CHAPTER – II
LITERATURE REVIEW

ABSTRACT

The literature review was carried out on the characterization of archeological artifacts like potteries, bricks and tiles in respect of the present study. The various research publications have been survived and enlisted relatively on the different scientific techniques adopted in analyzing the antiquities.
2.1. Introduction

In the present investigation spectroscopic, microscopic and X-ray diffraction methods (FTIR, XRD, SEM-EDS, and XRF) are employed in characterizing the archaeological artifacts excavated from five different sites of Tamilnadu. From these studies the mineralogical and chemical composition, degree of vitrification and elemental composition, crystalline mineral phases and the temper materials added in the samples of interest are determined. Besides that the firing temperature attained and the firing atmosphere adopted by the artisans during the manufacturing of the relics is established.

To achieve this goal different literature representing different studies from various publications has been reviewed. Since the precise characterizations and predictions cannot be made by implementing single scientific method, most of the reviewed literature shows the adaptation of more than one scientific method was presented briefly from the published research papers and books representing this field over the years until now. The literatures reviewed pertaining to the adopted scientific methods of the present work is given in the following paragraphs successively.

2.2. Review of literature

All the archaeological artifacts like potteries, bricks and tiles are made up of primary (clay) minerals, secondary and accessory minerals. They exhibit characteristic absorption bands in the mid IR region. Hence Farmer\(^1\) has employed
Infrared spectroscopy to investigate structure, bonding and chemical properties of clay minerals.

Tite and Maniatis\(^2\) have studied the progressive development of bloating pores in the calcareous clay with increasing firing temperature by scanning electron microscopy (SEM). From the microstructure it is revealed that the formation of bloating pore structures mainly depends on firing temperature, firing atmosphere and chemical composition of clay. Moreover the calcareous/non-calcareous nature of the relics can be obtained from elemental composition through EDS.

The technological examination of Neolithic-Bronze age pottery from central and south east Europe and from the near east has been done by Maniatis and Tite\(^3\) using SEM. Based on vitrification type (observed from SEM images) and chemical composition (from EDS), the type of clay and the firing procedure employed in the manufacturing process of artifacts are discussed.

In the field of ceramic analysis, scanning electron microscopy has been exploited by Tite\(^4\) to gain the knowledge about microstructure of shreds including the composition, shape and distribution of certain minerals. Furthermore, information provided by SEM can be used to locate the provenance of a ceramic assemblage and thus provide information on exchange networks.

Salah A. Tahoun\(^5\) has utilized X-ray diffraction (XRD) method for mineral composition of red bricks which is affected by raw materials used. The results suggested that bricks made out of bentonite sediments contain more quartz and less feldspar compared with those made out of soil materials. Moreover the intensity ratio of quartz reflection (3.34 Å) to that of feldspars (3.2 Å) forbids using soil materials for brick production that can be implemented on sound legal and technical grounds.
**Wagner et al** extensively used XRD and Mossbauer techniques on archaeological ceramics in order to characterize them. The presence of major minerals was identified by X-ray diffraction technique, whereas the firing temperature, firing condition and coloring mechanism of the above materials were assessed by Mossbauer measurements.

**Jordon et al** have studied the transformation of cretaceous clays during firing used in the manufacturing of ceramic tiles. They heated the clay samples for different temperature and corresponding transformations of minerals were studied using XRD analysis.

**Alia et al** have reported the mineralogical evolution of the three types of raw materials used for the brick industry in Spain during the firing process (800-1150°C) through XRF, XRD and FT-Raman spectroscopy. From the analysis, onset of early vitrification (at about 900°C), importance of Ca and Mg oxides for the synthesis of new mineral phases and its role in the vitrification process are discussed.

**Marcus Key and Emily Gaskin** analyzed Native American pottery from prehistoric Davis site in Lancaster County, Virginia using XRD. The role of particle size and furnace atmosphere on the sintering of clay powder for tile production was studied. The observations on various studies suggest that there is shrinkage of clay and increase in compressive strength with temperature.

**Tang et al** studied the mineral composition of the gloss/ body of Greek pottery by applying high resolution powder diffraction using synchrotron X-rays. The minerals quartz, illite, feldspar and hematite were found to be present in the respective samples and the supplementary SEM analysis revealed the gloss thickness of about 5 to 20µm.
De Benedetto et al\textsuperscript{11} have explored the FT-IR spectroscopy in assessing the chemical and mineralogical composition of ancient pottery shreds excavated from Canosa (Puglia). The IR vibrational assignment to different minerals was accomplished with the support of both literature references and standard materials. Besides that from FT-IR/XRPD data the most fascinating aspects of firing temperatures and provenance of pottery production were inferred.

Barone et al\textsuperscript{12} carried out the spectroscopic investigation on Greek ceramic artifacts by adopting FTIR, XRD and XRF techniques. From the infrared spectra obtained the ancient firing procedures, baking temperature and area of production were elucidated. In addition to that, crystalline mineral phases were identified through X-ray diffraction technique and chemical composition was done through X-ray fluorescence spectroscopy.

Kerstin Elert et al\textsuperscript{13} studied the mineralogical and textural evolution of calcareous and non-calcareous bricks during firing using XRD, SEM and porosity. The results showed that the vitrification degree, porosity and pore size distribution depends mostly on raw clay composition and firing temperatures. Besides that, high firing temperatures are required for NC to produce durable bricks that remain unaltered upon weathering.

Lara Maritan\textsuperscript{14} made characterization studies on the second Iron Age potteries using various spectroscopic techniques FTIR, XRD, XRF and Mossbauer studies. The addition of organic material by the potters to confer more plasticity to the clay paste during firing and the formation of graphite and amorphous carbon during firing of organic material is also reported by the authors.
Maravelaki Kalaitzaki et al\textsuperscript{15} have adopted XRD, TG-DSC, FTIR, SEM, AES and porosity (water absorption method and Mercury porosimetry) techniques to acquire information about mineral composition, thermal behavior and microstructure of hydraulic lime mortars. Further the capillary water absorption of a material is interpreted as the angular co-efficient of the rectilinear part of the capillary absorption curves.

Sridhar\textsuperscript{16} have reported that the Purananuru speaks about the Pandya kings who consumed the delicious wine imported from Rome. The close relationship of Pandyas with the Roman kings might have led to the establishment of a Roman colony and a seaport at Alagankulam.

Shoval and Beck\textsuperscript{17} employed FTIR spectroscopy to get the knowledge about ancient ceramic technology of the pot fragments from Tel Hedar, Galilee. The authors revealed that the potteries have high concentration of calcite temper materials which may be added at the time of manufacture to improve its quality.

Christina Papachristodoulou et al\textsuperscript{18} have done the characterization studies of ancient Hellenistic potteries using XRF, multivariate analysis and mineralogical analysis. Compositional and mineralogical data combined with archaeological and material science criteria allowed the various aspects of pottery making such as, selection of clays, tempers, firing temperature and firing condition.

Casaletto et al\textsuperscript{19} carried out a multi analytical investigation on medieval pottery from Italy by adopting different scientific techniques XRD, SEM-EDS and XRF. A similar chemical composition was found for all the shreds allowed them to infer the same provenance of the raw material.
SEM-EDS characterizations of ancient Bronze Age pottery from Lithuanian were carried out by Krapukaityte et al\textsuperscript{20}. The SEM morphology showed different vitrification stages and EDS revealed the percentage of calcium. From the results the firing temperature of the shreds and the calcareous nature of the source clay were determined respectively.

Roberts\textsuperscript{21} gave the idea of estimating firing temperature from thermal expansion measurements which is familiar in ceramic technology. This equivalent firing temperature is probably best index available for assessing the efficacy of ancient kilns and it is certainly important for attempts at imitation of ancient ceramics.

Palanivel and Velraj\textsuperscript{22} have analyzed the fired clay artifacts excavated from Tamilnadu, India using FTIR and FT-Raman spectroscopic techniques. From the IR absorption bands and their corresponding Raman shifts with relative intensity, the presence of minerals, traces of elements in the clay, the knowledge of artisans regarding the firing temperature and atmosphere conditions are well-established.

The techniques of Raman, FT-IR, XRF spectroscopy and XRD were used by Legodi and de Waal\textsuperscript{23} to examine the composition of ancient domestic clay pottery of South African origin. The results showed that South African potters used a mixture of clays as raw materials and the firing temperature for most samples did not exceed 800°C, which suggests the use of open fire.

The historical bricks of different age (from XII to XVIII centuries) recovered from medieval monastery in the Venice Lagoon have been characterized using FT-IR, TGA-DTG and DTA, XRD, SEM + EDS by Nick Schiavon et al\textsuperscript{24}. The relative abundance of primary (i.e. calcite and dolomite) and secondary firing minerals
(i.e. diopside and wollastonite) in the bricks coupled with their relative geometrical dimensions allowed placing the samples in a chronological sequence according to known historical changes in brick making firing temperatures and practices.

**Barilaro et al**\(^{25}\) applied FTIR along with XRD to identify the firing temperature and condition of a Sicilian Proto-Mojolica pottery. The firing temperature of the shreds were estimated from the thermal stability of mineral present, mainly from the mineral wollasonite, since it originate from the chemical reaction between quartz and carbonates when the temperature reaches 900°C.

Clay minerals, chemical composition of potteries and the atmosphere prevailed by the artisans belonging to the archaeological site Melchittamur were identified using Fourier Transform Infrared Spectroscopy and chemical analysis by **Velraj and Janaki**\(^{26}\). The analysis reveals that, the artisans of this site used various chemical compositions (potteries rich in Quartz, Alumina and Iron oxides), different minerals for making potteries and adopted open air or oxidizing atmosphere for firing the utensils.

**Eleni Nodarou et al**\(^{27}\) applied a combination of petrography, neutron activation (NAA), X-ray fluorescence (XRF), and X-ray diffraction (XRD) techniques on mud bricks from Vasiliki, Makrygialos and Mochlos in East Crete to examine the nature of the raw materials used, the technology of manufacture and the potential use of specific recipes. Furthermore a range of raw materials from the immediate vicinity of each site were sampled and analyzed in order to compare with the archaeological data and identify potential sources.

**Palanivel and Meyvel**\(^{28}\) have examined antique pottery shards of Tamilnadu, India, by employing Fourier Transform Infra Red (FTIR) and
Thermogravimetry- Differential Thermal Analysis (TGA-DTA) to infer the nature of the clay used, the textural and vitrification structures, the reactions associated with the mineral compositions to indicate the conditions of firing process adopted and firing temperature attained by the artisans at the time of manufacture.

Venkatachalapathy et al\textsuperscript{39} have scrutinized the magnetic mineralogy of a collection of archaeological potteries to obtain the actual magnetic carriers and the domain states of the constituent magnetic fine particles from the acquisition of isothermal remanence and low field susceptibility measurements. The magnetic mineralogy of all samples has been dominated by ferrimagnetic mineral (magnetite/magnetite with low titanium content) which is suitable for paleointensity measurement in determining the intensity of the ancient geomagnetic field.

Velraj et al\textsuperscript{30} have determined the firing temperature and atmosphere of the potsherds excavated from Tamilnadu, India using the spectroscopic technique FTIR and porosimetry studies by refiring it to different higher temperature. From the disappearance of octahedral sheet structure and the variations found in the dehydroxylation peak, the firing temperature was determined. They have also compared the porosity by water absorption method to the mercury intrusion method. The correlations were also made from porosity to firing temperature.

Cornlius Tschegg et al\textsuperscript{31} have analyzed Bronze age Cypriot ceramics by means of X-ray diffraction method (XRD) to study the thermally trigged reactions of carbonates in clay during ceramic firing. From the results, the occurrence of various crystalline phases (calcium silicates and calcium aluminium silicates) with respect to firing temperature was determined. Moreover, they have found that carbonate
temper materials were added during manufacture in order to improve the plasticity and mechanical strength of the material.

**Dhanapandian and Gnanavel** have investigated the effect of incorporation of granite and marble wastes in brick products collected from companies located in Salem District, Tamilnadu, using FTIR and Mossbauer spectroscopic techniques. The results showed that granite and marble waste can be added to an industrial clay mixture, already in use in the production of bricks with no major sacrifice on the properties of the final product.

Microstructure and chemical degradation of adobe and clay bricks was studied by **Juliana Calabria et al** by means of FTIR, XRD, SEM and nitrogen porosimetry. The mineral characteristics of the ceramics were done by FTIR and XRD. The study established the importance of porosity in determining the physical strength of the body and also porosity values were compared with the pores distribution observed in SEM morphology.

The characteristics of Roman period bricks and mortars in Serapis temple Pergamon (Bergama/Turkey) have been determined by **Ozlem Aslan Ozkaya and Hasan Boke** using XRD, SEM and a TG Analyzer to define the intervention materials for conservation works. From the analysis results they indicated that the mortars are stiff, compact and hydraulic due to the use of natural pozzolanic aggregates whereas the Roman bricks are of low density, high porosity and were produced from raw materials containing calcium poor clays fired at low temperatures.

**Ravisankar et al** carried out the qualitative analysis (FT-IR and XRD) to determine the major and minor constituent minerals in sediment samples from the
coastal area of Tuna, Gujarat. These results confirmed that the applied techniques are relatively quicker and more reliable in mineral analysis.

Mohamed Musthafa et al\textsuperscript{36} have estimated the temperature of the archaeological pottery sherds excavated from Tandikudi, India by comparing the porosity values with the pore system and vitrification observed in SEM morphology of the shreds. Chemical analysis gives the iron content hematite and magnetite which is directly connected with the atmosphere prevailed at the time of manufacture.

Venkatachalapathy et al\textsuperscript{37} have utilized Fourier Transform Infrared (FTIR) spectra and X-ray diffraction (XRD) analysis on archaeological pottery samples from Bhon, Maharashtra to establish the clay mineral type and its structural deformation due to firing, the maximum firing temperature attained during baking, firing conditions (open/reduced atmosphere) and iron mineral phase changes and also to identify mineral constituents.

An archeological funerary urn fragment from Campos dos Goytacazes, Brazil, was studied by Mangueira et al\textsuperscript{38} using electron paramagnetic resonance (EPR) and X-ray diffraction (XRD). The thermal stability of all paramagnetic species was calculated with isothermal treatment. The thermal stability of the three paramagnetic species indicates a firing temperature of around 500°C in the inner layer, between 400 and 500°C in the middle layer, an between 500 and 800°C in the outer layer. The presence of kaolinite structures only in the middle portion is consistent with the temperature values estimated.

Ljiljana Damjanović et al\textsuperscript{39} have made systematic archaeometric study of medieval pottery excavated Serbia including petrographic and chemical analysis,
powder X-ray diffraction and Fourier transform infrared spectroscopy. From the analysis they accomplished that Ras pottery, made of fine-grained clay with a rich mineral assemblage produced by firing in a temperature range between 800 and 900°C while Reljina Gradina ceramics were produced at similar temperatures from materials which, with regard to mineralogical and chemical composition, show clear similarity with local clay, suggesting that these samples are of local origin and production.

The ancient pottery samples excavated from Adichanallur (India) have been analyzed by FTIR, SEM and porosimetry methods by Velraj et al. The lower limit of firing temperature and firing atmosphere prevailed (FTIR), range of firing temperature (SEM) and upper limit of firing temperature (porosity) of the above artifacts during its manufacturing were assessed.
References


