Chemical and Mineralogical Characterization of Old Mortars from Jahangir Tomb, Lahore-Pakistan

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The 17th century tomb of the Mughal Emperor Jahangir has suffered several conservation and restoration interventions throughout its existence, without any systematic previous knowledge of the chemistry and mineralogy of old mortars. The characterization involved a multidisciplinary set of chemical, physical and micro-structural techniques, particularly X-ray fluorescence spectrometry and scanning electron microscopy equipped with EDS detector to determine the chemical composition of the mortars, which provides necessary information for compatible mortars as well as the state of conservation. The XRD and petrographic studies results show that the mortars were a variable mixture of burnt lime and fine Ravi river sand. The binder being the calcitic lime was obtained from kankar (lime nodules) abundantly found in zone B of Lahore soils and in its surrounding area which contains 70 % CaCO₃, 30 % clay and a fraction of sand with other impurities. The aggregates used were sand, brick pieces and broken kankar. Surkhi (pulverized brick) and rarely slag were used as pozzolanic materials to increase the strength of the mortars.

Key Words: Mughal architecture, Conservation, XRF, SEM-EDS, Petrography, XRD, Pozzolana, Kankar.

INTRODUCTION

Jahangir tomb is one of the majestic monuments, which represent the glorious architectural era of the Mughal architecture in the subcontinent (Pakistan and India). The construction of Jahangir tomb dates back to the 17th century and was built on Persian traditions¹ constituting of tomb building in the center of walled garden locally known as Chahar-Bagh². The garden is divided into four gardens which are further divided into four, thereby splitting the whole into sixteen segments. These segments are separated by walkways and water channels which are elaborately decorated with geometric patterns, fountains and cascades.

The walled tomb garden (Fig. 1a) is entered from the west through a monumental gateway in the eastern wall of a forecourt (Akbari Serai) that also featured external gates on the north and south with a mosque on the west in perfect symmetry³. At the center of the approximately 500 meters square garden (600 gaz square) is the stone-clad tomb itself, surmounted at each corner by a minaret reaching heights of approximately 30 m^{1,3}.

The aim of the present work is to characterize the old mortars for the development of integrated conservation strategies for future restoration works.

EXPERIMENTAL

Sampling methodology: The sampling of mortars is a crucial step that can influence the success of the characterization methodology⁴. The selection of the sampling sites was made with the help of in-charge conservator/technicians and according to the previous research methodologies⁵ adopted internationally⁶. Seven mortars were sampled from different sites as shown in Fig. 1b and described in Table-1.

Preliminary observations showed that samples SJT1, SJT3, SJT6 and SJT7, which were taken from wall sections composed of one layer with variable thicknesses. The samples were dirty white in colour and showed pieces of kankar, brick and fibrous material.

Megascopic observations of sample SJT2 revealed two distinct layers, one inner layer inside the masonry of dirty white colour with variable grain size distribution designated as SJT2-1 while the other layer facing outwards was of light reddish colour with fine grains with no fibers. This layer seems to be later addition as it was applied to fill the deteriorated masonry and designated as SJT2-2.

Samples SJT4 and SJT5 are composed of one uniform layer thickness with pale white colour. The samples showed small pieces of kankar, brick and slag strongly adhered to the substrate. The fibers were also found in these samples.

Fig. 1. (a) Aerial view of the Jahangir Tomb with Chahar-bagh setting

Fig. 1. (b) Plan of the Jahangir Tomb and sampling sites indicated on the plan

TABLE-1
DESCRIPTION OF THE OLD MORTAR SAMPLES

Sample	Mortar Function	Location
SJT1	Filler Mortar	Inner wall of archways
SJT2-1	Filler Mortar	External wall of archways
SJT2-2	Filler Mortar-new	External wall of archways
SJT3	Filler Mortar	Inner shaft wall of minaret
SJT4	Render	Exterior wall front facade
SJT5	Render	Interior wall rear facade
SJT6	Filler Mortar	Inner wall of rooms
SJT7	Filler Mortar	External wall of rooms

Characterization methodology: Prior to chemical characterization, the samples were studied petrographically and by XRD to determine the mineral phases present in thin sections⁶⁻⁸. The mortars were thoroughly examined using a petrographic polarizing microscope to determine the binder, aggregate and additives used in the past and to characterize them for future conservation works.

X-ray diffraction was carried out using Ni-filtered CuK α radiation, with a step size of 0.05° at 2 sec per point and 2 θ

ranging from 5° to 60°. Two types of fractions were analyzed, the fraction corresponding to the mortar as collected, designated as overall fraction and the other fraction designated as insoluble residues fraction, which was obtained after the acid digestion of the mortar samples to determine the siliceous aggregates from the lime paste.

The powdered samples were also used to make fused glass-beads (sample powder with lithium tetraborate in 1:5 ratios) for X-ray fluorescence spectrometry. The pills were analyzed with wavelength dispersive X-ray fluorescence spectrometer (WD-XRF, Axios, PANalytical), equipped with 5 diffraction crystals to establish the overall chemical composition of the mortars necessary for developing the compatible repair mortars.

In addition, polished sections were also prepared from vacuum impregnated samples in epoxy resin. They were analyzed by scanning electron microscope, JEOL JSM 6390 LA coupled with EDS (EDS, Oxford-1 NCA) spectrometer for texture imaging and bulk chemical compositional analysis.

RESULTS AND DISCUSSION

Preliminary observations of the polished sections under a petrographic polarizing microscope showed that all the mortars contains lime nodules (kankar), which indicate that the burnt lime was slaked with a minimum amount of water to convert CaO into Ca(OH)₂ with a release of energy⁹⁻¹¹.

The samples were heterogeneous and the aggregates showed different colour, shape and sizes⁹. The aggregate content was very small, approximately 1.5-2.5 times of the binder content as compared to the new modern mortars applied in 20th century where it is 5-6 times as determined in sample SJT2-2^{12,13}. The sand used was fine Ravi river sand in these mortars as observed under the microscope. Lime nodules in the form of broken kankar pieces were observed in all the samples with the exception of the sample of mortar added in later periods (SJT2-2).

Observation of thin sections (Fig. 2) showed that samples SJT1, SJT2-1, SJT3, SJT6 and SJT7 are almost identical with a binder composed mainly of a carbonate phase¹⁴. The largest fraction of the aggregates consists of kankar-carbonate, crushed bricks and sand with jute fibers added as an additive^{15,16}. This observation is further supported by the geology of the area. The kankar were extracted from the B-horizon of soil profile in addition to the high level river terraces around the cities of Lahore and Kasur^{15,16}. It was also possible to observe micro-crystalline phases of silica. The subangular to subrounded quartz grains embedded in the binder content are mainly silt sized to very fine sand sized as observed in the thin sections. The petrographic studies showed that the sand used in these mortars correlates mineralogically with Ravi sand which is the local source within the vicinity¹⁵. The fibrous material was also observed and found to be the jute fiber locally available and was used to increase the bonding strength of the constituents of the mortars.

The XRD results showed that the mortars are essentially composed of carbonate (calcite) and siliceous aggregates (Table-2) confirming the optical observations. The presence

TABLE-2
MINERALOGICAL COMPOSITION OF THE MORTARS ASSESSED BY XRD

Crystalline phases	SJT1	SJT2-1	SJT2-2	SJT3	SJT4	SJT5	SJT6	SJT7
Calcite	+++	+++	++	+++	+++	++/+++	+++	++/+++
Quartz	++	++	+++	++	++/+++	++	++/+++	++/+++
Illite/ Clay	++	+++	+/-	++	+/++	+	++	+/++
Albite	+/-	+	+/-	-	?	-/+	-/+	-/+
Haematite	+	+	+	t	+	t/+	+	+
Gypsum	+/-	+	t	t	t/-	+	+	+
Halite	?	+	-	?	t	?/t	?	-

+++Abundant, ++ present, + small amount, t traces, ? doubts in presence, undetected

TABLE-3
CHEMICAL COMPOSITION OF MORTARS DETERMINED BY XRF-SPECTROSCOPY (wt %) EXPRESSED IN OXIDES

Sample	wt%	SJT1	SJT2		SJT3	SJT4	SJT5	SJT6	SJT7
			SJT2-1	SJT2-2					
SiO ₂	wt %	23.742	22.893	44.718	25.916	21.342	20.984	22.836	24.042
TiO ₂	wt %	0.321	0.275	0.44	0.369	0.3	0.294	0.509	0.442
Al ₂ O ₃	wt %	6.706	5.174	8.106	6.007	5.49	5.282	6.837	7.361
Fe ₂ O ₃	wt %	3.507	2.301	3.507	2.757	2.551	2.285	2.675	2.322
FeO	wt %	0	0	0	0	0	0	0	0
MnO	wt %	0.082	0.066	0.082	0.06	0.028	0.068	0.096	0.057
MgO	wt %	2.404	1.61	2.304	1.954	2.36	1.555	2.177	1.364
CaO	wt %	30.039	33.804	20.139	30.505	35.34	35.932	32.304	31.067
Na ₂ O	wt %	1.245	0.883	1.245	1.855	0.866	1.249	1.549	1.68
K ₂ O	wt %	1.619	1.147	2.619	1.728	0.601	1.127	1.734	1.246
P ₂ O ₅	wt %	0.107	0.113	0.507	0.367	0.07	0.088	0.033	0.106
Cr ₂ O ₃	wt %	0.087	0.011	0.187	0.021	0.005	0.006	0.013	0.007
NiO	wt %	0.002	0.006	0.012	0.007	0.003	0.003	0.005	0
H ₂ O	wt %	0	0	0	0	0	0	0	0
CO ₂	wt %	0	0	0	0	0	0	0	0
LOI	wt %	29.894	31.55	15.894	28.454	31.044	30.785	29.232	30.306
Total		99.755	99.833	99.76	100	100	99.658	100	100

Fig. 2. Thin-section observation of the mortars with a stereomicroscope, showing brick pieces, kankar-lime and siliceous aggregates

of halite in the samples suggested its use as an accelerator for the mortars carbonation and gypsum in traces indicates the chemical deterioration of the mortar which is considered as the later developments^{17,18}.

The XRD analysis of insoluble residues fraction clearly indicated the fine sand content which supports the optical studies and the use of nearby Ravi sand which is mineralogically found the same as in the samples.

The samples SJT4 and SJT5, which were used as renders rich in recrystallized calcite also showed the presence of slag in addition to surkhi and kankar. These renders showed

recrystallized CaCO₃ as the carbonation can also occur as shown in the equations below when the render is hard as often observed in the old mortars and renders¹⁹⁻²¹.

These render mortars are very fine grained due to the fine aggregates used with fibers forming a monolithic mass. The slag used in these samples was brick kiln slag as described in textural references²²⁻²⁴.

For all the samples, the insoluble residues observed under a microscope together with XRD analysis confirmed that the aggregates used have uniform mineralogical composition. It was also possible to verify the presence of surkhi and fibrous material.

The chemical composition of mortars as oxides determined by XRF spectrometry (in wt. %) is shown in Table-3. The results revealed remarkably uniform composition for all the mortars except SJT2-2. The mortars were mainly composed of CaO ranging from 50-60 % and SiO₂ about 25 % with Al₂O₃ content of 7 %, which is higher due to the crushed brick added as an aggregate. The Fe₂O₃ content is 2-3 %, which is the characteristics of the historic lime mortars used in the past. The Na₂O and K₂O values range from 1-2 % while MgO content is almost 2 % in all the samples due to its occurrence as a subordinate phase with calcite (CaCO₃).

TABLE-4
CHEMICAL COMPOSITION OF MORTARS DETERMINED BY SEM-EDS AVERAGED (wt %)

Sample		SJT1	SJT2	SJT3	SJT4	SJT5	SJT6	SJT7
Oxides	wt %		SJT2-1	SJT2-2				
Na ₂ O	wt %	1	1.16	0.35	1.35	1	1.71	1.35
MgO	wt %	3	2.57	2.76	2.5	4	3.25	3
Al ₂ O ₃	wt %	6	6.55	9.52	5	4.5	7.7	6.75
SiO ₂	wt %	27	27.62	50.7	25	23	24.75	25.25
P ₂ O ₅	wt %	0.1	0.1	0	0.1	0.2	0.2	0.11
SO ₃	wt %	0.5	0.49	3.85	0.5	0.53	0.55	0.45
Cl	wt %	0.75	0.58	0.14	0.5	0.75	0.73	0.67
K ₂ O	wt %	2.5	2.35	0.73	2.25	3	3.27	2.25
CaO	wt %	55	54.9	26.95	58	60	53.13	56.55
TiO ₂	wt %	0.25	0.25	0.33	0.25	0.35	0.3	0.22
Cr ₂ O ₃	wt %	0.25	0.27		0.45	0.25	0.34	0.21
FeO	wt %	3	2.75	4.18	3.6	2.25	3.5	2.75
NiO	wt %	0.65	0.41	0.51	0.5	0.17	0.57	0.44
Total		100	100	100	100	100	100	100

The sample SJT2-2 was found different with all the other samples being rich in SiO₂ content of about 45 % and CaO 35 %, which further supported the petrographic analysis that showed higher aggregate to binder ratio for this particular sample. The Al₂O₃ content was found about 8 % with Fe₂O₃ content of 3.5 %, which is quite similar to the old mortars composition as described above. The Na₂O, K₂O and MgO were also found in similar proportions as in all other samples. The absence of fibrous material in this mortar is also one of the reasons of its low bonding strength.

The chemical analysis of the mortars clearly distinguished the new mortar from the old mortars and further elaborated the cause of its failure (cracks and falling materials at conservation locations) as observed at various locations during the survey. The results clearly depicted the higher binder aggregate ratio of new mortar which is not compatible with the old mortars and can be seen in further microstructure analysis done afterwards^{25,26}. This difference of binder aggregate ratio which can only be determined by the chemical analysis like XRF is the key step in developing the compatible mortars for future conservation works¹⁹.

The analysis of the mortars by SEM/EDS showed all the mortars (Figs. 3 and 4) have a compact microstructure typical of old lime mortars with aggregates well embedded in the matrices except SJT2-2 where they were found loosely bound²⁶⁻²⁸. The oxide compositional analysis of the mortar samples by EDS (Table-4) indicated that all the mortars were mainly composed of high amounts of Ca, Si and moderate amounts of Al, Mg and K.

However the amounts of Fe in all the mortars were found more or less to be the same (Tables 3 and 4). Abundant calcite crystal-amorphous hydraulic formations (CSH) cluster aggregates were determined in the matrix of the mortars²⁹⁻³¹, especially in the samples SJT2-1, SJT-6 and SJT-7 (Fig. 3a, c and d).

This reflected the use of abundant limestone fragments (locally known as kankar) as the main aggregate with sand and crushed bricks²⁹⁻³¹, which was most likely recrystallized

Fig. 3. SEM micrographs of the mortars a) SJT2-1 mortar is showing heterogeneous texture b) SJT2-2 mortar used as a filler afterwards different from the old mortars in texture as shown c) SJT6 showing typical heterogeneous texture d) SJT7 also showing the heterogeneous texture with CSH aggregates formations

as fine calcite and CSH aggregates and oriented as clusters following the hydration process:

The well to moderately uniform distribution of the porosity observed in the SEM images of old mortar samples is probably due to the homogeneous distribution of the hydration process *i.e.*, indicating most likely a uniform proceeding hydration process, mainly composed of calcite and CSH^{30,32}.

The presence of organic fibers also indicates an incremental increase in mortar durability as observed in the SEM image of the sample^{13,33}.

The presence of Cl (Table 4) which was also indicated in the XRD is not due to the deterioration but it was added as an accelerator for hydration process also supported by textural references^{17,18,34}.

Fig. 4. SEM micrographs of selected samples with corresponding EDS

Conclusion

This work for the mortars characterization of 17th century Mughal period has shown the calcitic lime was used as binder with broken kankar-limestone fragments, sand and crushed bricks as aggregates. The pulverized brick and sometimes brick kiln slag was added as pozzolanic materials to increase the strength of the mortars which also modifies the pore-structure subsequently increasing the durability. The addition of the jute fibers further improved the bonding strength and durability of these mortars. All the samples except SJT2-2 have similar

chemical compositions, which are in agreement with the previous historical research done on 17th century mortars of Mughal period in India (part of the subcontinent). The Sample SJT2-2 showed higher silica and low calcite content depicting it to be the new mortar having 5-6 times higher aggregate content (silica-sand) than the old mortars. The results proved it to be the later addition when the restoration works were first started on this monument in 19th century. Samples SJT4 and SJT5 are composed of one uniform layer consisting of fine aggregates applied as renders, which is finished with rich lime

finishing coats afterwards. This was a common technique to improve protection of ancient masonry against weathering agents.

The aggregates used in all the mortars have a mineralogical composition identical to the locally available materials supported by the geology of the area.

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