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FI-IR Spectroscopic and Porosity Studies to Estimate the Firing Temperature of the Clay Brick

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Abstract

The objective of this paper is to determine the firing temperature of burnt clay bricks, by means of infrared spectroscopic studies and apparent porosity measurements. To confirm the firing temperature, a re-firing study has also been conducted. The different heating temperatures (200,400,600,800 and 1000ºC) were used. Traditional brick production did not have firing temperature controlled devices or mechanism in the fired clay brick production. The kiln operator decides the firing temperature and duration. The absence of the device frequently results in the over or under firing of bricks greatly affecting in the engineering properties of this widely used construction material a re-firing study has also been conducted.

Keywords: Fired Clay bricks, firing temperature, thermal transformation, porosity, re-firing.

1. INTRODUCTION

In this paper the FT-IR used to determine the probable firing temperature of the brick with re-firing samples. And the ifferent temperature of these spectra were analyzed and compared with raw material.

The bricks were classified geologically, according to the clays from which they were made, and it was possible to estimate how completely the bricks made from each geological formation were represented by sample and these were fairly evenly distributed over the different types of clay, although unavoidably there were some gaps. It was shown that there are often close family resemblances between bricks made at different works for clays of similar origin.(1)

To determine much about the quality of a brick by physically analyzing individual brick and packages of brick at the discharge end of the kiln, it is necessary to separate firing problems from other problems affecting the quality of the brick. It is essential to understand the characteristics of the particular brick making raw material, because many firing problems were interested.

During the firing process of phyllosilicates and accompanying minerals like quartz, feldspar, calcite, dolomite and hematite, a series of transformations occur which will be decisive for establishing the final properties of the brick products(2). Through the brick making process, these crystalline structures, once they are being formed. An instantaneous destruction of the pre-existing structure does not occur.(3,4)

The firing temperatures achieved and firing techniques employed by the brick maker can reveal their technical skills. The firing is the production step that transforms the clay into an imperishable product. So scholars are interested in studying the firing process, temperature achieved in brick manufacture. From the knowledge of firing temperature value achieved and the method of firing one may be able to conclude how the process lavigated and tempered the raw clay used to make a brick. To estimate the firing temperature and limits of temperature of the bricks, different physico-chemical methods have been evolved in the study of brick samples. Various spectroscopic techniques such as Mossbauer spectroscopy, electron spin resonance spectroscopy, scanning electron microscopy and infrared spectroscopy and the other scientific methods such as thermal expansion, thermo luminescence and porosity measurements were employed by many researchers. (5-14)

The firing temperature achieved by ancient potters were estimated using Mossbauer spectroscopy (5-7) estimated the firing temperature of the pottery shreds using thermal expansion method. The firing temperature of the potteries were also determined by using scanning electron microscopy and porosity measurements(8-12) have used the thermoluminescence technique to determine the firing temperature of potteries made up of ceramic material.

Studied the temperature of firing and virification stage of archaeological materials using (FT-IR) Fourier Transform Infrared spectroscopic technique(13). The lower limit of firing temperatures for different archaeological artifacts like potteries, bricks and tiles from Tamilnadu and Andhra were carried out in order to find the type of clay, and translocation by using FT-IR (Fourier Transform Infrared spectroscopy by. (15-18) In the present study infrared spectroscopy and porosity value used to study the firing temperatures achieved by the bricks manufacturer.

2. EXPERIMENTAL

The determination can have a possible small error due to the fact that the original firing schedule and atmosphere could not be replicated exactly(17). The samples were re-fired at the rate of 200°C per hour using the muffle furnace in the laboratory

to the temperatures of 200, 400, 600, 800 and 1000°C. The temperature at each state was maintained for an hour as the soaking time in the furnace. It is known that once clay is fired at a certain temperature and then cooled down, it freezes at a stage which cannot be further altered by subsequent refiring unless the initial temperature is exceeded. The spectra were recorded in the explored range of frequencies 4000 – 400 cm^{-1} for the refired samples after cooling to room temperature using Nicolet Avatar 360 FT-IR spectrometer. The samples were palletized by mixing with spectra grade KBr at the ratio of 1:30 by weight. The KBr pellet of 13 mm diameter was kept inside the sample holder and scanned at 1 cm^{-1} resolution for the entire mid-infrared region till the identical charts were obtained in the consecutive trials. In the table 1 and Fig1 the as received state spectrum is placed as the lowest trace in the figure for the sample (TMB) and the other spectra of the same sample refired to different higher temperatures are vertically displaced with respect to the received state one in the ascending order of temperatures.

3. RESULTS AND DISCUSSION

Bricks are made of clay minerals and the common major clay mineral used in the making bricks is kaolinite. The firing temperatures of the bricks can be determined with the help of the study of thermal transformation of the clay minerals present in the brick samples.

The dehydroxylation of kaolinite minerals was first studied [19]. They reported, from the Differential Thermal Analysis (DTA) results that the dehydroxylation takes place at temperatures between 400°C and 525°C. A small amount of water can however persist up to 750°C or 800°C at which dehydroxylation is complete. Dehydroxylation of kaolinite was studied [20] also using infrared spectroscopy. They observed that among the hydroxyl bands in the 3700 – 3600 cm^{-1} region the bands at 3675, 3650 and 3630 cm^{-1} persist after firing to a temperature of 327°C. Reported that on heating[21], the intensity of bands due to kaolinite in the 3700 – 3600 cm^{-1} region decreases and at 500°C only a week broad band remains in the 3600 cm^{-1} region.

The thermal behaviour of the bands due to hydroxyl groups in clay artifacts were studied [22] also. They observed that the clay minerals begin to lose their crystalline hydroxyl groups and start disorganizing at temperatures in the range 400 - 600°C and for some minerals it can continue up to 800°C. According to them the persistence of a weak band around 3600 cm^{-1} indicates the presence of the iron hydroxyls still in the clay body and points to incomplete dehydroxylation.

The transformation of the clay minerals during the firing process is reflected in the 1100 – 1000 cm^{-1} region also. As the temperature is increased there is gradual destruction of the layer structures of the mineral. The bands at 1100 cm^{-1} and 915 cm^{-1} due to Al (OH) vibrations in the octahedral sheet structures begin to disappear with increasing temperatures and at 500°C the band at 925 cm^{-1} disappears completely.[21-23] This dehydroxylation is followed by crystal framework collapse and the tetrahydral sheet disorder can be seen from the broadening of the Si-O stretching bands in this 1100 – 1000 cm^{-1} region.

The weak shoulder band observed at 875 cm^{-1} in the spectra of iron rich clay minerals has been attributed to Fe (Al OH) group present in the clay mineral.[24] The presence of this group indicates the persistence of the octahedral sheet structures in the clay mineral, indicating incomplete dehydroxylation and thus firing temperatures below 800°C. Because at 800°C dehydroxylation of kaolinite minerals are completed but this band is observed only in the case of iron rich clay minerals. [20-22,25]

Another parameter that can help in determining the firing temperature of these materials is the porosity of the materials. The application of porosity studies to brick samples and its correlation with particle size and firing temperature is of recent interest. [26] Correlated the porosity values of punic amphorae with their firing temperature. They found that the samples made from noncalcareous clays of fine particles, subjected to a firing temperature below 900°C, according to them will have lower range of porosity values. Calcareous clays of coarse particles subjected to temperatures above 1050°C also will have lower range of porosities. The samples made from coarse particles of both calcareous and noncalcareous clays subjected to firing temperature below 1000°C have porosity values in the higher range.

With these observation regarding the thermal behaviour of absorption bands at around 3600 cm^{-1} region, at 915 cm^{-1} and the another one at 875 cm^{-1} along with the porosity studies an attempt is made to establish the firing temperatures of the brick samples in the present study.

Conventionally, temperatures above 900°C are taken as being the minimum for firing clay-based materials[27]. For building bricks the densification process during sintering is closely related to the grain growth therefore, the solid-state diffusion mechanism using geometrical parameters was proposed to explain the consolidation of clay particles into a dense body[28]. Furthermore, the mineralogical composition is considered a key parameter in achieving the desired microstructure of clay bricks, which in turn influences the mechanical properties and durability of the product. [29-32] Sintering at an appropriate temperature transforms as-formed / green clay bodies into a rigid, contiguous (although porous) product via a complicated succession of physical and chemical changes [33] can be briefly described as follows:

1. Dehydration occurs at ~ 100°C which is followed by dehydroxylation of clay minerals at~450-750°C, depending upon the type and purity of the clay. [34]
2. Decomposition of initial ingredients and loss of CO_2, S and hydrocarbons at ~ 400-650°C, again depending on the type of the flux and other raw materials used.
3. Alpha/beta-quartz conversion at 573°C
4. Solid state minerals reactions and the beginning of vitrification at ~ 1050-1150°C however, the melt formation may begin at temperature of the mixture. [35]

In clays, water may be bonded chemically in the form of OH ions or loosely attached surfacial water. The dehydration/dehydroxylation of comparatively pure clay minerals takes place over different ranges of temperature depending on the clay minerals present. Chlorite loses its water at a relatively higher temperature, usually beginning at ~750, but this temperature again depends on the mineral chemistry. The transformation of illite (mica) begins with its dehydration at ~ 600-700°C. The carbonate minerals lose carbon dioxide on heating, for example, calcite decomposition begins at ~650°C. The phase transformation temperatures vary from sample to sample because of its dependence on a number of factors such as the minerals grain size, heating rate, and how easily the evolved carbon dioxide can be lost to the atmosphere. [36]

At~573° alpha/low quartz undergoes a structural phase transition into beta / high quartz. As this transition involves rotation rather than the breaking of bonds, the reaction is reversible, so that any high quartz present after the high temperature soak will invert to low quartz during the cooling part of the firing cycle. The brick expands during the rise in temperature however, if the cooling rate is too fast, then highly damaging micro-cracking can occur [38].

Keeping in view the previous studies on bricks, it can be concluded that the main factors affecting the quality of bricks are related to the mineralogy of the constituent raw materials and processing conditions. An appropriate firing temperature and optimum particle size of the raw materials produce more vitrification, less porosity and relatively higher density. The higher the fired density of the product the better will be the mechanical properties; however, improvement in density is 'in general' accompanied by a reduction in the insulation behavior of the brick. [39]

3.1. Cauvery riverbed samples

The infrared spectra of a sample (TMB) from Cauvery riverbed refired to different temperatures (Fig 1 &Table 1). The sample (TMB) showed absorption peaks around 3600cm^{-1} (3698-3667 and 3626cm^{-1}) due to presence of crystalline hydroxyls in the clay mineral structure.

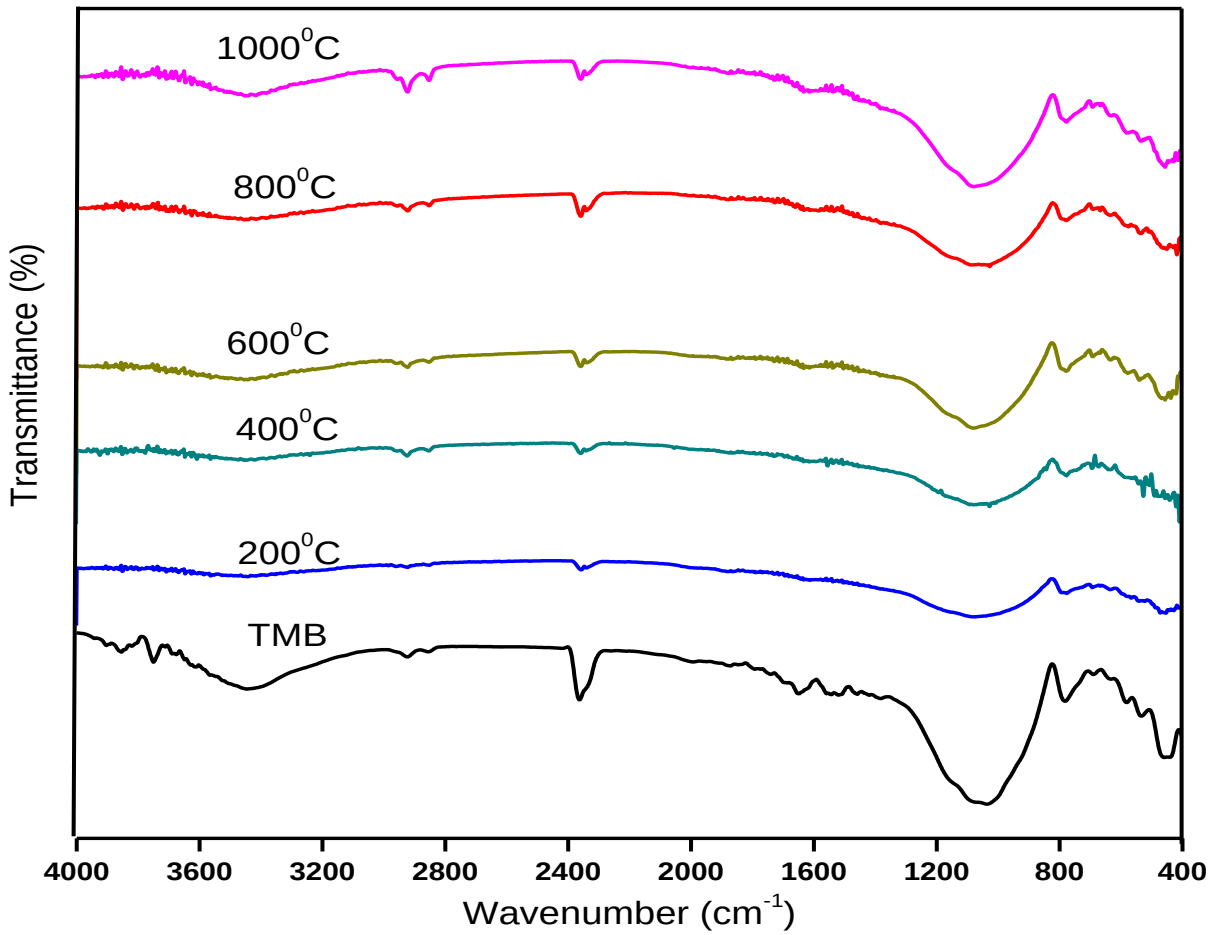

Figure 1: Infrared absorption spectra of Tharamangalam (TMB) sample at different refired temperatures

Table 1: The infrared absorption frequencies (cm^{-1}) of (TMB) brick refired at different temperatures

As received state		200 °C		400 °C		600 °C		800 °C		1000 °C		Tentative assignments	Name of the Mineral
Freq	**Int**	**Freq**	**Int**	**Freq**	**Int**	**Freq**	**Int**	**Freq**	**Int**	**Freq**	**Int**		
3687	vw	3698	vw	3667	vw	3667	vw	3666	vw	3667	vw	O-H Stretching of inner hydroxyl group	Kaolinite
		3674	vw	3646	vw	3625	vw	3625	vw	3453	vw		Kaolinite
3612	vw	3625	vw	3626	vw	3608	vw	3452	vw			Al-O-H Stretching inner hydroxyls	Kaolinite
3446	vw	3453	vw	3442	vw	3453	vw					V(OH) - Stretching	Kaolinite
1870	vw												
1845	vw												
1647	vw	1659	vw					1613	vw	1694	vw	O-H bending modes of molecular water	Kaolinite
		1564	w			1556	w	1537	w	1553	w	H-O-H Stretching	Illite
		1546	w			1537	w			1538	w		
1419	w	1537	w									Stretching vibration of CO_2	Carbonate minerals
1382	w											H-O-H Stretching (absorbed water)	Kaolinite
1253	w												
												Si-O Stretching (clay mineral)	Kaolinite
1035	vs	1027	vs	1076	vs	1027	vs	1080	vs	1081	vs	Si-O Stretching muscovite interference	Kaolinite
												Si-O quartz	Quartz
783	s	778	s	777	s	777	s	778	s	778	s	OH deformation linked to Al_3Mg_3	Quartz
690	w	667	w			693	w			692	w	Si-O Stretching	Kaolinite
632	w					639	w					Al_2O_3	
												Fe O of Fe_2O_3	
582	w											Fe-O of Fe_3O_4	Magnatite
												Fe^{3+} - O^{2-}	
534	w	534	w			526	w			540	w	Al-O-Si	Feldspar
						507	vw					Fe^{3+} - O^{2-}	
459	w	451	s	454	w	490	s					Si-O-Si	Quartz
441	v					476	s	456	w	457	s	Si-O-Al	Quartz
426	w	414	w	427	w	440	w			438	w	Si-O bending vibration	

v.w – very weak; v.s – very strong; m – medium; v.w.br – very weak broad; s – strong; sh – shoulder; v.w.sp – very weak & sharp; w.sh – weak & shoulder

The partial dehydroxylation of the clay mineral is thus reflected in the disappearance of the sharp band in this region. This is in agreement with the observation of prost et al., (1989). That among the hydroxyl group absorptions, only absorption around 3638cm^{-1} persists after heating to 500°C. this sample might have been thus fired to a temperatures of 500°C, as its lower limit. The Cavery riverbed sample TMB has absorption band around 3600cm^{-1} also indicating incomplete removal of crystalline hydroxyls on the a higher value for the lower limit for the firing temperature. None of the spectrum show any absorption in the 915cm^{-1} region, again indicating the lower limit of firing as 500°C. Because the band at 915cm^{-1} due to A1-OH persist only up to 500°C (Elsass, 1798, Maniatis, 1982 Miller, 1961). It is also observed that no band or shoulder appears of 875cm^{-1} in the TMB sample indicating that the dehydroxylation of Kaolinite minerals are completed are octahedral structure in the clay mineral disappeared (Ross and kerr, 1931; prost et al 1989, Maniatis, 1982).

A comparison of the spectra of refired samples with that of as received state sample reveals that the TMB sample of Cauvery riverbed showed absorption band around 3630 cm^{-1} indicating these samples would have been fired to a temperature below 800°C.

An attempt is made to relate the firing temperature with the porosity values in percentage. The porosity values of the Cauvery riverbed brick samples in the present study range from 16.21% to 20.31%.The porosity values are presented in table 2 .

Table 2: Porosity value and firing temperature of Tharamangalam (TMB) brick

Name of the sites	Estimated firing temperature in °C (using FT-IR)	Particle nature	Percentage of Porosity	Estimated firing temperature in °C (using porosity)
TMB	<800°C	Coarse	16.21	<900°C

For the samples that have low values (≤ 25%) of porosity the firing temperature determined for the sample were low maniates et al., (1984) also got lower values of porosity for (pottery) samples is fine particles size heated to temperature below 900°C. And another observation of maniatis was that coarse clay materials will have high porosity it fired to temperatures below 1000°C. The porosity value found to be 16.21% (TMB) form the porosity studies of the samples it was found that the bricks from Cauvery riverbed are made of course particles of clay materials as fired to higher temperature as have lower values or porosity. According to Maniatis (1984) sample made of coarse particles will have low value of porosity. The low value of porosity of the sample TMB indicates that the firing temperature of the sample in well below 900°C.

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