have been carried out on Honglvcai and the aim of this paper is firstly to determine the chemical formulation and microstructure of black decoration at the same time. All this information will contribute to extend knowledge on Honglvcai porcelain in northern China, poorly studied until now. Especially the structure of the black decoration technique has been identified and subsequently, this in turn will serve to clarify the link between Honglvcai and Doucai, and ultimately gives a clearer picture of the technique preparation and gradual evolution for the origin of Doucai.

2. Material and methods

2.1. Sample

Cizhou kiln system, as pointed in Fig. 2 with its primary sites, is one of the largest scale and highest level porcelain manufacturing centre in northern China, starting ceramic producing in late Tang (618–907 AD) dynasty and continued up to the early Ming dynasty. Unlike dogmatic and inflexible governing in official kiln, many breakthroughs regarding shaping, decorating methodology and thermal power revolution have been made by humble but wise artisans and impacted the later emerge of Jizhou and Jingdezhen kilns fundamentally.

Only few Honglvcai sherds with black outline have been excavated archeologically. For this paper, one precious Honglvcai sherd (archaeological code: 02LSH13), provided by Fengfeng District Conservation Institute of Cultural Relics and Archaeology, had been unearthed from the strata of Jin Dynasty at Sangong site, Linshui in 2002, and further confirmed as Jin dynasty product by archaeologists from its motif and style.

The photograph of the sample is presented in Fig. 3 with front and back views, where the body is sort of yellowish grey and the applied decoration colours are almost most comprehensive at that stage, including green, yellow, red, white and black. The sherd is probably part of the sculpture or porcelain pillow; both are representative artefacts of Cizhou polychrome objects.

2.2. Experimental details

Comprehensive visual, compositional and microstructural examinations were employed for this valuable sherd. The first survey was carried out non-destructively by energy dispersive XRF (Horiba XGT-7000, Japan). The instrument was performed at 30 kV and 0.062 mA in vacuum with 1.2 mm spot size and 150 s acquisition time for each point (approximately 30% dead time). The 1200 ℃ sintered National Soil Certified Reference (GSS-4), Corning Glass Standards D and C were used for constituent calibration of body, calcium glaze and lead glaze, respectively.

In order to explore the further information regarding microstructure and processing technologies utilised for polychrome decors application, a tiny but representative area, which contains both yellow, black and their boundary, was selected for cross section specimen preparation and initially studied by optical microscope (Keyence VHX-600E, Japan) after cutting, mounting in epoxy resin, grinding and polishing procedure. The interest piece was then coated with a thin carbon deposit for systemic SEM (Zeiss MA EVO 25, Germany) backscattered electron (BSE) mode observation and semi-quantitative area analysis by equipped EDS (Oxford Instrument X-act, UK) with corresponding appropriate parameters.

For systematic error minimising, standard cleaning procedures were executed strictly, including alternative de-ion water and acetone ultrasonic bath after cutting and rinsing or compression air blowing before each trial. In terms of XRF analysis, each region was tested four to seven individual points and each point was averaged by at least three trials. As for EDS measurements, area analysis mode was selected for every layer and the listed values contain the most of the interest areas. Moreover, the inclusions like quartz particles and bubbles are intentionally avoided in EDS line scanning. It is also worthwhile noticing that the curves of line scanning may fluctuate a little bit owing to the time and cost limitation (linescan binning factor 3, 200 points were collected within 320 micrometres in 50 min). Likewise, EDS mapping was acquired in 120 min for approximately 300*300 μm areas.

3. Results and discussion

3.1. The nature of the black decoration

3.1.1. High or low temperature fired?

With respect to non-destructive surface analysis, the mean values of calibrated EDXRF quantitative chemical constituents taken from potsherd body and typical coloured regions of the sherd are demonstrated in Table 1, where four points of black decoration are listed

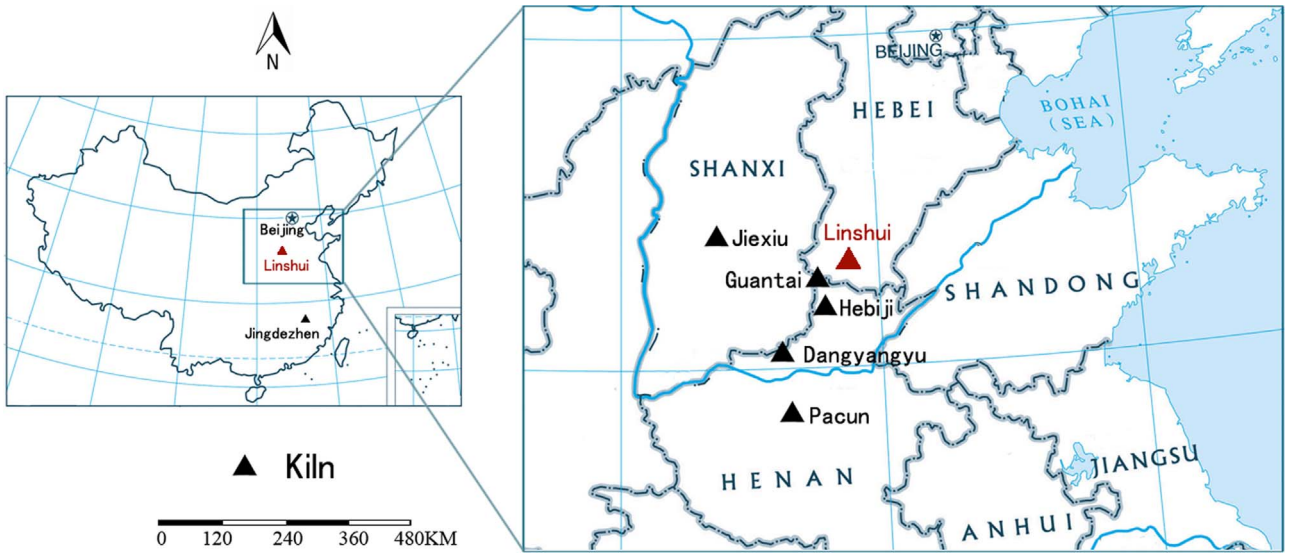

Fig. 2. Geographic distribution of major Cizhou kiln system sites in northern China, including Guantai, Linshui kiln in Hebei province, Pacun and Hebiji in Henan province and Jiexiu kiln in Shanxi province.

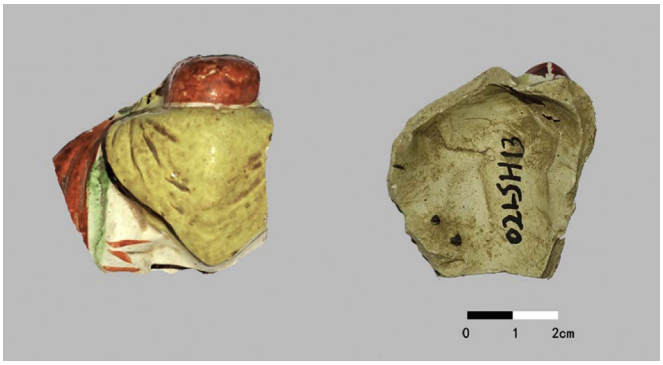

Fig. 3. Appearance of Honglvcai sherd (02LSH13) adopted in this study with front and back views. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

individually owing to the large dispersion of data.

Among all the results, black decoration is of interest due to the abnormal comparatively high lead content, 22 wt% in average and one peaking at 37 wt%, suggesting it seems very likely attributed to the SiO_2-Al_2O_3-PbO ternary system which is typically fired in low temperature with oxidising atmosphere like red, green and yellow over-glaze. The stereo optical microscopy investigation from cross section view, as validated in Fig. 4a, is involved as an assistant to support this standpoint since the right black decoration locates above a narrow light brownish yellow strip. Such characteristics are tremendously similar to the typical over-glaze structure to some extent where the pigments dissolute and a downward ion diffusion trend occurs during sintering and therefore cause colouration of the underneath transparent layer. All these evidence imply the black outline here seemingly belongs to low temperature fired over-glaze decoration.

Yet further SEM observation tends to convey a more complex viewpoint. Specifically, Fig. 4b, in which the high contrast bright top layer and the internal discrete zonal belt are diagnosed as lead-rich and iron-rich by EDS inspection whilst the surrounding dark band fall into traditional Chinese calcium-alkaline recipe range, which will be detailed discussed in following Section 3.1.2, demonstrates that the actual invasion degree of lead glaze (yellow decor) is much more serious than what has presented in optical observation due to the superior flowability and volatility of Pb and even at the far end of the black region there also exists a thin bright deposit which presumably results from lead vapour condensation during furnace cooling, rationalising the presence of small amount of lead all through the entire XRF examinations.

In sum, it could be concluded that instead of low temperature fired decor as high lead proportion indicated, the underlying black outline in fact belongs to high temperature fired decoration and, most importantly, the results remind further studies of the defect of non-destructive surface analysis like EDXRF with respect to Pb based samples, which are prone to deliver a deceptive illusion on account of its distinguished flowability and volatility.

Table 1
Chemical compositions of the body and primary decoration colours in Honglvcai sherd (wt%).

		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	TiO_2	MnO	Fe_2O_3	CuO	PbO
Yellow decoration	Mean	0.34	0.25	6.92	25.79	0.75	1.51	0.16	0.02	1.5	0.16	62.6
	S.D.	0.24	0.09	0.4	1.3	0.03	0.46	0.03	0.02	0.05	0.02	0.39
White (transparent) glaze	Mean	0.92	0.80	14.48	72.82	2.77	4.18	0.31	ND	1.15	0.03	2.86
	S.D.	0.31	0.58	0.57	1.49	0.19	0.36	0.04	–	0.14	0.01	0.41
Body	Mean	1.05	0.87	30.05	61.65	1.64	1.31	1.29	ND	2.01	ND	ND
	S.D.	0.32	0.28	0.25	0.13	0.06	0.11	0.04	–	0.03	–	–
Black decoration	Point 1	1.10	0.35	13.08	59.90	2.41	5.14	0.46	0.11	6.47	0.11	10.87
	Point 2	0.74	0.40	8.54	46.36	1.16	1.99	0.36	0.12	5.99	0.15	34.19
	Point 3	0.68	0.25	15.40	39.87	1.63	1.55	0.97	0.01	2.77	0.09	36.78
	Point 4	1.02	1.84	15.46	58.81	1.96	9.81	0.39	0.01	5.55	0.10	5.05
	Mean	0.89	0.71	13.12	51.24	1.79	4.62	0.55	0.06	5.20	0.11	21.72
	S.D.	0.21	0.76	3.25	9.75	0.53	3.81	0.29	0.06	1.66	0.03	16.10

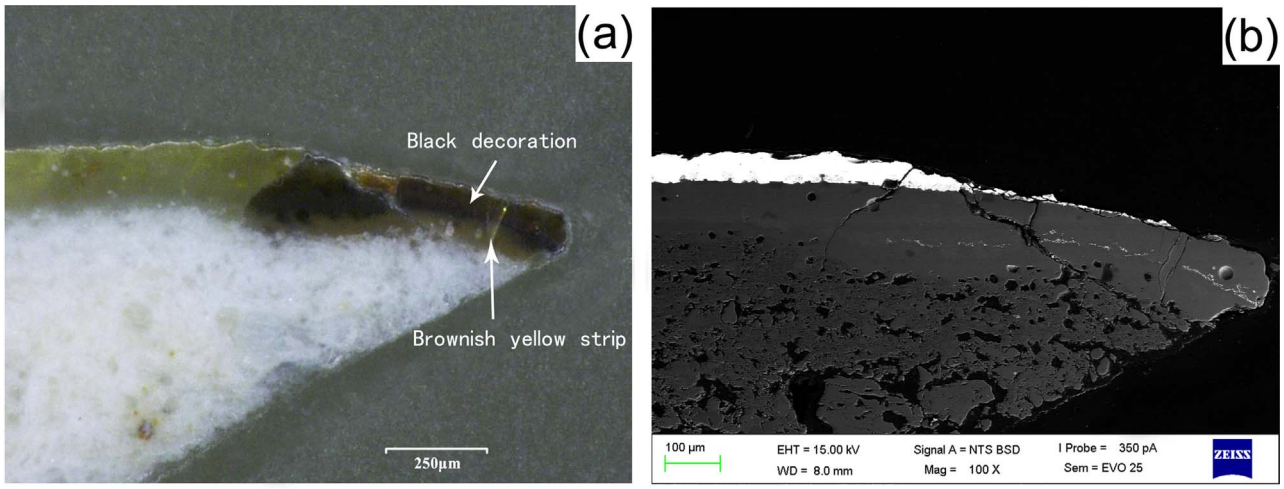

Fig. 4. (a) Stereo optical microscopy image of the sherd in cross section view, (b) the BSE image corresponding to (a) and the contrast essentially results from the compositional dissimilarity owing to different atomic number.

3.1.2. Over-glaze, sandwich glaze, or under-glaze?

As displayed in Fig. 4, a belt of the linear arrayed bright particles, corresponding to black decor in visual and confirmed as iron-rich pigment via EDS, are almost accumulated at the centre of the whole vitrified region. Such phenomenon has been extensively reported in worldwide under-glaze painted decoration, such as celebrated blue and white porcelain from Yuan dynasty, black and white porcelain produced in Cizhou system kiln during Song-Jin period. In order to address the intense dispute over the nature of the black decoration as summarised in Introduction, EDS line scanning and mapping was conducted for verifying the diffuse tendency originally.

The line scan survey, as illustrated in Fig. 5, from the invaded coloured lead glaze to inner body via whole glaze layer, initially indicates that the dividing boundary between glaze and body/slip locates at 190 μm in overview SEM image, but then the scanning curves of Fe, Ca, Al demonstrate a different scenario. Specifically, the counts of iron experience a gentle decline from invaded lead glaze toward the beneath calcium-alkaline layer until an abrupt bi-peak suddenly appears in approximately at 110 and 130 μm, which could be attributed to electron beam impinging cross the pigment region, and afterwards descend steadily to a negligible level. In contrast, the calcium concentration soars rapidly from lead glaze initially and swings seriously before 130 μm due to the heterogeneous distribution of precipitates, probably anorthite, whilst from this point onwards, the signal drops off dramatically from over 200 cps to less 50 cps, inferring that bounded by 130 μm where iron-rich particles accumulate, the former area with high content of alkaline earth like calcium can be regarded as glaze zone whilst the later area seems not because the extremely low concentration of flux. Besides, it is also supported by the curve of aluminium, which processes a similar upward trend within first 30 μm and subsequently keeps steady about 400 cps before 130 μm in spite of certain fluctuations, and then rises to form bimodal, peaking at 130 μm and 150 μm respectively, indicating that on one hand, the beneath vitrified layer has extremely high kaolin content as ordinary slip or body substrate and on the other hand, the region corresponding to two climaxes may be filled with alumina rich substance like mullite. After that, the curve levels off around 800 cps with certain dips owing to unmelted quartz particles and pores in body. All these three curves suggest the real dividing boundary of glaze and body/slip probably coincides with the pigment belt around 135 μm, even though it is visually indicated at 190 μm.

Moreover, aiming at better representative in larger scale, the EDS mapping trials are carried out and Fig. 6, including overlay and individual element distribution, demonstrate the results that divided by the colorant strip, the upper vitrified layer is full of exclusive short

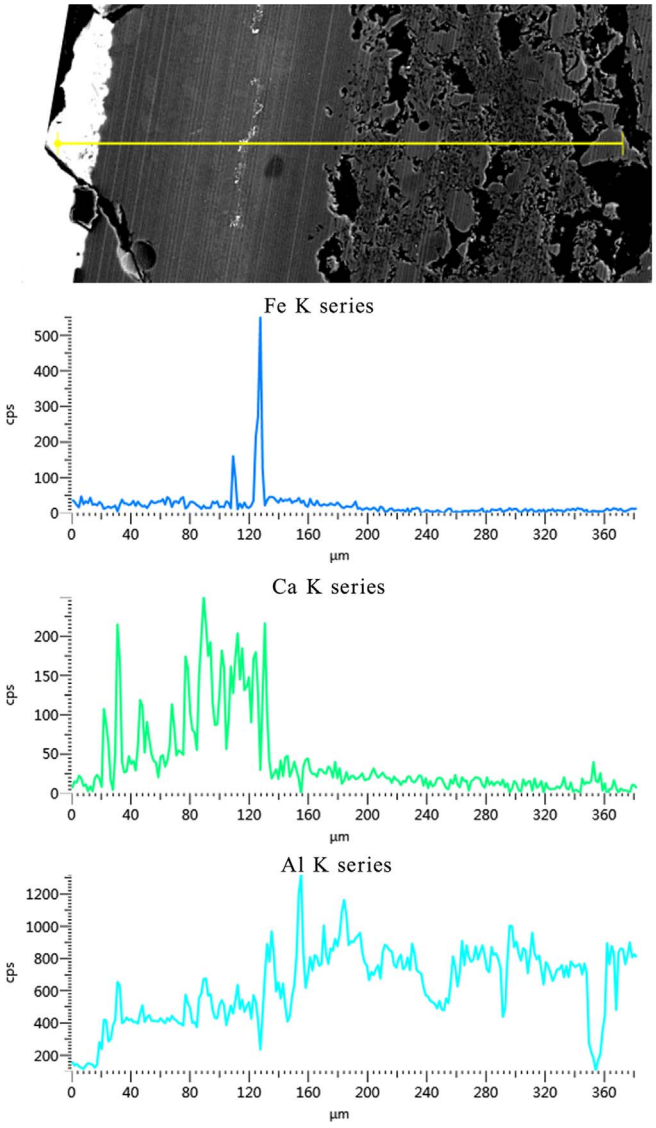

Fig. 5. A typical example of linescan curves with individual element counts verse distance.

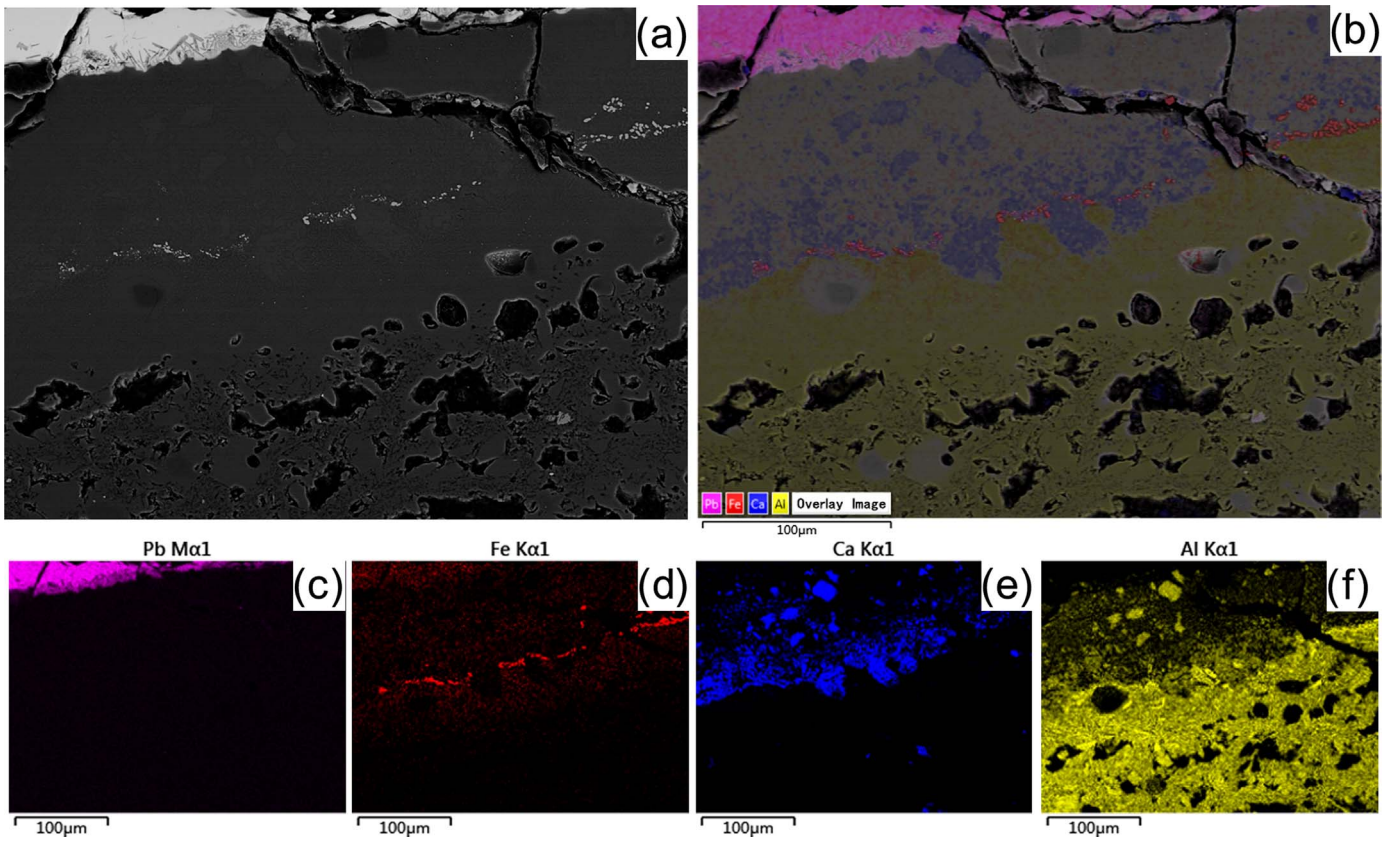

Fig. 6. The mapping image of black decoration, (a) original BSE image, (b) overlay mapping image, (c–f) individual mapping images, where the pink, red, blue and yellow colours exemplify the elemental distribution of Pb, Fe, Ca and Al respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

column or plate-like anorthite precipitates ($CaAl_2Si_2O_8$, inferred by the accordance of Al and Ca distribution and later stoichiometry estimation via EDS) which is common phase in ceramic glaze; whilst the lower vitrified layer, approximately 50 μm wide, contains few unmelted dissociated quartz particles with melting rims and certain amount of mullite crystals which should rarely be discovered in glaze but usually in the body part of ceramic. Such scenario is consistent with the line scanning results and therefore highly suggests that the vitrified layer under pigment zone was presumably not a part of glaze but of body/slip in this sherd.

Fig. 7 illustrates the laminated structure of overview (a), detailed yellow (b) and black (c & d) decoration in high magnification individually. The targeted EDS area analysis, as listed in Table 2, illustrate that the yellow decor consists of five laminas, from top to bottom these are low temperature fired lead glaze, calcium-alkaline glaze (labelled as layer A), well vitrified layer (labelled as layer B), slip (derived by higher Al and less impurities content than that in body) and body respectively. Likewise, the parallel phenomenon is observed under the black decoration notwithstanding there only exist four laminas, which comprise of calcium-alkaline glaze (layer A), thin pigment zone, well vitrified layer (layer B) and slip from top to bottom. The body part seems deformed probably due to sample incompleteness or preparation process.

It is worthwhile noticing that in yellow decor, the well vitrified layer (labelled as layer B in Fig. 7b) contains enormously high alumina content, up to 37.6 wt% in Table 2, confirming that this layer should originally belong to a part of slip indeed. Besides, there exist more Ca (about 1.7 wt%) and K (about 2.6 wt%) than that in beneath slip area (1.00 wt% of Ca and 2.29 wt% of K), suggesting that the vitrification results from larger flux concentration by diffusion during high temperature sintering probably.

As for black decor, interestingly, given the fact that the overall content difference of alkaline earth fluxes in yellow (where CaO $+K_2O$=4.36 wt%) and black (where $CaO+K_2O$=4.29 wt%) decoration seems negligible, the double wider vitreous layer (labelled as layer B in Fig. 7c & d) under pigment zone could only be attributed to iron content, indicating that even though the diffusion of flux elements from transparent underlying glaze does dominate the vitrification process, the ferric ions from pigments dissolution acts as an important role in this distinctive structure formation as well. Additionally, the hue difference between two vitrified layers, which may cause misleading delusion in optical observation, could also be explained by the difference of ion diffuse rate in solid and liquid state. To sum up, it could be concluded that the black iron pigments in fact located in the boundary between glaze and slip, and thus rather than the deceptive sandwich glaze structure, the black outline belongs to high temperature fired under-glaze decoration.

As for other possible interpretations proposed in previous studies [3–7], namely the over-glaze and Zhang's under-glaze theories, it seems that they all have their limitations to some extent as reviewed previously and more negative evidence has been exposed at this time. Specifically, for over-glaze hypothesis, it is hardly for the pigment particles to go through the entire precipitate-rich glaze from the surface and finally sink down homogeneously to form a straight zonal belt. As for under-glaze theory proposed by Zhang [3], although his conclusion is correct the underlying principles are probably not valid in this case since there only exist few bubbles in glaze and the distribution of pushed up pigments may not be straight either. Therefore, a combination of compositional and structural analysis would be recommended criteria for decorating techniques identification in the future study.

Overall, it is convinced that the well vitrified brownish yellow strip under the black decoration was part of slip due to compositional examination and consequently, the common reported scenario in both black and white porcelain and Honglvcai needs to be reassessed and

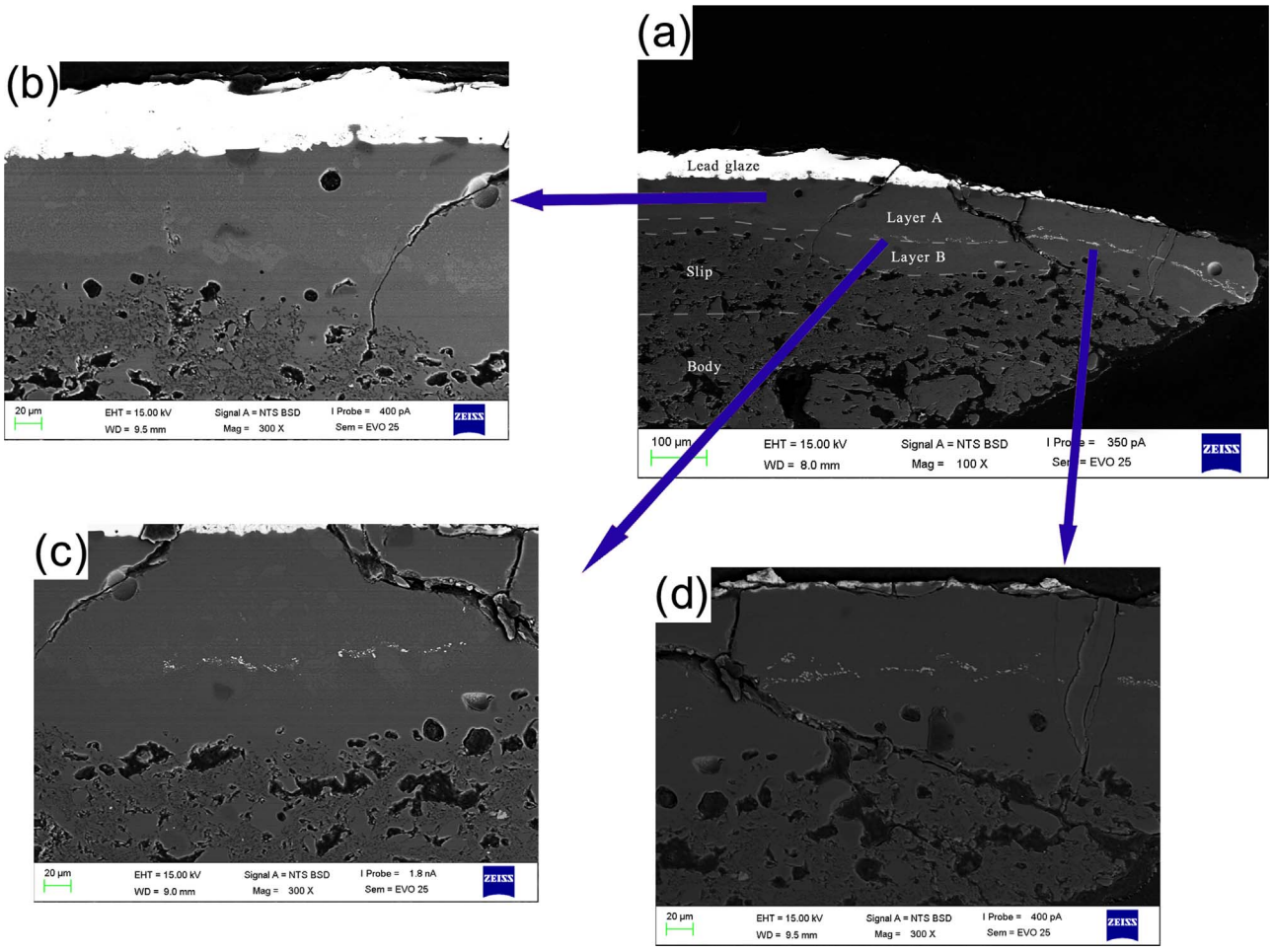

Fig. 7. The overview (a) and individual high magnification of BSE images of interest region, where (b) corresponds to yellow decor, which contains five layers: invaded lead glaze, Layer A, Layer B, slip and body; and (c) (d) correspond to black decor, which contains four layers: Layer A, pigment zone, Layer B and slip.

Table 2
Detailed chemical compositions of each lamella in yellow and black decors.

Yellow decoration		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	TiO_2	FeO
Layer A	Mean	1.92	0.96	21.07	65.73	2.88	6.13	0.56	0.75
	S.D.	0.09	0.11	0.85	1.38	0.27	0.63	0.32	0.05
Layer B	Mean	1.14	0.82	37.59	54.22	2.62	1.74	0.90	0.97
	S.D.	0.02	0.09	0.67	0.46	0.02	0.09	0.06	0.03
Slip	Mean	0.97	0.48	41.72	51.39	2.29	1.00	1.20	0.95
	S.D.	0.05	0.01	0.44	0.12	0.14	0.08	0.07	0.06
Body	Mean	0.67	0.56	34.85	58.70	1.35	0.75	1.16	1.96
	S.D.	0.04	0.07	0.18	0.13	0.08	0.12	0.01	0.12
Black decoration		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	TiO_2	FeO
Layer A	Mean	1.95	1.02	20.28	64.64	2.61	5.76	0.29	3.45
	S.D.	0.15	0.28	1.33	1.45	0.50	1.39	0.16	0.52
Pigment Layer	Mean	1.61	1.12	26.61	41.77	0.68	10.07	1.07	17.07
	S.D.	0.23	0.68	4.20	2.18	0.32	0.35	0.36	2.59
Layer B	Mean	1.22	0.92	34.63	53.89	2.21	2.08	0.76	4.29
	S.D.	0.03	0.13	1.38	1.17	0.14	0.25	0.06	0.26
Slip	Mean	1.01	0.41	41.19	51.82	2.18	1.06	1.05	1.28
	S.D.	0.09	0.06	0.76	0.55	0.08	0.17	0.26	0.15

presumably most of them belong to under-glaze concept in fact. Furthermore, it could be noticed that each single technique has their inherent drawback and may not eligible to propose comprehensive detailed pictures, sometimes may even provide the misleading information. At least for this kind of structure and related manufacturing identification, the combination of microscopy and chemical analytical strategy would be a promising candidate as powerful tools for more accurate judgement delivery for decorating determination.

3.2. The origin and development of Doucai

As for the origin of Doucai, Qin [2] had predicted that the Doucai wares which were produced in early Ming or even Yuan Dynasty (1271–1368 AD) must be found archeologically someday with the rise of blue and white porcelain in Yuan Dynasty. Moreover, Jiang believes one sherd in Ming Dynasty, Yongle reign (1403–1424 AD), which was excavated at Zhonghualu, Jingdezhen in 1984, is the *proto type* of Doucai since '*... in this sherd, the secondary fired green decoration was filled in brown* outline *which had been already fired at high temperature...and this sort of innovation initiates the later infilling technique in blue and white porcelain in Xuande reign (1426–1435 AD)...and it is no doubt that the development of Doucai must be on this basis.*' [1] According to his statement, the Honglvcai sherd with black outline analysed in this study, which has been confirmed as high temperature fired under-glaze before, should also be regarded as the proto type of Xuande Doucai but hundred years in advance.

With respect to the long incubation period (more than two hundred years from Honglvcai in Jin Dynasty to real Doucai in Chenghua reign)

and the sudden maturity of Doucai, various hypothesises were raised by specialists from different point of view. In general, those assumptions could be classified into two kinds of basic aspects: one is involved in cultural choice and social economy, especially influenced by imperial taste; the other tends to emphasise the importance of technique revolution in ceramic manufacturing. Specifically, it is reported that the financial accumulation since the gold age of '*Yongxuan Zhizhi*' (1403–1435 AD) and restability from war with Tartary laid the foundations for Doucai materially, at meantime the personal preference of Chenghua emperor (1465–1487 AD), who is famous for his femininity and outstanding artistic tastes, directly stimulated the generating and rapid development of Doucai [9]. On the other hand, some scholars argued that, rather than the beautiful subjective wish, the inherent driving force must be technical progresses [10] which are able to fundamentally push the promotion of polychrome porcelain from Honglvcai to Doucai; unless the severe halo effect of imported cobalt pigment (Sumali Blue) had been overcome, it is impossible to draw under-glaze blue outlines in Doucai like that of iron black in Honglvcai. Thus, ancient potters tried to seek other substitute cobalt ores for a long time, and once the domestic cobalt pigments, which usually express high viscosity due to low iron content, [10] was found and widely exploited in Chenghua reign for the finest painting, Doucai was eventually matured and flourished soon after.

4. Conclusion

In this study, the nature of black decoration on Honglvcai sherds excavated in Linshi kiln from Jin dynasty was determined by a series of scientific means. The controversial historical debate regarding whether the blackline belongs to over-glaze, sandwich glaze or under-glaze was eventually terminated by the combination of composition and microscopy analysis with reasonable explanations, therefore reveals technology evolution from Honglvcai to Doucai evidently. In other words, the low temperature fired over-glaze polychrome decorations with high temperature fired under-glaze blackline porcelain produced in Cizhou kilns system is the proto-type of Doucai in Ming Dynasty.

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