



Development of Chinese barium copper silicate pigments during the Qin Empire based on Raman and polarized light microscopy studies



Yin Xia ^{a,*}, Qinglin Ma ^b, Zhiguo Zhang ^b, Zhendong Liu ^c, Jian Feng ^d, Anding Shao ^e, Weifeng Wang ^a, Qianli Fu ^a

^a Key Scientific Research Base of Ancient Polychrome Pottery Conservation, SACH, Emperor Qin Shihuang's Mausoleum Site Museum, Xi'an 710600, China

^b Chinese Academy of Cultural Heritage, Beijing 100029, China

^c The Institute of Archaeology CASS, Beijing 100710, China

^d Xi'an Archaeology Institute, Xi'an 710068, China

^e Shaanxi Provincial Archaeology Institute, Xi'an 710068, China

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ABSTRACT

Ancient Chinese objects dating from the Warring States period (475–221BC) to the Han dynasty (206BC–220AD), including faience beads, wall paintings, polychrome decoration on pottery, stone and bronze objects, were studied using Raman microscopy and polarized light microscopy (PLM). Chinese Blue ($\text{BaCuSi}_4\text{O}_{10}$) and Chinese Purple ($\text{BaCuSi}_2\text{O}_6$) were identified, and additionally dark blue particles of a new pigment $\text{BaCu}_2\text{Si}_2\text{O}_7$ were also detected in purple decoration. Based on PLM observation, Chinese Blue, Chinese Purple and $\text{BaCu}_2\text{Si}_2\text{O}_7$ frequently coexist in single polychrome samples and cannot be individually identified using other analytical methods because of the minute amount that these chemicals present: sometimes comprising just one or two particles. The use of Raman spectroscopy can identify such small presences. This paper illustrates the man-made barium copper silicate pigments along the Qin historical development and shows the advantage of PLM combined with Raman spectroscopy to analyses pigments giving us an overall picture of the dates and the places where barium copper silicate pigments were used in ancient China. Their origin, decline and manufacturing techniques are also discussed.

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1. Introduction

Han Blue and Han Purple or Chinese Blue and Chinese Purple, chemical compounds $\text{BaCuSi}_4\text{O}_{10}$ and $\text{BaCuSi}_2\text{O}_6$ are two ancient Chinese man-made pigments named by E. FitzHugh (FitzHugh and Zycherman, 1983, 1992) and H. Berke (Berke and Wiedemann, 2000; Berke, 2002, 2007). Though natural $\text{BaCuSi}_4\text{O}_{10}$, a blue mineral, (Giester and Rieck, 1994) and natural $\text{BaCu}_2\text{Si}_2\text{O}_7$, a dark blue mineral, (Yang et al., 2013), were found in South Africa, these and $\text{BaCuSi}_2\text{O}_6$ are thought to have been artificially prepared by Chinese craftsmen. Based on recent research results (Thieme et al., 1995; Zuo et al., 2003; Ma et al., 2006, 2008; Cheng et al., 2007; Liu et al., 2007; Xia et al., 2008; Zhang et al., 2010; Cai et al., 2011), Chinese Blue and Chinese Purple were used between the later Western Zhou and the Eastern Han Dynasty (771BC–AD220). Large amounts of archaeological objects dating from the Warring

States period to the Han dynasty (476BC–AD220) have been excavated. It is clear from these finds that the use of barium-copper silicate pigments ceased at the end of the Eastern Han Dynasty, presumably because of a change of production techniques and preferred materials. Some previous research papers have offered incorrect analyses of these two pigments (Hui et al., 2007; Xi'an Archaeology Institute, 1999; Shaanxi Provincial Archaeology Institute and Xi'an Jiao-tong University, 1990). This paper presents new conclusions after studying twenty-two samples gathered from five provinces in China and the investigations using Raman microscopy and polarized light microscope.

2. Materials and methods

2.1. Archaeological samples

Twenty-two samples were selected for analysis, including faience beads and other decorative objects, namely wall paintings, polychrome decoration on pottery, and stone and bronze objects (see Table 1 and Fig. 2).

* Corresponding author.

E-mail addresses: xiayin2000@hotmail.com, xiayin2000lhj@163.com, 576510917@qq.com (Y. Xia).

Table 1
Sample description and PLM results.

No.	Sample no.	Time	Place	Object description length/diameter (cm)	PLM ^b Result
1	MAHP08-P	Warring State Period (476–221BC)	Gansu Province (Li et al., 2002)	Purple faience octagonal stick 2.3/1.2	CP(20–40%) + CB(<1%) + DB(<1%) + LW(A)
2	MAHP10-P	WSP ^a	Gansu Province (see above)	Purple faience octagonal stick	CP(20%) + CB(5%) + LW(A)
3	MAHB14-B	WSP	Gansu Province (see above)	Blue faience bead	CB(40%) + LW(A)
4	ZJC M6-P	late WSP	M6 Zhang Jiachuan, Gansu Province (Zhou et al., 2009)	Purple faience bead	CP(20–30%) + CB(3–5%) + DB(1–2%) + LW(A)
5	LY-OctS-P	late WSP	C1M3943 Xi-gong district, Luoyan, Henan Province	Blue-purple octagonal stick 7.5/0.5	CP(20–30%) + CB(3–5%) + DB(<1%) + LW(A)
6	YD M47:4-B	Late WSP (Qin state) to Qin Dynasty (476–210BC)	M47 Xi'an Post-telecom College construction site	Light blue earring 2.1	CB(40%) + LW(A)
7	YD M106:2-P	Late WSP (Qin state) to Qin Dynasty	M106 Xi'an Post-telecom College construction site	Dark-purple octagonal stick 4.1/0.9–1.0	CP(20–30%) + LW(A)
8	QXYG-B	WSP (Qin state) to Qin Dynasty	Qin Xian-yang Palace (Shaanxi Provincial Institute of Archaeology (2004))	Wall painting fragment with light blue hue	CP(20–30%) + CB(<1%) + DB(<1%) + LW(A)
9	QY-P	Qin Dynasty (246–210BC)	Qin terra cotta Army Pits (Xia et al., 2009)	Terracotta figure with purple under-robe	CP(20–30%) + CB(<1%) + DB(<1%) + LW(A)
10	HPP-P	Western Han Dynasty (206BC–AD 25)	Xi'an Archaeology Institute collection	Polychrome pottery	
11	HYL-M53-P	WHD(156–141BC)	M53:12, 2006 Hailang Chemical Industrial Factory construction site, Han Yang Mausoleum complex pits	Terracotta figure with purple	CP(20–40%) + CB(3–5%) + DB(<1%) + LW(A)
12	CLG-B	WHD	Chang-le Palace, Xi'an (Han Chang'an City Archaeological Team, 2006)	Wall painting fragment with light blue hue	CB(3–5%)
13	CLG-P	WHD	Chang-le Palace, Xi'an (see above)	Wall painting fragment with dark purple hue	CP(20–30%) + CB(3–5%) + DB(<1%)
14	TDG-P	WHD	Xi'an Archaeology Institute collection	Copper belt hook with purple decoration	
15	LGD-B	WHD	Xi'an University of Technology (The Cultural Relics Protection and Archaeology Institute of Xi'an, 2006)	Wall painting, light grey or blue hue	CB(2–3%) + CP(<1%)
16	MC-P	WHD King Zhong-shan (154–113BC)	Man-cheng Han tomb, Hebei Province	Polychrome pottery dish, light purple hue	CP(5–10%) + CB(3–5%)
17	BQQ-P	Mid-late WHD (86–49BC)	Bu Qian-qiu Han tomb, Luo-yang (The Luoyang Museum, 1977)	Wall painting, light purple hue	CP(30–40%) + CB(3–5%) + DB(<1%) + LW(A)
18	WSH-B	early WHD King Ji-nan (164–154BC)	Wei-shan Han tomb, Zhang-qiu, Shandong Province (Xia et al., 2008)	Polychrome pottery figure, light blue hue	DB(10%) + CP(3–5%)
19	QZ-P	early WHD (206BC–AD 25)	Qing-zhou Han tomb, Shandong Province	Polychrome pottery figure, light purple hue	CP(10–20%) + CB(<1%) + DB(<1%)
20	DP M1/2-B	early Eastern Han Dynasty (AD25–220)	Dong-ping Han tomb, Shandong Province (Xu et al., 2009)	Wall painting on stone ground, light blue hue	DB(10–20%) + CP(3–5%) + DB(<1%)
21	YL-M2-P	EHD (AD25–220)	Yang-Qiao-pan, Yu-lin, Shaanxi Province	Pottery bowl with purple polychrome	CP(3–5%) + CB(<1%) + DB(<1%)
22	05 MG M2-B	EHD Emperor An (AD107–125)	Guan-zhuang Han tomb, Mi-zhi, Shaanxi Province	Painting on stone ground, blue hue	Needlelike CB(50–60%) Plat

^a WSP=Warring State Period, WHD=Western Han Dynasty, EHD = Eastern Han Dynasty.

^b PLM=Polarized Light Microscopy, CP=Chinese Purple, CB=Chinese Blue, DB = BaCuSi₂O₇, LW = Lead White.

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