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CONTRIBUTION TO ACTIVE CONSERVATION: CHARACTERIZATION OF NEOLITHIC PERIOD MUD BRICKS REMAINS OF THE ARCHAEOLOGICAL SITE OF ÇATALHÖYÜK-TURKEY

Sina Noei

Kocaeli Archaeological Museum, Izmit, Turkey
(sina151@hotmail.com)

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ABSTRACT

In the 2014 excavation season, 6 mud brick samples were taken from different Neolithic layers of the Eastern mound of Çatalhöyük (C14 cal. 7100-5950 BC). For characterization of mud bricks, we carried out the following archaeometric techniques and analysis; hydrometer, atterberg limits, loss on ignition, deal with acid and sieve analysis, petrographic analysis (thin/thick sections, Stereo and Polarizing Microscope observations), X-Ray Diffraction (XRD) and Scanning Electron Microscopy and Energy Dispersive X-ray Spectroscopy (SEM-EDX). This methodology were performed for the first time in this study on mud bricks at Çatalhöyük.

According to results of this study, the chemical content of materials used in the mud bricks, the type/ratio of the binder, the particle size distribution, the effect of water content on volume change and soil consistency, as well as the type/ratio of the additives and filling materials could be determined. The results shed light as a reference point for the further mudbrick studies of the prehistoric period in Anatolia. As an interdisciplinary study, one of the goals of this research was to determine the chemical contents and physical properties of earthen building materials of Çatalhöyük to be able to make the same adobe mixtures for conservation activities. By using these compositions during the conservation treatments, original composition and characteristics of restoration mortars will not have been changed and physical stresses on original materials will be prevented.

Carried out in conjunction with the Konya Basin Paleoenvironments Project (KOPAL), the source of raw materials, binder and tempering agent used in each building from different layers were determined. The results of this study revealing clearly that the raw materials used for making mud bricks were derived from local resources nearby settlement. At the same time, the binders and additives were added to the raw materials in 4 different layers (VIA-IX layers) not only have been in each layer, but also in different buildings of the same layer have been changed (samples 1-2: buidings 80-76 and samples 3-4: buildings 7-11) in order to produce more robust and durable mud bricks.

The results proved that in the early layers of Çatalhöyük (layer IX/6800-6700 BC) only clay (muscovite) and organic additives (straw) were used for manufacture of mud bricks. But in VIII layer (6700-6600 BC) they were used two type of clay (kaolinit+muscovite). In VII Layer (6600-6500 BC) gypsum were added to mud bricks for the first time as binder. In VIA Layer (6500-6400 BC) firstly gypsum and slacked lime were used together and then only the slacked lime were used.

KEYWORDS: Earthen structures, Analysis, Mud brick, Neolithic, Central Anatolia, Clay, Lime, Gypsum, conservation

1. INTRODUCTION

This article provides a methodology for understanding the chemical content, physical properties, and behavior of mud bricks through various analyses compositions, using a case study from Çatalhöyük, Turkey. In the 2014 excavation season, 6 mud brick samples were taken from different Neolithic layers of the Eastern mound of Çatalhöyük. The East mound (Figure 1) is an optimal site for a systematic study of mud bricks because of its continual habitation sequence spanning 1400 years (Hodder, 2007).

Figure 1. East mound, South area.

The following analyses were carried out for the characterization of mud bricks: Hydrometer/sieve and Atterberg limits (soil mechanics tests), loss on ignition, deal with acid and sieve analysis, petrographic analysis (thin/thick sections, Stereo and Polarizing Microscope observations), X-Ray Diffraction (XRD) and Scanning Electron Microscopy and Energy Dispersive X-ray Spectroscopy (SEM-EDX). By carrying out this analysis, physical properties, chemical and mineralogical composition, the ancient technologies, and source of raw materials that were used for production of earthen building materials were identified.

In this study a package of analysis was conducted that contains simple chemical tests with soil mechanical tests besides detailed archaeometric analyses. Archaeometrical analysis (Liritzis et al., 2020) on an integrated approach is enough for characterization of mud bricks. Thus, besides the petrographic and simple chemical analysis, it is also necessary to carry out analysis that can determine the physical properties, the type/amount of binder, amount/distribution of clay, silt, sand and gravel by detail (hydrometer and sieve analyses together), type of soil and behaviour of mud bricks in contact with water (atterberg limits).

In the past years, there were a lot of scientific research for characterization of mud bricks. But in some cases, for characterization of earthen building materials some methods of analyses were used that did not give accurate and useful results. For example, analyses like magnetic susceptibility and pXRF (portable X-ray fluorescence). Portable X-ray fluorescence can give accurate result on metals and magnetic susceptibility can give accurate results on hard and homogeneous materials like stones, not on mud brick or plaster samples. Muddy mortars (mud bricks, pisse, cob, wattle and daub…) are made by hand and specially the tempering agents (mostly organic ones). The porosity/heterogeneity of earthen materials are not like stones or metals and these materials have a heterogeneous and soft texture. In these materials the lights and electrical waves cannot pass properly (will sheer away), and the results obtained will not be accurate and suitable for use in the conservation and restoration activities of earthen building materials and scientific interpretations. Thus, the most important point here is to choose methodology that can give reliable and accurate results. The results of this kind of studies cannot go further than the unreliable data and the results could not be used as characterization and scientific active conservation/restoration projects. This kind of scientific research must be done by expert scientists.

This interdisciplinary study at Çatalhöyük, not only enabled us to determine the chemical contents and physical properties of earthen building materials but also provided a scientific basis to reproduce the same adobe mixtures. By using these compositions during the conservation and restoration activities, original composition and characteristics of earthen restoration mortars will not have been changed and physical stresses on original materials will be prevented.

In situ conservation and exhibition of earthen structures (mudbrick) as a site museum is one of the most complex issues in site conservation and management of archaeological sites. In spite of various efforts worldwide, the end results are far from satisfying (Mazar, 1999). Conservation attempts of earthen structures in Çatalhöyük had already been made since the early 1990's and have continued up to the present day. Since the 1990's there have been various archaeological projects in Çatalhöyük including KOPAL (Konya Basin Paleoenvironments Project) that is a project for identifying the source of raw materials, scientific investigations, passive conservation activities such as temporary summer shelters during excavations, large permanent shelters, and active conservation treatments to preserve the earthen architectural remains. It is worth noting that the traditional pottery production in Konya covers the past 8000 years (Unal, 2021). Shelters constructed above the two main excavation areas (East mound) protect the archaeological structures from environmental damages. The shelters have allowed excavation, conservation, and exhibition to take place beneath them but the earthen building materials and the state of preservation of the mud bricks present serious problems for

preservation work. However, the desired successful results have not been achieved yet (Atalay et al. 2010).

Çatalhöyük is located at 10 kilometers Southeast of Konya's (ancient Iconium) Çumra Village in the Southern Anatolian Plateau (Turkey). The site is a hill with two mounds (east and west) with different altitudes (Figure 2). Since this mound is situated at a bifurcation, it is called "mound at the fork" hence Çatalhöyük in the Turkish language, where *çatal* means fork and *höyük* means cairn.

Figure 2. Air photo of Çatalhöyük, Neolithic East mound-right and Chalcolithic West mound-left (Konya-Çatalhöyük Research Project Archive, 2020).

In 1958 during the expedition of Southern Anatolia, Prof. James Mellaart discovered a huge mound consisting of neolithic settlement levels called Çatalhöyük. In 1961, Mellaart began its excavations which lasted until 1965 with a break in 1964. The next excavations were restarted in 1993, under the direction of the British archaeologist Prof. Ian Hodder. These excavations were projected for 25 years and are among the largest archaeological projects of our time in Turkey (Balter, 1998). Between the 2019-2020 seasons, Assoc. Prof. Çiler Çilingiroğlu from Ege University-Izmir, led the excavations for two years and finally since 2021, Assoc. Prof. Ali Uumut Türkcan from Anadolu University-Eskişehir has been leading the excavation.

James Mellaart's excavations (1961-1965) were confined mainly to the Southwest corner of the East mound (Figure 3), two small trenches were also dug on the Chalcolithic West mound. In fact, Çatalhöyük is late in the Neolithic sequence, occurring at the end of the Aceramic Neolithic and continuing through the Pottery Neolithic and into the Chalcolithic Period. The prehistoric mound settlements were abandoned before the Bronze Age (Hodder, 2007). In July 2012, the site was inscribed as a UNESCO World Heritage site.

Çatalhöyük consists of East and West mounds. The Eastern Mound consists of twelve building levels of a neolithic town which was inhabited from 7300-6100 BC, that is, 1200 years without interruption (Thissen et al, 2002) and the Western Mound contains Chalcolithic occupation levels from 6200-5200 BC, which reflect the continuation of the cultural practices evident in the earlier Eastern Mound. The settlement was neither destroyed nor looted. According to present-day estimates, up to about 10000 people lived together in Çatalhöyük (Hodder, 1998).

Figure 3. Southwest corner of the East mound-bottom of photo (Konya-Çatalhöyük Research Project Archive, 2020).

Central Anatolian Çatalhöyük is a rare example of well-preserved Neolithic settlement that is considered one of the key sites for understanding changes in Prehistoric ways of life, from the domestication of cattle and the adoption of a settled way of living to the invention of pottery and metallurgy (Hodder, 2007). It is also an optimal site for the study of mudbrick architecture because of its continual habitation sequence, as well as the extensive exposure of architectural remains. This deeply stratified sequence, with houses built one upon another, provides a unique opportunity to document temporal changes both within and between the houses through all the main occupational phases (Hodder et al, 2014). Moreover, people in Çatalhöyük usually painted pictures on two walls of their houses to document aspects of their lives and experiences (Gimbutas, 1990). They buried the deceased under the floors in their houses with characteristic grave goods (Brosius, 2005).

The site was set up as large numbers of buildings clustered together (Figure 1). The houses at Çatalhöyük through the early part of the sequence (the houses become multi-roomed complexes in the upper levels and in the West mound) consist of a main room with 1-3 side rooms that are used for storage and food preparation. The main rooms have walls there are more frequently replastered and normally contain the entrance ladder or stairs on the south wall, with the oven and hearth beneath the ladder. Houses with square walls were adjacent but the houses did not share common walls and every house had its own

wall. Houses were planed separately, and one house was built next to another. Sun-dried mudbricks, trees, and reeds were used in the making of the houses.

As mentioned before, there were a lot of interdisciplinary studies and projects in Çatalhöyük during the 1993-2018 period. Among these projects, the Konya Basin Paleoenvironments Project (KOPAL) conducted an extensive analysis of the geomorphology and geoarchaeology surrounding the Çatalhöyük mounds (Roberts et al, 2007). According to this project, the origins of the source material for mudbrick manufacture were determined. The results of the KOPAL research program established a stratigraphy with four primary units. The basal layer; contain fine-grained, carbonate-rich lake marl from the former Pleistocene Lake Konya (Roberts et al, 2009). The next layer includes dark organic clay up to 30 cm thick. The 3rd is the lower alluvium layer with a heavy dark grey-brown silty and smectite-rich clay with less than 5% organic matter and lacking a coarse fraction (Roberts et al, 1999). The 4th unit is the upper alluvium, reddish-brown silty clay with a significant coarse fraction (Roberts et al, 1996).

The aim of this study is to reveal the chemical composition and physical properties, beside the content of the mud bricks (binder, fillers, and additives) to reach the knowledge of the production process and have a better understanding about the causes of degradation processes of the earthen building materials in Çatalhöyük; that is, to identify the contents of the mud bricks in detail and have a better understanding of the interaction and behavior with their environment. On the other hand, thanks to the data of the KOPAL Project (providing the location of the raw materials) and in line with our analysis results, it is possible to make replicas of mud bricks with the same materials and mixture as the originals. This research offers an important opportunity and reference point for the use of modern scientific methods to analyses and reproduce the ancient technology used in mud brick buildings in Çatalhöyük.

2. MATERIALS AND METHOD

The accurate characterization of the Archaeological materials by various analysis methods has great importance in terms of active and passive conservation activities. As I mentioned before, the most important point is to choose analysis that can give reliable results not any type of analysis.

In order to understand the chemical content, weathering and decay processes, behavior of materials and technological processes and developments applied for manufacture of mud bricks, physico-mechanic analyses besides simple chemical tests, petrographic and archaeometric analyses provide very useful data in terms of determining the quality and quantity of archaeological materials.

In addition, soil mechanics research (in order to identify appropriate soil) is one of the fundamental activities in restoration and conservation of earthen building materials. The type of study recommended here is necessary for active conservation projects, specially to find access to suitable soil for manufacture of mud bricks in conservation and remedy projects of original and historical earthen structures.

As some of these analyses are not non-destructive applications (hydrometry, Atterberg limits, petrographic analysis, ignition loss, acid loss etc), so it is necessary to take samples in a way that does not inflict damage to the integrity and authenticity of the ancient archaeological constructions.

In some cases (e.g. Love, 2017), studies were carried out by taking very large amount of (40-50) mud brick/plaster samples. However, such a large amount of samples will not go beyond for only give damage to the integrity and authenticity of historical monuments. The number of samples is not important in studies that conducted without an accredited scientific methodology. According to the purpose of the research, by taking one sample from a wall of each building or one from a layer in a systematic way (by use accurate analysis), could be achieved much more reliable and scientific results.

2.1. Materials

In the 2014 excavation campaign, a total of 6 mud brick samples were taken under the auspices of the ex-directorate of Çatalhöyük excavation, Prof. Ian Hodder. All the samples were taken from the area that was excavated by the first director, Prof. James Mellaart. Six samples were taken from 4 different Neolithic layers of the East mound (South area). Two samples from the VIA layer (6500-6400 BC), two samples from the VII layer (6600-6500 BC), one sample from VIII layer (6700-6600 BC) and one sample from IX layer (6800-6700 BC).

The sampling of the mortars was carried out using a straw and a trowel, removing the first layer in contact with the atmosphere. Before conducting the samples to the physical, chemical, mineralogical, and other analytical investigation, samples were visually inspected by naked eye/magnifying glass for their conditions, texture, and nature of any visible aggregates. The Munsell Colour Chart was applied for describing mud bricks colours, allowing for direct comparison of mud bricks anywhere in the world. During the sampling latex gloves were worn and at no time were samples touched with the naked hand. The results of visual observations and features of the samples collected are reported in Table 1.

Table 1. Location and features of the samples.

Sample No	Dating, Layer	Location: East Mound, South Area	Visual Features of Samples	Munsell Color Chart
1. Sample Mudbrick	6500-6400 BC, VIA Layer	Building No. 80, Room 135, North wall No. 5036	Quite Solid Structure, Homogeneous, Non-Porous Building.	10 YR, V:4, C:2, Hue: 4/2, Dark Grayish Brown
2. Sample Mudbrick	6500-6400 BC VIA Layer	Building No, 76, Room 137, East Wall, No. 3401	Solid Structure, Homogeneous, Non-Porous Building	10 YR, V:6, C:3, Hue: 6/3, Pale Brown
3. Sample Mudbrick	6600-6500 BC VII Layer	Building No, 7, Room 487, South wall No. 3700	Weak Structure, Homogeneous, Non-Porous Building	10 YR, V:6, C:2, Hue: 6/2, Light Brownish Gray
4. Sample Mudbrick	6600-6500, BC VII Layer	Building No, 11, Room 169, North wall No.1122	Solid Structure, Heterogeneous, Fine Grained, Porous Building	10 YR, V:5, C:3, Hue: 5/3, Brown
5. Sample Mudbrick	6700-6600, BC VIII Layer	Building No, 4, Room 150, South wall No 263	Solid Structure, Heterogeneous, Very Fine Grained, Non-Porous Building	10 YR, V:5, C:3, Hue: 5/3, Brown
6. Sample Mudbrick	6800-6700 BC, IX Layer	Building No, 2, Room 116/117, North Wall No. 64	Quite Solid Structure, Heterogeneous, Very Fine Grained, Non-Porous Building, Fiber Additive	10 YR, V:7, C:2, Hue: 7/2, Light Gray

2.2. *Method*

The active and passive conservation treatments of archaeological materials require knowledge concerning the construction techniques and properties of the ancient materials as well as the deterioration factors and past interventions. A multi method approach that integrates different analytical techniques was adopted for the characterization of the physical and chemical properties of the mud brick samples of Çatalhöyük settlement. Hydrometer and sieve analysis, Atterberg limits, loss on ignition, deal with acid, petrographic analysis (thick and thin sections, observations with Stereo and Polarizing Microscope), X-Ray Diffraction (XRD) and Scanning Electron Microscopy and Energy Dispersive X-ray Spectroscopy (SEM-EDX) analysis were carried out.

In order to find the amount and type of clay size particles used in mud bricks, sieve analysis, hydrometer, and XRD analyses were performed together. In the continuation, Atterberg limits, deal with acid, loss on ignition analyses were performed to understand the physical/mechanical properties and the effect of water content on volume change and soil consistency of the collected samples. Petrographic investigations and Scanning Electron Microscopy (SEM) were conducted for studying the morphological characteristics of the samples. Parallel to petrographic investigations and X-Ray Diffraction (XRD) results, Energy Dispersive X-ray Spectroscopy (EDX) analysis helped us to determine the chemical composition of mud bricks.

Atterberg limit's (ASTM D4318) test was performed to determine the effect of water content on volume change and soil consistency (Howard, 1984). Moisture content is a measure of the shrink-swell and strength characteristics of cohesive soil as demonstrated in liquid limit (LL) and plastic limit (PL) testing. The difference between the plastic limit and liquid limit is defined as the plasticity index (PI). The liquid limit and plastic limit are water contents at which mechanical properties of soil change. The results of this tests are used to classify soil in accordance with ASTM D2487, and to estimate the swell potential of soil (Kalinski, 2006).

For the liquid limit test, soil (mud bricks) was passed through a #40 sieve (0.425 mm opening) and added distilled water to approximately 50 g of soil until it gained the consistency of peanut butter or frosting. A flat layer of soil was spread in the cup of *Cassagrande* device with the frosting knife and a grooving tool was used to cut a groove in the middle of the soil. The liquid limit device (*Cassagrande*) cranked at a rate of 2 cranks per second (for each crank the cup dropped from a height of 1.0 cm), the number of cranks that are required to close the groove over a length of 0.5 inch were recorded. The cup cleaned out and the same steps were repeated 4-6 times with the data recorded each time. The removed soil (from the cup) was heated in an oven (Heraeus) at 105 ± 5° C. After each heating the sample was cooled in a desiccator and then weighed (± 0,10 mg, Mettler H20). From the weight differences, the percent of moisture absorption (at 105° C), were calculated. This procedure provides a single data point corresponding to a single crank count and a single water content. Since we used the multi-point method to drive the liquid limit, the procedure was repeated at three different water contents and the data were plotted in a semi-log graph against the number of cranks. Liquid limit is defined as the water content at which the groove closes at exactly 25 cranks.

For the plastic limit test, soil (mud bricks) was passed through a #40 sieve (0.425 mm opening) and distilled water was added to make sticky little mud-balls. A pea-sized ball of mud was rolled on a glass plate to form a rod (with a diameter of 0.125 inch). This process was repeated until the soil crumbled to make a rod, which was quickly placed in an oven (Heraeus) for a moisture content reading. After each heating the sample was cooled in a desiccator and then weighed (± 0,10 mg, Mettler H20). From the weight differences, the percent of moisture absorption (at 105° C), were calculated (with the same method in loss on ignition analysis at 105° C). At this point the water content of the soil is the plastic limit. This process was repeated 3 times and the mean value was evaluated as the plastic limit.

Hydrometer (ASTM D422) and sieve analysis were carried out to determine the grain size distribution of soil and the percentage (%) of clay, silt and sand. This information is used to classify the soil in accordance with the Unified Soil Classification system (Kalinski, 2006).

For the hydrometer analysis, approximately 50 gr soil that passed from the #40 sieve (0.425 mm opening) was deal with 125 ml of the sodium hexametaphosphate (Calgon) solution in a 250 ml glass beaker and the mixture soaked for 24 hours. Then, all the mixture was transferred to specified dispersion cup and the mixture was stirred (with the specified stirring device at a rate of 10.000 rpm) for one minute. All the slurry was poured into a cylinder (1000 ml etched graduated) and filled with the distilled water to just below the etched mark. With help of a rubber stopper the cylinder was turned upside down and back, at a rate of 1 turn per second for 1 minute (60 turns). The hydrometer (152 H type hydrometer) was putted in the cylinder and a timer (timing device capable of reading to the nearest second) was started immediately. After 2 minutes the hydrometer readings were started with subsequent readings at 5, 15, 30, 60, 90, 120, 250, and 1440 minutes. At the same time the water temperature (thermometer capable of reading to the nearest 0.5° C) in the cylinder containing the soil slurry was recorded. According to these data (by Stoke's law) percentage of clay, silt and sand of the samples were calculated.

In the loss on ignition test a finely ground sample approximately 500 mg was placed in a porcelain crucible and weighed (± 0,10 mg, Mettler H20). The samples were heated in an oven (Heraeus) at 105 ± 5° C ±, 550 ± 5° C and 1050 ± 5° C. After each heating (24 hours), the samples were cooled in a desiccator and weighed. From the wight differences, the percentages of moisture absorption (at 105° C), the amount of organic materials (at 550° C) and calcium carbonate content (at 1050° C) of the samples were calculated (table 3).Deal with acid and then sieve analyses were carried out to determine the total content of the binding medium (carbonated material) and siliceous aggregates and other insoluble materials (organic materials) with acid were separated and the size grading of the siliceous aggregates was evaluated by sieve analysis. A dried sample (50 gr) was treated with HCl (10%) to dissolve the binding medium, and the acid-insoluble residue was filtered, washed and dried at 105 ± 5° C. The size grading of the acid- insoluble residue, being the siliceous aggregates, was sieved through different mesh sizes of, < 63, 63, 125, 250, 500 and 1000 microns, as well as 2.5mm and 5mm. The types, shapes, colors, and inclusions of material, as well as the approximate ratios of the different types of the aggregates were identified by means of a stereo microscope (Nikon SMZ 800 model) and the sieve analysis.

Petrographic analysis, thick and thin sections, Stereo and Polarizing Microscope observations were carried out with the aim of determining the mineralogical compounds of the samples and the substances within them in their approximate quantities. Thick section observations were carried out with a Nikon SMZ 800 model stereo microscope. Thin section studies were carried out using a Nikon Eclipse CI-POL model polarizing microscope to identify the minerals. For the petrographic analysis, samples molded in epoxy resin (Araldite AY103+HY 956) were cut with a low-speed saw (Buehler Isomet) to obtain thick and thin sections. The sample sections were stuck on petrographic slides and thickened first up to 1-2 mm and then down to 30 microns by using various sizes of silicon carbide powders (Buehler). The minerals were identified by a polarizing microscope (Nikon Eclipse CI-POL model) with transmitted light. Photographs were taken with an Olympus OM-1 camera.

Scanning Electron Microscopy and Energy Dispersive X-ray Spectroscopy (SEM-EDX) was carried out for the morphological description, determine the chemical composition, advanced and quantitative information and especially for the ratios of SiO_2, Al_2O_3 and CaO in mudbrick samples. A Carl Zeiss EVO LS 10 MODEL scanning electron microscopy equipped with a BRUKER and QUANTAX 200 Energy Dispersive X-ray Spectroscopy (EDX) spectrometer was used to investigate the micro structural and micro chemical properties of the mud bricks. The analyses were carried out on fresh-fractured sample, operates at 13 kV voltage, and current of 4 µA filament, 80 Pascal air vacuum, 225 magnification, and 12-13.2 mm working distances. Energy Dispersive X-ray Spectroscopy (EDX) was employed on the selected areas of Scanning Electron Microscopy (SEM) images (with the scales changing in the range of 50-100µm) belonging to mud bricks. EDX data were in the form of elemental concentration, and they were transformed

into oxides as wt. % which is conventionally used. For the SEM-EDX (Scanning Electron Microscopy-Energy Dispersive X-ray Spectroscopy) analysis, samples molded in epoxy resin (Araldite AY103+HY 956) were cut with a low-speed saw (Buehler Isomet) and polished with 3, 1 and 0.25 micron-sized diamond polishing compound (Metodi). After being polished with silicon carbide powders, the polished surfaces were covered with gold and the samples were analyzed both in Scanning Electron Microscopy (Carl Zeiss EVO LS 10 MODEL) and Energy Dispersive X-ray Spectroscopy (BRUKER and QUANTAX 200- Program; Espirit 1.8.5.).

Powder X-ray diffraction carried out to identify chemical compounds and particularly to identify the clay types used in earthen materials of Çatalhöyük. For the X-ray diffraction analysis, the samples were ground to below 90 microns and prepared in a special holder to identify the minerals. The mineral/phase contents of the samples were revealed by a GNR-APD 2000 Pro X-ray diffractometer using Cu-Kα with the working parameters of 30 kW, 15 mA. XRD analyses were carried out in the range of 0-60 2θ.

3. RESULTS AND DISCUSSION

A correct characterization of ancient materials can only be accomplished through interdisciplinary studies, which should include chemical, physical, petrographic and archaeometric analysis. According to analyses results, the chemical content, the type/ratio of the different types of binder, the amount of moisture, the particle size distribution (amount of the clay, silt, sand particles and also silicious aggregates), the type/ratio of the additives and filling materials used in the mud bricks can be identified.

3.1. Results of analyses

My approach is multidisciplinary and involves the chemical and physical characterization of mud brick samples. Samples were characterized using Hydrometer and sieve analysis, Atterberg limits, loss on ignition, deal with acid, petrographic analysis (thick and thin sections, observations with Stereo and Polarizing Microscope), X-ray diffraction (XRD) and Scanning Electron Microscopy and Energy Dispersive X-ray Spectroscopy (SEM-EDX) analyses. According to visual examinations all samples were made from fine aggregates and except the 3rd sample, all the others have solid structure. Only the 6th sample contain organic fiber additive and only this sample do not contain ash. According to Munsell Color Chart, the 4th and 5th samples are brown, and the color of other samples were all different from each other. The difference in colors is due to minerals and tempering agents used during manufacturing. Research from Çatalhöyük demonstrated that mud bricks of a similar color had different compositions and, inversely, different colored bricks had shared compositions (Love, 2017). According to Atterberg limits results there were two different types of soil, clay/loamy (CL) and high plasticity-Clay (CH). It should also be mentioned that there were 3 different types of binders that were used in samples as clay, slaked lime, and gypsum ($CaSO_4.2H_2O$). A detailed description of analyses results is broken down in the following paragraphs.

In the first sample (Figure 4), VIA layer (6500-6400 BC), according to hydrometer analysis (Figure 5) and atterberg limit's (Figure 6), 37.09 % of aggregates are clay size, 41.39 % silt size, 21.52 % sand size, gravel size aggregates rate is 0.00 %, soil type is clayey-loamy (CL), liquid limit rate is 49.10 %, the plastic limit is 22.80 %, and the plasticity index is 26.30 % (Table 2). According to the results of the loss in ignition analysis, loss at 105^0 C (moisture) is 5.23 %, loss at 550^0 C (organic materials) is 5.79 %, loss at 1050^0 C (calcium carbonate) is 28.34 %. According to deal with acid, 33.36 % of the sample reacted and 66.64 % retained (Table 3).

Figure 4. 1st sample.

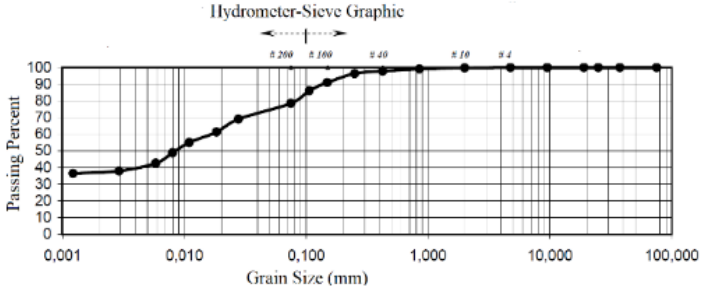

Figure 5. 1st sample Hydrometer and Sieve graphic.

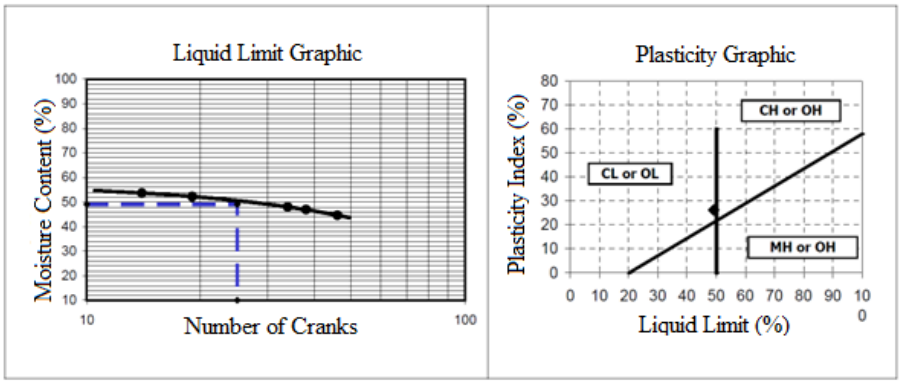

Figure 6. Liquid limit and plasticity graphics of the 1st sample.

Table 2. Hydrometry analysis and Atterberg limit's results.

Sample No.	Gravel (%)	Sand (%)	Silt (%)	Clay (%)	Liquid Limit	Plastic Limit	Plasticity Index	Soil Type
1	0,00	21,52	41,39	37,09	49,10	22,80	26,30	CL
2	0,00	24,94	34,45	40,61	48,40	20,80	27,60	CL
3	0,00	21,97	44,13	33,90	45,70	19,30	26,40	CL
4	0,73	19,19	33,19	46,89	50,40	21,90	28,50	CH
5	0,00	16,34	35,71	47,95	51,80	24,60	27,20	CH
6	0,00	13,74	34,47	51,52	59,10	22,40	26,70	CH

CL: Clay-Loamy, CH: High Plasticity Clay.

Table 3. Loss on ignition, deal with acid and sieve analysis results.

Sample No	Loss on ignition (%)			Acid loss (%)		Sieve (%)							
	105^0 c	550^0 c	1050^0 c	Lost	Retained	5000	2500	1000	500	250	125	63	<63
1	5,23	5,79	28,34	33,36	66,64	0,00	0,00	0,00	0,41	6,86	17,05	25,82	49,86
2	3,06	2,48	24,33	25,26	74,74	0,00	0,00	1,55	5,37	14,94	14,44	23,81	39,86
3	3,88	2,95	21,45	21,70	78,30	0,00	0,00	0,09	0,74	3,26	9,43	30,44	56,05
4	3,84	4,20	18,95	19,88	80,12	0,00	1,33	0,14	1,49	7,26	20,21	16,54	53,03
5	3,81	4,78	22,52	26,10	73,90	0,00	0,00	0,00	0,16	4,11	4,75	12,50	78,48
6	4,28	4,44	21,09	20,71	79,29	0,00	0,00	0,97	4,89	11,40	9,11	15,11	58,52

Petrographic investigations shows that the aggregates smaller than 125 microns consist of mica, a small amount of black slag powder, 2-3 % quartz and the rest is clay/silt size material. A small amount of aggregates between 125-500 microns are black slag powder, around 5% quartz and the rest are brown- colored undispersed masses. The rest of the aggregates larger than 500 microns are quartz, black slag powder and brown undispersed masses. The sample contain 5-10 % black slag fragments and the rest are quartz and feldspar minerals. As result of XRD analysis (Table 4), protoenstatite, quartz, albite, calcite, and dolomite were detected (Figure 7). According to SEM - EDX (Figure 8) analysis (Table 5), SiO_2 (50.09 %), CaO (11.24 %), Al_2O_3 (15.10 %), MgO (3.45 %), FeO (9.04%), K_2O (5.59 %), Na_2O (1.62 %), Ti_2O (0.38 %), SO_3 (0.25 %), and Cl^- (3.23 %) were detected (Figure 9).

Table 4. Minerals according to XRD analysis.

Sample No.	**Minerals**							
1	Quvartz	Calcite	Albite	Dolomite	Protoenstatite	-	-	-
2	Quvartz	Calcite	Albite	Dolomite	Gypsum	Hydrophilite	Graphite	Titano Magnetite
3	Quvartz	Calcite	Albite	-	Birnessite	Muscovite	-	-
4	Quvartz	Calcite	Albite	-	Gypsum	Hydrophilite	-	-
5	Quvartz	Calcite	Albite	Dolomite	Kaolinite	Muscovite	-	Low Cristobalite
6	Quvartz	Calcite	Albite	-	-	Muscovite	--	-

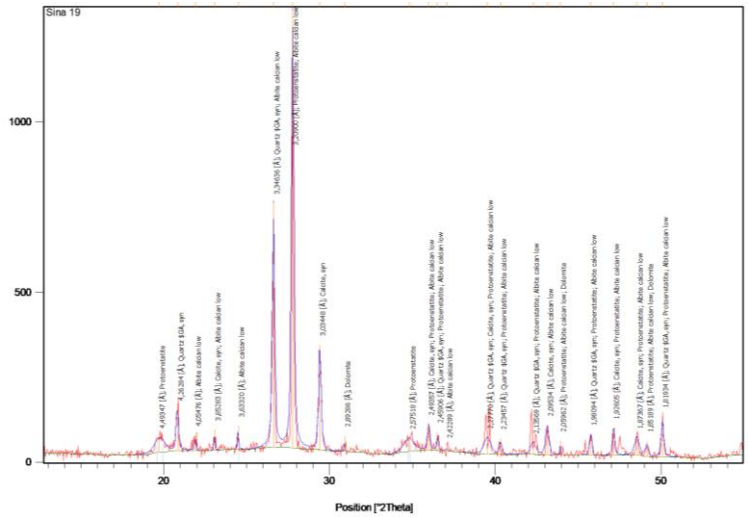

Figure 7. 1st sample XRD result.

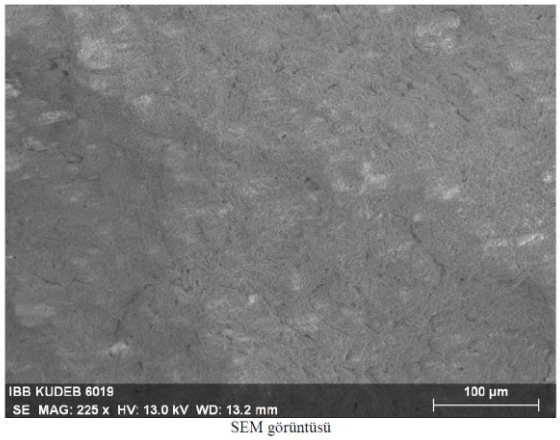

Figure 8. SEM images of micro cracks.

Table 5. EDX results of 1st sample.

Element	Norm. C (Wt. %)	Atom. C (At. %)	Compound norm.	Comp. C (Wt. %)
Oxygen	42.05	59.19		0.00
Sodium	1.21	1.18	Na_2O	1.62
Magnesium	2.08	1.93	MgO	3.45
Aluminium	7.99	6.67	Al_2O_3	15.10
Silicon	23.41	18.78	SiO_2	50.09
Sulphur	0.10	0.07	SO_3	0.25
Potassium	4.64	2.68	K_2O	5.59
Calcium	8.04	4.25	CaO	11.24
Iron	7.03	2.83	FeO	9.04
Phosphorus	0.00	0.00	P_2O_5	0.00
Chlorine	3.23	2.05		3.23
Titanium	0.23	0.11	TiO_2	0.38
Total: 100.00 100.00				

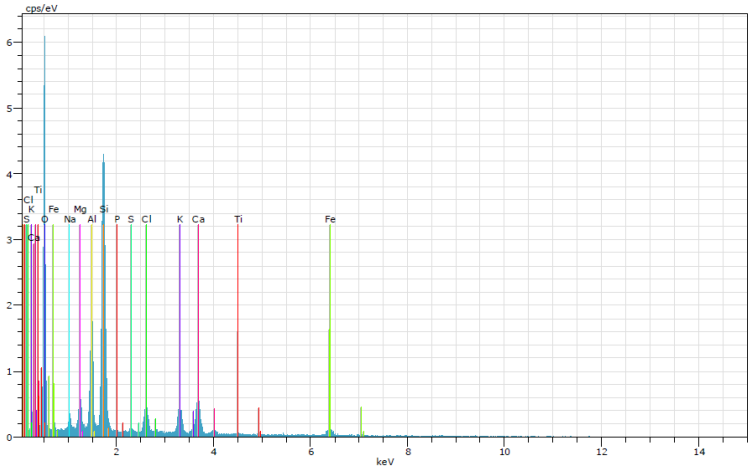

Figure 9. 1st sample EDX spectrum.

The first sample consisting of 10 % ash, a total of 30-35 % carbonate aggregates (calcite and dolomite), and 20 % lime as binder. A total of 60-65 % consists of siliceous materials, 20 % of cream-colored undispersed masses (fired bricks/burnt aggregates) defined as *secondary used* materials, 22 % of sand size, and the rest of silt/clay size aggregates. The aggregates are angular shaped and of terrestrial origin in 1 mm under sieve size (Figure 10).

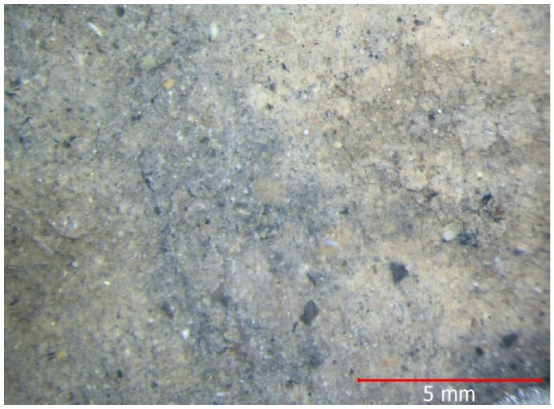

Figure 10. Thick section image of 1st sample.

In the second sample (Figure 11), VIA layer (6500-6400 BC), according to hydrometer analysis (Figure 12) and Atterberg limit's (Figure 13), 40.61 % of aggregates are clay size, 34.45% silt size, 24.94 % sand size, gravel size aggregates rate is 0.00 %, soil type is clayey-loamy (CL), the liquid limit rate is 48.40%, the plastic limit is 20.80%, and the plasticity index is 27.60 (Table 2). According to the results of loss in ignition analysis, loss at 105 ^{0}C (moisture) is 3.06 %, loss at 550 ^{0}C (organic materials) is 2.48 %, loss at 1050 ^{0}C (calcium carbonate) is 24.33 %. According to deal with acid, 25.26 % of the sample reacted and 74.74 % retained (Table 3).

Figure 11. 2nd sample.

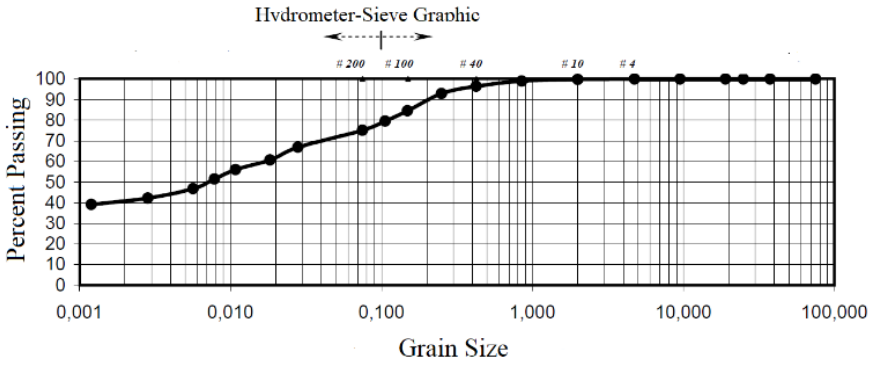

Figure 12. 2nd sample Hydrometer and Sieve graphic.

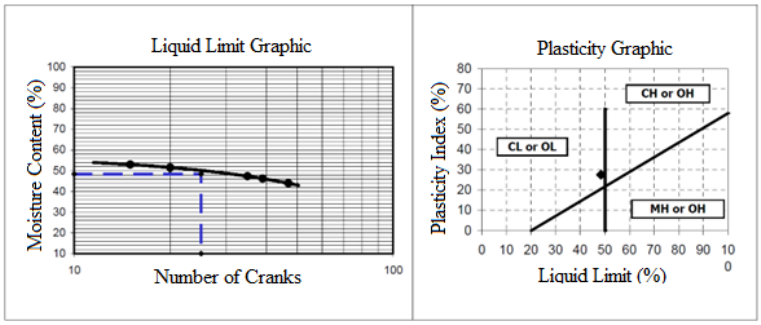

Figure 13. Liquid limit and plasticity graphics of the 2nd sample.

Petrographic investigations show that the aggregates smaller than 125 microns are black slag powder and the remainder is clay/silt size material. Aggregates between 125 and 500 microns are mica, a small amount of black slag powder and the remainder are cream-colored undispersed masses. About 20 % of the aggregates larger than 500 microns are quartz, the rest are cream-colored undispersed masses. The sample contain a few metamorphic fragments, biotite, quartz mineral and 3-5 % black slag particles. As a result of XRD analysis, gypsum, albite, hydrophilite, quartz, dolomite, calcite, graphite and titano magnetite were detected (Figure 14). According to SEM-EDX (Figure 15) analysis (Table 6), SiO_2 (52.74 %), CaO (10.78 %), Al_2O_3 (17.49 %), MgO (2.60 %), FeO ratio is (9.56 %), K_2O (3.92 %), Na_2O (1.18 %), Ti_2O (1.10 %), SO_3 (0.19 %), and Cl^- (0.42 %) were detected (Figure 16).

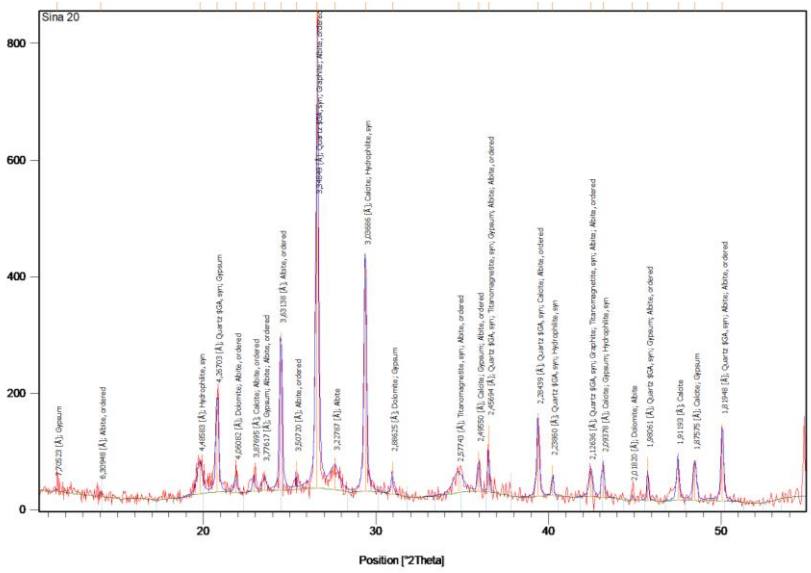

Figure 14. 2nd sample XRD result.

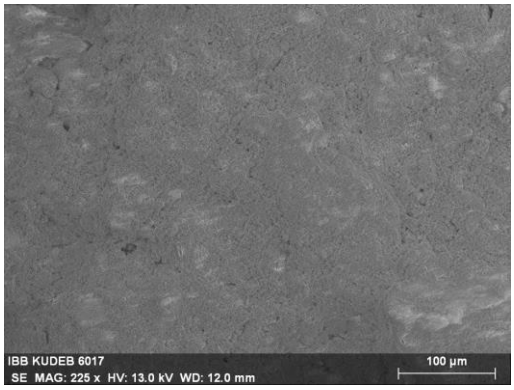

Figure 15. SEM images of micro porous and crack.

Table 6. EDX Results of 2nd sample.

Element	Norm. C (Wt. %)	Atom. C (At. %)	Compound norm.	Comp. C (Wt. %)
Oxygen	44.09	61.02		0.00
Sodium	0.88	0.84	Na_2O	1.18
Magnesium	1.57	1.43	MgO	2.60
Aluminium	9.26	7.60	Al_2O_3	17.49
Silicon	24.65	19.44	SiO_2	52.74
Sulphur	0.08	0.05	SO_3	0.19
Potassium	3.26	1.85	K_2O	3.92
Calcium	7.70	4.26	CaO	10.78
Iron	7.43	2.95	FeO	9.56
Phosphorus	0.00	0.00	P_2O_5	0.00
Chlorine	0.42	0.26		0.42
Titanium	0.66	0.30	TiO_2	1.10
Total:	100.00	100.00		

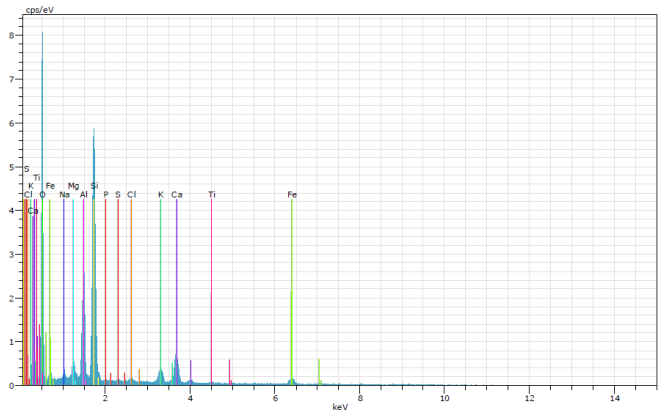

Figure 16. 2nd sample EDX spectrum.

The second sample consist of around 10-15 % ash and charcoal (graphite) as an additive, 25 % of carbonated aggregates (calcite and dolomite). A total of 65 % of the aggregates are siliceous materials, 10-15 % cream-colored undispersed masses (fired bricks/burnt aggregates), 25 % the sand size and the rest are silt/clay size aggregates. The aggregates are angular shaped and terrestrial in origin at 1 mm under sieve size (Figure 17). The binder is around 20 % lime, and the sample also contains gypsum and graphite. The presence of graphite in the XRD analysis indicates that the proportion of ash or charcoal pieces at least are more than 5 %.

In the third sample (Figure 18), VII layer (6600-6500 BC), according to hydrometer analysis (Figure 19) and Atterberg limit's (Figure 20), 33.90 % of aggregates are clay size, 44.13 % silt size, 21.97 % sand size, gravel size aggregates rate is 0.00 %, soil type is clayey-loamy (CL), the liquid limit rate is 45.70 %, the plastic limit is 19.30 %, and the plasticity index is 26.40 % (Table 2). According to the results of the loss in ignition analysis, loss at 105 ^{0}C (moisture) is 3.88 %, loss at 550 ^{0}C (organic materials) is 2.95 %, loss at 1050 ^{0}C (calcium carbonate) is 21.45 %. According to deal with acid, 21.70 % of the sample reacted and 78.30 % retained (Table 3).

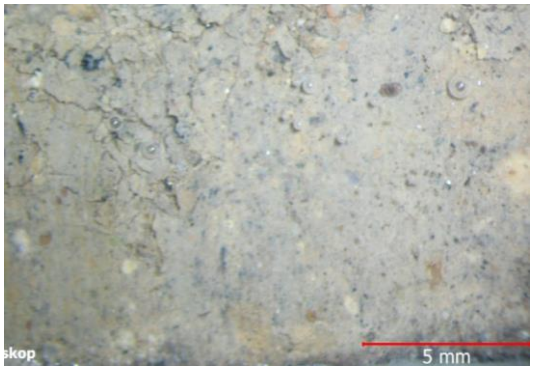

Figure 17. Thick section image of 2nd sample.

Figure 18. 3rd sample.

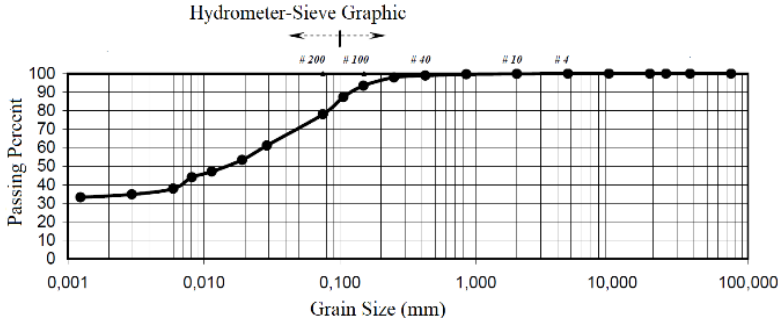

Figure 19. 3rd sample Hydrometer and Sieve graphic.

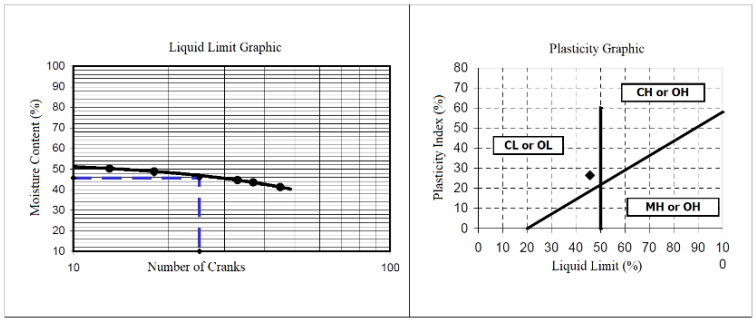

Figure 20. Liquid limit and plasticity graphics of the 3rd sample.

Petrographic investigations shows that the aggregates smaller than 125 microns are black slag powder and the remainder is clay/silt size material. Aggregates between 125 and 500 microns are mica, a small amount of black slag powder and the rest are cream-colored undispersed masses. About 20 % of the aggregates larger than 500 microns are quartz and the rest are cream-colored undispersed masses. The sample contains biotite, quartz mineral and 2-3 % black slag particles. As result of XRD analysis, muscovite, birnessite, quartz, albite and calcite were detected (Figure 21). According to SEM-EDX (Figure 22) analysis (Table 7), SiO_2 (52.46 %), CaO (10.18 %), Al_2O_3 (15.94 %), MgO (2.49 %) and FeO ratio is (11.21 %). K_2O (4.18 %), Na_2O (1.55 %), Ti_2O (0.56 %), SO_3 (0.00 %), and Cl^- (1.42 %) were detected (Figure 23).

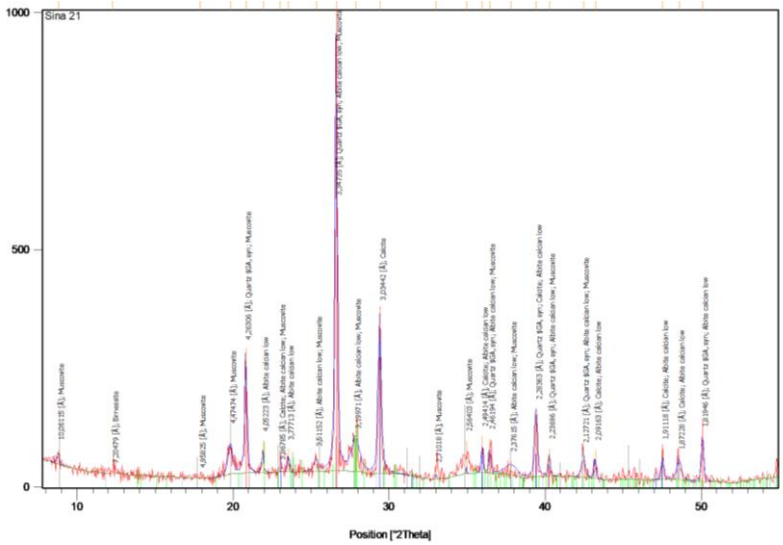

Figure 21. 3rd sample XRD result

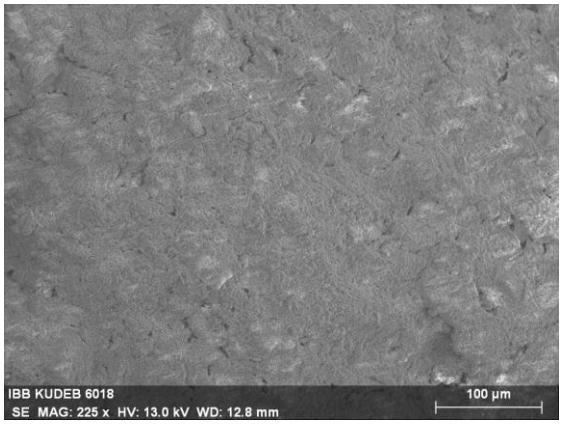

Figure 22. SEM images of micro cracks.

Table 7. EDX Results of 3rd sample.

Element	Norm. C (Wt. %)	Atom. C (At. %)	Compound norm.	Comp. C (Wt. %)
Oxygen	43.17	60.37		0.00
Sodium	1.15	1.12	Na_2O	1.55
Magnesium	1.50	1.38	MgO	2.49
Aluminium	8.44	7.00	Al_2O_3	15.94
Silicon	24.52	19.53	SiO_2	52.64
Sulphur	0.00	0.00	SO_3	0.00
Potassium	3.47	1.99	K_2O	4.18
Calcium	7.28	4.06	CaO	10.18
Iron	8.71	3.49	FeO	11.21
Phosphorus	0.00	0.00	P_2O_5	0.00
Chlorine	1.42	0.90		1.42
Titanium	0.34	0.16	TiO_2	1.56
Total:	100.00	100.00		

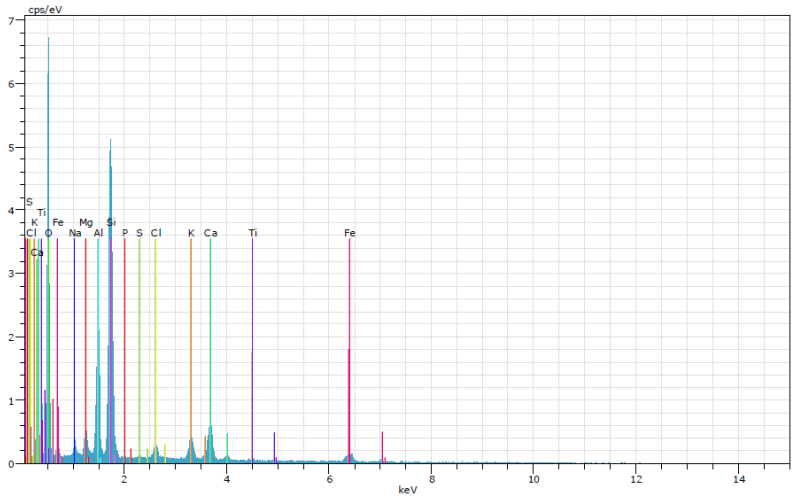

Figure 23. 3rd sample EDX spectrum.

The third sample consisting around 5% ash, 22 % carbonated aggregates. A total of 65-70 % is siliceous materials, 22 % sand size aggregates, biotite, quartz and 25-30 % cream-colored undispersed masses (fired bricks/burnt aggregates) as a filler and 2-3 % black slag fragments detected. The sample contains 30 % muscovite (expanded-clay type) as a binder, and the proportion of ash is less than the first and second samples. The aggregates are angular shaped and of terrestrial origin and aggregates are smaller than 1 mm (Figure 24).

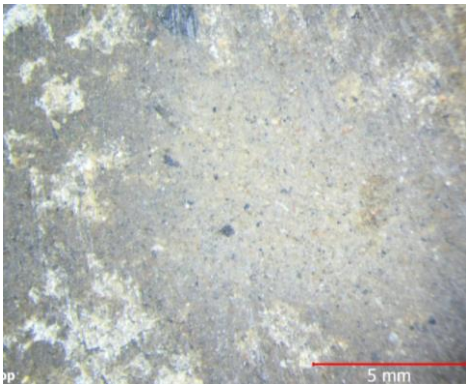

Figure 24. Thick section image of 3rd sample.

In the fourth sample (Figure 25), VII layer (6600-6500 BC); according to hydrometer analysis (Figure 26) and Atterberg limit's (Figure 27), 46.89 % of aggregates are clay size, 33.19 % silt size, 19.19 % sand size, gravel size aggregates rate is 0.73 % and soil type is high plasticity clay (CH), the liquid limit rate is 50.40 %, the plastic limit is 21.90 %, and the plasticity index is 28.50 % (Table 2). According to the results of the loss in ignition analysis, loss at 105 ^{0}C (moisture) is 3.84 %, loss at 550^0 C (organic materials) is 4.20 %, loss at 1050 ^{0}C (calcium carbonate) is 18.95 %. According to deal with acid, 19.88 % of the sample reacted and 80.12 % retained (Table 3).

Figure 25. 4th sample.

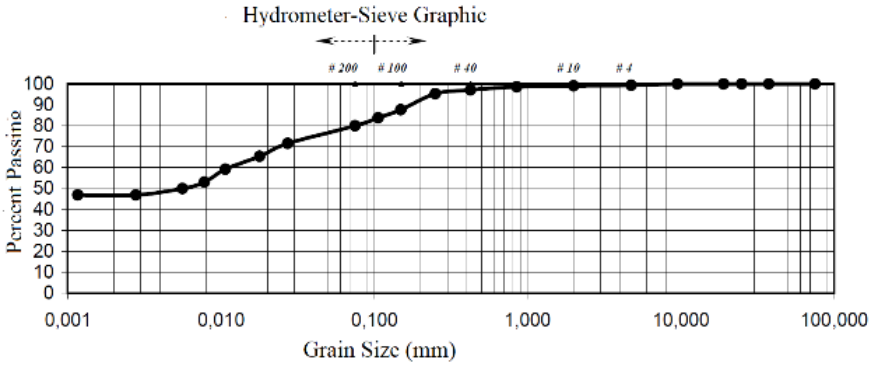

Figure 26. 4th sample Hydrometer and Sieve graphic.

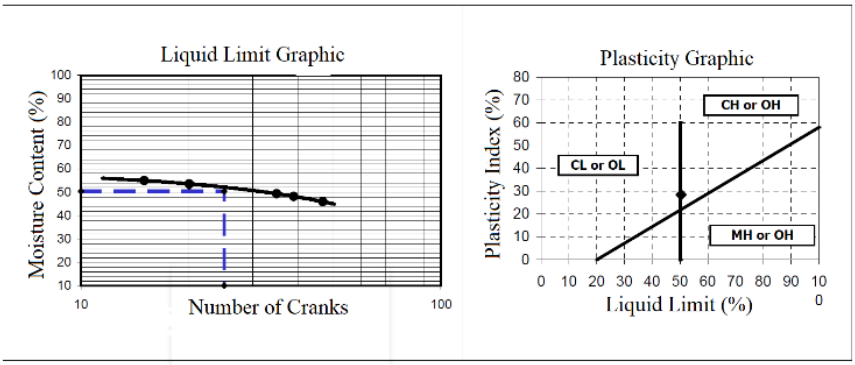

Figure 27. Liquid limit and plasticity graphics of the 4th sample.

Petrographic investigations shows that 3-5 % of aggregates smaller than 125 microns are black slag powder and the rest is clay/silt size material. Aggregates between 125 and 500 microns are mica, 2-3 % are black slag powder and the rest are cream-colored undispersed masses. About 20 % of the aggregates larger than 500 microns are quartz and the rest are cream-colored undispersed masses. The sample contain biotite, quartz mineral and 3-5 % black slag particles. As a result of XRD analysis, gypsum, albite, hydrophilite, quartz and calcite were detected (Figure 28). According to SEM-EDX (Figure 29) analysis (Table 5), SiO_2 (51.82 %), CaO (10.20 %), Al_2O_3 (15.65 %), MgO (3.04 %) and FeO ratio is (9.55 %). K_2O (4.60 %), Na_2O (1.69 %), Ti_2O (0.33 %), SO_3 (1.83 %), and Cl^- (1.30 %) were detected (Figure 30).

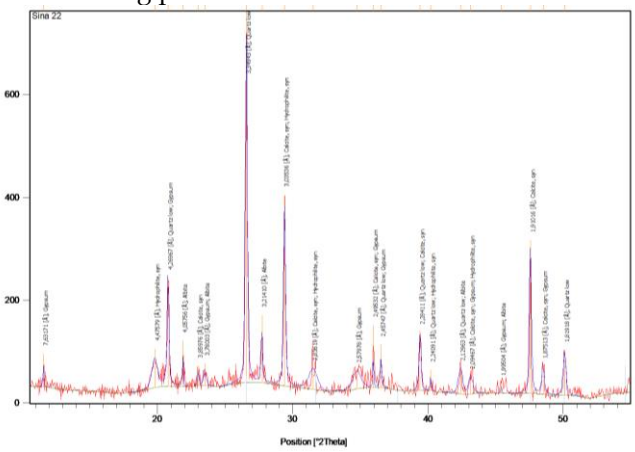

Figure 28. 4th sample XRD result.

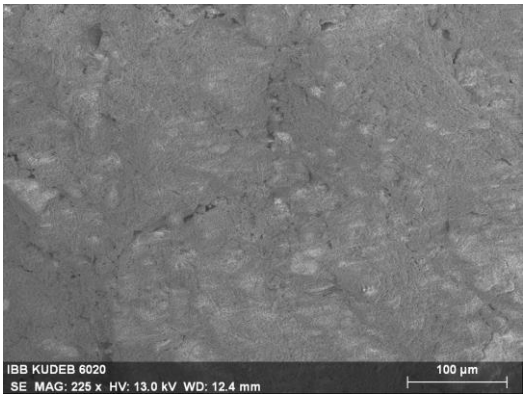

Figure 29. SEM images of micro cracks.

Table 8. EDX Results of 4th sample.

Element	Norm. C (Wt. %)	Atom. C (At. %)	Compound norm.	Comp. C (Wt. %)
Oxygen	43.65	60.58		0.00
Sodium	1.25	1.21	Na_2O	1.69
Magnesium	1.84	1.68	MgO	3.04
Aluminium	8.28	6.82	Al_2O_3	15.65
Silicon	24.22	19.15	SiO_2	51.82
Sulphur	0.73	0.51	SO_3	1.83
Potassium	3.82	2.17	K_2O	4.60
Calcium	7.29	4.04	CaO	10.20
Iron	7.42	2.95	FeO	9.55
Phosphorus	0.00	0.00	P_2O_5	0.00
Chlorine	1.30	0.81		1.30
Titanium	0.20	0.09	TiO_2	0.33
Total:	100.00	100.00		

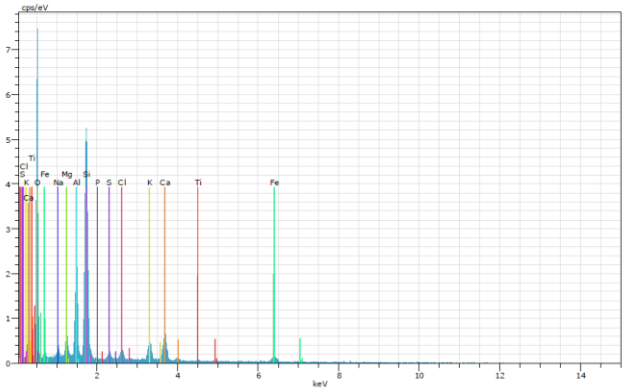

Figure 30. 4th sample EDX spectrum.

The fourth sample consists of around 10-15 % ash, 20 % carbonated (lime) material. A total of 80 % is siliceous materials, 20 % sand size, 1-2 % gravel size aggregates and the rest are silt/clay size material. The aggregates are angular shaped and of terrestrial origin in different shapes and the aggregates are smaller than 1 mm (Figure 31). In this sample the binder is gypsum and lime.

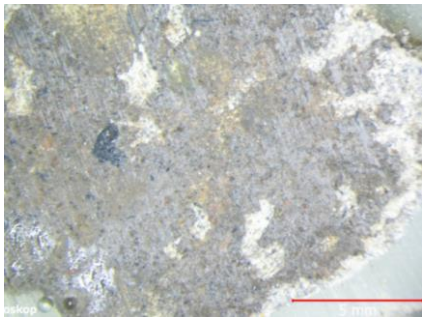

Figure 31. Thick section image of 4th sample.

plastic limit is 24.60 %, and the plasticity index is 27.20 % (Table 2). According to the results of the loss in ignition analysis, loss at 105^0 C (moisture) is 3.81 %, loss at 550^0 C (organic materials) is 4.78 %, loss at 1050^0 C (calcium carbonate) is 22.52 %. According to deal with acid, 26.10 % of the sample reacted and 73.90 % retained (Table 3).

Figure 32. 5th sample.

In the fifth sample (Figure 32), VIII layer (6700-6600 BC), according to hydrometer analysis (Figure 33) and Atterberg limits (Figure 34), 47.95 % of aggregates are clay size, 35.71 % silt size, 16.34 % sand size, gravel size aggregates rate is 0.00 %, soil type is high plasticity clay (CH), the liquid limit rate is 51.80 %, the

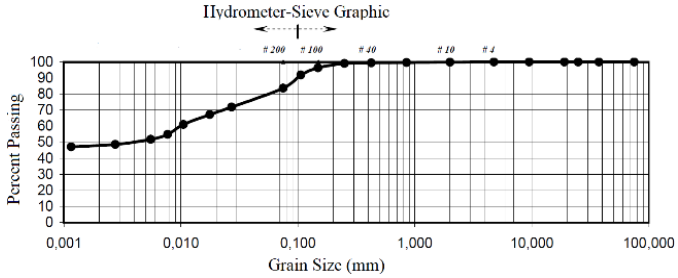

Figure 33. Liquid limit and plasticity graphics of the 5th sample.

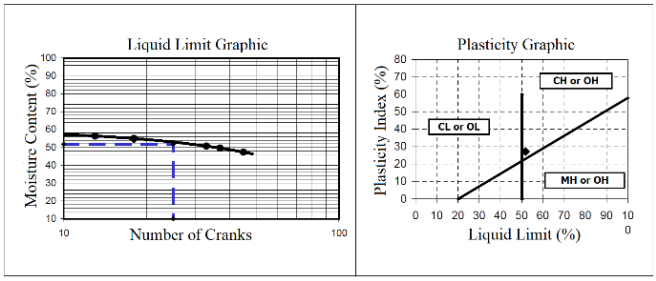

Figure 34. 5th sample Hydrometer and Sieve graphic.

Petrographic investigations show that, 2-3 % of aggregates smaller than 125 microns are black slag powder and the remainder is clay/silt size material. Aggregates between 125 and 500 microns are mica, a small amount of black slag powder and the rest are cream-colored undispersed masses. About 20 % of the aggregates larger than 500 microns are quartz, the rest are cream-colored undispersed masses. Sample contain biotite, quartz mineral and around 5 % black slag particles. As a result of XRD analysis, kaolinite, muscovite, quartz, low cristobalite, albite, dolomite and calcite were detected (Figure 35). According to SEM-EDX (Figure 36) analysis (Table 9), SiO_2 (52.48 %), CaO (10.22 %), Al_2O_3 (17.62 %), MgO (3.10 %) and FeO ratio is (9.06 %). K_2O (3.53 %), Na_2O (1.34 %), Ti_2O (1.77 %), SO_3 (0.48 %), and Cl^- (0.40 %) were detected (Figure 37).

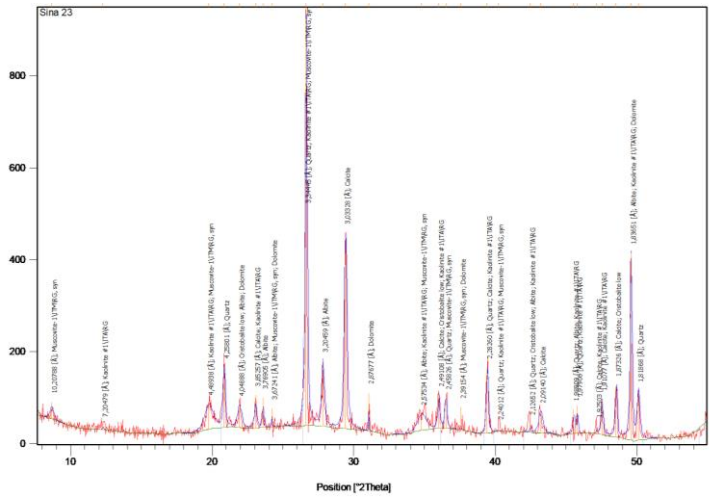

Figure 35. 5th sample XRD result.

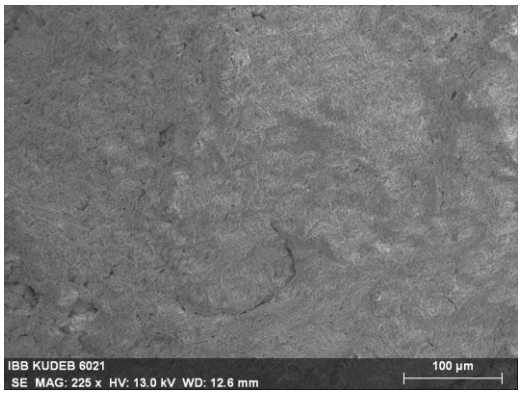

Figure 36. SEM images of micro cracks.

Table 9. EDX Results of 5th sample.

Element	Norm. C (Wt. %)	Atom. C (At. %)	Compound norm.	Comp. C (Wt. %)
Oxygen	44.35	61.14		0.00
Sodium	1.00	0.96	Na_2O	1.34
Magnesium	1.87	1.70	MgO	3.10
Aluminium	9.32	7.62	Al_2O_3	17.62
Silicon	24.53	19.27	SiO_2	52.48
Sulphur	0.19	0.13	SO_3	0.48
Potassium	2.93	1.65	K_2O	3.53
Calcium	7.30	4.02	CaO	10.22
Iron	7.04	2.78	FeO	9.06
Phosphorus	0.00	0.00	P_2O_5	0.00
Chlorine	0.40	0.25		0.40
Titanium	1.06	0.49	TiO_2	1.77
Total:	100.00	100.00		

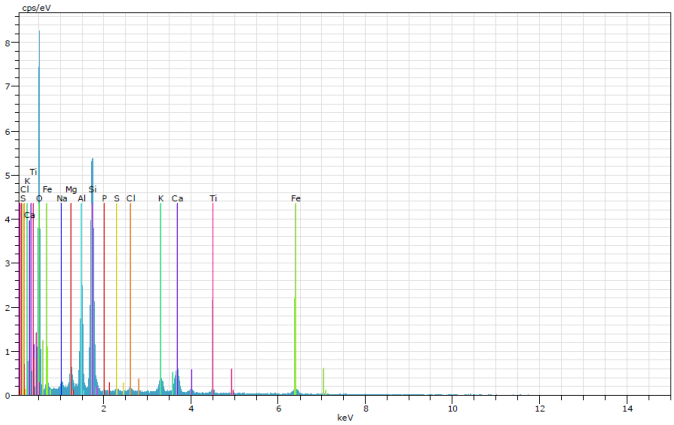

Figure 37. 5th sample EDX spectrum.

The fifth sample consists of 5-10 % ash, and 20-25 % carbonate particles (dolomite and calcium carbonate). A total of 70 % are siliceous materials, 17 % are sand size aggregates, and %10-15 cream- colored undispersed masses (fired bricks). The aggregates are angular shaped and of terrestrial origin and smaller than 1 mm (Figure 38). According to XRD, the sample includes around 40-45 % of two clay types (kaolinite and muscovite) as binder. It's so obvious that the mixture was prepared from different local clay resources (see Xanthopoulou et al., 2020).

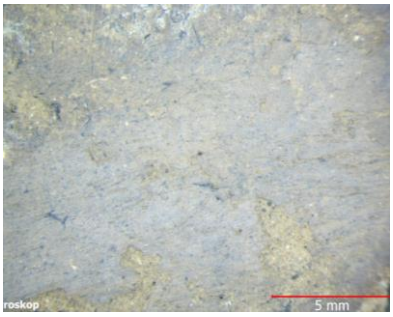

Figure 38. Thick section image of 5th sample.

In the sixth sample (Figure 39), IX layer (6800-6700 BC), according to hydrometer analysis (Figure 40) and Atterberg limit's (Figure 41), 51.52 % of aggregates are clay size, 34.47 % silt size, 13.74 % sand size, gravel size aggregates rate is 0.00 %, soil type is high plasticity clay (CH), the liquid limit rate is 59.10 %, the plastic limit is 22.40 %, and the plasticity index is 26.70 % (Table 2). According to the results of the loss in ignition analysis, loss at 105 ^{0}C (moisture) is 4.28 %, loss at 550^0 C (organic materials) is 4.44 %, loss at 1050^0 C (calcium carbonate) is 21.09 %. According to deal with acid, 20.71 % of the sample reacted and 79.29 % retained (Table 3).

Figure 39. 6th sample.

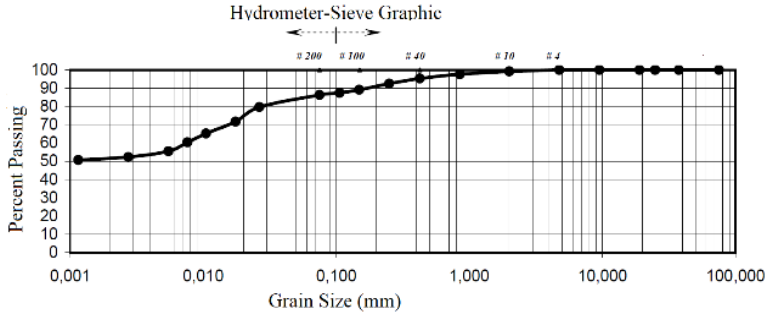

Figure 40. Liquid limit and plasticity graphics of the 6th sample.

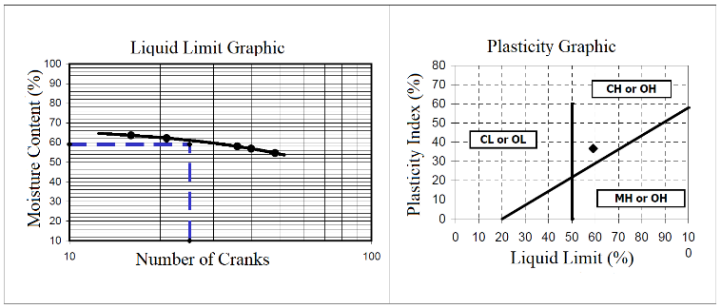

Figure 41. 6th sample Hydrometer and Sieve graphic.

Petrographic investigations show that, 2-3 % of aggregates smaller than 125 microns are clay/silt size materials. Aggregates between 125 and 500 microns are mica, and the rest is cream-colored undispersed masses. About 20 % of the aggregates larger than 500 microns are quartz, the rest are cream-colored undispersed masses. The sample contains biotite, quartz mineral and 3-5 % black slag particles. This sample contain organic additives, that it could be stems of sedge plants or animal hair. There is no trace of ash in this sample. As a result of XRD analysis, muscovite, quartz, albite and calcite were detected (Figure 42). According to the SEM-EDX (Figure 43) analysis (Table 10), SiO_2 (51.69 %), CaO (10.45 %), Al_2O_3 (17.70 %), MgO (3.33 %), FeO (9.24 %), K_2O (4.16 %), Na_2O (1.14 %), Ti_2O (0.79 %), SO_3 (0.71 %), Cl^- (0.66 %) and P_2O_5 (0.13 %) were detected (Figure 44).

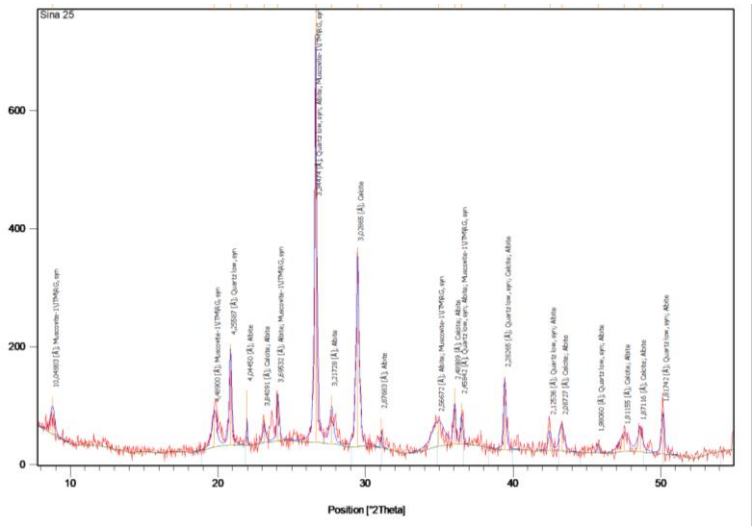

Figure 42. 6th sample XRD result.

Figure 43. SEM images of micro cracks.

Table 10. EDX Results of 6th sample.

Element	Norm. C (Wt. %)	Atom. C (At. %)	Compound norm.	Comp. C (Wt. %)
Oxygen	44.04	60.87		0.00
Sodium	0.84	0.81	Na_2O	1.14
Magnesium	2.01	1.83	MgO	3.33
Aluminium	9.37	7.68	Al_2O_3	17.70
Silicon	24.16	19.03	SiO_2	51.69
Sulphur	0.28	0.20	SO_3	0.71
Potassium	3.45	1.95	K_2O	4.16
Calcium	7.47	4.12	CaO	10.45
Iron	7.18	2.84	FeO	9.24
Phosphorus	0.06	0.04	P_2O_5	0.13
Chlorine	0.66	0.41		0.66
Titanium	0.47	0.22	TiO_2	0.79
Total:	100.00	100.00		

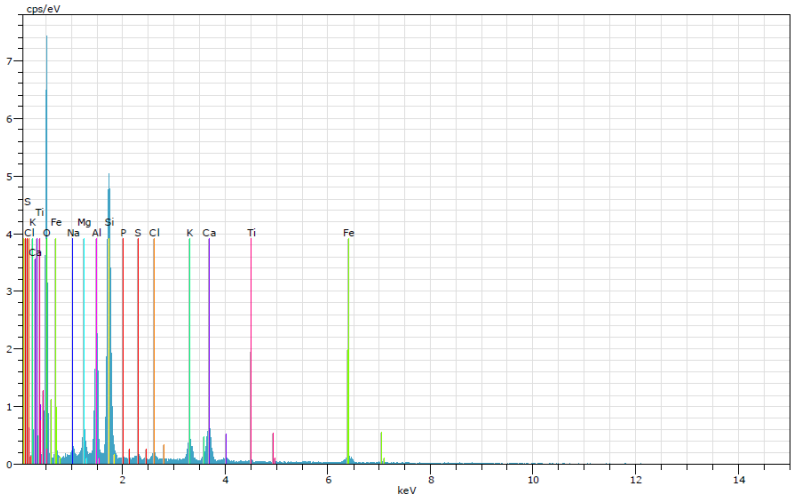

Figure 44. 6th sample EDX spectrum.

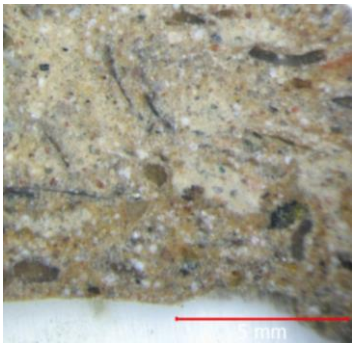

Figure 45. Thick section image of 6th sample.

The sixth sample consists of 20 % carbonate materials. A total of 70-75 % is siliceous materials, 10-15 % sand size aggregates, 10 % cream colored undispersed masses (fired bricks/burnt aggregates) and the rest are silt/clay size aggregates. The aggregates are angular shaped and of terrestrial origin under 1mm sieve size (Figure 45). The binder of sample is clay (muscovite, expanded-clay). The texture and mixture of this sample is totally different from the other samples and contain fibrous organic matters. This is the only sample that does not contain ash.

3.2. *Evaluation of results*

All samples contain 3 major minerals: Calcite, Quartz, and albite. Calcite ($CaCO_3$) is a major mineral resulting from adding dolomite fragments during the brick making processes, or from using lime as binding material. On the other hand, the presence of Quartz (SiO_2) and albite (Aluminosilicates of Na) are ascribed to the use of sand particles as an additive material in mudbrick manufacturing. Furthermore, protoenstatite, dolomite, minerals are frequently found in the soil of the region (Biricik, 1992). The second and fourth samples contain hydrophillit, which is a mineral usually found with gypsum, and these two samples contain gypsum.

The amounts of FeO indicate that the soil of the region contains ferrous minerals, and all samples contain it approximately at about the same rates (~10%). The simple fact of the presence of iron oxides is that they are very strong coloring agents. The reason why the colors are brown/dark brown in the samples is the color of the raw material and the high amount of iron oxide and charcoal. As the 6th sample does not contain charcoal, according to the Munsell Color Chart, the color of sample is light gray.

Some types of clay minerals have a special property that allows them to incorporate water molecules into their structures (these clay types, act as binder in earthen materials). These minerals are called expanding or swelling clays. Expanding clays have a 2:1 structure, with two tetrahedral and an octahedral layer. These swelling clays are called smectites (Velde, 2008). Other clays are called, by symmetry, non-expanding or non-swelling clays and have a 1:1 structure like Kaolinite (warren, 1999).

Two types of clays, muscovite and kaolinite were found in the 3rd, 5th, and 6th samples. According to the clay minerals classification, muscovite belongs to the smectites group as expanding clays and kaolinite is a non-expanding clay type. The third and sixth samples contain muscovite. The fifth sample contains both clay types and it indicates that raw materials of this sample were taken from different clay deposits.

The 1st and 2nd samples are from the same (VIA) layer and the soil type of both is same (clay-loamy). According to the loss on ignition, the 1st sample contains the highest amount of organic materials (5.79%) and calcium carbonate (28.34%). According to hydrometry and sieve analysis, they are similar in terms of particle size distribution, but the source of raw materials and the mixture of additives and fillers are different. The amount of organic matter in 2nd sample (2.48%) is almost half of the 1st sample (5.79%). The structure of 1st sample is harder than the 2nd and contains coarse siliceous aggregates. As both samples were belonged to layer VIA, in 1st sample (building 80), only slacked lime was used as binder, but in 2nd sample (building 76) it was understood that gypsum, which is known to increase strength and functionality, was also used by adding it to the mud bricks. This indicates that there is a development and an action towards better quality mud brick production at the same time.

The 3rd and 4th samples are from the same layer (layer VII/6600-6500 BC). The binder of the 3rd sample is clay (muscovite) and the binder of 4th sample is gypsum. The amount of organic matter in the 4th sample (4.20%) is higher than in the 3rd sample (2.95%). The structure of 4th sample is heterogeneous and harder than the 3rd sample. According to atterberg limits, the soil type of 3rd sample is clay-loamy, while the 4th sample is high plasticity-clay with a finer grain size and a higher liquid limit rate. The composition and soil type of samples are different from each other. Finally, it should be mentioned that the source of raw materials and mixture of these two samples are different from each other. In, 3rd sample (building 7) muscovite (expanding clay) was used as a binder and as its resistance to water was low and has a weak structure, in 4th sample (building 11) instead of muscovite, we see that the type of binder was changed and the gypsum, which is much more resistant to water than muscovite, was used. This revision of binder explains the hardness of the 4th sample. Also, it is so clear that the people of Çatalhöyük at that period (layer VII/6600-6500 BC) was developing his knowledge and experiences about the construction and building materials.

The 5th sample belongs to VIII layer. It contains two types of clay, muscovite, and kaolinite as the binder. The mixture of binder in this sample is totally different from other samples. The plastic limit rate (24.60%) is higher than all other samples. Since muscovite has low resistance to water and has a weak binding feature, it is obvious that Çatalhöyük residents (according to their experiences) by adding kaolinite they were wanted to produce stronger mud bricks. This mixture of 2 different clay type explains why the plastic limit of 5th sample is higher than all other samples.

The 6th sample belongs to the IX layer. It has the finest grain size distribution compared to the other samples. The 6th sample's binder is clay (muscovite) and has the highest rate of clay size and liquid limit rate (59.10%) and only this sample contains phosphorus. The texture and mixture of this sample is totally different from other samples and contain fibrous organic matter at the same time only 6th sample does not contain charcoal/ash. The structure of this sample shows us that organic materials such as clay and straw were used in the early periods. It means in Early Neolithic at Çatalhöyük only the clay and organic materials mixed for manufacture of mud bricks. We do not have any evidence yet that gypsum and slacked lime were used as binders between the 6800-6600 BC.

All the samples also contain *terracotta* (backed bricks) pieces that are defined as *secondary use* materials. The new buildings were built directly on the rubble of demolished old buildings. These materials belong to earlier phases of occupation that were reused during the renewal and reconstruction of new houses. The secondary materials not only save labor and time but also shows that consumed resources and raw materials are used correctly and consciously. On the other hand, such *secondary uses* probably combine the spirit of the old building with the energy of the new house, according to beliefs worlds. In other words, the continuation of a new life cycle on the old one reflects the endless cycle of life. It can also be said that the continuity achieved through belief systems helps to ensure the dialogue between generations and generations in a community, to transfer experience and knowledge, and to strengthen social ties. Another low possibility is that the reuse of the material used in old houses reflects a kind of recycling. In fact, such practices express the continuity in the society, the transition to settled life and the ability to hold on. At the same time, the level of knowledge about building materials is advanced and it is a sign of a strong system.

Based on data from the KOPAL Project, it could be said that the source of raw material of the 1st sample (VIA layer) is a basal layer with the addition of an organic clay. The 1st sample contains the highest amount of carbonated material and organic material. According to Munsell Color Chart, the color of this sample is Dark Greyish Brown.

In the 2nd and 4th samples, raw materials were taken from the upper alluvial layer according to color and silt/clay contamination. Additionally, these two samples contain gypsum and as far as grain size is concerned, the coarser fraction belongs to these samples.

The 3rd, 5th and 6th samples contain smectite and lack a coarse fraction. The raw materials of these samples are upper alluvial level. But the filler and additives of each one is different from the others.

The sixth sample is totally different in terms of additives. fillers and texture. It contains vegetal remains as organic binders and do not contain ash or charcoal.

4. CONCLUSION

As a rule, conservation on archaeological structures requires the use of repair mortars compatible with the original materials. Consequently, the complete characterization of the chemical, physical and mechanical characteristics of originally employed mortars is imperative to the success of the conservation process.

Since early 1990's there are many studies in Çatalhöyük and other prehistoric settlements in Anatolia and Mesopotamia about mud bricks. But none of these studies have specified the type and amount of different types of binders, fillers, additives by using qualitative and quantitative analysis methods. The biggest difference of this research with other mud brick studies is, beside the detailed characterization of the content of mud bricks, this study shed light to the determination of their physical/chemical properties, type and properties of soil and behaviour of mud bricks in contact with water (liquid/plastic limits and plasticity index of soil) and along with the ratios of the different type of materials used as binder in mudbricks.

In line with the results obtained, now we can produce the suitable mud bricks during conservation and restoration activities. In addition, the exact determination of contents of the materials used in mud brick manufacture helped us to fully understand the development process of adobe making technology and the diversity of materials, as well as the technological developments during the Neolithic Period in Çatalhöyük settlement.

According to analyses results, in layer IX (6800-6700 BC), only one type of clay (muscovite) and organic fiber (straw) were used for manufacture of mud bricks. But in VIII layer (6700-6600 BC) they were mixed two type of clay (kaolinit+muscovite). It is well known that kaolinite is one of the best clay types for manufacturing mud brink and it is more resistance to water. In VII Layer (6600-6500 BC) gypsum were

added to mud bricks for the first time. In VIA Layer (6500-6400 BC) first mix of gypsum and slacked lime and then only the slacked lime was used as binder.

The results showed us in the early layers (layer IX-6800) only clay and straw were used for manufacture of mud bricks. however, as time progressed, it can be said that after reaching the technology of making gypsum and slacked lime these two materials were added and were used both alone and mixed with each other during the production and manufacture of mud bricks in the late layers (layer VIIA-6400). It is so clear there is a development process and technological changes from 6800 BC to 6400 BC in the contents for production of better and durable mud bricks from early periods to the later periods.

This indicates to us that there is a search and an action towards for produce better quality mud bricks. In this context, this is an indication of how the technological development in the Neolithic period was the result of the use of correct knowledge, skills and experiences. At the same time, it shows us that there was a search for the materials that are more resistant to environmental deterioration factors. The use of lime and especially gypsum besides clays during the production of the mud bricks as binder indicate that the people of Çatalhöyük during the Neolithic period had an extensive knowledge of the raw materials and properties around them.

In the prehistoric period of Anatolia, the use of slaked lime, which was achieved by burning the limestone, was known only in plasters (Kingery et al, 1988). The most obvious use of the slacked lime is in the floor coverings called terrazzo (Hauptman et al), but the use of lime in mud bricks is not often mentioned. However, the use of gypsum in mud bricks in the prehistoric period has not been reported in detail in any research so far. It has been reported that lime or gypsum was used only in plasters in previous studies (Hauptman et al).

This research aims to provide a milestone in terms of a modern scientific approach to the detailed characterization of the content of prehistoric mud bricks from Çatalhöyük/Turkey as one of the principal sites in the world for prehistoric archaeology.

Finally, with this methodology of characterisation, in addition to determining the detailed content of the mud bricks and produce the mixtures suitable to the original mud bricks for conservation activities, also by a systematic sampling method we will be able to make a great contribution to the understanding the changes and technological developments.

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