Preliminary Study of the Characteristics of Injana Formation Clay Stone for Brick and Ceramic Industry

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ABSTRACT— ***The big problem that face the brick and ceramic industry in Northern Iraq is the lack of raw materials (washed clay) at the river banks. Many studies concluded to the high percent of carbonate in the clay stone of the geological formations in the area. The main attempt of the study was to find some treatment methods for clay stone belongs to beds among rock exposures in Northern Iraq to be used for brick and ceramic industry. This is depending primarily on the percent of $CaCO_3$ in the rocks. It is found that some clay stone beds within Injana Formation show Low Percent of $CaCO_3$ Clay stone (L.P.C.C.) compared with clay stone beds from Gercus and Fat'ha formations. Therefore, the work trend is shifted towards the study of their engineering properties and the effects of additives (crushed glass and sand) to its characteristics. Consequently, the study revealed that all the additives in general worsen some characteristics and improves others; otherwise, the natural sample without additives gives the best results. The natural samples have average values of 19 M Pa, which referring that the uniaxial compressive strength above the lowest value 16 M Pa according to Iraqi standards. The burned sample also shows 6.6% volume shrinkage without cracking or bloating, while the 21.6% low value of water absorption that falls within the Iraqi standards. The fluorescence is low; bulk density is 1.63 gm./cm^3 and the tensile strength is 1.2 M Pa.***

Keywords— Properties of clays, Injana Formation, North Iraq, Brick, Ceramic

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1. INTRODUCTION

Last decade, many studies were performed in Northern Iraq to explore clay stone beds suitable for industrial purposes, particularly brick and ceramic [1], [2], [3]. The study area is located approximately between E 42° 30′ 00″, E 43° 30′ 00″ and N 36° 10′ 00″, N 36° 45′ 00″. Calcium carbonate and gypsum were the main pollutant materials in brick industry, though; many efforts were done to reduce their percentages within the clay stone and/or mudstone beds from Gercus, Fat'ha and Injana formations [2], [3]. The first goal of this paper is to find ways for treating clay stone to improve their engineering properties. The alluvium sand [4] and grind glass were added in percentages reach to 30% to the clay stone to be used for manufacturing brick ceramic respectively. Adding of 30% of additives is directly reducing the percent of carbonate to the third. In natural clay stone, the carbonate is about 14%, while after adding 30% of sand or grind glass, the carbonate will be 10.5% that may change the plasticity [4], [5]. The lack of neither organic materials in clay stone nor the soil clay is one of the advantages of this material for brick and ceramic industry [4]. The most brick makers often used a mix of about 30% of sand and 70% of plastic clay [5]. The best additives that it cheap and enhance the brick properties like agriculture waste, mineral waste, rock sawing, ash and sand [5], [6], [7], [8], [9], [10], [11] and [12].

A reconnaissance survey including channel sampling has been carried out for clay stone outcrops of Gercus, Fat'ha and Injana formations in Nineveh Governorate (Figure 1). Gercus Formation is generally consisting in purple and red shale, mudstones, sandy and gritty marl. Fat'ha Formation is equivalent to sequence of carbonate, evaporite (gypsum and anhydrite), marl and clay stone. Injana Formation is a sequence of sandstone and clay stone [13]. An approximated percent of $CaCO_3$ in clay stone was checked in the field by reacting with 5% concentration of HCl. This method reaches to assign the relatively decline percentage of $CaCO_3$ within the upper layers of Injana Formation north of Mosul city. Consequently, the trend of the work was shifted towards the possibility of these Low Percent $CaCO_3$ Clay stone (L.P.C.C.) to usage for industrial purposes. Throughout, many geotechnical tests on (L.P.C.C.) were done to explain their utility as raw material for industry. The characteristics of these layers show good quarry condition, the average thickness is about 25 m. and their outcrops extend to several kilometers. The structural attitude is low dip angle not exceed to 9º. The wide apparent thickness due to the relatively low slopes and the accessibility is regarded as very suitable quarrying conditions. Consequently, this bed can be invested industrially.

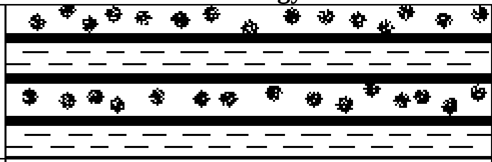
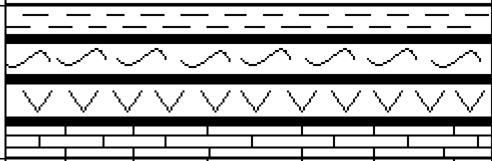
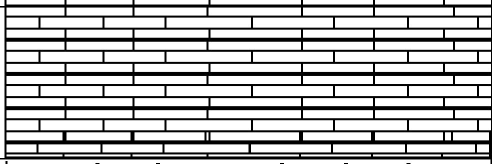
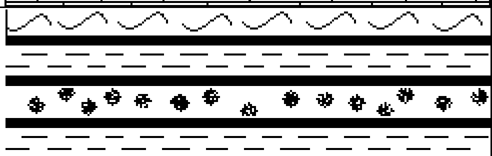
Formation	Age	Thickness m.	Lithology	Description
Injana	Upper Miocene	240-410		Sequence of sandstone and clay stone.
Fat'ha	Middle Miocene	305-610		Sequence of carbonate, evaporite, marl and clay stone.
Pila Spi	Middle Upper Eocene	85		Well bedded bituminous limestone.
Gercus	Middle Eocene	838		Generally purple and red shale, mudstones, sandy and gritty marl.

Figure 1: Descriptive succession of geological formations that exposed in study area.

2. PROCEDURE

2.1 Tests of clay sample

Channel sample of (L.P.C.C.) represented by 10 kilograms displays the total thickness of the bed. The sample was prepared in the Geotechnical Laboratory of Dams and Water Resources Research Center, Mosul University as the following procedure:

1. Measuring the percent of $CaCO_3$ by HCl of 5% concentration to dissolve carbonate materials, then filtration and drying the insoluble residue.
2. Sieve analysis to find the size distribution of coarse grains.
3. Hydrometer tests to find the size distribution of fine materials.
4. XRD analysis of natural sample, then ethylene glycol treatment was done; finally the sample was burned for two steps to 550°C and 1100°C. The equipment type that used for testing is XRD Spellman DF3 in the Geochemical Department of the Directorate General of Geological Survey and Mining, Baghdad, Iraq.
5. Major and trace elements analysis were performed including (SiO_2, TiO_2, Al_2O_3, Fe_2O_3, MgO, CaO, Na_2O, K_2O, Mn, Ni, Co) and L.O.I. The chemical tests were conducted in the Geochemical Department of the Directorate General of Geological Survey and Mining, Baghdad, Iraq.
6. Atterberge limits including plastic and liquid limit to get plasticity index were displayed [4]. The plastic limit device is consisting in glass plate 30X30X1 cm. with 130 mm. diameter porcelain dishes, spatula with blade 33X120 mm. and six aluminum moisture cans of 88cc. volume. The manual Casagrande liquid limit device is used to obtain liquid limit. Plastic limit was used as suitable water content to form the samples that will be used to get engineering properties [5].

The tests in steps 1,2,3 and 6 are checked for three times and the results of them represent the mean value, while in steps 4 and 5 the results are for represented sample.

2.2 Samples forming

Seven dry samples of clay stone were prepared each has 0.5 Kg. weight, which are passing through sieve 0.6 mm. from (L.P.C.C) sample. Flood plain sand sampled from Said Hammed area about 15 Km. south of Mosul Dam were added to the three samples (10S, 20S and 30S) in ratios (10, 20 and 30) % respectively. The grind glass also added to the three other samples (10G, 20G and 30G). The seventh sample (L.P.C.C) is still without any additives to see its prime behavior.

Water was added in about 25% ratio to all seven samples which are almost reach to plasticity limit. Seven small disc sets (A) were performed for each sample with a diameter of about 30.6 mm. and a height of 11.6 mm. for physical tests.

Other disc sets of samples (B) were prepared with a diameter of about 63.6 mm. and a height of 36.6 mm. for tensile strength test by Brazilian Method. Cylindrical sample sets (C) were shaped according to available mold that having dimensions 36 mm. heights and 30.6 mm. diameter for compressive strength (Figure 2). The quantity of clay stone for each type of samples were calculated according to the mold volume to reach the density ranged between 1.6-1.7 gm./cm^3. The material was put in the mold at whole and the gradual compaction pressure for 5 minutes give mastery over to reach 6 ton.

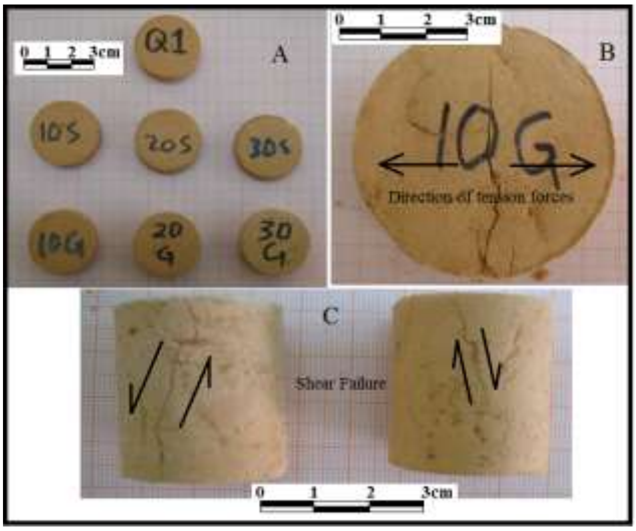


Figure 2: Three types of prepared samples for engineering tests, A: small disc has diameter 30.6 mm., B: large disc has diameter 63.6 mm. and D: cylinder has diameter 30.6 mm., Q1 is the natural clay material (L.P.C.C) without any additives, the numbers 10-30 means the percent of additives of sand S or glass G, note photos B and C after test.

A compression instrument was used by exposing the same weight samples to 6 M Pa. pressure. The prepared samples were left for about 48 hours in laboratory condition, dried in oven on 105 °C for 24 hours.

The samples were burned to 950°C for two hours of maturity in a raising rate of 200°C/hour. The samples were left in the oven for 24 hour to be cooled (figure 2). Samples burning were done by the Turkish Furnace type PLF120-7. The procedure for drying and burning approach many studies, but some stopped in 900°C after one hour of maturity [14].

2.3 Samples tests

Physical and mechanical tests are done as the following steps:

1. Measure the dimensions in accuracy 0.01 mm. and weight in accuracy 0.001 gm. three times for each sample in three conditions, wet, dry and burn.
2. Bulk densities of wet, dried and burned samples are calculated according to dimensions and weight.
3. Shrinkage, bloating and weight loss after drying and burning are calculated.
4. Immerse the small discs (set A) by water for 24 hours to measure the water absorption, and leave them in water for seven days to check the fluorescence.
5. Mechanical strength were done on sets B and C. Tensile strength by Brazilian Method was done on set B, while uniaxial compressive strength was done on set C. The samples were tested in the laboratories of Mosul Technical College – Department of Building and Construction by the MATEST compression machine. The Brazilian Method and the direct method is more confidence than the flexural that gave results in tensile strengths much higher than the real values [15].

3. DISCUSSION OF RESULTS

The low percentage of $CaCO_3$ within the concerned clay stone layers of uppermost Injana Formation in comparison with other adjacent clay stone beds of other exposed geological formations were explained in (Table 1) by the percent of CaO. The insoluble residue of Injana Formation Clay stone is greater than other formations, which mean the low percent of soluble material that represent by calcium carbonate (Table 1). The insoluble residue is consist of clay minerals (Kaolinite, Chlorite, Montmorillonite and Illite) and other minerals (Quartz, Plagioclase and Muscovite), which are explicate by X.R.D analysis (Figure 3) [4]. The presence of suitable flux oxides percent (Table 2) like SiO_2, Al_2O_3 and Fe_2O_3 encourage to study these layers in details. The majority of phyllosilicate minerals and flaky mica that shows in figure 3 refers to clay stone richness by clay and silt grains [4]. Mica minerals are more easily disintegrate than feldspar and quartz, so in this case the grain size of mica may be reach to silt size, while feldspar and quartz remain in the size of sand. The bulk sample of clay stone reflects the quartz and feldspar (Figure 3A), while the treated sample shows the reflection of clay minerals and muscovite (Figure 3B). When (Fe – Al) oxides is exceed to (15.5) % and the CaO is still low, this can be regarded as good indication for brick industry comparable with other clay stone beds in nearby areas [3]. In addition, the loss of silica ratio and loss on ignition components increased the relative porosity and the presence of free potassium oxide aids in lowering burning temperature [1].

Table 1: Calcium oxide percent and insoluble residue for clay stone of geological formations that obtained in study area.

Geological formations	CaO %	Insoluble residue %
Gercus	18.87	78.14
Fat'ha	19.6	79.58
Injana	13.98	84.11

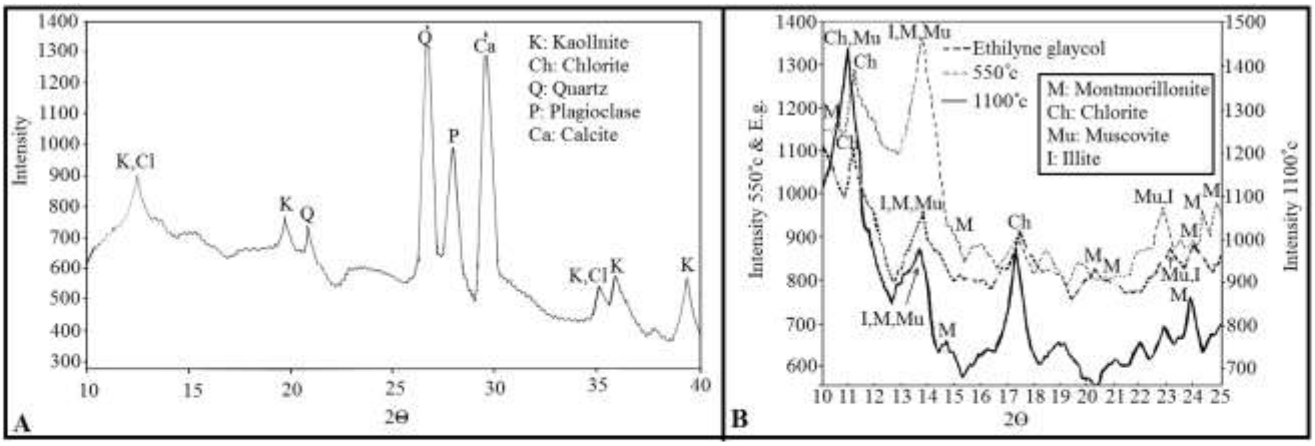


Figure 3: XRD chart of A: bulk sample (L.P.C.C), B: treated sample by heating to 550°C, 1100°C and ethilyne glaycol.

Table 2: Chemical analysis for sample (L.P.C.C).

Co ppm	Ni ppm	Mn ppm	L.O.I (wt%)	K_2O (wt%)	Na_2O (wt%)	CaO (wt%)	MgO (wt%)	Fe_2O_3 (wt%)	Al_2O_3 (wt%)	TiO_2 (wt%)	SiO_2 (wt%)
20	155	600	16.41	1.2	0.82	13.98	6.2	4.7	10.86	0.53	44.52

In engineering classifications both particle size and, plasticity properties are considered in soil or clay stone [4], [16]. Grain size gradation of the clay stone bed is graded from fine sand to silt and clay with weighted ratios of (15, 44, 41) % respectively (Figure 4). This gradation is suitable for brick and ceramic industry; this is due to its sand and silt grains which shear in making partial glass cementing material through burning, and the clay content helps for brick forming [17]. The present results can be compared with previous [8], [14], [17], and [18], which some got approach gradation of sand, silt and clay (12.8, 44, 43.2) % respectively [18]. The grain size gradation show mediatory position of present study in fine grain size less than 0.005 mm. (Figure 4). The three studies show the close of results in the size less than 0.075 mm, which approach the size of silt [4]. The mediatory position of clay and colloid size [4] is reflect in the suitability for brick and ceramic manufacturing on the base of plastic limit and plasticity index (Figure 5) [19]. The values of plastic limit in all three studies are closed; while the difference is appear in the plasticity index. The plasticity index depends on both plastic and liquid limits; the value of liquid limit is moderate so the position is optimal in comparison to other studies [17], [18]. The high value of liquid limit may led to increase shrinkage during drying, while the convergence in values of plastic limit and plasticity index compile the material to optimum extrusion (Figure 5).

The physical tests results (Table 3) revealed that the bulk density of the burning samples rang from (1.51 – 1.64) gm./cm^3, these values are almost close to the common densities of brick. The additives are not effect to the bulk density, meanwhile, addition of kraft pulp decrease the density from (1.82) gm./cm^3 to (1.5) gm./cm^3 [7]. The accepted water absorption in the Iraqi Specification for brick manufactured from clay [20] is reach to 28% for type C, while type A must not exceed to 22%, so the (L.P.C.C), 20G and 30G only are fall within the range of type A. The cause of increase water absorption especially when adding sand may due to increase of primary porosity. All samples are shown low fluorescence which fall within type A [7]. The less reduction in volume and lower weight losing happened in sample (L.P.C.C) by drying, while the greater loss in weight happened in sample (L.P.C.C) also. The weight loss can produce mostly from the loss of forming water throughout drying process in addition to lose of crystalline water during burning process. The most accepted exegesis of this phenomenon that the larger part of forming water in this sample goes in the structure of clay minerals, so the water loss in drying asymptotic to firing [4]. During samples burning, the shrinkage happens through most samples except (20S, 30G) that bloated, this is may be related to the mineralogical changes [9]. It is worth to mention in this context that, the shrinkage and/or bloating and the weight loss didn't lead to any fracturing in the samples through drying and burning process. This is may be due to (48) hours shadow drying before burning and the gradual increase of temperature and (2) hours of burning maturity as well as the good properties of the studied clay.

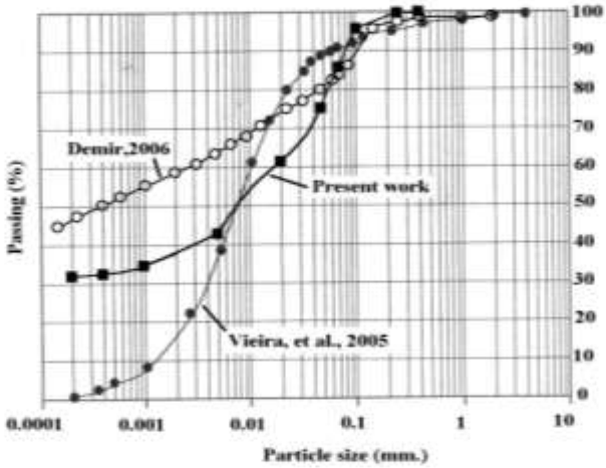


Figure 4: Grain size distribution diagram for sample (L.P.C.C) compared with previous studies.

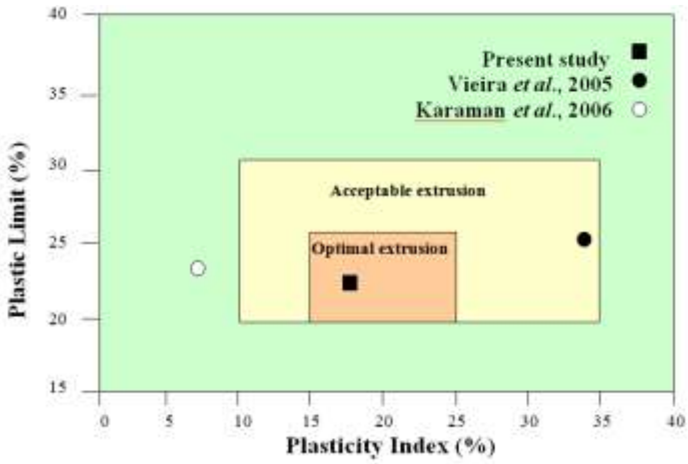


Figure 5: Natural sample (L.P.C.C) optimization according to Atterberge limits comparing with previous studies.

Table 3: Physical properties and the effects of drying and burning on dimensions and weights.

Sample no.	Bulk density (gm./cm^3)	Water absorption (%)	Fluorescence	Reduction by drying (%)			Reduction by firing (%)		
				length	volume	weight	length	volume	Weight
(L.P.C.C)	1.63	21.6	low	1.8	8.3	17.3	2.3	6.6	17.4
10S	1.55	28.1	low	2.8	13.8	18.9	1.1	3.6	16.8
20S	1.63	24.1	low	5.5	22.1	18.6	0.0	-3.6	16.3
30S	1.53	26.0	low	4.3	15.4	19.8	0.7	1.1	16.9
10G	1.62	22.9	low	3.6	11.6	18.8	2.2	8.0	15.7
20G	1.64	19.6	low	3.6	13.6	19.3	2.1	5.3	13.4
30G	1.51	18.7	low	2.8	10.2	18.8	-6.4	-2.0	11.7

The additives are down shot the compressive and tensile strength of the produced samples from 19.0 M Pa for compressive strength to 13.4 in sample 10S, and from 1.2 M Pa to 0.4 M Pa for tensile strength (Table 4). That's mean the additives change the product from type A to type B and C [20]. The average percentage of tensile strength of the respective compressive strength is 6.25%, and the lowest percentage is for sample 20S, while the highest is for sample 10S. Generally, the percentage of all samples ranged between 3% and 10%, the excepted range usually is near the value 2.8% of the sample 20S [15]. The sand and glass additives are decrease the calcium carbonate to reach the optimum percent that utilize as fluxing agent, herewith using the rice husk with non-carbonate clay enhance the characteristics of product brick [10], [21].

Table 4: Mechanical properties measured for burned samples.

Sample no.	Compressive strength (M Pa)	Tensile strength (M Pa)	(Compressive/Tensile)%
(L.P.C.C)	19.0	1.2	6.3
10S	13.4	1.2	8.9
20S	14.0	0.4	2.8
30S	15.7	0.6	3.8
10G	14.3	1.0	7.0
20G	14.5	1.1	7.6
30G	16.2	1.2	7.4

4. CONCLUSIONS

The initial spotlight of the study is that the studied clay stone layers can be regarded as suitable raw material for brick industry through its appropriate mining conditions and good engineering properties. The additives leads to reduce; density and water absorption, but in the same time would reduce the mechanical strengths. The study is concluding to use the raw materials of clay alone for brick production without sand and glass additives. The price of raw material without additives is assumed 0.3 cent for one brick, while with sand additives may reach to 0.5 cent and with grind glass may reach to 1.1 cent. From the commercial viewpoint, using the (L.P.C.C) material without any additives reduce the manufacturing cost. This study is recommended to possibility delineate stratigraphically for discover other clay stone beds in other places, and trial to use other additives like thrash and straw that is available in the area.

5. ACKNOWLEDGEMENT

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