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Tracing the origin of blue and white Chinese Porcelain ordered for the Portuguese market during the Ming dynasty using INAA

M. Isabel Dias [a,*], M. Isabel Prudêncio [a], M.A. Pinto De Matos [b], A. Luisa Rodrigues [a]

[a] Campus Tecnológico e Nuclear/Instituto Superior Técnico, Universidade Técnica de Lisboa, EN 10 (Km 139,7), 2686-953 Sacavém, Portugal
[b] Museu Nacional do Azulejo, Rua da Madre de Deus no 4, 1900-312 Lisboa, Portugal

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ABSTRACT

The existing documentary history of Chinese porcelain ordered for the Portuguese market (mainly Ming dynasty.) is reasonably advanced; nevertheless detailed laboratory analyses able to reveal new aspects like the number and/or diversity of producing centers involved in the trade with Portugal are lacking.

In this work, the chemical characterization of porcelain fragments collected during recent archaeological excavations from Portugal (Lisbon and Coimbra) was done for provenance issues: identification/differentiation of Chinese porcelain kilns used. Chemical analysis was performed by instrumental neutron activation analysis (INAA) using the Portuguese Research Reactor. Core samples were taken from the ceramic body avoiding contamination form the surface layers constituents. The results obtained so far point to: (1) the existence of three main chemical-based clusters; and (2) a general attribution of the porcelains studied to southern China kilns; (3) a few samples are specifically attributed to Jingdezhen and Zhangzhou kiln sites. In a chronological point of view, for the studied samples we assist to an increasing improvement of the production procedure from late 15th till the 17th centuries of the Chinese porcelains sent to Portugal, especially enhanced by the association of late porcelains with refining processes of the original raw material, consistent with removal of more heavy minerals. In the case of some samples a kiln attribution was possible, but for the majority of the samples we haven't found yet the specific kilns.

1. Introduction

Chinese porcelains and Portuguese faience wares were the main subject of a FCT research project (Dias, 2006, in press), which had as major achievement the increase of the knowledge of the Portuguese movable assets from the 16th–18th centuries. In this project public research laboratories and museums worked together in an interdisciplinary approach. In the present work results obtained for Chinese porcelains ordered for the Portuguese market (15th–17th cent.) are presented. The first stage of the work comprised a detailed typological/decorative classification of porcelain shards which was used for a first chronological and provenance approach.

Nevertheless we are aware that this customary method for dating ancient Chinese porcelain based on shapes and decorations by its changes in the different dynasties, it is sometimes problematic, as visual features may vary only a little. Also they may be imitated in later dynasties. Therefore, chemical analyses have played an important role in the study of these ancient ceramics. As enhanced by Li et al. (2003) most ancient Chinese kilns used clays mined from local areas and some geochemical and mineralogical differences may be expected. On the other hand, also other variables may influence composition, like production techniques, including processing, washing and mixing diverse raw materials.

Regarding the Chinese porcelain ordered for the Portuguese market, the existing documentary history is reasonably advanced; nevertheless it is necessary to proceed with detailed laboratory analyses so that new relevant aspects can be unraveled like the number and/or diversity of producing centers involved in the trade with Portugal. The spreading of Chinese porcelain in Europe is a consequence of the Portuguese naval expansion to the Orient. Returning from the first great trip, Vasco da Gama will have brought exemplars to offer to king D. Manuel who will have become interested in fine porcelains; thereafter, in 1507 the Portuguese Monarch ordered several objects. In this way, the armillary sphere, the royal weapons and religious symbols like the monogram IHS (Iesus Hominum Salvator) encircled by a crown of thorns, were immortalized on the first porcelains manufactured specifically for the Portuguese market and dated about 1520. Between 1540 and

* Corresponding author. Tel.: +351 219946222; fax: +351 219946184.
E-mail address: isadias@ctn.ist.utl.pt (M.I. Dias).

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1552 objects with registrations in Portuguese were ordered, and the difficulties felt by Chinese craftsmen in copying alphabetic inscriptions, strange for them, are perceptible in those pieces. They certify that a clandestine trade proceeded between Portugal and China despite the cut of official relations. To these decorative patterns others were joined like the armory of noble families and emblems of religious Orders dedicated to consolidate the missions of the Portuguese empire in the Orient. This particular anthology of objects, expressing that the Portuguese were pioneers in the commerce of porcelain, is spread all over the world and treasured in national and foreign museums, and private collections (Harrison-Hall, 2001; Pinto de Matos, 2002/03, 2003; Pinto de Matos and Salgado, 2002).

Using instrumental neutron activation analysis (INAA) major (a few), minor and trace elements contents of around 30 porcelains wares excavated from different sites in central Lisbon and Coimbra were determined. By virtue of various statistical methods we attempt to find out the provenance characteristics/kilns attribution of the Porcelain wares ordered for the Portuguese market, found in different Portuguese archaeological excavations, in terms of trace elements composition.

Blue and white porcelains are undoubtedly considered the most important of the Chinese porcelains produced along the various dynasties, from the Yuan, Ming to the Qing. One of our purposes is to locate the origin(s), at a regional scale, of the porcelains found in Lisbon and Coimbra archaeological excavations. To do so, it is assumed that there is already a chemical fingerprint of Chinese kilns. On the other hand, if not possible to attain such an ambitious goal, at least contribute to figure out the diversity of productions centers involved in the commercial trade to Portugal.

In ancient times (usually before 1900 AD), it was very difficult to transport raw materials from one place to another in China, so potters usually employed local materials to manufacture pottery and porcelain and also employed specific raw materials and/or batch composition in certain time periods, which provides a very good scientific basis for employing element characterization in provenance and dating studies (Leung and Luo, 2000). Another important feature is that chemical composition difference between samples from different kiln sites is usually much greater than that between samples from the same kiln but different time periods, because a change of emperor did not influence the raw materials and/or batch composition significantly. So, in this work, considering that same materials might have been used in a certain kiln along dynasties, we will also compare chemical composition of our samples with others from Yan to Qing dynasties, in certain kilns. So, we can better ascertain kiln(s) identification of porcelains ordered for the Portuguese market.

We are aware that, as previously noted by Leung and Luo (2000), in provenance studies, comparing data from different researchers/and institutions/and methods is always a challenge, with risks, because the differences in equipment, measuring conditions, standard samples, etc., strongly influence the measurement results. So, in order to reduce and eliminate the influence of these difficulties and to better establish a comparison between chemical results obtained from diverse methods, and in different laboratories, the use of element ratios is recommended, as well as normalized values for example relatively to the mean composition of the continental crust. It is the recommended way, complementing typological classification studies, to attribute provenance.

Thus, the main goals of this work are: (i) the chemical characterization by INAA of the ceramic body of Chinese porcelains found in Lisbon and Coimbra (Portugal) archaeological excavations (15th–17th centuries, Ming–early Qing dynasties); (ii) establishment of correlation between chemical composition and typological/decorative features; and (iii) identification of the Chinese kilns/region of production.

2. Materials selection and methods

The sample selection methodology for the present work was as following:

Fifty excavation reports were consulted and around 100 boxes of ceramic material found in various parts of Lisbon were selected for detailed macroscopic study. After the inventory of this huge amount of material, about 50 shards of different objects were selected for compositional analysis. Thus the selection took into account the representativeness of major historic-stylistic-chronological-decorative features macroscopically identified, and also the potential production centers attribution and historic problematic.

The materials studied comprise samples of Chinese porcelain (15th–17th cent.) selected from archaeological findings from Central Lisbon excavations (yielded by the company Era Arqueologia SA) and Coimbra excavations (at the Museum of Santa Clara a Velha). Samples from the National Museum of Ancient Art collection, as well as from excavations taken in this Museum were also studied.

In Table 1 together with photograph, sample reference and archaeological site, the typological/stylistic/visual data are presented, as well as geographical and chronological frameworks.

For the study of compositional ceramic body of porcelain, sampling was conducted with a diamond drill with a small diameter (2 mm $< \varnothing <$ 5 mm), which obtained cores depends on the thickness of the fragments. This was done to avoid any contamination resulting from the composition of the glaze and decoration, as well as from the extraction tool itself (Fig. 1). It is important to enhance that only for a few shards it was possible to obtain appropriate cores to further analyses. So, only 24 shards were sampled for analysis, because of the thinness and hardness of most of them.

Chemical analyses were done by instrumental neutron activation analyses (INAA), determining major and trace elements contents with very good accuracy and precision (in general <5%). Core samples were ground into a fine powder in an agate mortar and then dried in an oven at 100 °C for 24 h and stored in desiccators until the samples can be weighed. Irradiations were done in the core grid of the Portuguese Research Reactor (Sacavém), as neutron source. GSD-9 (sediment) and GSS-1 (soil) of the "Institute of Geophysical and Geochemical Prospecting" (IGGE) were used as reference materials. The reference values were taken from data tabulated by Govindaraju (1994). The following elements were determined: Na, K, Mn, Fe, Sc, Cr, Co, Zn, As, Ga, Br, Sb, Rb, Cs, Ba, La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, Lu, Hf, Ta, Th, U. Samples and standards were irradiated together in the core grid of the Portuguese Research Reactor (ITN, Sacavém) for 2 min (short irradiation) and 7 h (longer irradiation). The irradiations are carried out in two positions: A short, pneumatic-tube irradiation is 2 min long, at a thermal neutron flux of 2.6E12 n/cm^2/s; the long irradiation is 7 h long, at a thermal neutron flux of 4.0E12 n/cm^2/s, epithermal 3.7E10 n/cm^2/s and fast flux of 1.6E10 n/cm^2/s. The gamma-ray analysis is performed using a Ge γ spectrometer consisting of a 150 cm^3 coaxial detector and a low energy photon detector (LEPD), connected through Canberra 2020 amplifiers to Accuspec B (Canberra) multichannel analyser. This system has a FWHM of 1.9 keV at 1.33 MeV (coaxial Ge detector), of 300 eV at 5.9 keV and of 550 eV at 122 keV (LEPD) (more details can be found in Dias and Prudêncio, 2007).

A multivariate statistical analysis of chemical data was done by using the Statistica data analysis software system (StatSoft, Inc., 2011).