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RESEARCH ARTICLE

Microscopic analysis of "iron spot" on blue-and-white porcelain from Jingdezhen imperial kiln in early Ming dynasty (14th–15th century)

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Abstract

"Sumali," as an imported cobalt ore from overseas, was a sort of precious and valuable pigment used for imperial kilns only, which produces characteristic "iron spot" to blue-and-white porcelain in early Ming Dynasty (A.D. 14th–15th century). Although there were some old studies on it, the morphology and formation of iron spot has not been fully investigated and understood. Therefore, five selected samples with typical spot from Jingdezhen imperial kiln in Ming Yongle periods (A.D. 1403–1424) were analyzed by various microscopic analysis including 3D digital microscope, SEM-EDS and EPMA. According to SEM images, samples can be divided into three groups: un-reflected "iron spot" without crystals, un-reflected "iron spot" with crystals and reflected "iron spot" with crystals. Furthermore, 3D micro-images revealed that "iron spots" separate out dendritic or snow-shaped crystals of iron only on and parallel to the surface of glaze for which "iron spot" show strong metallic luster. Combining with microscopic observation and microanalysis on crystallization and non-crystallization areas, it indicates that firing oxygen concentration is the ultimate causation of forming reflective iron spot which has a shallower distribution below the surface and limits crystals growing down. More details about characters of "iron spot" used "Sumali" were found and provided new clues to coloration, formation mechanism and porcelain producing technology of imperial kiln from 14th to 15th centuries of China.

KEYWORDS
blue-and-white porcelain, iron spot, microstructure, Sumali

1 | INTRODUCTION

Cobalt in oxidation states will show noticeable blues in glasses and glazes (Scholes, 1946). Because of this characteristic, cobalt ores have been used as a glass colourant which can be traced to Iraq about B.C. 2000 (Kaczmarczyk, 1986; Moorey, 1999). In China, a kind of cobalt-blue pigment imported from Persia in West Asia, also called "Sumali", was taken to China for producing blue-and-white porcelain with blue decorations, when a famous Chinese navigator, Zheng He, sailed westwards seven times by the order of emperor for trade and exploration in the years between 1405 and 1433 of the Yongle and Xuande reign (A. D. 1403–1435) of Ming Dynasty (Kerr, Needham, & Wood, 2004; Wen & Pollard, 2016). Because of the lack of exploitation and utilization of native cobalt ore, this kind of imported cobalt-blue pigment was

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so rare and precious at that time that only imperial kilns or namely Guan kiln were able to use it to produce blue-and-white porcelain for royalty, which represented the royal aesthetic and the highest quality of ancient Chinese ceramics (Chinese Silicate Society, 1996).

It is generally believed that a special feature with high-iron and minimal manganese of imported cobalt-blue pigment “Sumali” distinguishes early 15th century blue-and-white porcelains from their equivalents after that times (Cowell & Zhang, 2001; Li, 1998; Wu, Li, Deng, & Wang, 2004), which bases on a conclusion that native Chinese cobalt ores contain high manganese component content, and conversely, that the absence of manganese indicates the ore was imported (Banks & Merrick, 1967; Chen, Guo, & Zhang, 1978; Garner, 1956; Watt, 1979; Young 1956). However, besides chemical composition, this precious cobalt material compared with native Chinese asbolite also has its own characteristics that are still lack of the researches in its appearance characteristics and formation mechanism, both in practice and in theory.

A distinguished feature of blue-and-white porcelains used “Sumali” is the celebrated “heaped and piled” quality. The high-iron cobalt used during the Yongle and Xuande reign periods tended to diffuse through the glazes to develop dark flecks especially in the gathering place of pigment and always show dark grey, brown and rusty color on the surfaces of glaze which are called “black spot” or “iron spot.” Spots with metallic luster are commonly known as “Tin light” (Chen, Guo, & Zhang, 1990; Zhang & Guo, 1989). As a unique characteristic of blue-and-white porcelain produced at that time, these spots are so much admired by later connoisseurs that it was sometimes laboriously simulated by Guan kiln in the Qing Dynasty (A.D. 1644–1912) using manganese-cobalt ores applied in a myriad of tiny dark-blue dots to cobalt painting before glazing, but without success (Zhou, 1958). On the contrary, “iron spot” is not a typical effect associated with high-manganese native cobalt ores due to different raw materials and firing technology (Wood, 1999; Zhang, 2000).

Even though predecessors had some researches on “iron spot” of its chemical composition, micro-structure and formation mechanism (Chen, Guo, & Zhang, 1986; Wu, Li, & Guo, 1999; Zhang & Guo, 1989), it is difficult to form systematic classification and explanation on reflection principles and formation mechanism due to the lack of samples and single method. Thus, the objective of this study was to find the diversities of different spots for further understanding the coloration mechanism and formation mechanism of “iron spot” and possible firing technology. Moreover, it is also expected to provide new clues for identification of blue-and white-porcelain using “Sumali” manufactured in Guan kiln in early 15th century.

So, in this article, we analyzed five samples which excavated from Guan kiln by various microscopy methods to give comprehensive studies. The macro-features, microstructure and chemical composition of samples were observed and analyzed by 3D digital microscope, scanning electron microscopy energy dispersive X-ray analysis (SEM-EDS) and Electro-Probe Microanalyzer (EPMA) sequentially. Traditional optical microscopes can only obtain two-dimensional (2D) patterns while the materials we study are in three-dimension. This severely limits the ability in the microstructure and distribution characterization of the materials. Recently, with significant advancement of 3D digital imaging techniques, what is difficult to observe in the past can be well observed now and images of the samples in digital form make quantitative measurement of important microstructure parameters possible and accurate through 3D visualization (Hu et al., 2016).

By using various micro-analysis methods including 3D microscopy, SEM-EDS and EPMA, this article evaluates the micro-morphological features, microstructure and composition of “iron spots.” The different “iron spots” will be classified systematically to know its coloration and formation mechanism, then further the understanding of production technology and identification features of blue-and-white porcelain used Sumali-based cobalt-blue pigments in the early Ming Dynasty of ancient China.

2 | MATERIALS AND METHODS

As the provenances of cobalt pigment in Ming Hongwu (A.D. 1368-1398) and Xuande (A.D. 1426–1435) reign are under debate (Du & Su, 2008), it is more appropriate to select samples from Yongle (A.D. 1403–1424) period. Thus a series of blue-and-white porcelain sherds using imported cobalt ore made in Jingdezhen imperial kiln in Yongle reign (hereafter referred to as YL) provided by the Jingdezhen Ceramic Archaeological Institute was analyzed in this study. Five selected samples with characteristic spot were considered to be representative (Figure 1).

The samples were prepared by cutting with a silicon carbide (SiC) saw blade. First, samples were observed and photographed under Keyence digital microscope VHX-600 with low-magnification to see morphology features of spot like, size, shape, color and glossiness on glaze surface; then microscope VHX-600 with leading-edge RZ optical design zoom lenses with up to 5,000× magnification was used to obtain 3D composite images and to aid in the collection of the diameter of spots and their spacing, the crystallization width or crystallization depth, and so forth.

Crystallization conditions of “iron spot” and Quantitative elemental analysis of crystals were carried out using a scanning electron microscope (SEM, Hitachi S-3600 N, Japan) with an Genesis 2000XMS EDS system and X-max 80 mm^2 EDS detector. SEM was operated at an acceleration voltage of 20 kV with samples coated with a thin layer of gold (~20 nm) in vacuum to guarantee electrical conductivity. EDS was applied to initially analyze elements of crystallization, and non-crystallization areas of the samples were tested and analyzed comparatively to find further information. Then, the chemical composition of cross-section of the sample was tested by electron-probe microanalyzer (EPMA, EPMA-1720, Shimadzu, Japan) operating at 15 kV. The spot diameter of the electron beam was 1μm. Specimens for EPMA analysis were prepared in cross section view by resin embedding then mechanical polishing with fine grades of SiC paper and Micromesh polishing cloth. The specimen surface was coated with carbon film of approximately 20 nm in thickness for prevention of charge accumulation.

3 | RESULTS

From microscopic observation of characteristic spot on glaze surface and chemical composition analysis of EDS, results showed that

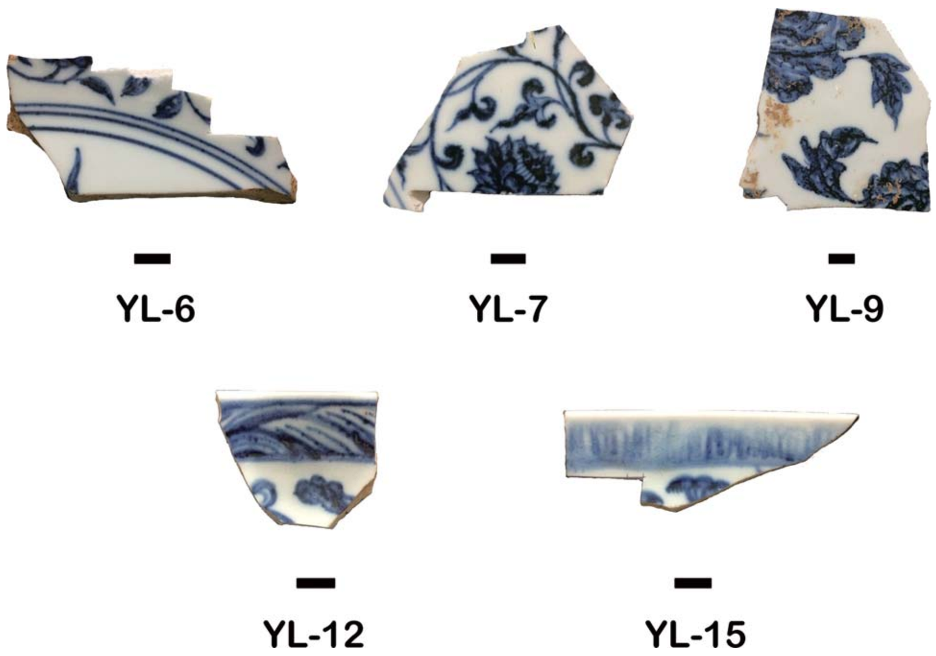

FIGURE 1 Photographs of samples (scale bar: 1 cm). [Color figure can be viewed at wileyonlinelibrary.com.]

characteristic spots are more than one condition. There are at least three different morphologies and distributions based on observation and testing. The different microstructures lead to different spot appearances of "Sumali." The three kinds of spot are depicted as follows:

3.1 | Unreflected "iron spot" without crystals

The first kind of spot, YL-9, an un-reflected spot which has long strips dark spots of 0.7-mm long without glisten phenomenon under the naked eyes or the microscope (Figure 2), shows no crystal in the region of spot under the SEM.

3.2 | Unreflected "iron spot" with crystals

The second is un-reflected "iron spot" with crystals typical as YL-7 and YL-12. "Iron spots" of YL-7 is circular with a diameter of 0.4 mm. Central grey area is about 0.2 mm in diameter, which reflected phenomenon cannot be observed under the microscope and naked eyes. Some

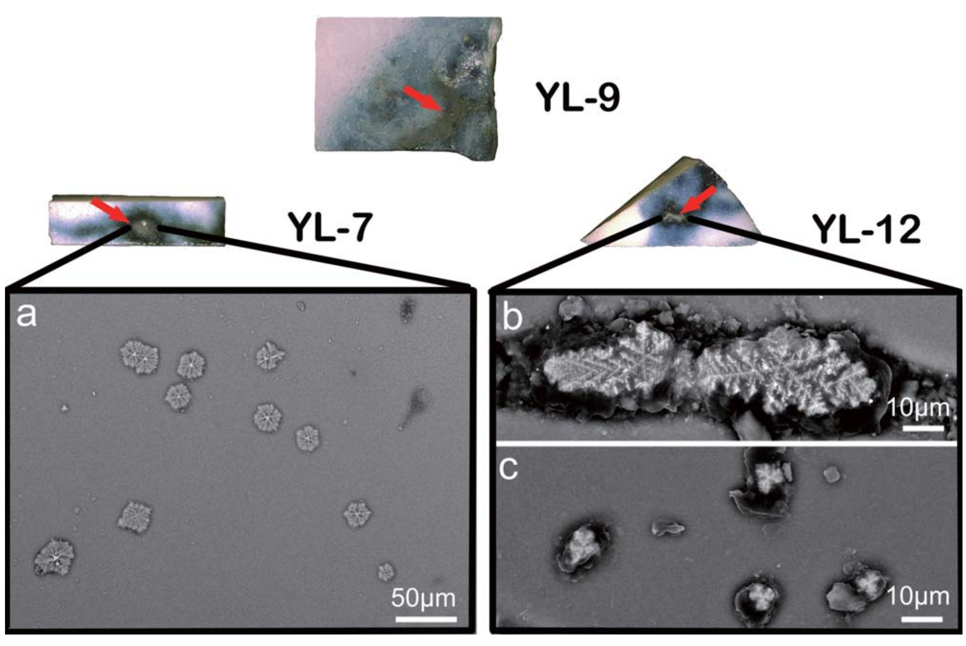

FIGURE 2 Microscopic images of YL-7, YL-9, YL-12 (The sampling zones are indicated with red arrows) and SEM images: (a) Snow-shaped crystals of YL-7 (×300) (b), (c) Snow-shaped crystals of YL-12 (×1k). [Color figure can be viewed at wileyonlinelibrary.com.]

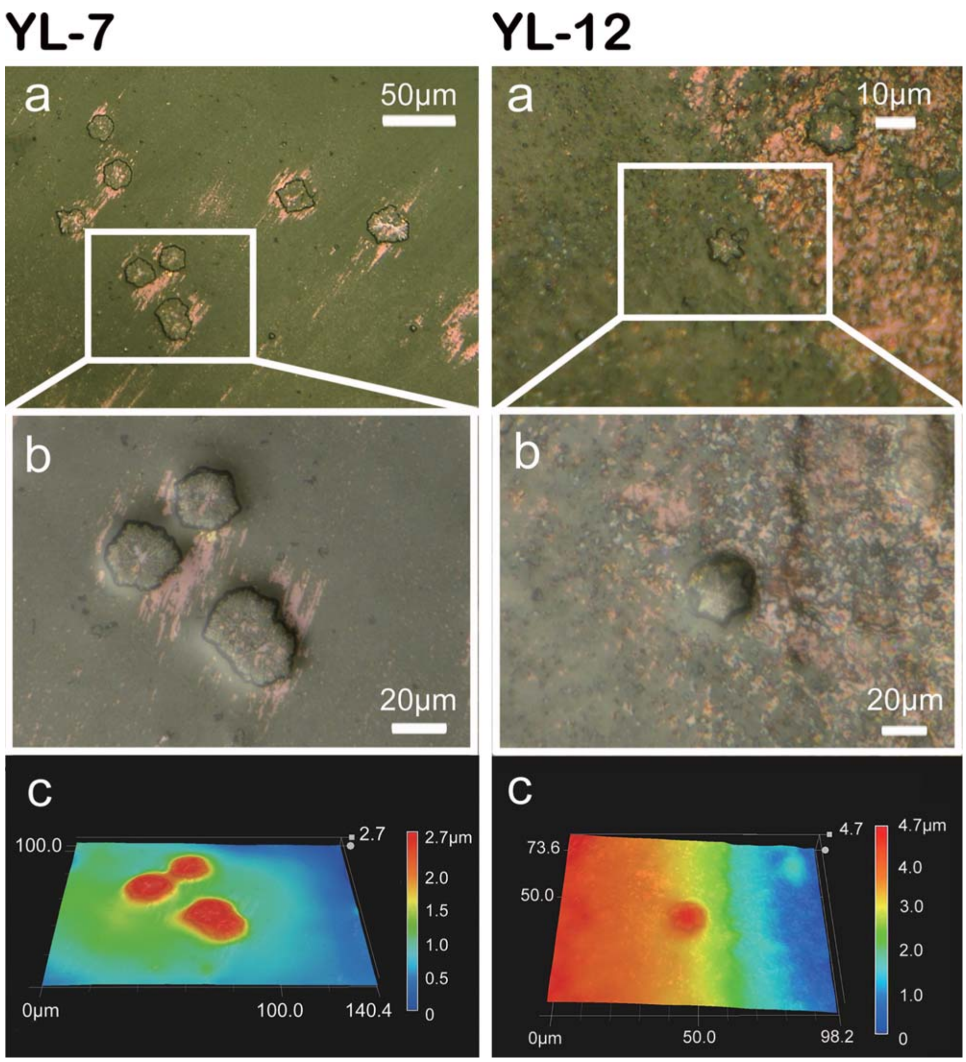

FIGURE 3 3D image of YL-7 and YL-12 obtained by digital microscope (From top to bottom: (a) Optical microscopic images; (b) 3D zooming image of crystals with sidelight; (c) Color contour map of crystals). [Color figure can be viewed at wileyonlinelibrary.com.]

irregularly dark spots appear about 0.5-mm long on the surface of YL-12, while the central grey region surrounded by dark blue is 0.3-mm long and shows no metallic luster under microscope (Figure 2).

By observing SEM photos, it is interesting to find more about micro-structure of this kind of "iron spot." Although no glitter is found by microscope in the YL-7, there are still few hexagonal snow-like crystals space which are about 20 μm in diameter scattered in the grey area of the glaze surface with large intergranular (Figure 2a). Comparatively, YL-12 is special where hexagonal snow-shaped crystals seem only growing in the voids but not on the surface of glaze (Figure 2b,c), and crystals appear in the grey area as well as YL-7.

The growth condition can be further understanding from 3D composite images (Figure 3). It is intuitive that crystals of YL-12 are higher than the surface of glaze under sidelight and, in the color contour maps, crystals are measured 1.53 μm on the surface of glaze. It is interesting that we found protruding crystal of YL-12 in 3D composite images is opposite to that observed in SEM images, which can be explained by the contrast of backscattered electron imaging in SEM depended on atomic number but not morphology. And crystal of YL12 is 1.87 μm on the surface of glaze eliminating interruptions of surface inclination.

3.3 | Reflected "iron spot" with crystals

The last one is the reflected "iron spot" with crystals, also called "tin light," like YL-6 and YL-15. There are platinum metallic spots about 3 mm in diameter on the glaze surface of YL-6. For purpose of comparing "iron spot" on and under the surface, part of glaze of "iron spots" was ground. It is found that silver precipitates only float on surface of the glaze and grow in directions parallel to the glaze surface. There are platinum metallic spots about 0.3 mm in diameter on the surface of YL-15. Like YL-6, in order to compare "iron spots" on and under the surface, part of glaze of "iron spots" was ground and silver precipitates are found only on surface of the glaze (Figure 4). Both reflected areas of YL-6 and YL-15 are within the zones of the dark blue.

SEM photos of YL-6, a large area covered with large branch-like crystals which is dense and interlaced, extends up to 100 μm (Figure 4a). Surrounding those, small branch-like or snow-shaped primary

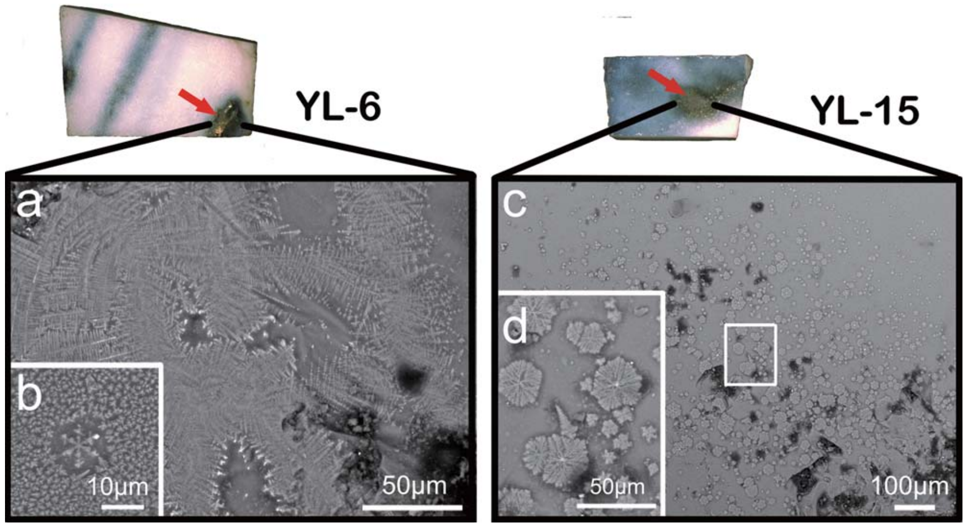

FIGURE 4 Microscopic images of YL-6, YL-15 (The sampling zones are indicated with red arrows) and SEM images: (a) Branch-like crystals of YL-6 (×500); (b) snowflake and snow-shaped primary crystals (×5k); (c) Snow-like crystals of YL-15 (×100); (d) A zoom in view of a selected region (×400). [Color figure can be viewed at wileyonlinelibrary.com.]

crystals are <2-μm long and intergranular space is <1-μm long. There are several full-grown hexagonal snow-like crystals among the primary crystals, which vary from 10 to 30 μm in diameter (Figure 4b). It is speculated that the large branch-like crystals could be overgrown snowflake crystals because of a gradually visible increase of crystals size. Observing ground areas, nothing can be found under glaze but bubbles. For YL-15, a large number of hexagonal snow-like crystals grow on the glaze surface. The size of crystals gradually grows up from white glaze area to "iron spot" area which ranges from 5 to 50 μm. Intergranular space is <50 μm (Figure 4c, d).

By observing 3D composite images of YL-6 and YL-15, all of snowflake crystals float on the surface of glaze, and crystal is 1.75 and 1.90 μm higher than the surface of glaze, respectively eliminating interruptions of surface inclination (Figure 5). In all, there is no exception that the crystals of spots are all growing on the surface of glaze in 3D images.

To verify that crystals only grow on the surface of the glaze, EPMA was used here to observe cross-section under crystallization area of YL-15. No crystals were found under the glaze except few bubbles and some unmelted pigment particles (Figure 6). In addition, minimal manganese was detected in the glaze, which conforms to characteristic of the imported cobalt pigment (Chen, Guo, & Zhang, 1978).

3.4 | Chemical composition

Crystallization (c) and non-crystallization (g) areas of samples were tested by EDS. Fe and Co contents of crystallization (c) and non-crystallization areas are shown in the Figure 7 below. Because crystals were not found in the YL-9, only contents of the non-crystallization area are listed in the figure. Comparing all samples, Fe and Co content of YL-9 is lowest. Only Fe, Co contents of crystallization areas are higher than that of non-crystallization areas without exception, which speculated that crystals of these samples are all the crystallization of Fe (Li, Luo, Li, Li, & Guo, 2008; Liu et al., 2012).

4 | DISCUSSION

According to the experimental results, samples are divided into three categories depending on reflective phenomenon and different crystal structures. The first category is un-reflected "iron spot" without crystals. Such as the YL-9, no crystal is observed on the glaze surface with SEM. The second category is un-reflected "iron spot" with crystals which has few snow-like crystals on the surface of glaze, including YL-7 and YL-12. Crystals of YL-7 and YL-12 are much less and smaller, which presents no luster in central grey regions of "iron spots" on the macro-level. The last category is reflected "iron spot" with crystals, such as YL-6 and YL-15 with a number of hexangular snow-like crystals on the surface of glaze.

By observing both crystallization condition above and below the glaze in 2D and 3D microscope images, the precondition of forming "tin light" is that all crystals are on the surface of glaze and always grow parallel to the surface no matter how rough it is, because crystals form a reflective layer whose reflectivity is much stronger than the glaze. YL-6 sample has a flat glaze surface on which large crystals line up tightly and small crystals distribute evenly, so "iron spot" show continuous silver shimmer with high brightness (Wang, 2012). Likewise, the YL-15 with same snow-shaped crystals also presents dark silver reflective light. But they are not as bright as YL-6 because crystals grow small and untight. So without a layer of uniform crystal, small intergranular space and flat glaze surface, it is unable to develop highly reflective surface on a large scale, even if single crystal is big and its structure is conducive to the reflection of light.

Above all, "tin light" must form a layer of crystals on the surface of glaze, but it is uncertain whether "iron spots" without gloss grow crystals. There are always some factors like size, shape, distribution and intergranular space which influence the reflectivity of crystals and the appearance of "iron spot" to varying degrees. Moreover, the snowflake crystals whose branches grow along parallelly the

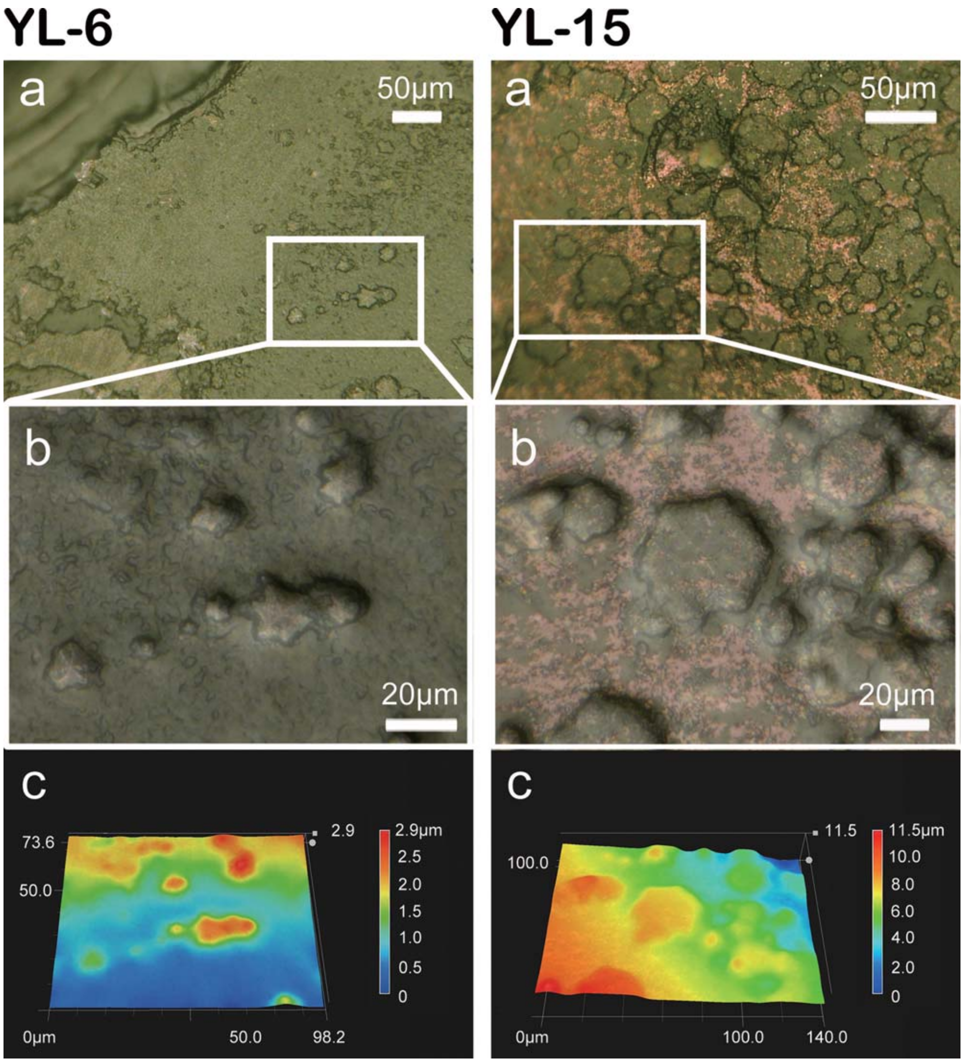

FIGURE 5 3D image of YL-6 and YL-15 obtained by digital microscope (From top to bottom: (a) Optical microscopic images; (b) 3D zooming image of crystals with sidelight; (c) Color contour map of crystals). [Color figure can be viewed at wileyonlinelibrary.com.]

surface of glaze should belong to the crystallization of iron according to the chemical composition and micro-morphology (Bharathi et al., 2010).

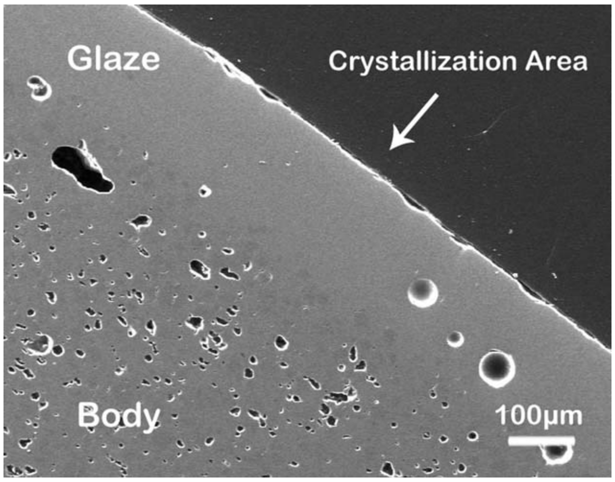

FIGURE 6 Cross-section of YL-15

After knowing more about morphology, coloration mechanism of "iron spots" is discussed here. Crystals are only found within the areas of "iron spot" and never beyond the range. It infers that cobalt-blue

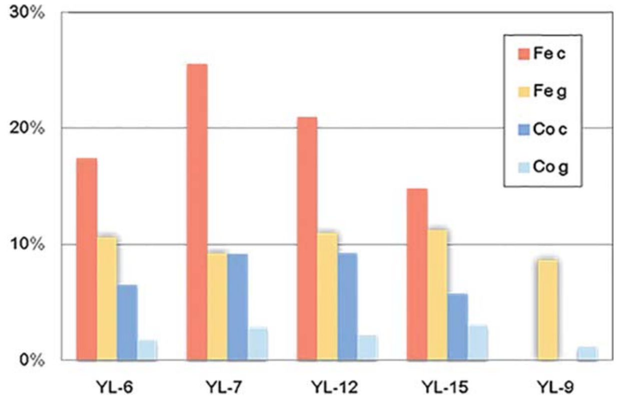

FIGURE 7 Comparison histogram of Fe, Co concentration in crystallization (c) and non-crystallization (g) areas of samples. [Color figure can be viewed at wileyonlinelibrary.com.]

pigment, not glaze, provides the crystallizing agent to form crystals. Only when crystallizing agent is saturated in the molten glaze, crystals could be precipitated. "Sumali" in the early times was always ground inadequately and mixed with coarse particles so that some residual crystal nucleuses were retained to participate in the formation of more crystals. However, it seems contradictory that crystals of "iron spots" only grow on the surface of glaze and blue-and –white porcelain belongs to under-glaze ceramic. It can be explained by the diffusion of the cobalt-blue pigment. The craftsmanship of blue-and-white porcelain in the early Ming Dynasty is immature. To increase yield and facilitate coloration, a large amount of flux was added into pigment. In addition, iron in the cobalt-blue pigment also has fluxing effect to reduce melting point. Cobalt pigment used in early Ming Dynasty exactly enrich in iron, which promotes diffusion of pigment from coloring district to non-coloring district and to the surface of glaze (Zhang, 2000). Once it reaches a right condition of nucleation, it is easy to generate crystal nucleuses on the surface (Zhang, 2009). Finally, under certain conditions of temperature and supersaturation, crystal nucleuses begin to grow into crystals on the surface of glaze because oxygen concentration in the glaze is lower and have a shallower distribution below the surface, which limits the crystals growing to a much shallower region (Dejoie et al., 2014). According to Tammann crystallization theory, different holding time and degree of supercooling both impact on the growth of crystals, so firing temperature and holding time also have an effect on the shape and the distribution of crystals (Schultz, 1975).

5 | CONCLUSIONS

Characteristic spot of blue-and-white porcelain using "Sumali" in early Ming Dynasty have different morphological characteristics. In this article, it can be divided into three groups: un-reflected "iron spot" without crystals, un-reflected "iron spots" with crystals and reflected "iron spots" with crystals or "tin light". "Iron spots" tend to grow dendritic or snow-shaped crystals of the gathering place of cobalt-blue pigment and crystals only grow on and parallel to the surface of glaze, which is the main reason why iron spot show metallic light. It also indicates that firing temperature, holding time and local oxygen pressure result in varieties of size, shape, distribution of crystals of iron spot. To sum up, by the means of analyzing micro-morphology and the components information of "iron spots," the morphology of "iron spots" is closely tied to the raw material, firing temperature, atmosphere and multiple factors. It may offer references to further researches on the coloration mechanism of blue-and-white porcelain in early Ming Dynasty and provides a potential possibility to identify blue-and-white porcelain using imported cobalt pigment made by imperial kiln in 14th to 15th centuries.

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