

# THE CHEMICAL COMPOSITION AND PRODUCTION AREA OF EARLY WESTERN ZHOU PROTO-PORCELAIN UNEARTHED FROM YEJIASHAN CEMETERY, SUIZHOU, CHINA\*

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*The bodies and glazes of 27 early Western Zhou proto-porcelain samples from Yejiashan cemetery, Hubei Province, were analysed using LA-ICP–AES, SEM, XRD, a thermal expansion instrument and other analytical methods. The results indicated that the bodies of all samples were characterized by high silicon and low aluminium, and were made with porcelain stone raw materials found in the south of China. The glazes are typical of high-temperature calcium glazes of the CaO (MgO) – K<sub>2</sub>O (Na<sub>2</sub>O) – Al<sub>2</sub>O<sub>3</sub> – SiO<sub>2</sub> series, with relatively high Mn and P content, which was probably caused by the addition of plant ashes. The physical properties and phase compositions of Yejiashan proto-porcelain show that firing processes were still in the early stages of development in ancient China. Multivariate statistical analyses indicated that Yejiashan proto-porcelain might have come from the Deqing area, in Zhejiang Province. These results provide new archaeological evidence for research on issues related to material flow in the Western Zhou dynasty.*

**KEYWORDS:** PROTO-PORCELAIN, YEJIASHAN CEMETERY, CHEMICAL ANALYSIS, HIGH-TEMPERATURE CALCIUM GLAZE, PROVENANCE ANALYSIS

## INTRODUCTION

Yejiashan cemetery, located in Suizhou, Hubei, is a high-grade noble burial site dating to the early Western Zhou dynasty (Fig. S1). With its unique funerary system, clear burial layout, and the great quantity and variety of buried artefacts, the site has attracted extensive scholarly interest. Artefacts unearthed at Yejiashan cemetery include many bronze vessels with Chinese inscriptions such as *Zeng* (曾) and *Zeng Hou* (曾侯; Marquis of Zeng), as well as more than 70 pieces of precious proto-porcelains, such as zuns, tall covered jars, vases, drinking vessels, urns, guis, wide covered jars, and tall column-footed discs (Institute of Cultural Relics and Archaeology of Hubei Province *et al.* 2011a,b, 2012, 2013a,b). Most of the proto-porcelains are highly ornamented, with wavy, concave and chequered patterns, and decorated with cloud and lightning patterns, and all are glazed in celadon or dark green. Yejiashan proto-porcelain from the early Western Zhou period is distinctly identifiable in terms of its exact burial time and its clear combination and coexistence relationship, making it highly valuable for researchers. It provides important material specimens for studying the craft and production area of early Western Zhou proto-porcelain from the Jiangnan Plain in Hubei.

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Yejiashan proto-porcelains have been excavated from large- and medium-sized tombs; they have not been found at minor burial sites. Huang (2014, 48–52) and Guo and Hu (2014, 220–34) note that these specimens embody the syntagmatic relations of the early Western Zhou proto-porcelains, and are suggestive of whether being buried with proto-porcelain was related to the identity and status of the people entombed. Moreover, there are signs of intentional pre-burial damage to certain pieces, possibly reflecting specific burial customs. Huang (2014, 48–52) and Guo and Hu (2014, 220–34) further note that there are unique markers of proto-porcelain production (carved ornamentation, S-shaped appliqué, circumcrescent cake-like ornamentation etc.) that show similarities with specimens found in the Jiangsu and Zhejiang regions.

Previous research has noted that Yejiashan proto-porcelain is ‘great in quantity and variety, and its origin is a subject worthy of study’ (Li *et al.* 2013), and that ‘further consideration is needed to determine whether it was produced locally or transported from faraway areas’ (Duan and Chen 2014). To that end, we conducted sampling analysis on proto-porcelains unearthed from Yejiashan cemetery. Moreover, through a comparative study of proto-porcelains from other areas, we aimed to interpret technological and cultural connotations in relation to Yejiashan proto-porcelain processes and production areas.

## SAMPLES AND METHODS

### *Samples*

Twenty-seven porcelain specimens excavated from seven tombs in the Western Zhou cemetery of Yejiashan, Suizhou County, Hubei Province, were selected for this study. Twenty were complete artefacts, and seven were collected from the excavation sites (for examples, see Fig. 1). According to archaeological reports, all samples date to the early Western Zhou dynasty and were unearthed from M27, M28, M50, M51, M65, M111 and M126 (Table S1).

### *Methodology*

The chemical compositions of all the samples’ bodies and glazes were quantitatively analysed using laser ablation inductively coupled plasma–atomic emission spectrometry (LA-ICP–AES). A LEEMAN-Prodigy ICP–AES with a NEW-WAVE laser ablation system was used for this process. Nine elements were identified in the bodies (Al, Fe, Mg, Ca, Na, K, Ti and Mn). The data were quantitatively controlled using the Corning B and NIST 610 glass standards. The analytical details and calibration method are presented elsewhere (Cui and Zhang 2007). SiO<sub>2</sub> data were calculated by subtracting the sum of all other elements in weight per cent oxides from 100%. For the major and minor elements, most relative standard deviations were less than 1%. The data recoveries for the major elements were commonly 100 ± 5%. Table S2 presents the results for some major and minor elements with three repeat analyses of the NIST 610 and Corning B glass standards. The body tests used a sample cross-sectional area, and the glaze tests used an area with a thick glaze layer. Samples were swabbed with alcohol before the tests, and each sample was tested two or three times and then averaged. The results are shown in Tables S3 and S4.

Full cross-sections of the vessel fragments (including both internal and external surfaces) were mounted as polished blocks for optical microscope and scanning electron microprobe analysis to determine the body and glaze compositions, pore distribution, glaze-layer thickness, glaze craze and mineral crystals. The combined layers of body and glaze of typical proto-porcelain samples were observed and analysed using a Hitachi TM3030 tabletop scanning electron microscope



Figure 1 Examples of the proto-porcelains unearthed from Yejiashan cemetery: (a) three-line tall covered jar M65:23; (b) porcelain urn M65:19; (c) porcelain urn M27:82; (d) porcelain wide covered jar M27:99; (e) porcelain urn M28:243; (f) porcelain zun M28:242. [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

(Fig. 2 and Fig. S3) and a Keyence HDS-5800 extended depth of field 3D digital microscope (Fig. 3). The phase compositions of typical proto-porcelain samples were analysed using a Rigaku D/max 2550 V X-ray diffractometer (Table 1). The bibulous rate was measured using the water-boiling method (Table 1). The thermal expansion curves of some samples were measured using a Bahr Thermo Analyse DIL 806 optical dilatometer, and the firing temperatures were estimated (Tite 1969) (Table 1).

## RESULTS

### *Ceramic bodies*

There were slight variations of most elements in the major and minor chemical compositions of proto-porcelain bodies from different tombs, but the scope for content change of some elements is greater, given the significant variation both in terms of starting materials, the expected refractoriness of firing and the glaze fit. The  $\text{Al}_2\text{O}_3$  content ranged from 13.50% to 24.90%, and the  $\text{SiO}_2$  content ranged from 67.79% to 80.51%. Sample M111:160 contained a high  $\text{R}_x\text{O}_y$  content of 12.99%, while the  $\text{R}_x\text{O}_y$  content for other samples ranged from 4.83% to 8.16%. The  $\text{R}_x\text{O}_y$  content was mainly composed of  $\text{K}_2\text{O}$ ,  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$ . The  $\text{K}_2\text{O}$  content was between 1.62% and 4.55%, the  $\text{Fe}_2\text{O}_3$  content was between 0.96% and 2.36%, and the  $\text{TiO}_2$  content was between 0.58% and 1.21%. Such proximity in composition suggests that these proto-porcelains have experienced exactly the similar degree of modification by raw material processing. Eliminating the results for M111:160, the values for which seemed to be outliers,

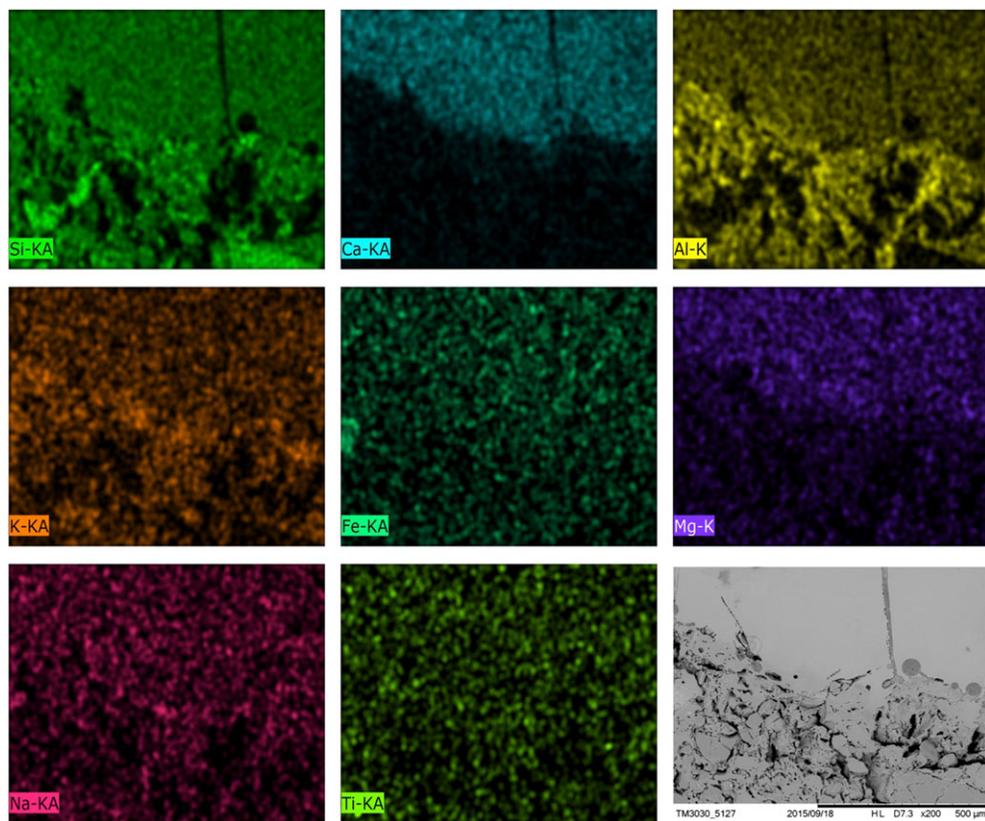


Figure 2 The elemental map distribution of the interaction zones between the body and glaze of sample M65:19. [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

the molar ratio of  $R_xO_y/Al_2O_3$  in the other samples was between 0.25 and 0.56, and the molar ratio of  $SiO_2/Al_2O_3$  was between 4.63 and 10.14. Colouring elements such as  $Fe_2O_3$ ,  $TiO_2$  and  $MnO_2$  were low, with bodies being mostly grey or beige, and only a few samples had high  $Fe_2O_3$  and  $TiO_2$  contents, making for darker grey bodies.

### Glazes

Chemical composition data were obtained for 23 proto-porcelain glazes (four samples were excluded due to thin glaze layers). The results are shown in Table S4. The  $Al_2O_3$  content of the glazes was between 11.53% and 17.11%, and the  $SiO_2$  content ranged from 40.25% to 64.32%. The  $Al_2O_3$  and  $SiO_2$  content of the glazes was significantly lower than in the bodies, while the CaO content was significantly higher (10.69–25.43%). The  $Fe_2O_3$  content (the main colouring element of the glazes) was between 1.38% and 4.46%; thus, in a reducing atmosphere, the glaze colours were mostly green or dark green. The MgO and  $K_2O$  contents were also higher (MgO 2.18–7.47%;  $K_2O$  2.39–5.49%). The  $P_2O_5$  content was between 0.5% and 3.69%, significantly higher than in the bodies. Overall, the proto-porcelains were high in body fluxes. The  $R_xO_y$  content was between 19.5% and 41.48%, the molar ratio of  $R_xO_y/Al_2O_3$  was between 1.92 and 4.95, and the molar ratio of  $SiO_2/Al_2O_3$  was between 4.53 and 9.48.

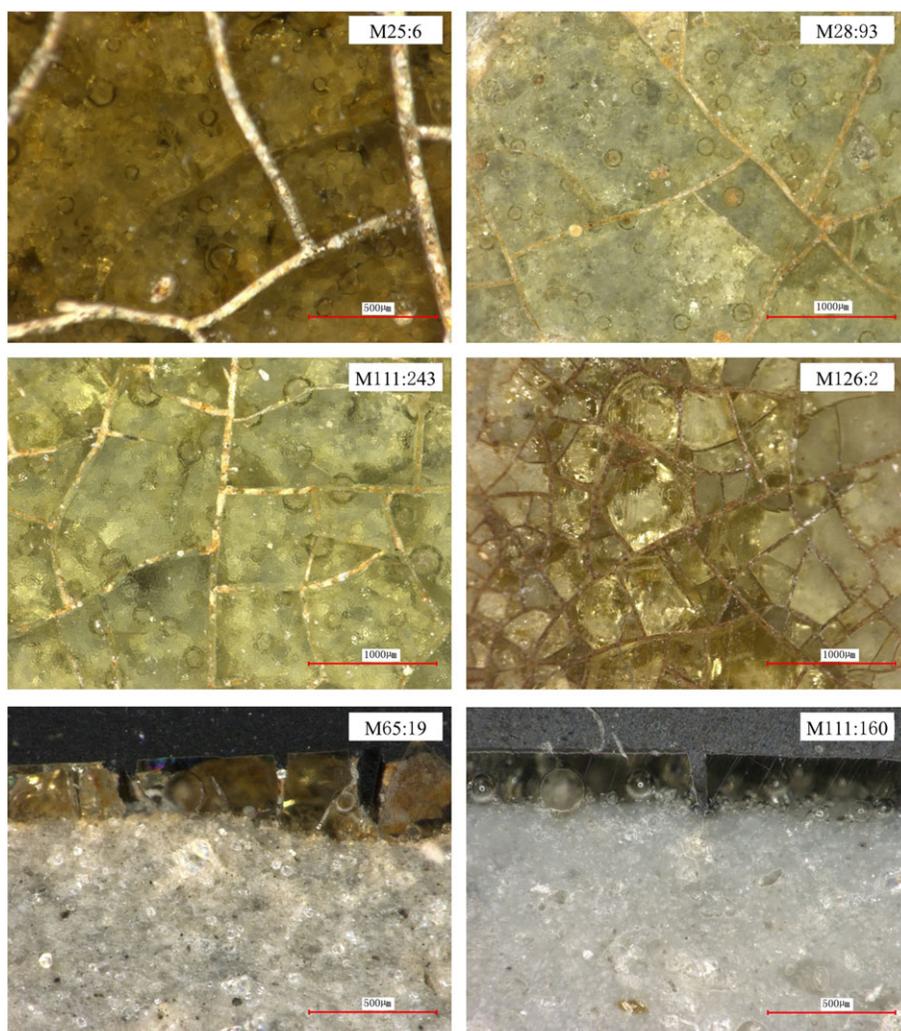


Figure 3 Microscopic images of typical bubbles and cracks found on proto-porcelain glazes from Yejiashan cemetery. [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

### *Physical properties and phase composition*

The samples were tested for phase composition. Firing temperature tests and microstructure observation were performed, while related physical properties such as porosity were tested as well. The results are shown in Table 1.

## DISCUSSION

### *The body-making technology*

The chemical compositions of the proto-porcelain bodies from Yejiashan reflect the characteristics of body-making processes in the initial stages of porcelain development (Wu

Table 1 The physical properties and phase compositions of the proto-porcelain samples from Yejiashan cemetery

Sample	Number	Bibulous rate (%)	Firing temperature (°C)	Mineral composition
Porcelain jar	M25:6	2.11	1116	$\alpha$ -quartz, a little cristobalite and mullite
Porcelain jar	M50:7	2.08		$\alpha$ -quartz, a little cristobalite and mullite
Porcelain jar	M51:5	2.01		$\alpha$ -quartz, a little cristobalite and mullite
Porcelain urn	M65:19	2.04	1220	$\alpha$ -quartz, some cristobalite and a little mullite
Three-line tall covered jar	M65:23	2.05	1285	$\alpha$ -quartz, some cristobalite and mullite
Porcelain jar	M111:160	2.03	1164	$\alpha$ -quartz, a little cristobalite and mullite
Porcelain jar	M111:165	2.03		$\alpha$ -quartz and a little mullite
Porcelain jar	M111:166	2.03	1212	$\alpha$ -quartz, some cristobalite and a little mullite
Porcelain zun	M28:242	2.06	1189	$\alpha$ -quartz, a little cristobalite and mullite
Porcelain urn	M28:243	2.05	1081	$\alpha$ -quartz, a little cristobalite and mullite
Porcelain urn	M27:15	2.09	1143	$\alpha$ -quartz, some cristobalite and a little mullite
Porcelain urn	M27:92	2.04		$\alpha$ -quartz, a little cristobalite and mullite
Porcelain tall column-footed disc	M126:2	2.05		$\alpha$ -quartz and a little mullite
Porcelain zun	M126:3	2.02		$\alpha$ -quartz, some cristobalite and a little mullite

Jun *et al.* 2011a). In fact, the  $\text{Al}_2\text{O}_3$  content of proto-porcelain bodies from areas such as Shaanxi, Shanxi, Henan, Hebei, Jiangxi and Zhejiang is generally about 15%, while the  $\text{SiO}_2$  content is between 70% and 80% (Pollard and Hatcher 1986, 1994; Li 1998, 87–91). On average, the  $\text{Al}_2\text{O}_3$  content of proto-porcelain bodies from Yejiashan is 17.36%, and the  $\text{SiO}_2$  content is 76.09%. The results fall within the variation ranges of the main elemental contents of proto-porcelain bodies found in most parts of China. Furthermore, based on the proto-porcelain body chemical formula (i.e., the molecular molar ratio of the oxide content), it can be determined that the  $\text{R}_x\text{O}_y/\text{Al}_2\text{O}_3$  ratio in the proto-porcelain samples from Yejiashan cemetery is 0.25–0.56, and the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio is 4.63–10.14. These generally maintain the same ranges of variation for proto-porcelains from other parts of China (0.5–0.7 for  $\text{R}_x\text{O}_y/\text{Al}_2\text{O}_3$  and 5–11 for  $\text{SiO}_2/\text{Al}_2\text{O}_3$ ; Wu Jun *et al.* 2009). The chemical compositions of proto-porcelain bodies are similar and largely depend on the materials that are used (Li 1998, 87–93). The chemical composition of the Yejiashan proto-porcelain bodies is characterized by high silicon and low aluminium, similar to the porcelain stone found in the south of China. Zhejiang is the birthplace of early green-glazed porcelain, while white-glazed porcelain came from Jiangxi, and both regions are rich in minerals such as porcelain stone (Guo 1987; Li 1998, 86–104). This high-silicon/low-aluminium feature of the proto-porcelain bodies suggests that the Yejiashan proto-porcelain raw materials might have been similar to clayey raw materials made of porcelain stone.

In terms of ceramic development, the changes in chemical composition from pottery to porcelain in most areas of southern China were from high- $\text{SiO}_2$ , low- $\text{Al}_2\text{O}_3$  and high-solvent  $\text{R}_x\text{O}_y$  (mainly  $\text{Fe}_2\text{O}_3$ ) to low- $\text{SiO}_2$ , high- $\text{Al}_2\text{O}_3$  and low-solvent  $\text{R}_x\text{O}_y$  (also mainly  $\text{Fe}_2\text{O}_3$ ) (Li 1998, 77). Such materials can withstand higher firing temperatures. The solvent  $\text{R}_x\text{O}_y$  content in the proto-porcelain bodies from Yejiashan range from 4.83% to 8.16% (Table S3 and Fig. S2)—mainly because of the  $\text{K}_2\text{O}$ ,  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  contents—but the overall contents of  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  are low, averaging 1.41% and 0.86%, respectively. At that time, the lower-iron and lower-titanium raw materials were gradually selected by preference. This content level and

the relative stability of the content suggest that potters intentionally selected materials with low iron and titanium to make Yejiashan proto-porcelain. A low iron and low titanium content tends to make the body whiter in colour after firing, which is a landmark change in the success of proto-porcelain firing. Minor fluctuations exist in the ratios of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  and  $\text{R}_x\text{O}_y/\text{Al}_2\text{O}_3$  (Fig. S2) in some proto-porcelain bodies, suggesting that proto-porcelain production processes were still at an early stage of development and that the processing of the raw materials was not yet refined.

### *The glaze-making technique*

As the earliest form of enamel, proto-porcelain glaze provides a historical record of the glaze-forming processes (Li 1978). It has been the subject of extensive research, and significant hypotheses have been proposed regarding the origin of the proto-porcelain glazes. The general consensus is that the glazes originated from fusible clay, plant ash, 'pottery coating', 'kiln sweat' and so on (Luo *et al.* 1992, 1995; Wood 1999). The hypotheses propose the following. (1) When the kiln was built, raw materials with low refractoriness were used, and the parts directly exposed to fire in the inner wall of the kiln melted. When low-melting materials were accidentally used for making proto-porcelain bodies, due to excessive wind or firewood, the surfaces of products near the fire melted into the glassy material in the case of fierce firepower (Chen and Zhang 2002). (2) It originated from plant ash, or ash mixed with the right amount of clay (Wood 2009; Wu Jun *et al.* 2011b), mainly because high-temperature glazes from the Shang and Zhou dynasties have high CaO,  $\text{P}_2\text{O}_5$  and  $\text{MnO}_2$  contents, similar to the chemical composition of plant ash. Ash was easy to obtain, and since the ancient potters used firewood as fuel, they were probably inspired by the fact that the ash layer transformed into a glassy substance at high temperatures. (3) Proto-porcelain glazes were formed by the vitrification of 'pottery coating' (Li 1983) and black clay glaze pottery (Li *et al.* 1987); the raw materials for 'pottery coating' were clays similar to the raw materials used for pottery bodies, but their particle size was much smaller. (4) It was inspired by kiln wall vitrification. One option is a natural glaze-forming process, which Zhang called 'fly-ash' (Zhang 1986, 41); this process will form a recognizable lime-rich ash glaze. The other is the volatilization of potassium, over the lifetime of the kiln; with countless successive firings, this leads to the build-up of high levels of potash on the surface of the kiln walls, leading to relatively thick glassy layers (Yin *et al.* 2011).

The major and minor elemental compositions of the proto-porcelain glazes from Yejiashan cemetery (see Table S4) place them in the class of high-temperature calcareous glazes found in ancient China. The CaO levels in the glazes have a mean of 17.12%, which is significantly higher than the content of alkali metal oxide in the glaze. The MgO and  $\text{K}_2\text{O}$  contents are also high (averaging 3.38% and 3.51%, respectively), while the  $\text{Al}_2\text{O}_3$  content is relatively low (mean 14.58%). Thus, it is clear that the Yejiashan proto-porcelain glaze is a high-calcium glaze mainly taking CaO, MgO and  $\text{K}_2\text{O}$  as the flux (Fig. 2). The proto-porcelain glazes are high-temperature glazes that can be broadly divided into two categories. The first contains a high flux content—in particular, higher levels of CaO—while the  $\text{Fe}_2\text{O}_3$  content is generally low. It is mainly a cyan-greenish high-calcium glaze with Ca, K, Mg and so on as the flux, with high transparency. The second type has a low CaO content, and its main co-solvents are  $\text{Fe}_2\text{O}_3$  and  $\text{K}_2\text{O}$ ; this glaze is a deep caramel or black, with high opacity (Li 1998, 86–104). In terms of aesthetic effects, each glaze has its own characteristics and functions. The proto-porcelain glazes from Yejiashan cemetery are mostly green or dark green; a few are green with some yellow, caused by the incomplete reduction of  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$ . It is clear, therefore, that the proto-porcelain glazes from Yejiashan are probably high-temperature glazes of the

CaO(MgO)–K<sub>2</sub>O(Na<sub>2</sub>O)–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> series, typical of high-temperature calcium glazes found in ancient China (Chen and Zhang 2002; Yin *et al.* 2011).

Overall, the content of the flux R<sub>x</sub>O<sub>y</sub> is high in the Yejiashan proto-porcelain glazes, averaging 29.33%. In the firing process, the glaze is sufficiently melted with a high degree of vitrification and glossiness, and running occurs in the part of the thicker glaze layer. The proto-porcelain glazes from Yejiashan take CaO (MgO) as the main flux, with relatively high Mn and P contents. The sum of both elements in most samples is between 1% and 3%, with a maximum of 4.32%. The use of high-calcium minerals such as limestone does not improve the Mn and P contents (which are usually less than 0.1%) in porcelain glaze (Misra *et al.* 1993; Wu Jun *et al.* 2011b). Since ash was easy to obtain in southern China, the high calcium level in the sample glaze was probably produced by ash, which contains high Mn and P contents (Zhang 1986; Rehren and Yin 2012). It is worth noting that the glaze in several of the Yejiashan artefacts is high in Fe<sub>2</sub>O<sub>3</sub> content. For example, the Fe<sub>2</sub>O<sub>3</sub> content of sample M27:92 is 4.46%; since Fe<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> are the main colouring agents, the glaze colour is darker on these proto-porcelains with higher iron content.

### *The firing process*

The phase analysis of X-ray diffraction and the analysis of firing temperatures for some representative Yejiashan samples (Table 1) found firing temperatures ranging from 1081°C to 1285°C, which were measured precisely using a Bahr Thermo Analyse DIL 806 optical dilatometer. In addition to  $\alpha$ -quartz, high-temperature cristobalite and mullite, which are important typical characteristics of porcelain or porcelain bodies in their original form, are evident in the phase compositions.

The proto-porcelain jar M25:6 has a body firing condition of 1116°C, its phase composition is mainly  $\alpha$ -quartz, and there are small amounts of cristobalite and mullite. The three-line tall covered jar M65:23 has a body firing condition of 1285°C, its phase composition is mainly  $\alpha$ -quartz, and there is some cristobalite and mullite. The presence of high-temperature cristobalite and mullite indicates that the firing temperatures of these pieces are high; however, there are some fluctuations in the firing temperatures of the Yejiashan proto-porcelains. This is possibly because the proto-porcelain pieces were placed in different locations in the kiln, where temperatures might not have been constant.

Nevertheless, the rate of water absorption in the analysed samples with numerous pores is more than 2%, indicating that there was no sintering and that the body was not dense enough except for the individual samples that have a higher firing temperature. Therefore, there was still a technological gap in comparison with mature porcelain (Li 1978). In appearance, the glaze layers are thin and have poor uniformity. The glazes are severely crazed, with bubbles of varying sizes, and the body and the glaze are poorly combined, with severe shedding (Figs 2 and 3 and Fig. S3). The cracked glazes indicate that the porcelains should be single-fired.

This reflects the primitiveness of the glazing and firing techniques in the early Western Zhou. In terms of mineral composition, although a certain amount of mullite reformed within the proto-porcelain body due to the high firing temperatures of certain samples, there is relatively less glass-phase material (Fig. S3). This is one reason why the glaze layer is easily peeled off, due to the low degree of integration between body and glaze.

Generally speaking, the difference between porcelain and pottery is that the body surface of porcelain is solid and compact—mostly white or greyish white in colour—and the cross-section has a glassy lustre. In terms of performance, porcelain is strong, with very low porosity and water

absorption. There are numerous glassy states and a certain amount of mullite in its body, and it also has a layer of glass glaze that has a certain thickness and good adhesion (Li 1978). We can say, therefore, that the proto-porcelain unearthed from Yejiashan cemetery and its firing processes were still in the early stages of porcelain development in ancient China.

### *Provenance analysis*

Issues of proto-porcelain provenance are of great concern to scholars and regional differences have arisen, such as a 'southern version' (Li 1998; Chen *et al.* 1999, 2003), a 'northern version' (An 1960, 1978) and a 'multicenter version' (Zhu and Wang 2004). The source location of the proto-porcelain unearthed at Yejiashan cemetery has scholarly significance. First, these pieces belong to an earlier era—the early Western Zhou dynasty. Second, their body-glaze formula, decoration, moulding and other processes have distinctive features (Huang 2014). Third, they were unearthed in tombs with clear combinatory relations and comprise the largest quantity of Western Zhou proto-porcelain found in the Hubei region. Lastly, as yet, no samples have been found in the archaeological discoveries of kiln sites for proto-porcelain in Hubei Province. Hence, analysing the sources of these proto-porcelain pieces is of great significance for the scientific understanding of cultural relics unearthed from Yejiashan cemetery, and it provides new information for research on material flow during the Western Zhou dynasty.

Process analyses have shown that the proto-porcelain from Yejiashan cemetery should have come from the same area, but the specific location is in question. For comparison, we selected proto-porcelain from four southern kiln sites (Huoshaoshan, Zhejiang; Kuzhuwu, Zhejiang; Huangmeishan, Zhejiang; and Yingtan Jiaoshan, Jiangxi) for multivariate statistical analysis together with proto-porcelain from Yejiashan cemetery. The proto-porcelains from these four kiln sites are well represented. The Jiaoshan site in Yingtan is considered the earliest large-scale kiln factory (Li *et al.* 1990) found in China thus far, specializing in firing stamped hard pottery and proto-porcelain. The Huangmeishan site in Huzhou is the oldest proto-porcelain kiln site found in Zhejiang Province (Pan 1999). Meanwhile, the Huoshaoshan site in Deqin, Zhejiang Province, was the first kiln site formally excavated in China (Li 2008; Zheng *et al.* 2008). The major and minor chemical composition data (Table 2) for the proto-porcelain from these four kiln sites were measured in the same way as the Yejiashan proto-porcelain was measured, which avoids the systematic errors caused by testing with different instruments. It should be noted that the elementary compositions of the proto-porcelain from the four kiln sites were measured using the acid dissolution method, while the Yejiashan proto-porcelain was measured using the laser ablation method. However, the systematic errors between these two methods are relatively small, and the statistical analyses of those data can be merged (Cui and Zhang 2007).

Since the test data are multidimensional, it is difficult to describe or compare the similarities and differences in an intuitive graphical way. In this study, factor analysis was used for data processing in the multivariate statistical analysis. This method uses several minimum integrated factors, such as F1, F2 and F3 (where  $F_i = x_1^i A + x_2^i B + \dots$ ,  $A, B, \dots$  are element contents, and  $x_1^i, x_2^i \dots$  are positive and negative weighting factors calculated by the appropriate procedure) to extract as much information as possible characterizing the research object and achieve dimensionality reduction. This reflects not only the relationships between the sampling points (i.e., that neighbouring sampling points have similar properties and belong to the same class) but also the relationships between the variables (elements) and the samples (i.e., that sampling points of the same type will be characterized by adjacent variables) (Luo 1997). SPSS 18 was used for the analysis, and the results are shown in Tables S5 and S6.

Table 2 The means and standard deviations (in wt%) of the major and minor elements of the proto-porcelain samples excavated from various sites

Site	Number		Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>
Yejiashan, Hubei	27	Mean	0.23	0.56	17.36	76.09	2.71	0.45	0.86	1.41
		SD	0.14	0.13	2.87	3.38	0.68	0.88	0.15	0.31
Huoshaoshan, Zhejiang	13	Mean	0.13	0.53	15.50	78.23	1.97	0.24	0.59	2.40
		SD	0.05	0.08	1.51	1.89	0.37	0.06	0.06	0.40
Kuzhuwu, Zhejiang	12	Mean	0.18	0.58	15.90	78.07	1.57	0.26	0.78	2.45
		SD	0.06	0.10	1.82	2.31	0.40	0.07	0.08	0.28
Huangmeishan, Zhejiang	8	Mean	0.34	0.54	16.59	76.31	2.21	0.32	0.69	2.69
		SD	0.05	0.05	1.08	1.01	0.11	0.04	0.02	0.35
Yingtian Jiaoshan, Jiangxi	10	Mean	0.08	0.84	18.32	73.49	1.39	0.32	0.74	4.56
		SD	0.03	0.13	1.77	2.57	0.28	0.07	0.05	0.88

The Kaiser–Meyer–Olkin (KMO) value for the sample PCA analysis of proto-porcelain from Yejiashan and the four kiln sites was 0.602, indicating that this set of data was suitable for factor analysis. Factors one and two ( $F_1$ ,  $F_2$ ) can explain the total variance of PCA analysis at 38.29% and 26.76%, respectively; the sum of both is 65.06%, thus retaining most of the information. Therefore, analysing these samples using  $F_1$  and  $F_2$  is reasonable. Figure 4 is a scatter diagram with  $F_1$  and  $F_2$  as coordinates.

As shown in Figure 4, the major and minor elemental factor analysis scatter points for the proto-porcelain bodies unearthed from Yejiashan cemetery and the four kiln sites fall within the three ranges. This shows that the Yejiashan proto-porcelain does not overlap with that from Jiaoshan, while there are some similarities with the proto-porcelain from Huoshaoshan, indicating that the Yejiashan proto-porcelain might have come from the kiln factory area of

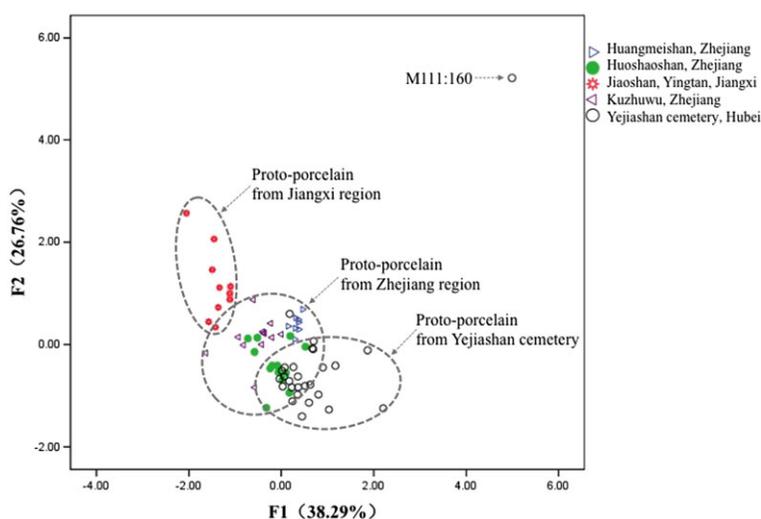


Figure 4 The major and minor elemental factor analysis of sample proto-porcelain bodies from Yejiashan cemetery and four kiln sites. [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

Huoshaoashan, where there was proto-porcelain firing from the late Western Zhou up to the 'Spring and Autumn' period (Zhu 1989; Zhu and Zhu 2008). However, the body materials used during the second part of the late Spring and Autumn period differ from those used during the late Western Zhou and the early Spring and Autumn period, but they are the same as those used in the second part of the middle Spring and Autumn period (Zhang *et al.* 2014).

A possible reason for part of the overlap between Huoshaoashan and Yejiashan is that the Huoshaoashan kiln also fired proto-porcelain during the early Western Zhou period, but the body materials used during the early Western Zhou differ from those used in the Spring and Autumn period. Alternatively, there were other kilns firing proto-porcelain near Huoshaoashan using the same or similar materials as the Huoshaoashan kilns to supply the Zeng state in Yejiashan. Deqing, in Zhejiang Province, was one of the earliest regions for proto-porcelain firing in China, and from the Shang to the Western Zhou dynasty, and then to the Spring and Autumn period, there are some corresponding proto-porcelain kilns that have been discovered or excavated (Zhu 1989; Zhu and Zhu 2008), therefore, this case is possible. Overall, from evidence gathered through the scatter-point analysis of major and minor elemental compositions of proto-porcelain unearthed from Yejiashan cemetery, it is likely that these specimens came from the area of Deqing, Zhejiang Province.

It should be pointed out that the proto-porcelain glazes from the Western Zhou kilns at Deqing analysed by Yin *et al.* (2011) contained less than 2% K<sub>2</sub>O, while the average K<sub>2</sub>O content of the Yejiashan proto-porcelain glazes is 3.5%, with none falling below 2%, leading us to speculate on the low potassium oxide levels in the Deqing glazes, which are slightly unusual for Chinese proto-porcelain glazes of this period (Li 1998). This may be because there are, of course, many kilns in the Deqing region, the time span involved is long and the sample-size is fairly small, so some anomalies might arise from slight differences in the potters' manufacturing practices, which suggests that the provenance analysis of Yejiashan proto-porcelain calls for further research, combined with investigation and research into the Shang and Western Zhou kiln sites.

#### CONCLUSION

Our analysis of the proto-porcelain unearthed from Yejiashan cemetery provides an initial understanding of the following aspects:

- (1) The differences among the major and minor elements of the proto-porcelain bodies unearthed from the different Yejiashan tombs are insignificant; therefore, there it is not likely that they came from multiple areas. The body materials are characterized by high silicon and low aluminium, similar to porcelain stone from southern China. High Fe<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> contents result in a grey–white or deeper grey colouring.
- (2) The proto-porcelain glazes in Yejiashan belong to the typical high-temperature calcium glaze in ancient China, and the glazing colours are mostly celadon and dark green. CaO is taken as the main flux, and there are relatively high percentages of manganese (Mn) and phosphorus (P) in the glaze, which were probably caused by plant ash with high manganese and phosphorus contents. The Fe<sub>2</sub>O<sub>3</sub> content in some of the glazes is high, causing the glaze colour to be darker.
- (3) The firing temperatures of the Yejiashan proto-porcelains are between 1081°C and 1285°C. In addition to  $\alpha$ -quartz, high-temperature cristobalite and mullite, which are the important typical characteristics of phase composition for porcelain in their original form, are found in Yejiashan proto-porcelain bodies. Water absorption is more than 2%. The glazes are severely crazed, the body and glaze are poorly combined, and the shedding phenomenon

is serious. This reflects the primitiveness of the glazing and firing techniques in the early Western Zhou period.

- (4) Proto-porcelain unearthed from four southern kiln sites was selected in order to perform multivariate statistical analysis along with proto-porcelain from Yejiashan cemetery. Factor analysis based on the major and minor elemental chemical elements indicates that the Yejiashan proto-porcelain might have come from the Deqing area, in Zhejiang Province.

These results provide new archaeological evidence for research on issues related to material flow in the Western Zhou dynasty.

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## SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

Table S1. Sample information for proto-porcelains unearthed from Yejiashan Cemetery.

Table S2. Results for some major and minor elements with three repeat analyses of NIST 610 and Corning B glass standards by LA-ICP-AES. Published values are from Pearce *et al.* (1997) and Brill (1999) for NIST 610 and Corning B, respectively. (W%).

Table S3. Major element compositions of sample bodies unearthed from Yejiashan Cemetery (w%).

Table S4. Major element compositions of the sample glazes from Yejiashan Cemetery (w%).

Table S5. Total variance of explanation for factor analysis.

Table S6. Factor load matrix.

Figure S1. The location of Yejiashan Cemetery, Suizhou City.

Figure S2. Scatter diagram showing the molar ratio of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  and  $\text{R}_x\text{O}_y/\text{Al}_2\text{O}_3$  for proto-porcelain bodies and glazes in the Yejiashan samples.

Figure S3. Backscattered electron images of combined body and glaze layers of typical proto-porcelains from Yejiashan Cemetery.

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