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# Microscopic study of Chinese bronze casting moulds from the Eastern Zhou period

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ABSTRACT

Piece mould casting technology, as a hall mark of the central plains of China during the Bronze Age, has attracted scholars' interest globally. Bronze ritual vessels found in this area were produced in large quantities and generally cast with the moulds composed of three or more sections. This enormous industry certainly required workers to have had professional knowledge to ensure the success of every single cast. Mould making technology was one of its most important parts. This research looks into the microstructure of bronze casting moulds of the Eastern Zhou period, which was a rarely studied topic in previous research. Through comparison with local pottery, it is shown that casting moulds in this period were produced with quite specialized sand-rich material, and clay was only a minor component. It is then discussed how this unique material can be beneficial to the bronze casting process and how this technology was integrated as a crucial part of the Chinese bronze casting system. These analyses might be able to portray mould makers in the ancient bronze foundries as a group of specialized people with their own traditions and professional knowledge.

## 1. Introduction

The sophisticated piece mould casting technology is taken as a hall mark of the central plains of China during the Bronze Age. Bronze ritual vessels, which were used for serving food and wine for ancestors during worship rituals, and for the legitimization of power, were largely cast with the mould composed of three or more sections. Motifs are divided by mould divisions and usually organized symmetrically. All these patterns were either impressed from a model or carved directly onto the mould. When these sections were dried and fired to an adequate strength, they would be assembled together with an outer casing. A core was set inside the mould, leaving a gap as thick as the bronze object. The molten bronze would then be poured into the mould. When the metal cooled, the mould and core could be broken up and removed. Based on this unique technology, ancient people in China developed a whole set of bronze ritual vessels which have no counterparts in other regions of the world. They usually have symmetrical shapes as well as intricate patterns. Some can be quite big and weigh over 800 kg. The whole process of manufacture, from making the mould, alloying the metal, to finishing the surface of the object was labour intensive and required specialized cross-craft knowledge (Bagley, 1995; Franklin, 1983). The whole foundry therefore needed to be organized accordingly (Li, 2007, 2003).

Bronze casting in the Chinese central plains probably started in the late Neolithic period (2500–2000 BC), but the earliest identified evidence of widespread bronze use appears during the Erlitou culture (around 1900–1500 BC) located in Henan, southern Shanxi and southern Hebei (An, 1993). Size and complexity of bronze vessels gradually increased in the following periods, and it is generally agreed that the bronze casting industry culminated in the central plains of China during the Late Shang to Early Western Zhou period (around 1300–977 BC). Huge ritual vessels in this period were cast with numerous mould sections divided both vertically and horizontally. Motifs often fill the entire body of these vessels and in some cases decorations were made in high relief. The patterns of this period look much more expressive than in earlier periods. After a transition period of around 100 years, the Late Western Zhou (877–771 BC) and Eastern Zhou periods (770–256 BC), witnessed a considerable simplification in the design of the section moulds and wide adoption of welding and casting-on techniques. The production efficiency of the bronze objects was much improved during this period, and the archaeological findings far outnumbered those of earlier periods. The focus of the industry shifted to surface decoration techniques such as tinning, gilding

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and various inlays (for a general review of this topic see Bagley, 1999; Dong, 2007; Von Faulkenhausen, 1999; Hua, 1999; Rawson, 1987; Zhang, 2011). As the central political power collapsed in this period, the bronze casting industry was also decentralized, as evidenced by the emergence of huge foundries serving local states such as at Houma in Shanxi province and Xinzheng in Henan province.

Currently, scholars are still discussing controversially on some technical issues concerning the piece mould casting technology of the Chinese Bronze Age. For example, it is unclear whether the design of the section moulds was made using a model of the complete object or a single repeatable section of it. The pattern and inscription making technologies are also in debate (See Bagley, 1987, 37–45; Chase, 1983; Meyers and Holmes, 1983; Nickel, 2006). Another matter for discussion is the possible use of the lost wax casting technique, which is generally recognized as a western tradition and might act as a supplementary to piece mould casting in China (Chen et al., 2009; Hua, 2010; Tan, 2007). Once all these technical details are sorted out, we will be able to reconstruct a comprehensive picture of the bronze casting system of ancient China. This would also facilitate our ongoing discussion on craftsmen's technical choices, the labour organization within the foundry and knowledge transmission between cultural groups.

In parallel, some scholars have been studying mould-making technology, which has been recognized as a crucial part of Chinese bronze industry. To ensure the success of every cast, the mould material should bear some special properties such as low shrinkage during drying, high resistance to thermal shock, low thermal conductivity and good ventilation during the casting (Tan, 1999). Many chemical analyses have been conducted on mould fragments found in Anyang in Henan province, Zhouyuan in Shaanxi province, Houma in Shanxi province and some other Bronze Age sites (Lian, 1996; Liu et al., 2007; Liu and Yue, 2005; Tan, 1999; Wang, 2002; Zhou et al., 2009). These analyses have confirmed that casting moulds were manufactured with silica-based materials and usually have silica contents higher than normal ceramics. Chinese scholars have suggested that they were made by fine clay tempered with silt, sand and plant ash (Tan et al., 1993). Research has also been conducted to identify the firing temperature of bronze casting moulds. Some scholars have suggested that this temperature should be higher than the decomposition temperature of calcite (898 °C), while other studies on thermal expansion tests have revealed that casting moulds may not have been fired over 700 °C (See Liu et al., 2008; Tan, 1999).

However, the microstructure of this material never seems to have received much attention in these studies. Indeed, published microscopic observations made by Stoltman et al. (2009) and Freestone et al. (1989) form the entire body of literature on this topic. However, these two papers have actually demonstrated the significance of microscopic study, which provides essential information about the specific physical characteristics of this material and whether specialized technology and knowledge were needed for bronze workers to make the mould. Explored in detail this will contribute to the reconstruction of the whole bronze manufacturing system in the Bronze Age of China and explain in part why those bronze vessels could be made with such high quality. It will also help in the discussion surrounding the specialization of bronze craftsmen. Moreover, microscopic observations, when quantified by point-counting (Stoltman et al., 2009), can be compared with data from other sites of different periods to shed light on the initiation and development of the bronze casting system of ancient China.

## 2. Materials

All the materials in this project were excavated from bronze foundries in a site located at Xinzheng, Henan province (Fig. 1) and dated to the Eastern Zhou period which can be subdivided into the Spring and Autumn (770–476 BC) and the Warring States periods (475–221 BC). The site is named *Zheng Han gu cheng* and was the capital city of the ancient *Zheng* and *Han* states.

This site was firstly occupied by the State *Zheng* during the early Spring and Autumn period. State *Han* brought down *Zheng* in 375 BC and set its capital at Xinzheng until it was conquered by the First Empire Qin in 230 BC. The bronze casting activities in this site nearly covered the entirety of the Eastern Zhou period. The ritual vessels of *Zheng* and *Han* states are characterized by their massive size, intricate patterns and novel designs, and may represent the highest level of casting skills during the Eastern Zhou period. The

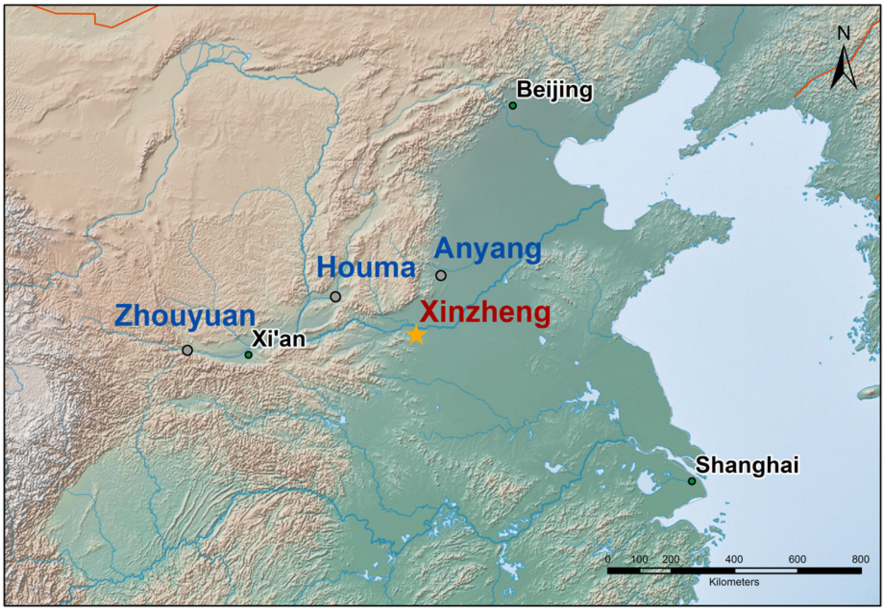

**Fig. 1.** Map shows the location of Xinzheng, in the central plains of China. Several other Shang and Zhou bronze casting foundries are also labelled in the map.

foundry area was first excavated in the 1960s, and archaeological work on this site is still ongoing today. Four bronze casting foundries have been identified at the site and appear to have been used at different times. Huge amounts of mould fragments have been uncovered. While some of these can be identified as mould fragments associated with ritual vessel, artefact, and tool making, the majority of them cannot be traced back to their intended product (Cai, 2011).

For this research, forty eight samples were collected from these foundries to cover the whole lifespan of the bronze casting industry of this site. The chronology of these samples was determined by stratigraphy and associated pottery sherds. They are small fragments and the intended products of them are usually difficult to identify (Fig. 2). Local pottery samples were also collected from every time period of the site for comparison. Two types of vessels, *dou* and *li*, were selected as representative of local ceramics. *Dou* was used as a container for food during meals and was assumed to have a fine fabric. *Li*, as cooking pot, was usually tempered with sand and flint.

It is expected that this research can tell us the material characteristics of bronze casting moulds used in this period and shed new light on the study of ancient mould making technology. Moreover, the comparison between mould and local pottery may show whether specialized crafts knowledge was employed in producing moulds, and whether this was beneficial to the casting of bronze.

## 3. Methodology

Being too fragile for cutting and polishing without treatment, the samples were first consolidated with epoxy resin. All the samples were then mounted as blocks and polished to 0.5 micron. A Leica DM4000M microscope was used for optical observations. Samples were then coated with gold and examined in a Scanning

**Fig. 2.** Some of the bronze casting mould pieces selected and analysed in this project. Most of them are small fragments and hard to trace to their intended product.

Electron Microscope (SEM) for high quality electron imaging. A Hitachi S-3600N SEM equipped with EDAX energy dispersive spectrometer (EDS) was employed for detailed observation and chemical analysis. The acceleration voltage was set at 20 kV and the working distance kept at 12–15 mm. The back scattered electron (BSE) image detector was mainly used to study the microstructure of the samples. The bulk chemical composition was obtained by averaging three random analyses of areas of 870 μm × 1270 μm. Considering the variable beam intensity of the SEM system and the porosity of the samples, all the data have been normalized to 100% and should be taken as semi-quantitative only. It is understood that gold coating is not ideal for chemical analysis and may affect the accuracy of the result, but considering the importance of high quality images in this research, we still chose to coat samples with gold. As all the analytical work was done with the same instrument, the data should be considered as internally consistent and the comparison of chemical data between different samples should be reliable.

The electron images were then analysed with the point counting method for a semi-quantitative estimate of the content of particles, voids and matrix (Stoltman, 1989). Ninety seven points were set in each image. Due to the difficulty in counting small particles under 10 μm, these were taken as being part of the matrix. The size of counted particles was recorded for detailed discussions. In some tempered pottery, large inclusions could even be identified with the naked eye. These inclusions were not counted in the final result, since they were consciously added temper by humans, and do not reflect the property of the raw clay (Table 1).

## 4. Results

### 4.1. Chemical composition

The concentrations of seven major elements (Na, Mg, Al, Si, K, Ca, Fe) were obtained by SEM-EDS analysis and are presented in Table 2. Mould samples from different periods do not show any significant variations in chemical composition, but the differences between moulds and local pottery can be clearly seen in binary diagrams (Fig. 3). For mould samples, the silica content is relatively high and usually above 75%, while for pottery samples, it is usually around 60%–70%. Only rarely does it exceed 70%. On the other hand, the pottery is richer in alumina, iron oxide and magnesia. For lime and potash, the difference between moulds and pottery is negligible. The lime content of moulds seems more variable. The average potash content of local ceramic pottery is slightly higher than in the moulds. In addition, for pottery samples, the content of alumina is positively correlated with iron oxide and magnesia while negatively correlated with silica and soda. This may suggest that in ceramics, iron oxide, magnesia and alumina are contained within the same phase while silica and soda come from a different one. However, in moulds, these correlations are difficult to identify as all plots are usually concentrated in a single area. All correlation factors were calculated separately for moulds and pottery, and are presented in Table 3. It should be noted that no correlation factor above 0.7 can be found for the moulds. This will be discussed further combined with the microstructure.

It is not surprising to find increased silica content in the moulds for they would have needed high refractive properties to hold molten metal during casting. Indeed, extra silica in bronze casting moulds has been detected and reported by many scholars. It is usually interpreted as phytoliths introduced by adding plant ash into the mould fabric (Tan, 1999). Numerous spot analyses have been done on sand particles in these samples, and the results of several individual analyses are shown in Table 4. Apart from the dominant quartz particles, several other minerals such as calcite, potassium feldspar, and albite (sodium feldspar) were regularly identified. However, as mentioned before, these data should only be considered as semi-quantitative and, without petrologic and mineralogical analysis, the specific names of some minerals are hard to determine. These minerals and the clay matrix are the main sources of alkali oxides, magnesia, lime and iron oxides for these samples.

### 4.2. Microstructure

The microstructure of casting moulds and pottery was revealed by high resolution BSE imaging. For semi-quantitative point counting analysis, the fabric is classified into three phases: sand particles, voids, and matrix. Though the voids are sometimes counted as matrix in pottery analysis, their function should be emphasized here because of their critical role in the casting process for the venting of gases.

The point counting results are presented in Table 5. Typical BSE images of mould and local ceramics are shown in Fig. 4. Once again, mould samples are similar to each other in spite of the various dates and the difference between the moulds and domestic ceramic can be easily noticed. The fabric of the moulds is composed of angular particles with small amounts of clay matrix as interconnecting material. The porosity of these samples is quite high and many of these pores are connected with each other. In contrast, ceramic samples are dominated by a vitrified clay matrix with some particles and few voids. This observation is demonstrated by point counting results illustrated in Fig. 5. The three apexes of the ternary diagram represent the sand particles, matrix and voids. It can be noticed that mould samples are concentrated in the apex of sand, while ceramic samples are usually in the region of the matrix apex. Void content of moulds is generally between 15% and 40% while pottery samples usually contain less than 10% voids. Only in three cases, pottery samples (Po2, Po3, Po7) have more than 15% voids and they are joined by another mould sample (CM19) located in the transition zone between the major pottery and mould groups. The three pottery samples are all *dou* container and not heavily tempered. The mould sample is macroscopically similar to others moulds, despite its different structure. These four samples form only a small subset of the total assemblage and may represent sandy ceramics and clay rich moulds, and do not detract from the larger picture.

Moreover, the particles in the moulds are generally bigger than those in the pottery. Big quartz particles can be identified as temper in some pottery samples, but the bulk of the fabric is still dominated by vitrified clay matrix and small sand inclusions. The measurements of particles' long axes are also presented in Table 5 and illustrated in a histogram (Fig. 6). The particle length of sand in pottery is around 40–50 μm. One exception is Po10, a ceramic *li* with a mean particle length of 105 μm. BSE imaging reveals that its fabric is heavily tempered with large quartz particles but, compared with moulds, these particles are still far less in number. Two categories may be found in mould samples in terms of particle size. The first group is characterized by particles around 60–70 μm in length which is approximately 10 μm larger than those found in pottery, while the second group generally have a mean length around 100 μm. The mean values of particle length were also calculated respectively for the four different periods in Eastern Zhou (the Early Spring and Autumn, the Late Spring and Autumn, the Early Warring States and the Late Warring States). From the Early Spring and Autumn period to the Late Warring States period, there is no considerable change of particle size detected in casting moulds. Samples from the Early Warring States period have the largest mean particle size of 84 μm, but considering the limited sample number, there is no statistically significant difference detected between these groups (Fig. 7).

**Table 1**
Description of mould and pottery samples.

| Lab code | Description | Context | Period | Intended shape of mould | Colour and fabric |
|---|---|---|---|---|---|
| CM1 | Casting mould | T514(4):81 | Early SA | – | Red fabric with thin black layer on the casting surface, voids caused by burning organic fabrics can be identified |
| CM2 | Casting mould | T514(4):83 | Early SA | – | Grey fabric |
| CM 3 | Casting mould | T514(4):78 | Early SA | – | Light grey fabric |
| CM 4 | Casting mould | T514(4):79 | Early SA | – | Brownish fabric, elongated pores identified |
| CM 5 | Casting mould | T185H710:299 | Late SA | – | Dark grey fabric and brownish casting surface, high physical strength |
| CM 6 | Casting mould | T185H710:8 | Late SA | – | Red fabric |
| CM 7 | Casting mould | T185H710:300 | Late SA | – | Grey fabric and brownish casting surface |
| CM 8 | Casting mould | T185H710 | Late SA | Ring-head artefact | Grey fabric, tiny pores identified in cross-section |
| CM 9 | Casting mould | T183(4):22 | Early WS | Knife | Grey fabric |
| CM 10 | Casting mould | T186(3):79 | Early WS | Ring-head knife | Grey fabric, loose and fragile body |
| CM 11 | Casting mould | T183(4):235 | Early WS | – | Orange fabric, tiny pores identified in cross-section |
| CM 12 | Casting mould | T187H715:(83) | Early WS | – | Grey fabric and brownish casting surface, |
| CM 13 | Casting mould | H717:92 | Late WSI | – | Steel grey fabric, tiny pores identified in cross-section |
| CM 14 | Casting mould | T187(3):28 | Late WSI | – | Light grey fabric |
| CM 15 | Casting mould | T191(2) | Late WSI | – | Brick-red fabric, black core identified, elongated pores identified in cross-section |
| CM 16 | Casting mould | T191(2) | Late WSI | – | Steel grey fabric |
| CM 17 | Casting mould | T191(2) | Late WSI | – | Brownish fabric and light grey casting surface, elongated pores identified in cross-section |
| CM 18 | Casting mould | T191(2) | Late WSI | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 19 | Casting mould | T191(2) | Late WSI | – | Brick-red fabric, elongated pores identified in cross-section |
| CM 20 | Casting mould | T31(3) | Late WSI | – | Grey fabric, elongated and tiny rounded pores identified in cross-section |
| CM 21 | Casting mould | T31(3) | Late WSI | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 22 | Casting mould | T31(3) | Late WSI | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 23 | Casting mould | T31(3) | Late WSI | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 24 | Casting mould | T31(3) | Late WSI | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 25 | Casting mould | T4H38 | Late WSI | – | Light brownish fabric and dark grey casting surface, elongated pores identified in cross-section |
| CM 26 | Casting mould | T4H38 | Late WSI | – | Brick-red fabric, tiny pores identified in cross-section |
| CM 27 | Casting mould | T4H38 | Late WSI | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 28 | Casting mould | T4H38 | Late WSI | – | Brick-red fabric |
| CM 29 | Casting mould | T188H676:152 | Late WSII | – | Brownish fabric, elongated pores identified in cross-section |
| CM 30 | Casting mould | T183(3):38 | Late WSII | Knife | Orange fabric, tiny pores identified in cross-section |
| CM 31 | Casting mould | T17(3) | Late WSII | – | Brick-red fabric, loose and fragile body |
| CM 32 | Casting mould | T17(3) | Late WSII | – | Brick-red fabric, rounded pores identified in cross-section |
| CM 33 | Casting mould | T17(3) | Late WSII | – | Brownish fabric, pores created by burning organic fibres can be identified in cross-section |
| CM 34 | Casting mould | T17(3) | Late WSII | – | Brownish fabric, rounded pores identified in cross-section |
| Po1 | Ceramic *dou* | T552Y36 | Western Zhou | – | Grey fabric and brownish core |
| Po2 | Ceramic *dou* | T506Y29 | Early SA | – | Dark grey fabric |
| Po3 | Ceramic *dou* | T515(3) | Middle SA | – | Grey fabric |
| Po4 | Ceramic *dou* | T41(5) | Late SA | – | Dark grey fabric, elongated pores identified in cross-section |
| Po5 | Ceramic *dou* | T26H10 | Early WS | – | Dark grey fabric |
| Po6 | Ceramic *dou* | T40(3) | Middle WS | – | Grey fabric, fragile body |
| Po7 | Ceramic *dou* | T34(2) | Late WSII | – | Grey fabric with whitish surface |
| Po8 | Ceramic *li* | T552Y36 | Western Zhou | | Brownish fabric |
| Po9 | Ceramic *li* | T506Y29 | Early SA | – | Brownish fabric |
| Po10 | Ceramic *li* | T515(3) | Middle SA | – | Brownish fabric tempered with large grey particles (around 1.5 mm) and small white particles, elongated pores identified in cross-section |
| Po11 | Ceramic *li* | T41(5) | Late SA | – | Red fabric with grey core, tempered with small white particles, elongated pores identified in cross-section |
| Po12 | Ceramic *li* | T26H10 | Early WS | – | Red fabric tempered with large white particles (around 1.5 mm), elongated pores identified in cross section |
| Po13 | Ceramic *li* | T40(3) | Middle WS | – | Brick-red fabric tempered with large white, grey and pink particles (the largest is around 4 mm) |
| Po14 | Ceramic *li* | T18H18 | Late WSI | – | Brick-red fabric, elongated pores identified in cross-section |

*SA stands for the Spring and Autumn; WS stands for the Warring States.

The coefficient of variance (CV) of particle size shown in the last column of Table 5 demonstrates how well these sand particles were sorted. The mean CVs for moulds and pottery were calculated as 0.52 and 0.64 respectively. It is argued that sand particles in moulds were better sorted than in pottery, although the limited sample size should be noted as a possible source of error.

High magnification images show the details of the matrix as well as the interaction region between sand particles and clay matrix (Fig. 8). In ceramic samples, the clay matrix is vitrified and forms a network which holds all the sand particles together as a consolidated structure. The chemical interaction between sand particles and matrix is widespread and some small particles may have been absorbed by the matrix. However, the mould matrix shows that clay minerals were just gently sintered with each other. In some cases, isolated clay particles can still be identified. The boundary between the matrix and individual sand particles is clear,

**Table 2**
Bulk chemical composition of mould and pottery samples.

| Lab code | Description | Period | Composition % | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | $Na_2O$ | MgO | $Al_2O_3$ | $SiO_2$ | $K_2O$ | CaO | FeO |
| CM1 | Casting mould | Early SA | 1.9 | 1.6 | 11.6 | 76.4 | 3.0 | 1.0 | 4.5 |
| CM2 | Casting mould | Early SA | 1.7 | 1.3 | 11.3 | 77.2 | 3.3 | 1.4 | 3.7 |
| CM 3 | Casting mould | Early SA | 1.9 | 1.6 | 11.4 | 77.0 | 2.8 | 1.2 | 4.0 |
| CM 4 | Casting mould | Early SA | 1.9 | 1.7 | 12.0 | 75.4 | 3.1 | 1.3 | 4.5 |
| CM 5 | Casting mould | Late SA | 1.5 | 1.5 | 11.5 | 75.6 | 3.2 | 1.9 | 4.7 |
| CM 6 | Casting mould | Late SA | 1.7 | 1.4 | 11.3 | 77.2 | 2.6 | 1.6 | 4.1 |
| CM 7 | Casting mould | Late SA | 1.3 | 1.4 | 11.7 | 77.3 | 3.3 | 1.0 | 4.0 |
| CM 8 | Casting mould | Late SA | 1.8 | 1.5 | 12.3 | 76.6 | 2.7 | 1.3 | 3.8 |
| CM 9 | Casting mould | Early WS | 1.6 | 1.4 | 12.3 | 76.0 | 3.0 | 1.1 | 4.5 |
| CM 10 | Casting mould | Early WS | 1.8 | 1.4 | 11.2 | 77.8 | 2.6 | 1.3 | 3.8 |
| CM 11 | Casting mould | Early WS | 1.5 | 1.4 | 11.2 | 77.7 | 3.4 | 0.9 | 3.9 |
| CM 12 | Casting mould | Early WS | 1.6 | 1.5 | 11.7 | 77.4 | 2.7 | 1.2 | 3.9 |
| CM 13 | Casting mould | Late WSI | 1.7 | 1.2 | 11.1 | 78.9 | 2.9 | 0.9 | 3.3 |
| CM 14 | Casting mould | Late WSI | 1.9 | 1.3 | 11.3 | 77.9 | 2.8 | 1.2 | 3.5 |
| CM 15 | Casting mould | Late WSI | 1.5 | 1.5 | 10.7 | 75.7 | 2.6 | 4.3 | 3.6 |
| CM 16 | Casting mould | Late WSI | 2.2 | 1.5 | 10.9 | 78.1 | 3.1 | 1.1 | 3.0 |
| CM 17 | Casting mould | Late WSI | 1.7 | 1.7 | 11.3 | 77.2 | 2.8 | 1.5 | 3.8 |
| CM 18 | Casting mould | Late WSI | 0.9 | 1.0 | 11.9 | 76.1 | 3.0 | 1.5 | 5.6 |
| CM 19 | Casting mould | Late WSI | 1.7 | 1.2 | 12.7 | 76.6 | 2.7 | 2.7 | 2.5 |
| CM 20 | Casting mould | Late WSI | 1.8 | 1.5 | 10.4 | 77.7 | 2.8 | 1.4 | 4.4 |
| CM 21 | Casting mould | Late WSI | 2.1 | 1.6 | 11.4 | 75.5 | 2.9 | 2.1 | 4.3 |
| CM 22 | Casting mould | Late WSI | 2.1 | 1.5 | 11.2 | 73.6 | 2.6 | 4.6 | 4.3 |
| CM 23 | Casting mould | Late WSI | 2.0 | 1.5 | 12.6 | 75.7 | 2.8 | 1.3 | 4.0 |
| CM 24 | Casting mould | Late WSI | 1.8 | 1.6 | 11.4 | 76.7 | 2.7 | 2.3 | 3.4 |
| CM 25 | Casting mould | Late WSI | 2.1 | 1.6 | 10.1 | 72.4 | 2.4 | 8.2 | 3.1 |
| CM 26 | Casting mould | Late WSI | 2.1 | 1.8 | 11.4 | 76.4 | 2.7 | 2.1 | 3.4 |
| CM 27 | Casting mould | Late WSI | 2.1 | 1.5 | 11.0 | 77.0 | 3.0 | 1.6 | 3.8 |
| CM 28 | Casting mould | Late WSI | 2.0 | 1.3 | 10.5 | 78.5 | 2.8 | 1.3 | 3.6 |
| CM 29 | Casting mould | Late WSII | 2.0 | 1.6 | 11.9 | 75.9 | 3.2 | 1.2 | 4.1 |
| CM 30 | Casting mould | Late WSII | 1.7 | 1.4 | 11.3 | 77.6 | 2.9 | 1.0 | 4.1 |
| CM 31 | Casting mould | Late WSII | 2.1 | 1.9 | 11.3 | 75.1 | 2.9 | 2.8 | 4.0 |
| CM 32 | Casting mould | Late WSII | 2.2 | 1.7 | 12.4 | 74.7 | 2.4 | 2.6 | 4.0 |
| CM 33 | Casting mould | Late WSII | 2.1 | 1.8 | 11.2 | 76.4 | 2.6 | 2.1 | 3.8 |
| CM 34 | Casting mould | Late WSII | 1.9 | 1.0 | 10.6 | 78.0 | 2.1 | 2.3 | 4.0 |
| Po1 | Ceramic *dou* | Western Zhou | 1.3 | 2.0 | 14.6 | 70.8 | 3.2 | 1.7 | 6.3 |
| Po2 | Ceramic *dou* | Early SA | 1.4 | 1.7 | 13.3 | 74.3 | 3.1 | 1.2 | 4.9 |
| Po3 | Ceramic *dou* | Middle SA | 1.7 | 1.9 | 13.9 | 73.6 | 2.9 | 1.4 | 4.5 |
| Po4 | Ceramic *dou* | Late SA | 1.1 | 2.2 | 16.3 | 67.4 | 3.9 | 1.2 | 7.8 |
| Po5 | Ceramic *dou* | Early WS | 1.0 | 2.2 | 16.2 | 67.6 | 3.8 | 1.6 | 7.6 |
| Po6 | Ceramic *dou* | Middle WS | 1.2 | 2.0 | 14.1 | 72.1 | 3.1 | 1.7 | 5.7 |
| Po7 | Ceramic *dou* | Late WSII | 1.5 | 1.9 | 13.2 | 73.3 | 3.4 | 1.4 | 5.2 |
| Po8 | Ceramic *li* | Western Zhou | 1.4 | 2.2 | 14.1 | 69.8 | 3.8 | 2.3 | 6.3 |
| Po9 | Ceramic *li* | Early SA | 0.9 | 2.0 | 14.5 | 69.6 | 5.0 | 1.4 | 6.5 |
| Po10 | Ceramic *li* | Middle SA | 0.6 | 2.3 | 17.7 | 61.7 | 5.6 | 2.8 | 9.3 |
| Po11 | Ceramic *li* | Late SA | 0.8 | 2.8 | 17.4 | 63.7 | 3.7 | 3.5 | 8.1 |
| Po12 | Ceramic *li* | Early WS | 1.3 | 2.3 | 15.1 | 69.4 | 3.6 | 1.5 | 6.8 |
| Po13 | Ceramic *li* | Middle WS | 1.3 | 2.4 | 16.3 | 67.6 | 3.7 | 1.2 | 7.4 |
| Po14 | Ceramic *li* | Late WSI | 1.2 | 2.0 | 14.9 | 70.2 | 3.6 | 1.3 | 6.8 |

*SA stands for the Spring and Autumn; WS stands for the Warring States.

which suggests that the reaction between them was minor. The clay matrix just glues these big particles together, and the whole structure is much looser than the ceramic. This may be the reason why mould samples are much less resistant to water, as noted during sample preparation. These findings may also be due to the different firing temperature for these two types of materials. The degree of vitrification in the ceramic matrix is higher than in the moulds, but their chemical compositions are not much different (1%–2% $Na_2O$, 2% MgO, 15%–20% $Al_2O_3$, 60–70% $SiO_2$, 2%–5% $K_2O$, 1%–4% CaO and 4–8% FeO). Therefore, the firing temperature of the pottery should be higher than that of the moulds. For pottery in this period, researchers have pointed out that the firing temperature is generally around 800–1000 °C ( Li, 1998, 68).

Another interesting finding is the identification of large calcite particles in the mould fabric. In Fig. 9, one calcite particle over 500 μm in length is labelled and it is obviously not created by post-depositional process. If these calcite particles can be counted as part of the raw material for casting moulds, two interesting things should be noted. Firstly, the decomposition temperature for calcite is around 898 °C, which means that these moulds were never heated above this temperature for any extended period of time. Secondly, as only few calcite particles were found in the pottery samples, their main source of lime should be clay minerals. In contrast, the lime content in the moulds largely depends on the number of calcite particles and therefore is more varied. This is consistent with the observation that the plots of mould samples are more dispersed in their lime content.

## 5. Discussion

The microscopic study has revealed the characteristics of bronze casting moulds as low clay content and high porosity (Figs. 4 and 5). In contrast, clay matrix is the dominant phase in all pottery. The bulk chemistry obtained during this study has revealed the same result. The clay matrix is a major alumina, iron oxide, and magnesia bearing phase in this case, and when above 60% in volume, such as

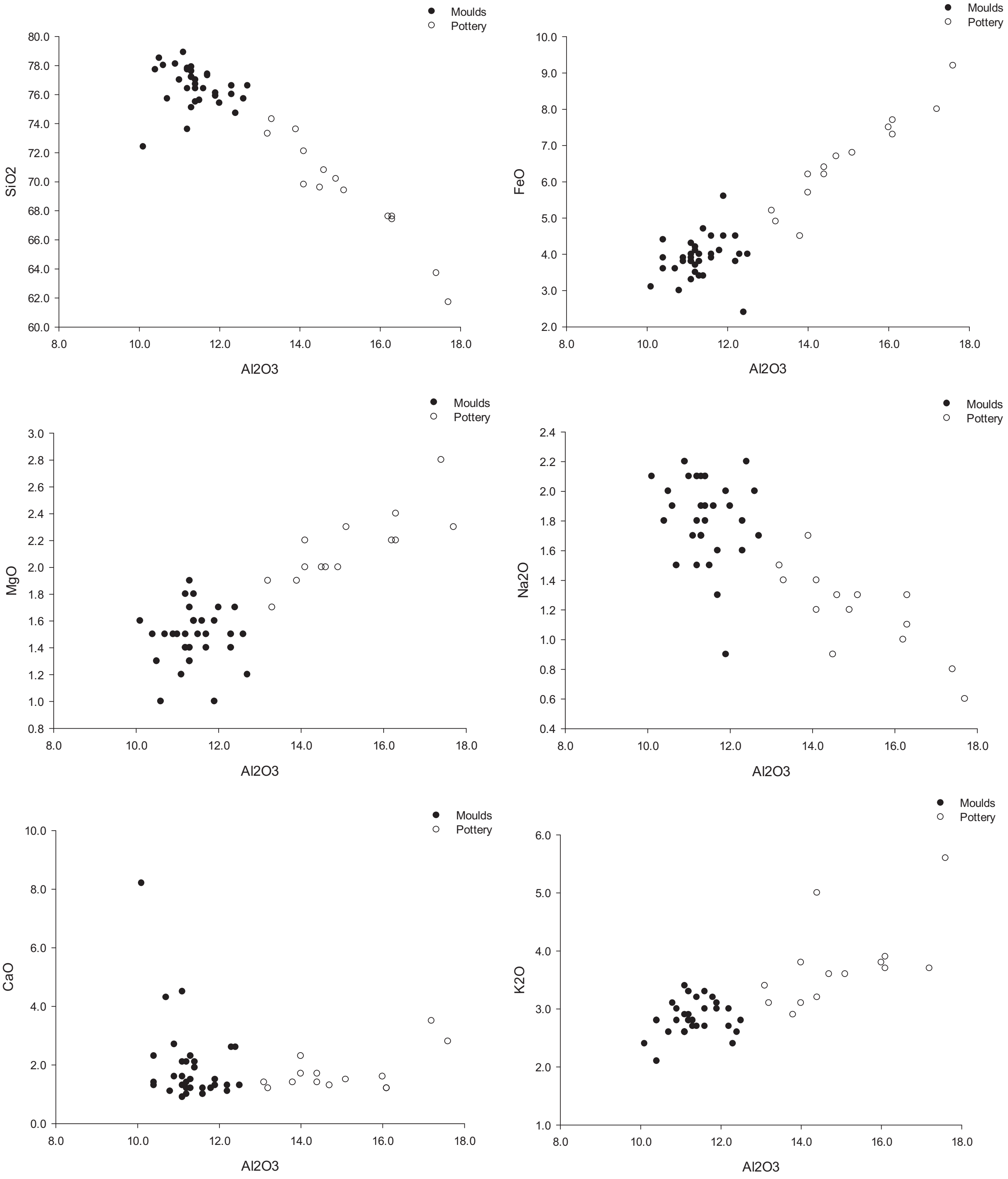

**Fig. 3.** Binary diagrams of bulk chemical results. Black dots represent mould samples while white dots stand for pottery samples. It can be noticed that white dots show correlations in many diagrams but black dots does not have this characteristic.

**Table 3**
Correlation factors between seven oxides components.

| | | Moulds | | | | | | | Potteries | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | $Na_2O$ | MgO | $Al_2O_3$ | $SiO_2$ | $K_2O$ | CaO | FeO | $Na_2O$ | MgO | $Al_2O_3$ | $SiO_2$ | $K_2O$ | CaO | FeO |
| Pearson correlation | $Na_2O$ | – | 0.54 | −0.21 | −0.29 | −0.36 | 0.25 | −0.41 | – | −0.56 | −0.79 | 0.86 | −0.79 | −0.58 | −0.85 |
| Pearson correlation | MgO | 0.54 | – | 0.05 | −0.40 | 0.00 | 0.18 | −0.06 | −0.56 | – | 0.83 | −0.82 | 0.32 | 0.70 | 0.76 |
| Pearson correlation | $Al_2O_3$ | −0.21 | 0.05 | – | −0.15 | 0.21 | −0.37 | 0.21 | −0.79 | 0.83 | – | −0.96 | 0.58 | 0.57 | 0.95 |
| Pearson correlation | $SiO_2$ | −0.29 | −0.40 | −0.15 | – | 0.27 | −0.69 | −0.07 | 0.86 | −0.82 | −0.96 | – | −0.71 | −0.67 | −0.97 |
| Pearson correlation | $K_2O$ | −0.36 | 0.00 | 0.21 | 0.27 | – | −0.52 | 0.29 | −0.79 | 0.32 | 0.58 | −0.71 | – | 0.36 | 0.71 |
| Pearson correlation | CaO | 0.25 | 0.18 | −0.37 | −0.69 | −0.52 | – | −0.26 | −0.58 | 0.70 | 0.57 | −0.67 | 0.36 | – | 0.53 |
| Pearson correlation | FeO | −0.41 | −0.06 | 0.21 | −0.07 | 0.29 | −0.26 | – | −0.85 | 0.76 | 0.95 | −0.97 | 0.71 | 0.53 | – |

**Table 4**
Chemical composition of sand particles.

| Lab code | Description | $Na_2O$ | $Al_2O_3$ | $SiO_2$ | $K_2O$ | CaO | $TiO_2$ | FeO |
|---|---|---|---|---|---|---|---|---|
| CM18 | Fe mineral | bdl | 4.9 | 40.2 | 0.3 | bdl | 10.2 | 44.4 |
| CM25 | Calcite | bdl | bdl | bdl | bdl | 100.0 | bdl | bdl |
| CM28 | Ca–Fe mineral | bdl | 23.0 | 42.4 | bdl | 23.6 | bdl | 11.0 |
| CM34 | Albite | 9.2 | 17.5 | 71.9 | bdl | 1.4 | bdl | bdl |
| CM34 | Potassium feldspar | bdl | 16.4 | 67.5 | 16.1 | bdl | bdl | bdl |

bdl stands for below detection limit.

in the ceramic fabric, the bulk composition contains relatively high quantities of these oxides. Moreover, the low quantity of sand particles present in the pottery means that they likely have little influence on the bulk composition, and therefore the content of these oxides is mostly determined by the amount of clay present within the matrix. This explains why there is a positive correlation between these elements. In contrast, as clay is just a minor phase in mould fabrics, the alumina, iron oxide and magnesia contents are much lower in these samples and, because of the influence from feldspars and iron rich minerals, the positive correlations are eliminated.

According to the research of Freestone et al. (1989) on moulds from Anyang, the probable raw material is loess, which is similar to that of the local pottery. However, according to the result of this research, it must have been sourced either from carefully selected loess deposits with much sand, or the loess was processed by ancient workers to reduce their clay content. Kerr and Wood (2004, 103) have suggested that mould makers may have deliberately washed away some clay from the loess and increased its porosity. Voids caused by burning out organic temper were rarely identified in our samples, which may indicate that the creation of porosity by the addition and burning of organic materials was not the method used by mould makers, though it is a technique mastered by potters since the Neolithic period in China (Li, 1998, 32). Moreover, the consistency of particle size in moulds also reveals the careful processing of raw materials in mould making.

The fabric of the moulds shows it may have had low strength and plasticity before firing. This means that moulding rather than

**Table 5**
Point counting result for mould and pottery samples.

| Lab code | Description | Period | Sand % | Matrix % | Voids % | Average size of sand (μm) | Std of size (μm) | CV |
|---|---|---|---|---|---|---|---|---|
| CM1 | Casting mould | Early SA | 54 | 19 | 27 | 68 | 38 | 0.56 |
| CM2 | Casting mould | Early SA | 44 | 18 | 39 | 92 | 51 | 0.56 |
| CM 3 | Casting mould | Early SA | 50 | 30 | 20 | 67 | 41 | 0.60 |
| CM 4 | Casting mould | Early SA | 64 | 17 | 19 | 64 | 36 | 0.57 |
| CM 5 | Casting mould | Late SA | 53 | 36 | 11 | 96 | 82 | 0.86 |
| CM 6 | Casting mould | Late SA | 42 | 39 | 19 | 64 | 43 | 0.68 |
| CM 7 | Casting mould | Late SA | 53 | 25 | 22 | 73 | 37 | 0.50 |
| CM 8 | Casting mould | Late SA | 49 | 19 | 32 | 105 | 55 | 0.52 |
| CM 9 | Casting mould | Early WS | 47 | 28 | 25 | 76 | 40 | 0.52 |
| CM 10 | Casting mould | Early WS | 55 | 19 | 26 | 99 | 48 | 0.48 |
| CM 11 | Casting mould | Early WS | 51 | 21 | 28 | 74 | 38 | 0.51 |
| CM 12 | Casting mould | Early WS | 48 | 26 | 26 | 99 | 43 | 0.43 |
| CM 13 | Casting mould | Late WSI | 58 | 15 | 27 | 121 | 59 | 0.48 |
| CM 14 | Casting mould | Late WSI | 52 | 31 | 17 | 78 | 35 | 0.46 |
| CM 15 | Casting mould | Late WSI | 49 | 26 | 25 | 71 | 43 | 0.61 |
| CM 16 | Casting mould | Late WSI | 51 | 24 | 25 | 100 | 43 | 0.43 |
| CM 17 | Casting mould | Late WSI | 37 | 35 | 28 | 57 | 25 | 0.43 |
| CM 18 | Casting mould | Late WSI | 35 | 31 | 33 | 55 | 28 | 0.51 |
| CM 19 | Casting mould | Late WSI | 33 | 49 | 18 | 54 | 33 | 0.61 |
| CM 20 | Casting mould | Late WSI | 66 | 14 | 20 | 98 | 44 | 0.45 |
| CM 21 | Casting mould | Late WSI | 59 | 12 | 28 | 59 | 29 | 0.49 |
| CM 22 | Casting mould | Late WSI | 38 | 27 | 34 | 73 | 28 | 0.38 |
| CM 23 | Casting mould | Late WSI | 60 | 22 | 18 | 69 | 37 | 0.53 |
| CM 24 | Casting mould | Late WSI | 62 | 13 | 25 | 62 | 29 | 0.46 |
| CM 25 | Casting mould | Late WSI | 47 | 16 | 37 | 73 | 33 | 0.45 |
| CM 26 | Casting mould | Late WSI | 63 | 7 | 30 | 61 | 35 | 0.57 |
| CM 27 | Casting mould | Late WSI | 64 | 9 | 27 | 63 | 26 | 0.42 |
| CM 28 | Casting mould | Late WSI | 57 | 14 | 28 | 72 | 42 | 0.58 |
| CM 29 | Casting mould | Late WSII | 62 | 13 | 25 | 76 | 39 | 0.52 |
| CM 30 | Casting mould | Late WSII | 51 | 27 | 22 | 81 | 39 | 0.47 |
| CM 31 | Casting mould | Late WSII | 32 | 24 | 44 | 64 | 33 | 0.52 |
| CM 32 | Casting mould | Late WSII | 57 | 18 | 25 | 57 | 26 | 0.44 |
| CM 33 | Casting mould | Late WSII | 43 | 29 | 28 | 63 | 34 | 0.54 |
| CM 34 | Casting mould | Late WSII | 59 | 25 | 16 | 66 | 39 | 0.58 |
| Po1 | Ceramic *dou* | Western Zhou | 24 | 69 | 7 | 49 | 40 | 0.80 |
| Po2 | Ceramic *dou* | Early SA | 26 | 55 | 19 | 56 | 32 | 0.57 |
| Po3 | Ceramic *dou* | Middle SA | 25 | 52 | 23 | 55 | 34 | 0.61 |
| Po4 | Ceramic *dou* | Late SA | 10 | 82 | 7 | 54 | 37 | 0.69 |
| Po5 | Ceramic *dou* | Early WS | 20 | 77 | 3 | 42 | 22 | 0.51 |
| Po6 | Ceramic *dou* | Middle WS | 27 | 67 | 6 | 48 | 31 | 0.65 |
| Po7 | Ceramic *dou* | Late WSII | 28 | 56 | 16 | 38 | 23 | 0.61 |
| Po8 | Ceramic *li* | Western Zhou | 22 | 73 | 5 | 52 | 34 | 0.65 |
| Po9 | Ceramic *li* | Early SA | 18 | 81 | 1 | 38 | 23 | 0.61 |
| Po10 | Ceramic *li* | Middle SA | 16 | 79 | 5 | 105 | 66 | 0.63 |
| Po11 | Ceramic *li* | Late SA | 9 | 80 | 11 | 34 | 19 | 0.57 |
| Po12 | Ceramic *li* | Early WS | 18 | 79 | 3 | 47 | 34 | 0.72 |
| Po13 | Ceramic *li* | Middle WS | 16 | 82 | 2 | 34 | 22 | 0.66 |

*SA stands for the Spring and Autumn; WS stands for the Warring States.

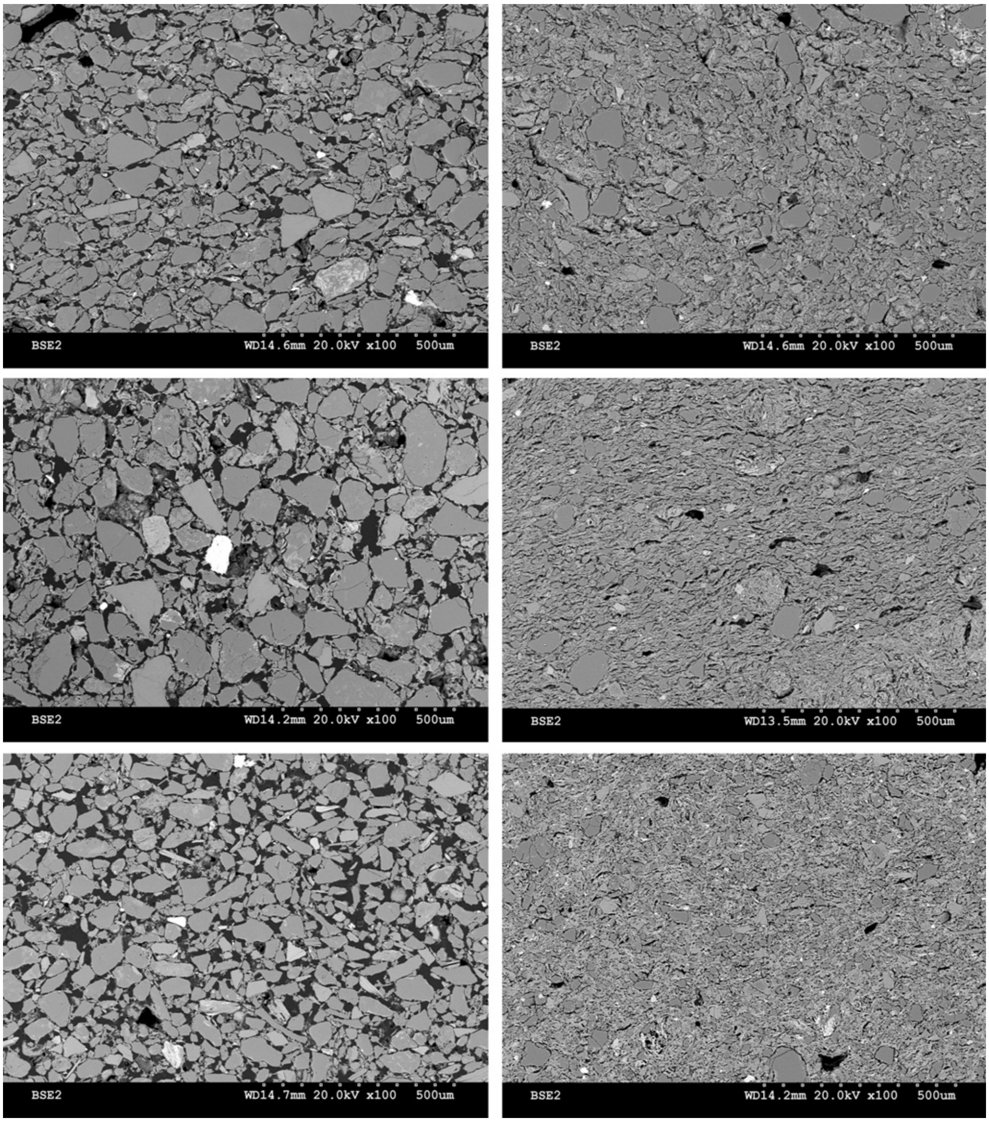

**Fig. 4.** BSE images of mould and pottery samples. The three images in the left are for moulds and the three in the right are for pottery. Left side from top to bottom: CM9, CM10, CM18; right side from top to bottom: Po1, Po4, Po9. The magnification, in all cases, is ×100.

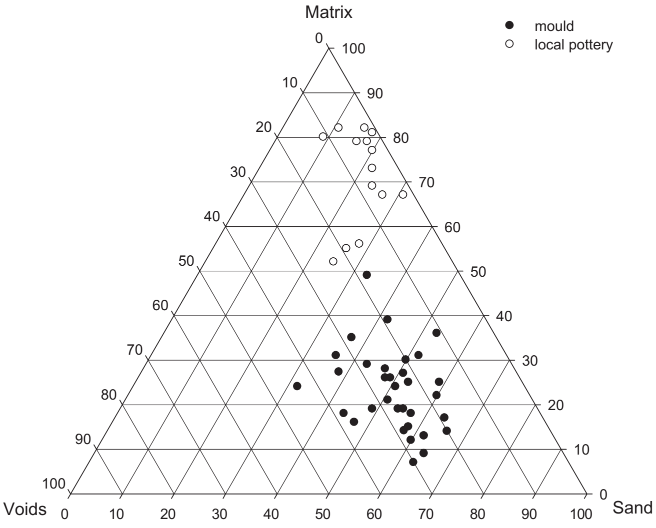

**Fig. 5.** Ternary diagram made for point counting result.

hand shaping or wheel making was the most effective way to shape them. It would have made it easy to carve motifs on the mould surfaces, but, for impressing, the lack of strength and plasticity may have caused cracks when the moulds were removed from the model. It is still not possible to say that those intricate patterns found on the mould pieces were all carved. The plasticity and strength of the moulds may have been increased by adding organic glues such as animal dung and honey (Freestone et al., 1989). More microscopic study in the future can certainly contribute to our knowledge on this aspect. According to Tan (1999), phytoliths added in with plant ash were crucial property modifiers for the casting moulds and caused the elevated silica content in their bulk composition. It is hard to comment on this statement based on the analytical results of this research, as SEM-EDS analysis of polished blocks is not the standard method for identifying phytoliths. However, no phytoliths were identified in our samples during BSE imaging, and it can be argued that phytoliths do not explain the excess silica found in mould samples as had been previously suggested. It is the high sand/clay ratio that causes the extensive high silica content found in these moulds. The exact firing temperature

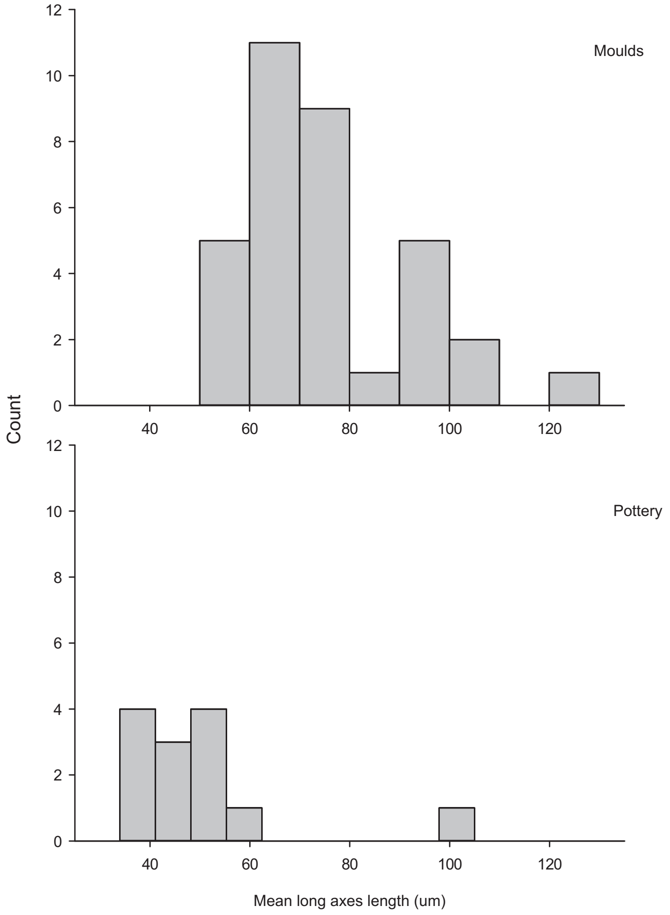

**Fig. 6.** Histogram for mean long axes length of sand particles found in moulds and pottery.

of casting moulds is hard to estimate. However, the low degree of vitrification of the matrix and the existence of large calcite particles in moulds may limit the firing temperature below that of local pottery, to possibly less than 800 °C.

The unique microstructure of casting moulds may impart them with some excellent physical properties which may have been critical for the successful casting of bronze artefacts with intricate patterns. Firstly, the shrinkage of this material during the drying and firing process is much lower than in clay-rich ceramics. The shrinkage of clay-based material is mainly caused by the loss of water held within the clay itself. However, for moulds, clay is just a minor phase in the fabric and the volume of sand particles tends not to be considerably affected by gaining or losing water. This stability in shape is crucial for casting artefacts with fine and complex patterns, which need to be made while the mould is still wet and workable. On the other hand, the use of clay-rich fabrics would cause the patterns to be distorted during the drying process. Therefore, a low shrinkage fabric is generally preferred for mould-making materials. Secondly, highly porous moulds tend to have good venting properties during casting, which can help reduce casting porosity. Despite being dried and fired, some gas and vapour will form when the molten metal is poured into the mould. If these gases cannot escape before the consolidation of the metal, they form small holes on the surface of or within the object. Some of these holes may be removed through polishing but when too many of them are present on a complicated decorated object, it will be quite hard to polish them away and the appearance of the artefact can be badly affected. Pores in the mould fabric can be an important channel for escaping gases, especially when they are interconnected. Thirdly, the high porosity combined with low firing temperature can improve the mould's resistance to thermal shock because voids and particles can stop the expansion of cracks and prevent the failure of whole structures (Kilikoglou et al., 1998; Tite et al., 2001). These properties are quite similar to those associated with modern sand casting moulds. It seems that Chinese craftsmen were aware of some of the benefits of modern moulds already more than two thousand years ago. They must have had a clear understanding of the need to acquire specific raw materials, and used their skills and knowledge to ensure that most mould piece would lead to a successful bronze cast. It can be said that other than the complex design of piece moulds, specialized mould making technology is also a cornerstone of the high quality and large scale bronze industry of the Eastern Zhou period.

This whole process requires professional knowledge and is labour intensive. Proper organization of production and division of labour should be essential for this industry. Franklin (1983) has raised the concept of co-craft production to emphasize that the bronze casting system in ancient China was not only about the production of metal but also based on the mastery of clay based material. She also proposed a model to interpret the bronze producing system in Shang period of China as the collaboration of craftspeople from different units of the bronze workshop with various expertises. Li (2007) polished her hypothesis and suggested that in the second half of the Anyang period, the central court of Shang controlled the flow of raw materials and final products but used secondary elites to organize the production of different units for the same crafts. The moulds involved in this paper are from the Eastern Zhou period, which is much later than the Shang period, and therefore it is inappropriate to apply the organization models reconstructed for Shang period directly in this case. However, the analytical result presented in this paper may still be able to shed some new light on the effort in constructing a theoretical model of bronze producing in ancient China. The use of specialized material for mould making certainly supports the hypothesis that bronze casting was based on a close cooperation between workers with different knowledge. But, in terms of techniques, mould-making should not be treated as a branch of pottery manufacture but as an integral part of the Chinese bronze casting tradition. Mould makers might use the loess similar to potters but they processed it in a quite different way to acquire a specialized raw material for making moulds. Moreover, the shaping, pattern making and firing

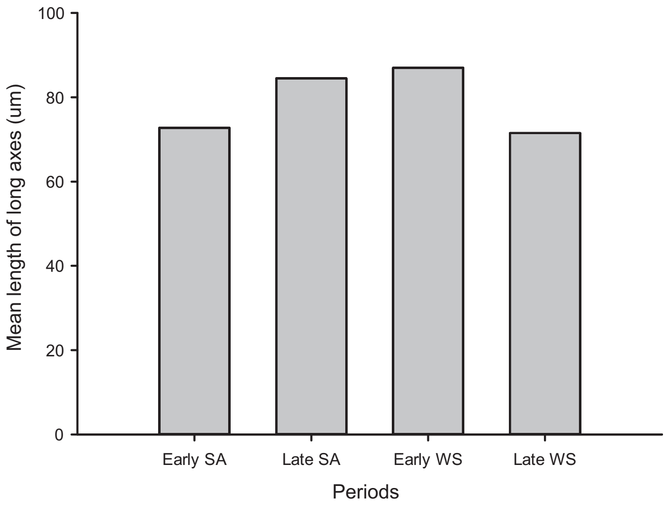

**Fig. 7.** The mean length of particles' long axes in four different periods. SA stands for the Spring and Autumn period and WS stands for the Warring States period.

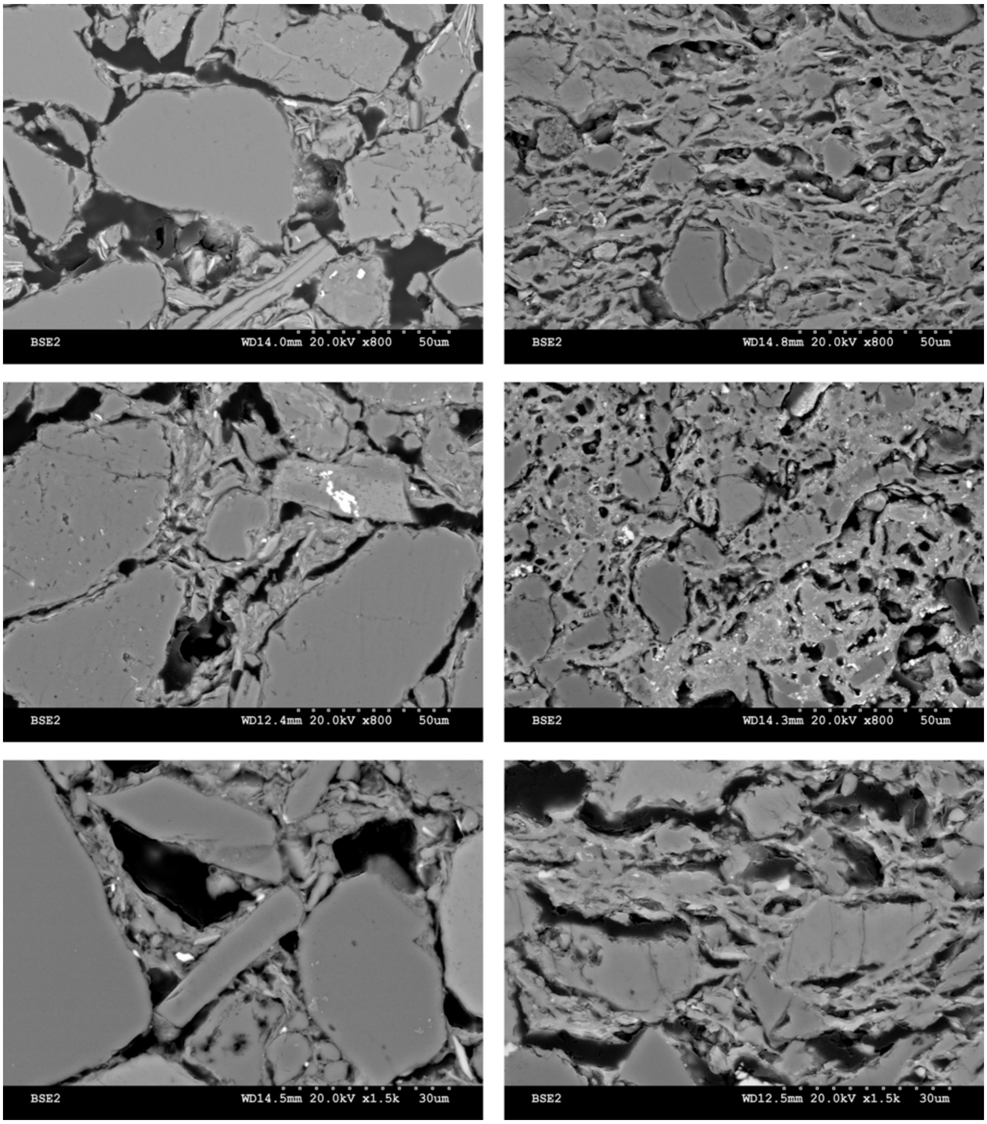

**Fig. 8.** High magnification BSE images of moulds and pottery. Left side from top to bottom: CM1, CM5 and CM34. Right side from top to bottom: Po5, Po9, Po14. The magnification of top two pairs is ×800 and for the last pair ×1500.

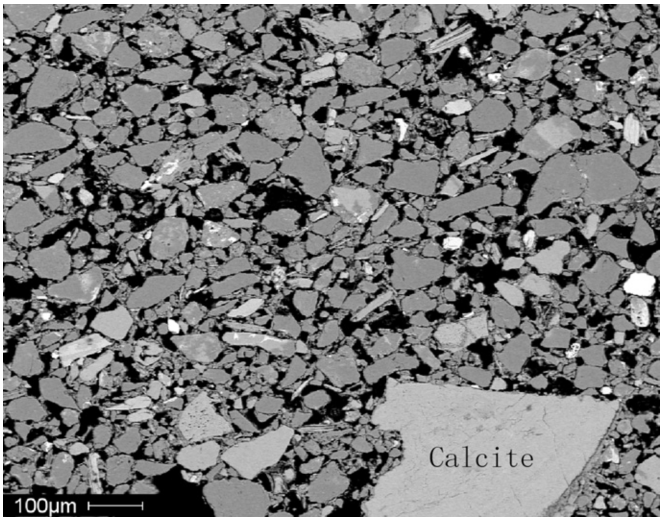

**Fig. 9.** BSE image of CM25. A calcite particle over 500 μm long can be identified at the bottom part.

technique for these two crafts appear to have been different from each other. The present data shows that within this period, most of the mould samples are similar to each other in terms of chemical composition and microstructure. This implies that the technical tradition of mould making was fully developed before the Eastern Zhou period. Therefore, it may be reasonable to suggest that mould makers were a group of independent craftsmen who held their technical traditions and passed them down through generations.

## 6. Conclusions

Through all these analyses and discussions, it can be said that bronze casting moulds from the Xinzheng site dated to the Eastern Zhou period of China were made with unique materials and using specialized technology. Low clay, high porosity, and high sand content were all beneficial to bronze casting, and mould making formed a crucial part of the ancient casting technology of China. The manufacturing of bronze ritual vessels was a prestige industry without doubt, which involved many specialized workers and complex labour organization. The importance of mould makers in this system should not be underestimated. With their professional knowledge of both mould design and raw material preparation, they may be identified as an independent group that held long-lasting technical traditions.

This research presents a new perspective on the microstructure of ancient Chinese bronze casting moulds and shows in what ways they affected the quality of bronze objects. However, this study also brings forward a number of new questions. The Eastern Zhou was certainly not the earliest period in which moulds were made with this type of material. Two previous analyses (Freestone et al., 1989; Stoltman et al., 2009) of moulds from Anyang have shown that no later than the Anyang period of the Shang Dynasty (14th century to 11th century BC), low clay and sand-rich material had been used for bronze casting moulds. Sand particles in these moulds seem finer than those in the Eastern Zhou moulds presented here.[1] However, the origin of this mould making technology at Anyang remains ambiguous. The earliest piece mould technology in China was discovered in the Erlitou period (18th century BC), but scholars still have little understanding of how those moulds were made. None of the materials that we are familiar with in ancient China since the Neolithic period share the particular characteristics of these casting moulds, which were likely either developed indigenously during the Early Bronze Age through experimentation or introduced from outside. The Bronze Age material most similar to these moulds can only be found in the faience of ancient Egypt (Freestone et al., 1989; Kleinmann, 1987; Kingery and Vandiver, 1987; Tite et al., 1983). However, these areas did not make use of this type of material to make casting moulds. There is no doubt that piece mould casting was first invented in the central plains of China, but the initial use of such a mould making material remains open to discussions.

## Acknowledgements

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[1] Judging from the BSE image in Freestone et al. (1989) and personal communication with Ian Freestone, the mean length might less than 50 μm.

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