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ANALYSIS AND CHARACTERISATION OF THIRD CENTURY ANCIENT MORTARS AT SUBRAMANYASWAMY TEMPLE REDISCOVERED AFTER THE 2004 TSUNAMI NEAR MAMALLAPURAM SHORE, INDIA

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Abstract

A characterization study on lime mortar samples from the Subramanya Swamy temple, Saluvankuppam, rediscovered after the 2004 tsunami, was carried out to understand the raw materials used and the application technology. The binder is air lime with binder to aggregate ratio of 2:3. Calcitic crushed sea shells along with siliceous river sand were used. The particles are poorly graded with large amount of nano-particles in the form of silt and clay minerals. The major peaks identified in XRD are Calcite, Quartz and Kaolinite. The presence of Geothite and Gibbsite in the mortar is due to chemical reaction between the mortar and the laterite bricks. The mortar is not affected by salt crystallization which is evident from the low sodium chloride content of 0.32%. The organic test results indicated the presence of carbohydrates and protein that supports the traditional practice of adding herbs into mortars. TGA point out structurally bound water of 1.5% and carbon dioxide 28.88%. The SEM images show the presence of microbial colonies in the mortars. EDX indicates the complete formation of Calcite.

***Keywords**: Archaeological site; Coastal ancient temple; Old lime mortar; XRD; SEM-EDX; TGA-DTA*

Introduction

Lime is the most widely used binder in historical masonry units such as bricks and stones. The raw material composition, the physical, mechanical and durability properties, can play significant roles in the structural behavior of historic structures. Therefore, determining the characteristics of the lime mortar is important for the production of restoration mortars of ancient buildings.

The intervention should be carried out with the aim of safeguarding their authentic values such as aesthetic, historical, social, and cultural values. The characteristics of ancient materials are of particular importance in conservation studies, since they provide information on traditional material characteristics. This requires an interdisciplinary collaboration that deals with the type and characteristics of the original building materials, their mineralogical composition for compatible intervention materials that would be used in preservation and restoration studies.

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On December 26th, 2004, the tsunami along the Indian Ocean shoreline exposed an inscription from the 10th century CE engraved on a rock boulder at Saluvankuppam, 6km north of Mamallapuram, Tamil Nadu, India. The area was excavated by the *Archaeological Survey of India* (ASI), Chennai Circle [1]. Based on the clue, a shrine dedicated to the Hindu deity Murugan temple (Figs. 1a and 2c) was discovered by a team of archaeologists from the ASI. The excavation exposed the entire Subramanya temple complex showing different phases of temple building architecture over a period of time from the 4th–5th century CE to the 11th–12th century CE. The entire temple was constructed with lime mortar (Fig. 1b) and laterite bricks (Fig. 2a and b) during the Sangam period (the 3rd century BCE to the 3rd century CE) and a granite Pallava temple (Fig. 1) dating from the 8th century CE and constructed on top of the brickwork (Fig. 1c and d). The ASI team considered that brick temple could be the oldest of its kind to be discovered in Tamil Nadu. Initially, excavations revealed an 8th century CE Pallava era shrine. Further excavations revealed that the 7th century CE shrine was built on the brick foundation of an earlier shrine. The shrine has been dated to the Sangam period (4th century CE). The lime plasters from Sangam age were considered for the study. The main objective of the study was to characterize the ancient mortar and find its application technology.

Fig. 1. Images of the Salvankauppam Temple and sampling areas:
a. rediscovered; b. sea shells as aggregate in mortar;
c. Laterite wall at bottom; d. brick work at top

Fig. 2. Images of the Salvankauppam Temple and sampling areas:
a. Laterite block with bedding lime mortar; b. rebuilt portions with granite stones;
c. another view of remains of Temple; d. lime mortar sample

Experimental part

The characterization study was carried out in two phases, namely mineral and chemical characterization. The mineralogical characterization of historic mortars was performed for a number of reasons related to the conservation of traditional structures. It is difficult, and perhaps unwise, to analyze a mortar with only one method of characterization. Corroboration of evidence of identification and quantification for mineralogical composition is best supported by a combination of methods, including traditional chemical analysis. A wide range of mineralogical characterization methods were chosen for the study of historic mortars namely XRD, electronic particle size distribution, SEM-EDX, TGA–DTA, and FT-IR [2].

In addition to the mineralogical characterization of components of binder and aggregate, it is necessary to perform a chemical analysis on the acid separated binder in historic mortars. Wet chemical analyses allowed the determination of the chemical composition of the acid-soluble binder and after separation, information on the mortar's aggregate was obtained. The wet chemical analysis was performed on the acid filtrate for the determination of soluble oxides of Fe, A1, Ca and Mg. The filtrate of acid digestion was utilized to find the organic substances (carbohydrates, fats and protein) and salts (Na, Cl, S and K) within a historic mortar [3].

Sampling

Samples of Subramanya Swami temple mortar were taken and preserved by ASI, Chennai circle and labelled as *LM-SK*. The mortar sample was sieved through 75µm (Fig. 2d). The powder sample passing through the 75µm sieve was taken and examined.

Acid loss analysis

The ratio of lime as the binder and aggregates as filling material in mortars were determined by treatment of mortar samples with dilute hydrochloric (0.1N) acid [4, 5] and the results are presented in Table 1.

Table 1. Acid loss analysis

Sample Description	Initial weight (g)	Weight of sand retained (%)	Weight of binder (%)	B/A Ratio
LM – SK-1	30	40.3	59.7	3:2
LM – SK-2	30	44.0	56.0	2.8:2.2
LM – SK-3	30	47.4	52.6	2.6:2.4

The mortar samples were prepared, dried and weighed (m_M) by a precision balance. Then, the mortar samples were left under the solution of dilute hydrochloric acid (10%) until all carbonated lime ($CaCO_3$) in the samples entirely dissolved. Aggregates that remained insoluble were filtered through a filter paper, rinsed with distilled water, dried in an oven, and then weighed (m_A). Acid soluble and insoluble ratios were calculated using equations (1) and (2) respectively:

$$Insoluble\ (\%) = [(m_M - m_A)/m_M] \times 100 \quad (1)$$

$$Soluble\ in\ Acid\ (\%) = 100\% - Insoluble\ (\%), \quad (2)$$

where: m_M - weight of mortar sample and m_A - weight of the aggregates.

Acid soluble ratio is not the exact ratio corresponding to lime ratio since both lime and calcareous aggregates that could be used in the mortars were dissolved in the solution of dilute hydrochloric acid. Therefore, lime/aggregate ratio was calculated by the formula as in equations (3) and (4) respectively:

$$Aggregate\ (A\%) = (100 \times IS)/[(AS \times 74)/100) + IS] \quad (3)$$

$$Lime\ (\%) = 100\% - Aggregate\ (A\%), \quad (4)$$

where: IS - is insoluble part and *AS* - is soluble part in acid.

Molecular weight of $Ca(OH)_2$ and $CaCO_3$ are 74 and 100g respectively. The binder part resulting in acid loss analysis is used for chemical analysis.

Chemical analysis

Chemical analysis (Table 2) of the mortar samples were carried out by traditional wet chemical analysis [6].The calcium and magnesium were estimated on titration of the sample with ethylene diaminetetraacetic acid (EDTA), with murexide and eriochrome black T (eBT) as indicators. The amounts of Al_2O_3, Fe_2O_3, Cl and Na were determined by Atomic Absorption Spectroscopy (AAS). Gravimetric Method was used to find the amount of silica (SiO_2) present in the sample.

Table 2. - Weight percentage (%) by chemical analysis for binder, clay minerals, loss on ignition, hydraulic index, cementation index and other chemical components (SO_4^{2-} and SrO)

CaO	MgO	Fe_2O_3	Al_2O_3	K_2O	Na_2O	Cl^-	SiO_2	SO_4^{2-}	SrO	LOI	HI	CI
47.35	0.30	0.58	0.51	0.02	0.15	0.11	8.87	0.07	0.27	39.2	0.21	0.54

CaO+MgO - *Binder*, **Al_2O_3+Fe_2O_3+SiO_2 -** *Clay Minerals*,
LOI - Loss on ignition at 400°C, **HI -** *Hydraulic Index*, **CI -** *Cementation Index*

The *Hydraulic Index* (HI) and *Cementation Index* (CI) is calculated as per equations (5) and (6) respectively:

$$\text{Hydraulic Index (HI)} = m(Al_2O_3+Fe_2O_3+SiO_2)/m(CaO+MgO) \quad (5)$$

$$\text{Cementation Index (CI)} = m(1.1Al_2O_3+0.7Fe_2O_3+2.8SiO_2)/m(CaO+MgO) \quad (6)$$

The identification of lime based on *Hydraulic Index* ranges [7] is:

$0.30 < HI < 0.50$ - weakly hydraulic
$0.50 < HI < 0.70$ - moderately hydraulic
$0.70 < HI < 1.10$ - higher the index, more hydraulic properties

The classification of lime based on *Cementation Index* developed by *E.C. Eckel* [8] is:

$0.00 < CI < 0.15$ - air lime
$0.15 < CI < 0.30$ - sub hydraulic lime
$0.30 < CI < 0.50$ - weakly hydraulic
$0.50 < CI < 0.70$ - moderately hydraulic
$0.70 < CI < 1.10$ - higher the index, more hydraulic properties

Organic test

In order to confirm the presence of organics, the historic mortars were tested for carbohydrate, protein and fatty acids on a dry basis. The presence of fat and protein in the samples were tested using *crude fat method* and *Kjeldhal method* respectively in accordance to IS: 7874 (Part I) -1975 [9]. The amount of carbohydrate was calculated from the percentage of calcium oxide and carbon dioxide. Further FT-IR analysis was carried out as the confirmation test.

Gradation of fine aggregates

Gradation of aggregates was done in two stages on the residue of acid loss test. The particle distribution of more than 75µm was carried out in normal sieve shaker whereas the aggregates of particle size less than 75µm was done with electronic particle size distribution (CILAS 1064 liquid series). It helps to find the proportion of clay, silt and sand particles with XRD analysis.

Mineral characterization

XRD analysis of finely ground mortar samples was done using Bruker Desktop-Diffractometer working with the Cu K_α radiation (K_α = 1.54182), and graphite monochromator in the diffracted beam, at 1.5kW and interpretation was done by Bruker DIFFRAC.SUITE EVA Software. It gives a qualitative result on the possible presence of minerals in the mortar samples.

Thermal Gravimetric Analysis (TGA) was done to determine the structural and hydration water contents of lime mortar and the possible thermal decomposition of other mineral phases. The analysis was carried out in a flushed air atmosphere using alumina (Al_2O_3) cells at a heating rate of 20ºC/min and at intervals of 30ºC up to 1000ºC. Here the mass of the sample is monitored [weight loss] as a function of temperature.

Differential Thermal Analysis (DTA) was used to identify various component materials and to observe the reactions associated with controlled beating of the mortar sample, this method reveals thermal transformation, which includes dehydration, oxidation and decomposition. In addition, crystalline transitions have been observed, which is exothermic or endothermic in nature. DTA was used to identify various component materials of a non-fractioned mortar and to observe the reactions associated with controlled heating of the mortar [10, 11].

Infrared spectroscopy (FTIR), performed on Perkin-Elmer 1000) was used to obtain qualitative information, from a chemical point of view on some of the characteristic compounds contained in ancient mortar (calcium and magnesium hydroxides and carbonates, gypsum, etc.)

and for determining the presence of salts (nitrates, sulfates, oxalates, etc.) as well as organic compounds. FTIR analysis provides supplementary information to XRD. The principal constituents like Calcite, Aragonite, Vaterite, Magnesite, Portlandite, CSH (calcium silicate hydrate) and CASH (calcium alumina silicate hydrate) phases, and organics used could be identified in the aged lime mortars [12].

Scanned Electron Microscopy (SEM) was used to determine the surface morphology of the mortars and it provides information on the morphology and texture of the minerals formed. Energy Dispersive X-Ray analysis (EDX) was used to determine the elemental composition of mortars.

Result and Discussion

Raw material characteristics and its composition

From acid loss analysis as presented in Table 1, lime to aggregate ratio (L/A ratio) is 3:2 based on the following calculations for equations (1) and (2) respectively:

$$\textit{Insoluble (IS)} = [(30\text{-}20)/30] \times 10 = 33.33\%$$
$$\textit{Acid Soluble (AS)} = 100 - 33.33 = 66.66\%$$

Aggregate (*A*) = 40%
Lime (*B*) = 100 – Aggregate = 60%
Binder to Aggregate (*B/A*) ratio = 3:2

L/A ratio in mortars is an important parameter influencing physical, mechanical and durability characteristics of the mortar. Generally the ratio varies from 1:1 to 1:6 for mortars made up of lime and quartz as aggregates. How ever in Saluvankuppam mortars, the ratio is different from conventional, as sea shells (Calcareous) are used in addition with quartz aggregates. Hence, the lime content from sea shell aggregate will also contribute to binder content which is reflected in higher binder compared to aggregate. Hence, ratio 3:2 may be classified as air lime mortars. The lime and aggregates should be in the right proportion so that the mortar with good workability, durability and strength is produced [8, 13]. These ratios were in similar ranges with several historic lime mortars [14] and as said by *A. Moropoulou et al.* [15], typical lime mortar will have ratio more than 1:2.

Chemical test (Table 2) was used to confirm the air lime characteristics as found out in acid loss analysis. As mentioned in Table 2, the chemical analysis indicates 47.35% of high calcium lime with very small amount of magnesium oxide. The classification of hydraulic or air lime is based on hydraulic and cementation index (*HI and CI*) and percentage of clay impurities ($Fe_2O_3 + Al_2O_3 + SiO_2$**)**. The hydraulic index (*HI)* and cementation index (*CI*) is 0.21 and 0.54 respectively which indicates the lime is weakly hydraulic binder. Hence, the lime is typical air lime with sea shell aggregate as shown in Figure 1b. Moreover, the presence of strontium oxide (SrO) about 0.027% in chemical analysis confirms the role of sea shells as strontium mineral present in sea shells in natural form. The NaCl salt was about 0.32%. The sea shells are calcareous in nature and on the long run the shells would have dissolved with binder showing the feeble hydraulic character. High calcium air lime does not exhibit the hydraulic character.

Hence, the use of sea shells (crushed) would have exhibited the hydraulic properties to some extent. Also, loss on ignition (LOI) of about 39.20% demonstrates the high rate of carbonation. The *LOI* is less than that of CaO + MgO, meaning the binder content is on the higher side as the calcitic aggregates would have combined with silica to form calcite. Further the maximum LOI for eminently hydraulic lime is around 20% and thus, air lime with calcitic coarse aggregates and quartz as fine aggregates was used. The reason for the addition of calcitic aggregates in proportion with quartz is the calcitic aggregates along with high calcium binder in the presence of organics resist to salt crystallization and acts as sacrificing layer to protects the structure [16]. The amount of carbohydrates, protein and fat in the range of 1.48%, 0.85% and 0.12% respectively as presented in Table 3 ascertain the role of organics.

Table 3. Organics analysis of mortar

Parameter	Result	Procedure
Fat	0.12	DGHS LAB Manual 3
Protein	0.85	Kjeldhal Method
Carbohydrate	1.48	Calculation Method

The different plant extracts in form of carbohydrates, protein and fats would have been added to improve various properties of mortar. The organic admixture modifies the fresh and hardened properties and finally helps in carbonating of lime mortar resulting in higher strength and durability [17]. Still in India, there is a traditional practice of adding the cocktail of fermented plant extract to mortars. Organics rich in carbohydrates will reduce to carbon dioxide to enhance the carbonation within the mortar. In the same way, as stated by *J. Jasiczak and K. Zielinski* [18], the addition of proteins acts as air entraining agent in fresh mortars and increases the workability. It also acts as waterproof to mortar and controls the water movement. The temple being situated on seashore facing marine environment and, these plant extracts were added to protect from environmental deterioration [19].

Organics added would have acted as acrylic coating and water repellent to resist salt crystallization [20]. When it comes to aggregates, the particle size distribution greater than 75µm shows higher the percentage of coarse than the fine sand (Fig. 3). The sand used in the temple is poorly graded with low shear strength, high permeability. As already mentioned, large quantity of ground fine sand or sea shell dust (around 1.75mm range) have been added. Figure 4 give the gradation of aggregate less than 75µm done using electronic particle size distribution. The representation of all ranges of particle is very low and between 500 to 600µm the curve is very steep. The curve above 53-500µm is well graded. Results also reveal the presence of 7% clay particles, 53% silt particles and 40% fine sand particles The analysis from both the graph indicate the presence of more fine grained particles which point that the grinding of the lime mortar mix would have been carried out prior to casting [21].

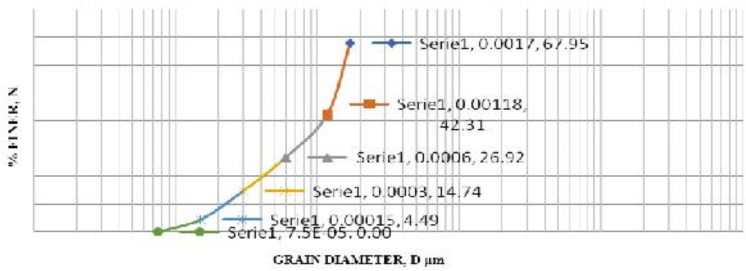


Fig. 3. Grain size distribution of particles greater than 75µm

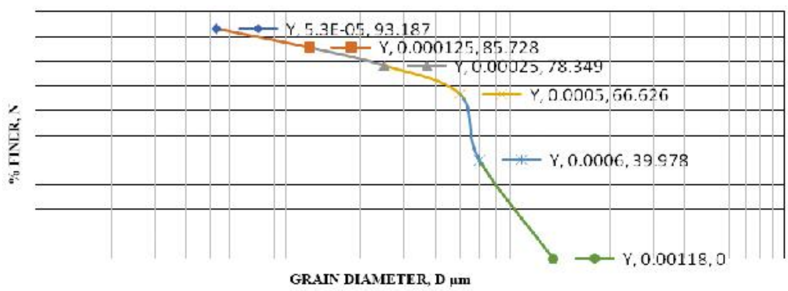
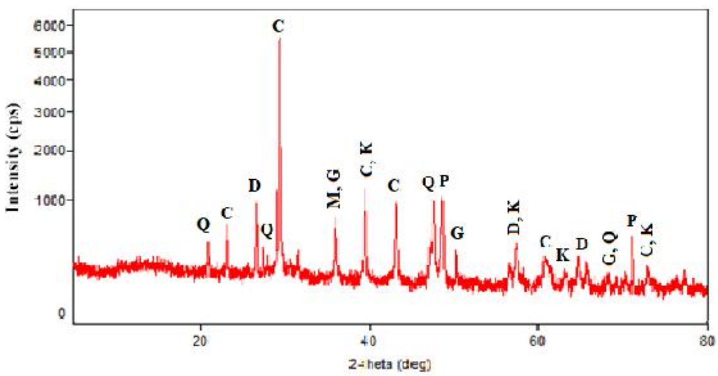


Fig. 4. Grain size distribution of particles less than 75µm

Discussion

Mineralogical interpretation

XRD is in line with chemical analysis. XRD pattern as presented in Figure 5 indicates the major peaks of calcite in form of Aragonite and Quartz and minor peaks of Geothite, Gibbsite, Kaolinite, Doylite and Magnetite. Major formations of Aragonite occur near seashore. The Calcite peaks were higher in composition when compared to the Quartz peaks and hence there is a complete transformation of CaO to Calcite. Gibbsite, Magnetite and Doylite presence were due to the Laterite bricks as they were rich in iron oxide as the temple was built on the cushion of alluvium. *Satyabhama Bahreenath et al* [22] has indicated that chemical reactions occurred between the mortar and the bricks and hence it resulted in these peaks. The Kaolinite peaks are due to the hydration phases of calcium silicate hydrat (*CSH*) and calcium aluminate hydrat (*CAH*) from the calcitic aggregates, however these peak values are very smaller in composition. Lime plaster were prepared in the kiln at the site with air lime, whole and crushed sea shells (Shale), and river sand. The shell beds and river sand at Muthukadu (very near to Saluvankuppam) is confirms the usage of quartz sand and kiln at the site indicate the slaking of sea shells with lime. Pure air lime would have been prepared either by reducing the burning temperature of lime or extracting the milk of lime after wet slaking.



Q - Quartz, G - Goethite,C- Calcite, K - Kaolinite, M - Magnetite, D - Doyleite, P - Periclase

Fig. 5. Illustration of XRD peak

TGA with DTA interpretation

As presented in Figure 6, weight loss of 1.11% at the temperatures between 200° and 600°C was mainly due to loss of structurally bound water (H_2O) of hydraulic reaction products of CSH, CAH, etc., from Kaolinite. Weight loss (Fig. 5) at the temperatures over 600°C was mainly attributed to the release of carbon dioxide gas (CO_2) during the decomposition of calcium carbonates. The ratio of CO_2:H_2O is inversely proportional to hydraulicity of the mortar. The percentage of H_2O and CO_2 is 1.11 and 28.77 respectively and the ratio is CO_2:H_2O is 25.91 which is greater than 10 and hence the mortar is regarded as non-hydraulic mortar or air lime [23]. The burning temperature during the production of lime will be less and lime would have been sieved to remove impurities.

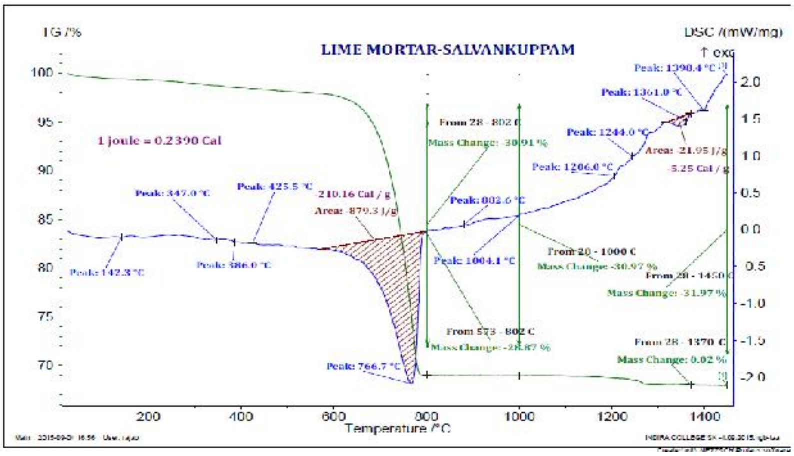


Fig. 6. TGA peaks

The greater presence of nano level particle in particle size analyses point to the milk of slaked lime was used as the binder. The presence of clay minerals as discussed in XRD may be due to the addition of sea shell aggregates. Being non-hydraulic lime mortar, they have the capacity to hold much water in their pastes and retain water against the absorption of masonry units. This derives from the property of non-hydraulic lime to be fatter than hydraulic lime, which provides good workability for lime mortars.

FTIR Interpretation

The FTIR results of the ancient sample are given in Figure 7. In FTIR spectrum, the very deep and narrow intense peaks at 2512cm^{-1} are for Calcite.

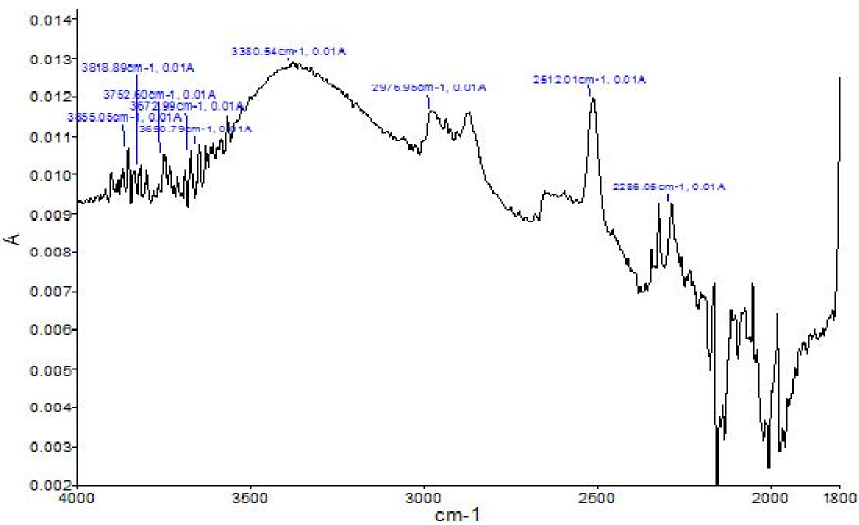


Fig. 7. FT-IR Graph of LM-SK mortar sample

The absorption peak at 2977cm^{-1} justifies that the organics in the form the carbohydrates or polysaccharides has been used in the production of lime mortar and it is in par with organics analysis. Hence, there is a feasibility of addition of fermented jaggery (Unrefined Sugar) with kadukai (Terminalia Chebula) into the mortars at fresh state. Peaks for protein at 1630cm^{-1} present in organics analysis. The very narrow peak at 3650cm^{-1} possibly free Portlandite. The two continuous stretching peaks 3650 and 3672cm^{-1} of Kaolinite validate results of XRD. The high frequency of low mass hydrogen atom can be possible between 3600 to 4000cm^{-1} [24] specify the low amount of compounds of CH stretch with chlorides in the presence of water which would have formed as temple was under sea water for several centuries in this range.

SEM - EDX analysis

Analysis of the mortars by SEM-EDX showed that mortars have a compact microstructure with aggregates well embedded in the matrix. The structure is amorphous in nature with less than 5% of crystalline substance. Fig. 8a possible identifies aggregate heaped with white calcite and other minerals on the top. Inner mortars as in Fig. 8a, b and c shows the formation of gel in the matrix be evidence for the kaolinite in confirmation with XRD. Some location in the mortars shows porous structure (Fig. 8d).

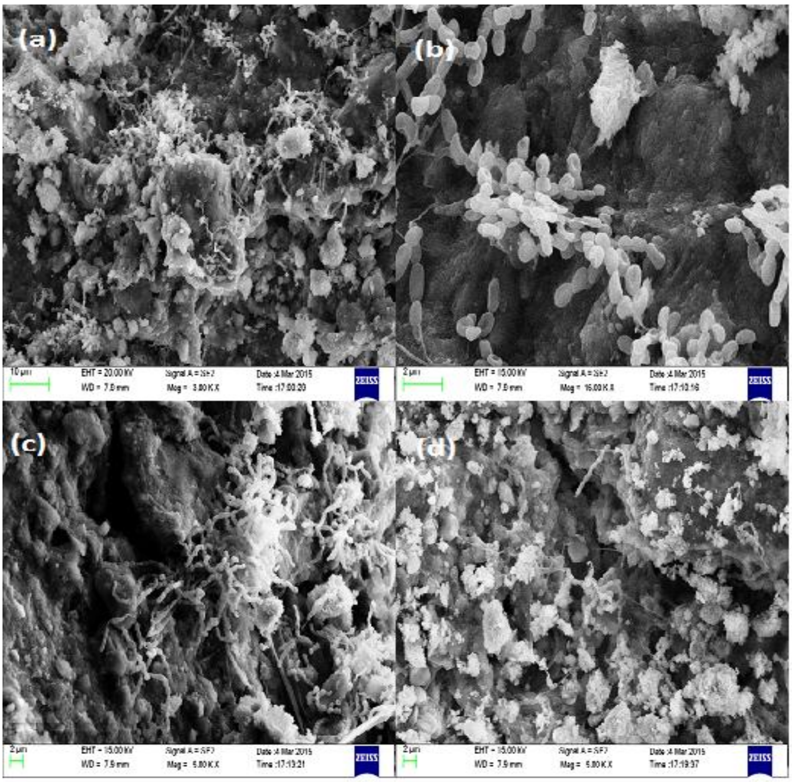


Fig. 8. SEM images of samples: a. Lime and sand Matrix; b. and c. Colonies of microbial formation; d. formations of porous structure

In par with chemical analysis, there is a minimum formation of halite crystals. The elemental analysis (EDX) on SEM image also demonstrates the complete formation of calcite taken on Figure 8a and whereas EDX on image Figure 8b imply the presence of calcium, ferrous, aluminum and silicates (Kaolinite).

All images in Figure 9 give you an idea about the inner structure of the mortar. The mortar is heterogeneous and amorphous in nature with the mixture of minerals like calcite and clay minerals and quartz etc. Large fibers structures indicate the calcite in the form of Aragonite in line with XRD. Also, Figure 9b and c illustrate the biological organism colonies [25]. Figure 9d demonstrate the cleavage within the mortar structure.

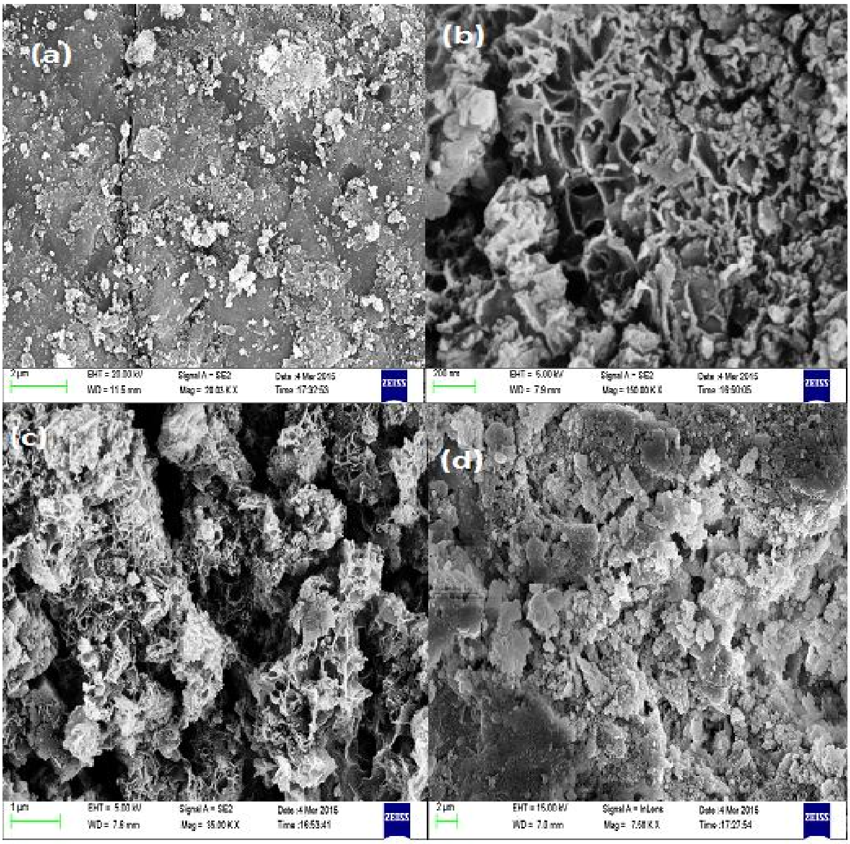


Fig. 9. SEM images of samples: a. Solid mass of lime sand matrix; b. Kaolinite gel; c. White Calcite layer heaped on top of fibres CSH; d. Porous structures.

Production technology of mortar

Traditional lime mortar is produced by wet or dry slaking. If the slaking of lime is with excess water it is called wet, whereas with minimum amount of water it is termed dry slaking. In the case of the Salavankuppam temple, the shell lime was nearest sources (sea shore) would have been wet slaked in open tanks. Based on XRD and TGA result, it is found that only the milk of lime rich in calcium oxide was extracted, leaving the clay impurities at the bottom. The process of slaking would have been done for few months. After getting enough lime by slaking, calcitic aggregates and river sand in the presence of organics would have been grinded for few hours just before placing the mortar. The finer silica after grinding along with clay minerals in shell lime will accelerate the reaction rate less than a day to the hydrated products without change in the microstructure structure [26].

Conclusions

The mortars of the Saluvankuppam Subramanya Swamy temple has a binder to aggregate ratio of 3:2 and has been identified as air lime (high calcium content) based on hydraulic and cementation index. However, the clear binder to aggregate was not accurately predicable due to the presence of calcitic aggregate and it may be equal to 2:3. Organic test along high percentage of loss on ignition confirms the carbohydrate, protein and fat derived from extract of plants. Mortars are free from salts though immersed in sea water for centuries. The rich binder content along with shell aggregates in the presence of organics has resisted salt crystallization of plasters for centuries. The moderate values of calcareous sand or crushed sea shells coarse aggregates in proportion with fine sand has been used to prepare the lime-mortar. The presence of SrO (Strontium mineral) in the form of binder or aggregate as Strontium mineral occur in shells in meagre values. The XRD peaks of $CaCO_3$ indicate Aragonite in composition as shells being Aragonite in natural composition. Other major minerals present in the mortar were identified as Quartz, Goethite, Gibbsite, Kaolinite, in XRD peaks. These peaks were due to the presence of rich iron oxides in the Laterite bricks that were used in the temple, organics in the form of carbohydrates had improved the process of carbonation. The formation of biological colonies by decomposition of organics in mortar would have resisted the salt crystallizations over ages.

Despite the tsunami of temple under the sea for 10 centuries and its recent rediscovery, the ancient temple still exists in good condition, on account of the technology of production and materials added in the mortar.

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