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CHARACTERIZATION OF MORTARS FROM THE NUR JAHAN TOMB LAHORE, PAKISTAN

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Abstract

This work describes the chemical and mineralogical characteristics of historic pozzolanic lime mortars and modern cement mortars sampled from the Nur Jahan Tomb in Shahdara complex, Lahore, Pakistan. The mortars were studied through petrographic (polarized optical microscopy), mineralogical (XRD) and elemental (SEM-EDS, XRF) analyses. Crushed brick as pozzolanic additive created hydraulic mortars in the Mughal and British period. We also detected the prevalent use of cement-based mortar for the repairs. These compositional differences highlight the complexity of hydration processes. The formation of different hydration products in historic and modern mortars makes them incompatible and further accelerated deterioration and loss of historic fabric.

Keywords: *Mortar; Mughal; Pozzolana; C-S-H phases; XRF; SEM-EDS; LA-ICPMS*

Introduction

Mortars have been widely used for bedding, jointing and rendering of brickwork and stonework since remote times. Mortars are composite materials made of one or more types of binders, different kinds of aggregates, water and optionally additives- natural or artificial ones [1, 2]. The mortars are mostly classified according to the type of binder. Mortars based on lime prevailed from Roman times until the 18th century, when they were substituted by new hydraulic binders (such as soil cement binders) and by Portland cement in the 19th century [1, 3].

The 17th century Nur Jahan tomb is one of the three most beautiful monumental tombs, in the Shahdara complex, built during the reign of Emperor Shah Jahan. The tomb was constructed in Mughal characteristic style of brick and lime mortar masonry core faced with red sandstone and white marble inlay works. The tomb has undergone many interventions that determined it's deterioration (Fig. 1a and b). The incompatibility of the new materials used for restoration has altered the original features and deteriorated the facades as shown in Figure 1.

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Fig. 1. Deteriorated façades of Nur Jahan Tomb: a) Rear façade with cracks, b) Biological growths, c) Jointing locations of historic and repair masonry, d) Lime-Cement mortar interface, e) Cracks in repair masonry and f) Cracking and rusting of repair works with cement mortar.

The chemical and mineralogical characterizations were carried out to investigate and compare the binder, aggregate and additives of the historic lime mortars of the tomb with the mortars added later for restoration. Initial observations categorized three major types of mortars, which were further investigated to specify their characteristics. The chemical analysis for the major and trace elements was carried out by XRF and LA-ICPMS techniques. These were complemented with SEM-EDS for chemical composition of products generated during hydration processes (reaction rims, pozzolanic products). XRD and petrography were done for mineralogical characterization.

Experimental

Sampling

Mortars sampling was carried out under the supervision of conservators in charge. The size of each sample was worked out in order to complete all analyses and to reserve some material for future studies. 5 samples, representing different periods, were collected from the tomb building. Three major periods were identified:

(a) 17th century mortars - originally used during the Mughal period construction;
(b) 19th century mortars - collected from the places restored under the British rule;
(c) 20th century mortars - used for post 1947 restoration works.

Analytical Techniques

The samples were prepared according to the prescribed standard procedure for mortar analysis [2, 4]. 30μm thick sections were prepared for petrographic microscopy to ensure the identification of all constituent phases.

The JEOL JSM-6300 LA scanning electron microscope coupled with energy dispersive X-ray analyzer (EDS) was used for studying the texture, morphology and alteration products in the selected samples.

Samples were fixed in epoxy resin (Laromin C 260-40 and Harter DY 26 SP-60) using a vacuum chamber during drying process. Afterwards, samples were heated up to 50°C in oven, to make epoxy plugs with Labo Press-3. These plugs were finely polished and carbon coated to avoid the interference with the electron beams during SEM-EDS measurements.

Parts of the sample were grounded in an agate mill to provide powders for XRD and XRF analysis [5-8]. The diffraction patterns were collected using Bruker, AXS D8 Advance powder diffractometer (CuKα radiation) equipped with the Lynxeye super speed detector system.

Glass beads were casted with the homogeneous mixture of dehydrated sample powder with a flux ($Li_2B_4O_7$) by Perl 'X3 (automated glass bead casting machine) using platinum crucibles. The fused glass beads were analyzed with wavelength dispersive X-ray florescence spectrometer (WD-XRF, Axios, PANalytical) equipped with 5 diffraction crystals for major elemental analysis. The laser ablation microprobe with inductively coupled plasma-mass spectrometry (LA-ICPMS) was performed on the glass beads for the trace elemental composition. The instrumentation used was a Coherent CompexPro Excimer ArF laser with beam homogenization and the Elan 6100 DRC ICP-MS (Perkin Elmer) equipped with the software SILLS.

Results and Discussion

Macroscopic Description of Mortars

Mortar samples were categorized into types A, B and C according to their macroscopic observations. The three types represent three major periods in the tomb history: 17th century, the original construction in Mughal times; 19th century, restoration intervention during the British rule; and 20th century, the recent restoration works.

Type A mortars includes dull, white-coloured calcitic fragments (locally known as Kankar). Crushed brick pieces (locally known as Surkhi) and brick kiln slag occasionally used as aggregates, it is characterized as pozzolanic mortar [4]. Two types of A samples (#17 and #19) were characterized in this study.

Type B mortars is also a pozzolanic mortar with light brownish colour and is characterized by a high amount of calcitic nodule fragments that can be seen with the naked eye, with a small fraction of sand and finely crushed bricks as aggregate. One sample of type B (#48) was analyzed for the study.

Type C mortars are Portland cement mortar based on cement paste with sand as aggregate. Two type C samples (#107 and #108) were studied, one dark reddish and the other dark grey, respectively.

Microscopic Petrographic Description of Mortar

The binder aggregate ratios are 1:1 for the Mughal, 1:3 for British and 1:6 for the modern mortars [5, 7, 9]. The petrographic observations were compared and correlated with the X-Ray diffraction analysis. The crystalline phases are identical in type A and B samples. Gypsum, found in type A and C samples, is probably due to weathering process of carbonate sulphatation [1, 3, 4].

Type A mortar

Petrographic observation of the type A lime mortars (locally known as red lime mortars made from Kankar burning) shows that the aggregate is mainly composed of crushed brick and few calcitic fragments (Kankar). The crushed brick also possesses the pozzolanic properties that can be identified from reaction rims at the interface of the lime binder and brick elements. The calcitic fragments (Kankar) are unburnt pieces left after the burning of calcite for lime production [4]. Brick kiln slag, found in sample #17, suggests its use as filler in addition to jute fibers. The addition of fibrous material was a traditional technique in the Indian subcontinent to provide adhering strength to the mortar constituents [4, 7]. The mineralogical composition included calcite as the main binder in addition to clay, silica quartz, albite, dolomite, chlorite, hematite and K-feldspar. Rare biotite and muscovite were found.

Type B mortar

The observation of the type B sample revealed high ratio of calcitic fine aggregates (in the form of crushed Kankars) and lime as binder with finely crushed brick fractions used as a pozzolanic material. No fibrous material or slag was found. The mineralogical phases were the same as in type A mortars.

Type C mortar

The mineralogical phases in type C cement are mainly quartz and few calcite crystals embedded in the cement paste. Hematite was observed in the dark reddish sample (#107). Reaction rims were noted in all the samples around the pozzolanic materials. These rims are due to the formation of morphological types of C-S-H (calcium silicate hydrates) gels which were further studied in detail with SEM-EDS.

Chemical Analysis and Mortar Composition

The old mortar samples were analyzed by XRF to determine the major and trace element contents [7, 8]. Samples were further subjected to LA-ICPMS for accurate concentration measurements [9, 10]. The chemical data clearly distinguished the different mortars [11].

Type A mortars were composed of 32-35% calcite (CaO + LOI) and over 40% silica with 8% alumina. The Iron content is 3-4%, characteristics of the historic lime mortars used in the past [4, 11]. The soda and potash values range from 1 to 3% while the magnesia content is about 2%.

Type B mortar is rich in calcite (about 65%-CaO + LOI), has a silica content of about 21% and alumina 5%. The iron content is 2.3%, soda and potash range from 1-1.3% and magnesia 1.5%.

Type C mortar contains 20-25% (CaO + LOI) and about 60% silica. Alumina ranges from 6-7% with variable amounts of soda (1.5-1.7%) and potash (1.5-2.7%). The hematite content was 4.6% in sample #107 and 2.8% in sample #108.

The chemical nature of trace elements and their relative concentrations (Fig. 2) indicated that there is a not a significant compositional difference between the three studied mortar types. Therefore, it is possible that their raw materials have the same origin and zones were employed. The trace elemental composition can be further used for detailed studies on the provenance of the raw materials [12, 13].

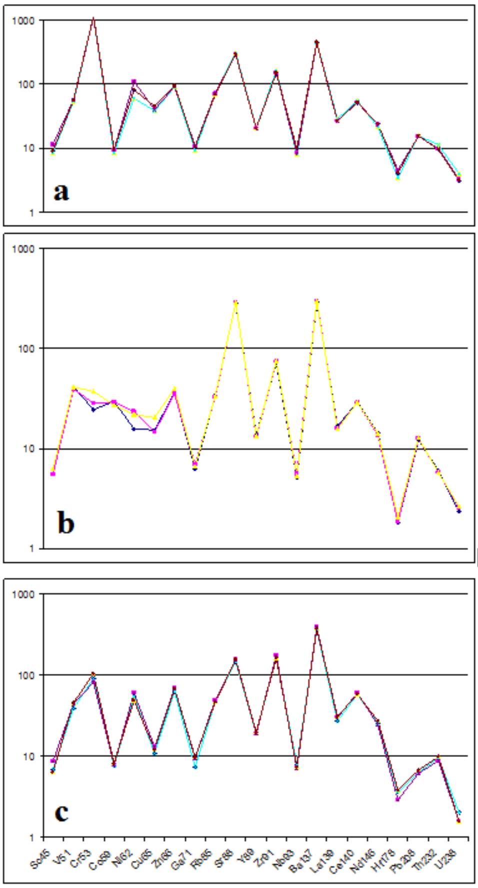

Fig. 2. Trace element composition determined from LA-ICPMS analysis: a) Type-A, b) Type-B, c) Type-C

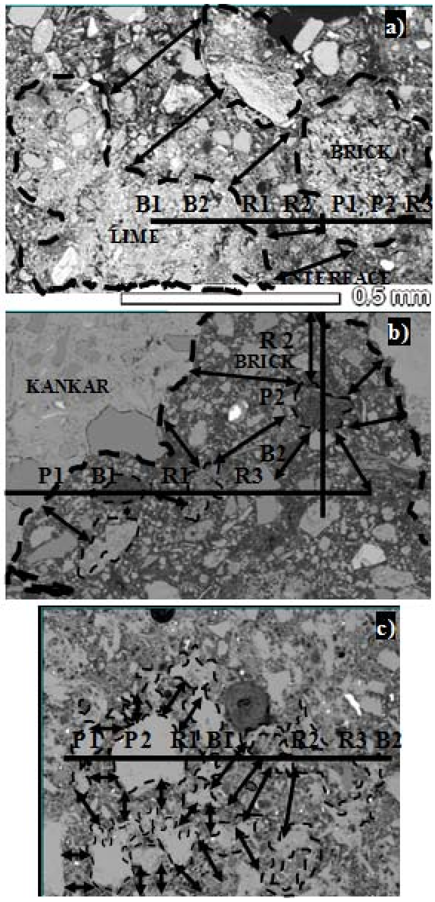

Fig. 3. SEM image of reaction rims formed around pozzolana fragments with pointed EDS microanalysis locations for a) type-A mortars, b) type-B mortars and c) type-C mortars showing; binder (B1 and B2), Reaction Rim Interface (R1, R2 and R3) and Brick Pozzolana (P1 and P2)

Hydration Process and Products in Lime and Cement Mortars

The Mughals used pozzolana (crushed bricks) and lime (Kankar pieces) as aggregate in hydrated lime mixtures to produce highly durable resulting mortars. In general, mortar resistance and durability is attributed to the presence of components that react with calcium hydroxide base to form calcium silicate hydrates and calcium aluminates hydrates (stable and naturally occurring hydration products), commonly called C-S-H and C-A-H phases [13, 14]. This durability is documented on studies of Byzantine, Roman and Ottoman mortars [15-17]. The formation of these phases also provides strong adhesion bonds between binders and aggregates to keep constituents intact. The traditional techniques of lime mortars were followed during the British period but with different proportions of aggregates and binders.

The migration of SiO_2, Al_2O_3 and Fe_2O_3 from the pozzolana fragments to the binder and the consequent formation of C-S-H and C-A-H phases which creates reaction rims around the pozzolona (Fig. 3a and 3b) in type-A and B mortars [18-21]. The EDS spot analysis of binder, pozzolana and reaction rims (Fig. 4a and 4b) shows that concentrations in SiO_2 and Fe_2O_3 concentrations increase significantly from binder to rim [20-23]. The alumina content increases in the reaction rims, except in the old mortars (#17 and #19) in which clay was additional binder [4, 20].

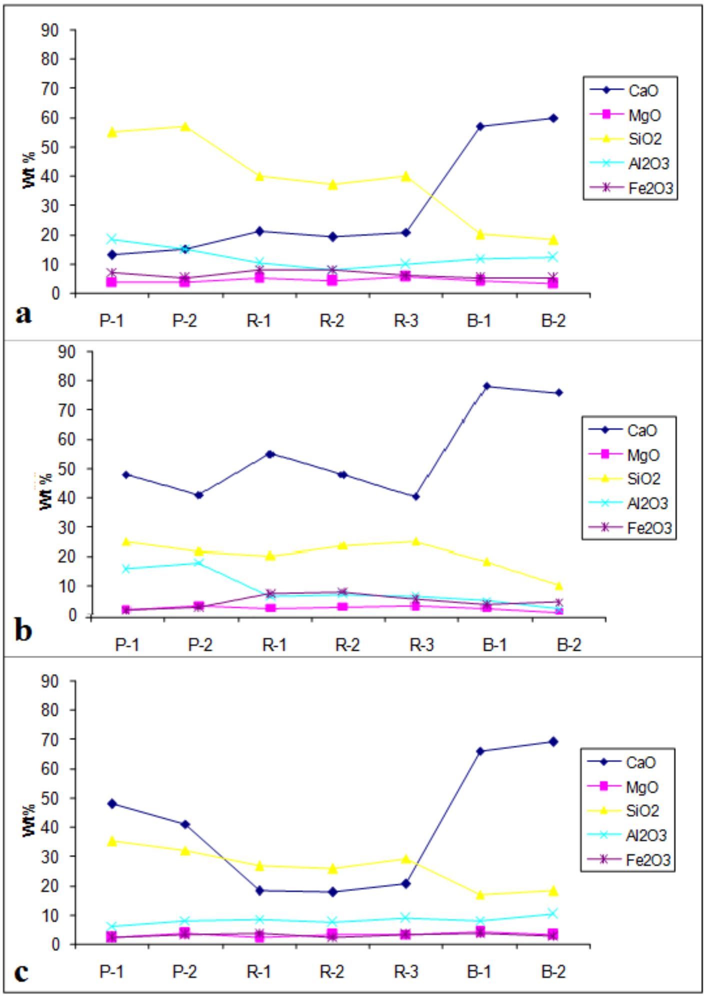

Fig. 4. EDS microanalyses comparison of reaction rims around pozzolana fragments formed in: a) Mughal period mortars, b) British period mortars, c) Recent cement mortars.

The hydration products in type C mortars consist of Calcium hydoxide Portlandite ($Ca(OH)_2$) and Calcium aluminate trisulphate hydrate, or ettringite ($Ca_6Al_2(OH)_{12}\cdot(SO)_4\cdot 26H_2O$) which may transform into Calcium aluminate monosulphate hydrate ($Ca_4Al_2(OH)_{12}\cdot(SO)_4\cdot 6H_2O$) in addition to the C-S-H gel [24-26]. The micro-structural network of C-S-H covers the whole surface of mortar (Fig. 3c) [27]. The SiO_2 content is high in the C-S-H reaction rims while the Al_2O_3 and Fe_2O_3 remain almost constant with small variations from binder to aggregate (Fig. 4c). The binding of cement-based mortars is due to the C-S-H gels but the hydration products are unstable (these minerals are not found in nature) [1-4]. In case of lime the early strength is low but it increases with time while other properties like porosity and permeability decrease. That can explain the long time durability of lime-based mortars. Portland cement has an early high strength (almost 80% of it is obtained within 28 days). Its strong adhesion, compression and tension properties produce mechanical stresses on the surrounding materials [28, 29]. Historic materials are deteriorated at different locations of the historic structures (Fig. 1c and f) by these stresses.

Comparison of Lime and Cement based Mortars for Repairs in Historic Structures

The porous and vapor permeable lime mortars are more durable for restoration [30-32]. The cement-based mortars leads to cracking while settling with the neighboring materials (Fig.1c and d). The cure process of cement based mortars implies its mass reduction and causes cracks-which facilitate the penetration of water and migration of soluble salts inside the historic masonry (Fig. 1e). This is one of the common deterioration patterns, found in all locations of Nur Jahan tomb where cement-based mortars were applied.

The other significant agent of deterioration was sulphate attack that results into mortar disintegration due to the decomposition of C-S-H adhesion bonds. The corrosion of the historic materials (Fig. 3f) was observed at some restored parts of the historic structures where cement-based mortars were used for pointing.

Limes built structures are flexible and can tolerate minute structural and seasonal movements [33]. Rigid materials like cement, instead, cannot tolerate similar movements because of its hardening [34, 35]. When lime based mortars are used in repair works, they ensure a degree of continuity that cement mortars, which lack elasticity, do not allow [36, 37].

The other significant reason that compels to avoid the use of cement based mortars is based on the aesthetic quality, which is important while dealing with the historic fabrics. The colour of cement based mortars is grey (sample #108). Coloured mortars similar to the dark reddish stone (sample #107) where obtained by the addition of pigments to ordinary or white portland cemnet.

Conclusions

This study is the first to present a petrographic, mineralogical and geochemical characterization of the Nur Jahan Tomb mortars. Three main mortar phases were identified in the Nur Jahan Tomb: the oldest dates from the Mughal period (1628-1658 A.D); the second represents repairs during the British period (1847-1947) and the last were used after the independence from British rule (occurred in 1947).

The crushed bricks used as aggregate in historic lime mortars served as pozzolana attributing the hydraulic characteristics to the mortar. The studied reaction rims indicate that brick lime interfaces are largely composed of calcium, silica and alumina due to the formation of hydration products (calcium silicate hydrates and calcium aluminate hydrates). These hydration products create strong adhesion bonds that make mortars more durable. On the other hand, cement-based mortars used for repairs proved to be incompatible due to their hydration process and products that resulted into disintegration.

The comparative studies of historic and repair mortars clearly show different chemical nature and characteristics based on binder and aggregates. These new data will be useful to prepare mortars of high compatibility for future restorations.

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