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Characterization of early Chinese northern celadon with lead glaze from Caocun kiln within Yecheng site

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ABSTRACT

Archaeologists found a number of ceramic shards dating to the Northern Dynasties (386–581 CE) in the Caocun kiln within the Yecheng site (Hebei Province, China). A small amount of samples with grey body and green glaze are considered as representative early northern celadon. In order to investigate the raw materials, firing process and product quality about them, we collected some shards with reddish and green glazes from this kiln site, and analyzed the chemical composition, firing temperature and phase composition. The data show that the raw materials used in this kiln can be divided into two categories according to the contents of calcium and iron. The green wares have the elemental characteristics of northern ceramic products, higher level of aluminum in the body compared to the southern. The results of phase composition indicated that the quality of bodies from Caocun kiln in this work is poor, which might be related to raw materials of high-alumina and relatively low firing temperature. All the samples are covered by lead glazes of different categories, which is significantly different from most of Chinese ancient celadon. Based on the results, it can be speculated that the manufacture of ceramic wares in Caocun kiln might be complicated.

1. Introduction

Celadon is a term for a family of colored glaze itself and for ceramic wares so glazed. For them, iron generally acts as the coloring agent. However, the glaze ranges in color from yellow to green with brown. The earliest mature celadon wares were considered to be produced in Yue kiln from Zhejiang (southern province of China) during Eastern Han dynasty (24–220 CE) (Li, 1998). Since then, it became the main type of ceramic product for a long time in China. For the Chinese ancient celadon, there are obvious differences in the decoration and modeling between wares produced in south and north (Li, 1981). In addition, scientists also found that these two categories of ceramic products also differ in composition due to use of different type of raw material (Li, 1998; Guo, 1987). An accepted view is that the manufacture of northern celadon was not as prosperous as that in the southern region. In history, there are many famous representatives of celadon products from the south, such as wares from Yue (Eastern Han to Song dynasties, about 24–1279 CE) and Longquan kiln (Song to Qing dynasties, about 960–1722 CE). The southern celadon wares were usually fired in the dragon kiln with pine as fuel, while the northern were fired in the mantou kiln (smaller and more compact than the southern dragon kiln) with coal as fuel (Li, 1981; Feng, 1982). According to archaeological excavations, the production of northern celadon began in Northern Dynasties (386–581 CE) later than the southern (Li, 1981; Liu and Yuan, 1999). The development during the period from Northern Dynasties to Sui (581–618 CE) and early Tang (618–907 CE) is the early stage of northern celadon. It laid the foundation for the production of celadon from Huangbao kiln (later called Yaozhou, Shanxi province) during Tang dynasty, and even the northern Song (960–1279 CE) white porcelain. However, people know little about the early northern celadon, because there were few found kiln sites from this period and related studies.

Yecheng located in Hebei Province served as the capital of six dynasties about 370 years (204–577 CE). During this period, it was the political, economic and cultural center of northern China. From 1983, archaeologists from Institute of Archeology, Chinese Academy of Social Sciences and Hebei Provincial Cultural Relics Research Institute jointly carried out survey, exploration and excavation work. In 2014–2015, archaeologists took excavation at Caocun kiln within this site in order to study the regional ceramic manufacture. Following the past view, archaeologists thought that this kiln mainly produced low-temperature pottery during Northern Dynasties. However, archaeologists unearthed some shards of rare earliest northern celadon in this site. In this paper, we analyzed their chemical composition, firing temperature and phase

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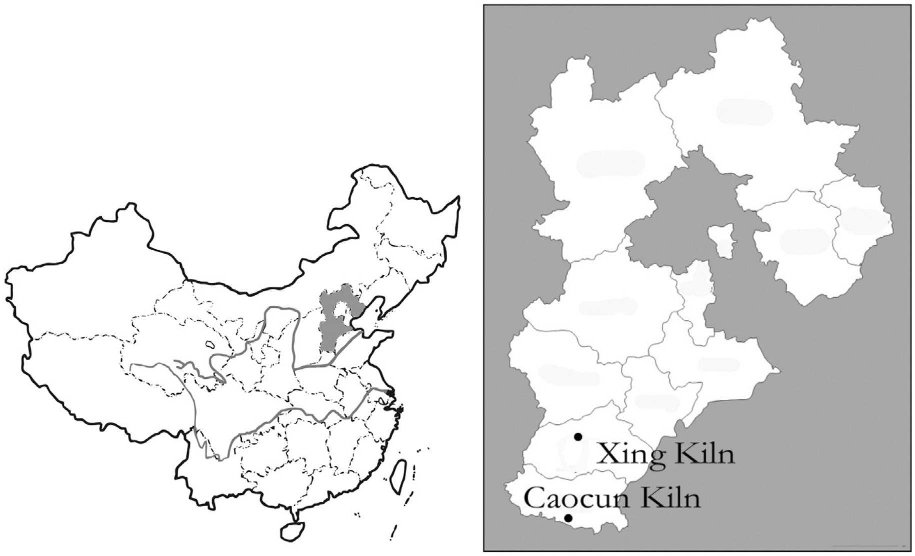

Fig. 1. The geographical location map of Hebei province in China (left) and Caocun and Xing kiln in Hebei (right).

composition. The results might help to learn more about the manufacture technologies of earliest northern celadon, such as use of raw material and glazing. In addition, we think that this paper could also provide reference data for the provenance research on early northern celadon.

2. Materials and methods

2.1. Samples and preparation

The kiln within Yecheng site was found in Caocun village located in Linzhang County, Hebei Province (Fig. 1). The collected samples include 13 pieces of ware shard (HBYC), 6 glazed supporting nails and 2 pillars of large objects (HBYCZ). The information and images of samples were shown in Table 1 and Fig. 2. Some samples have a reddish body, and the glaze is reddish/brown. They are the commonest unearthed ceramic shards in this kiln site. The rest covered by green glazes are considered by archaeologists as the early northern celadon. They are marked as green wares in this paper.

All the samples were washed three times in an ultrasonic cleaner

Table 1
Basic information for samples from Caocun Kiln.

Sample	Type	Color of glaze	Color of body
HBYC01	Bowl	Reddish/brown	Reddish
HBYC02	Bowl		Reddish
HBYC03	Bowl	Green	Grey
HBYC04	Bowl	Green	Grey
HBYC05	Bowl	Green	Grey
HBYC06	Bowl	Green	Grey
HBYC07	Bowl	Green	Grey
HBYC08	Bowl	Green	Grey
HBYC09	Bowl	Green	Grey
HBYC10	Bowl	Green	Grey
HBYC11	Bowl	Green	Grey
HBYC12	Bowl	Green	Grey
HBYC13	Bowl	Green	Grey
HBYCZ01	Supporting nail	Green	Grey
HBYCZ02	Supporting nail	Reddish/brown	Reddish
HBYCZ03	Supporting nail	Green	Grey
HBYCZ04	Supporting nail	Green	Grey
HBYCZ05	Supporting nail	Green	Grey
HBYCZ06	Supporting nail	Reddish/brown	Grey
HBYCZ07	Pillars of large objects	Green	Grey
HBYCZ08	Pillars of large objects	Green	Grey

with deionized water and dried at 95 °C. After the composition measurement of body and glaze, the sample was polished to remove the surface glaze with emery grinding wheel. Then, some samples were ground into powders for diffraction experiments, while the bulk samples were used to measure the thermal expansion curve and water absorption.

2.2. Methods

The composition of samples was analyzed with the Energy Dispersive XRF (EDXRF, Eagle III μProbe, EDAX, USA) at the Institute of High Energy Physics. The spectrometer was equipped with a Mo tube and a 125 μm Be window. It worked at a voltage of 40 kV, current of 250μA for body and 120μA for glaze. The X-ray tube generating beam spot size was selected to be $\Phi = 1$ mm. The spectrum of each sample was recorded with a live time of 300 s. The detector is a liquid-nitrogen-cooled Si (Li) crystal with a 160.3 eV at Mn Kα. The analysis was carried out in a vacuum environment. The software employed for analysis is the program VISION32 associated with the instrument, and we used in-house ceramic reference material for quantitative analysis of the body and lead-rich glaze (Ma, 2014; Zhu, 2008). The precision of the elemental measurement data varies from several (MnO) to hundreds (Al_2O_3, SiO_2, K_2O, CaO, TiO_2, Fe_2O_3, CuO, PbO_2) of PPM. The data of Na_2O and MgO is provided as references because of the poor fluorescence yields and low counting obtained for characteristic X-ray radiation. The composition value (Tables 2 and 3) was taken the mean data of at least 3 different measurement points.

The X-ray diffraction (XRD) patterns were obtained on a X-ray diffractometer (D8 Advance, Bruker, Germany) with Cu-Kα radiation ($\lambda = 1.5406$ Å). Operating conditions for the XRD was 40 kV of voltage and 40 mA of current. The samples were continuously scanned from 10°–60° (2θ), with a step size of 0.02° and a time per step of 1.0 s. To prepare the sample, the bodies were ground into powder. The minerals were identified using the ICDD (International Center for Diffraction Data) database.

The dilatometer used in this research was a DIL402PC (NETSCH, Germany) thermal expansion instrument at The Palace Museum. The samples were cut into pieces with size 25 × 5 × 5 mm. Samples were heated at a rate of 5 °C/min in N_2 atmosphere. The sample was washed in the ultrasonic cleaning machine. Then, it was dried in oven at 110 °C to constant weight. Water absorption was measured via boiling in water for 3 h, using a digital scale with a resolution of 0.01 g.