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Effect of the Change of Firing Temperature on Microstructure and Physical Properties of Clay Bricks from Beruas (Malaysia)

I. Johari[1], S. Said[1], B. Hisham[1], A. Bakar[1], Z. A. Ahmad[2*]

[1]School of Civil Engineering, University Sains Malaysia, Penang, Malaysia

[2]School of Material and Mineral Resources Engineering, University Sains Malaysia, Engineering Campus, 14300 Nibong Tebal, Penang, Malaysia

Abstract:

This study is focused on the behaviour of fired-clay brick from the area around Beruas (Malaysia) that is known for it brick industries. The firing temperatures were set from 800°C to 1250°C and soaking time was fixed for an hour. The effects of firing temperature on the phase changes, microstructure, compressive strength, water absorption and porosity of the bricks were investigated. Test results indicate that the optimum firing temperature was found to be 1200°C. The percentage of porosity significantly reduces from 39.33% to 5.87% when sintered from 1000 °C to 1250 °C. Bricks sintered at 1200°C exhibited the highest strength of 89.5 N/mm^2. The effect of firing temperature significantly improved the microstructure in terms of porosity and the quality of physical properties of fired-clay bricks.

Keywords: *Fired-clay brick, Microstructure, Porosity, Phase analysis, Compressive strength, Water absorption*

1. Introduction

During firing of fired clay brick, a series of transformation occurs which determine the final properties of the brick product. The main factors involved in manufacturing bricks are the type of raw materials, fabrication method, drying procedure, firing temperature and firing profile. These factors will affect the quality of the final product [1]. However, [2] suggested that the durability and strength of bricks are related to their microstructure and mineralogy. In unfired clay bricks, the strength and water permeability are related to the size and shape of the particles present and the forming process, but upon heating, the nature of the mineral comprising the mass has a very important influence because of the chemical reactions and partial fusings which occur then [3].

The porosity in brick unit depends on the type of clay used in manufacturing and temperature of firing. According to [4], the porosity of the brick influences its compressive strength, water absorption and permeability. During the sintering process in the brick manufacturing process, stable initial raw materials transform into complex compounds at high temperatures. New compounds are also formed due to chemical reactions that take place. These compounds have impacts on the stability of the material due to the decrease or increase in the volume of the system. A previous study by [5] showed that vitrification of ceramic material is an important factor that influences the quality and physical properties of the end product such as strength and permeability.

*) Corresponding author: zainal@eng.usm.my

The objective of this research is to study the effect of the change of firing temperature on microstructure and physical properties of clay bricks from Beruas (Malaysia). Beruas is known as one of the important clay brick manufacturing areas in Malaysia, located on the west coast of the Malaysian Peninsular as illustrated in Fig. 1. Beruas is rich in ball clay which is suitable in producing fired-clay bricks and is thus the major producer for the northern part of Malaysia. Bricks in Beruas are traditionally produced using wood firing kilns.

Fig. 1. Location of the clay pits and the brick factory in Beruas, Malaysia

Most of the factories have been in production for more than 30 years. To date, there is no systematic scientific information published regarding clays from Beruas. The method of manufacturing fired-clay brick was adopted from the factories but the firing process is different. Heating rate, soaking time and firing schedule were constant. The firing temperatures used are varied, from 800°C to 1250°C.

2. Materials and method

2.1. Preparation of test specimens (before firing)

The clay was supplied by brick factories around Beruas, Malaysia. The material collected was wet and in large chunks, and was then exposed to ambient sun and dried at maximum temperature of 35°C, with minimum 6 hours exposed time for 7 days. Identification of clay was carried out by the XRD technique using a Bruker D8 ADVANCE machine. XRD

patterns were scanned in steps of 0.034° in a range of diffraction angles from 10° to 58° of 2θ° for clay using Copper (CuKα) as X-ray source with a wavelength of 1.5406 Å.

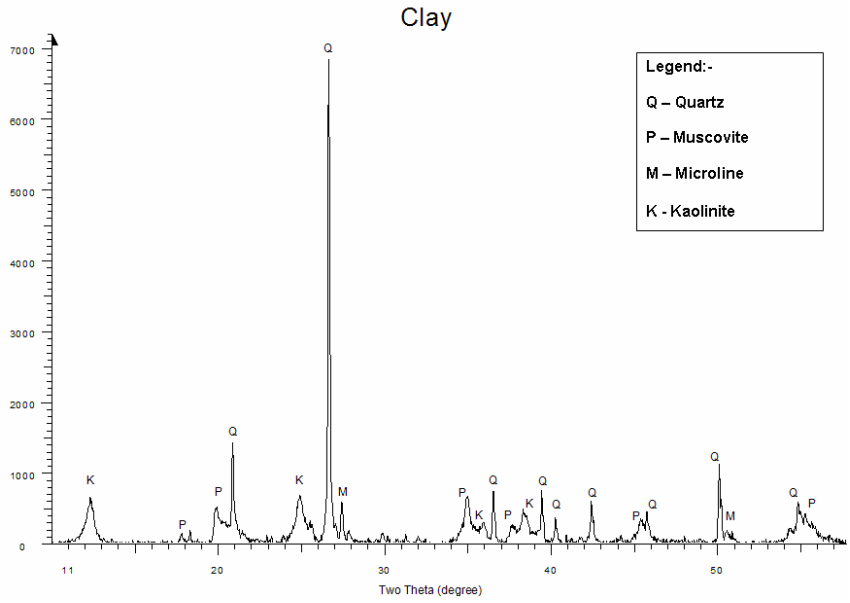

Fig. 2. XRD diffractogram of clay taken from Beruas, Malaysia

The results are shown in Fig. 2. An X-Ray spectrometer machine, Rigaku RIX 3000, was used to determine the chemical composition of the clays. Samples were sieved passing 75μm before testing was conducted. 25g of a particular sample was collected in a dried state and then compacted inside a sample holder. Loss of ignition value for the sample was prepared separately. For the Atternberg limit test and density test, the clays were sieved passing 425μm and the test was performed in accordance to BS1377: Part 2 Clause 4.3 (1990) and BS 1377: Part 2 clause 8.3 (1990), respectively. Using a suitable amount of water for mixing, the sieved clays were extruded and wire cut to form brick shapes in the size of 67 x 33 x 20 mm^3. The green bricks were air-dried for 24 hours in an ambient room temperature of 27°C and oven dried at 105°C for another 3 days. Before the firing process, the simultaneous thermogravimetric analysis (TGA) and differential thermal analysis (DTA) techniques that permit the continuous weighing of samples as a function of temperature at desired temperature were carried out [6-8]. The machine used was NETZSCH model STA 409 PC LUXX.

2.2. Preparation of test specimens (after firing)

The dried green bricks were fired in a muffle furnace, model Carbolite 1400. 10 samples were fired for each firing temperature, from 800°C to 1250°C, with 1 hour soaking time, respectively. After the sintering process, the fired-clay bricks' physical properties were observed. The particle morphology of the materials was performed by scanning electron microscopy (SEM). In this study, field emission scanning electron microscope (FESEM) with high resolution imaging was used to characterize the samples. The phase changes after firing were investigated using XRD technique. XRD patterns were scanned in steps of 0.017° in a range of diffraction angles from 6° to 90° of 2θ° for clay bricks fired at different temperatures. Then the fired-clay brick samples were subjected to compressive strength, water absorption and porosity tests. It was performed according to BSEN 772-1 (2000), BSEN 772-7 (1998) and ASTM C20 respectively.

3. Results and Discussion

3.1. Properties of clay

The physical colour of clay supplied was grey. After the sintering process, the colour of fired-clay brick turned into dark red (Fig. 3) at the temperature of 1200°C and indicated that there is a Fe_2O_3 content. It is shown from the XRF analysis as tabulated in Tab. I. The clay contained less than 3 wt% fluxing components (K_2O, Na_2O and CaO). Fig. 2 presents the XRD patterns of clays. The clay is contains two major minerals, kaolinite $Al_2(Si_2O_5)(OH)_4$ (ICDD 01-079-1570) and quartz SiO_2 (ICDD 00-046-1045). Other minerals such as microline $KAlSi_3O_8$ (ICDD 01-084-0709) and muscovite $K(MgAl)_{2.04}(Si_{3.34}Al_{0.66})O_{10}(OH)_2$ (ICDD 00-040-0020) were detected. Tab. II shows the Atterberg limit of clay. The value of plasticity index, plastic limit and liquid limit were 24.14%, 25.56% and 49.70%, respectively.

Tab. **I**. XRF analysis of clay from Beruas, Malaysia (wt %)

Component	wt%
SiO_2	67
Al_2O_3	26
Fe_2O_3	2.9
K_2O	2.1
MgO	1.2
Na_2O	0.069
CaO	0.11
P_2O_5	0.036
Loss on ignition (LOI)	8.75

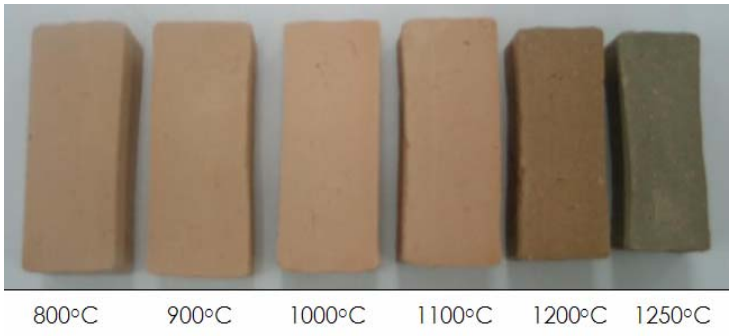

Fig 3. Effect of firing temperature on the colour and shrinkage of the clay

Tab. II. The properties of clay from Beruas, Malaysia

Properties	Result
Natural water content (%)	16.30
Liquid limit (%)	49.70
Plastic limit (%)	25.56
Plasticity index (%)	24.14
Density (g/cm^3)	2.60
Colour	Reddish grey

3.2. Thermal analysis (TG/DTA) on raw clays

From Fig. 4, the first stage of firing clay is a completion of drying pore water. From the TG curve, it can be seen that the initial weight of the sample was reduced by 0.62% when

the temperature was increased up to 289.8°C. At the temperature of 350°C, the chemical combination water of the clay started to be driven off. This chemically combined water is not to be confused with pore water and water of plasticity, which escapes during early stages of drying. This chemically combined water is part of the molecular structure of the clay and is only affected by temperatures above 350°C. The weight of the sample was reduced by 1.43% when the temperature increased to 658.8°C where the dehydroxylation of clay minerals occurs. The effect of flux components such as K_2O, Na_2O and CaO can be seen when the clay started to have a reaction at around 900°C. This also marks the beginning of the sintering process for the clay.

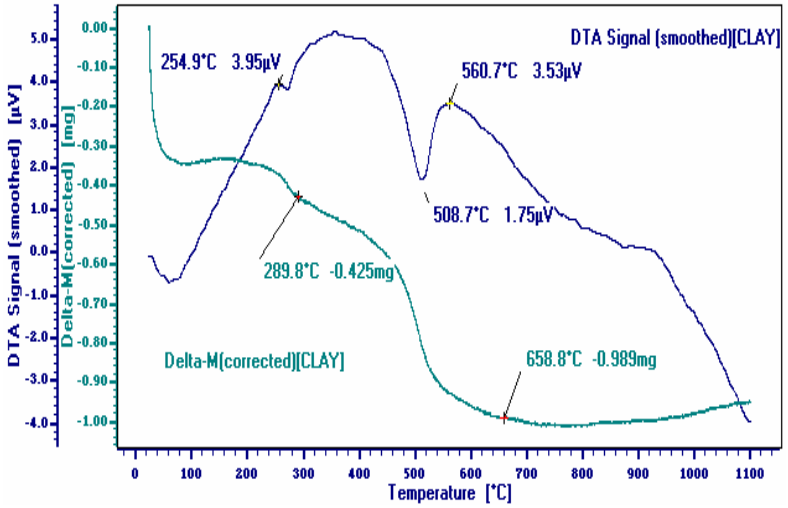

Fig. 4. Thermogravimetric and differential thermal analyses of the raw clay

3.3. Phase identification

In Fig. 5, changes of diffractograms of fired-clay bricks with increasing sintering temperatures are illustrated.

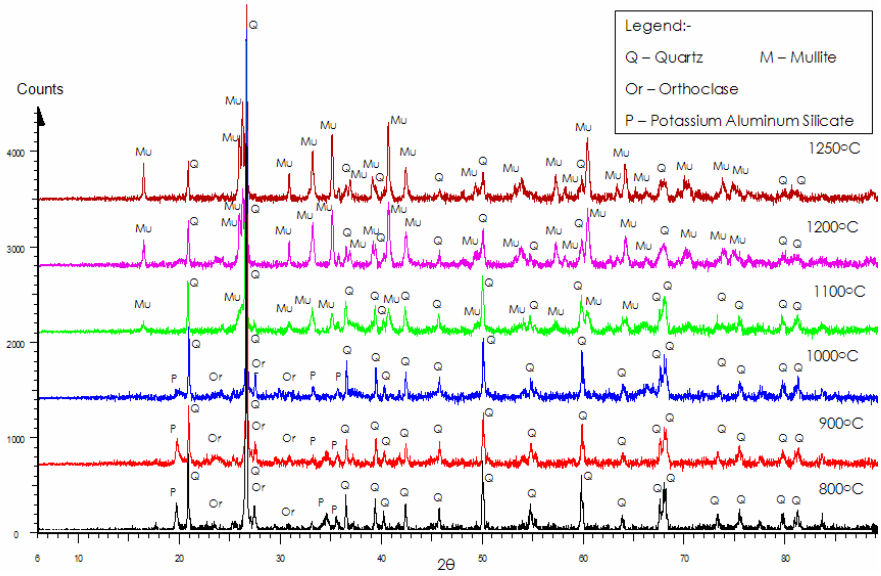

Fig. 5. XRD diffractograms of the clay fired at different temperatures

Based on the results, quartz SiO_2 (ICDD 00-046-1045), potassium aluminum silicate $KAl_3Si_3O_{11}$ (ICDD 00-046-0741), orthoclase $KAlSi_3O_8$ (ICDD 01-075-1592), and mullite $Al(Al_{1.272}Si_{0.728}O_{4.864})$ (ICDD 01-083-1881) were detected. It shows a significant change of clay phase when sintered from 800°C to 1250°C. Along with the disappearance of the lines of quartz, a decrease in the feldspar content and the formation of aluminosilicate is noted (Fig. 5). Most of the quartz phases and peaks disappeared and changed form when temperature rises to 1250°C. The orthoclase peak was slowly diminishing when sintered from 800°C to 1100°C and completely disappeared at 1200°C. This is due to the formation of mulite phase. A new phase of mullite developed between the temperatures of 1100°C to 1250°C as explained by [9]. The peak also became sharp, showing a highly crystalline compound [10].

3.4. Microstructure of clay brick

Fig. 6 shows the microstructure of the fracture surface of the clay bricks sintered from 800°C to 1250°C for 1 hour. The microstructure changes with the sintering temperature. At 800°C and 900°C, the brick has not yet experienced full solid state sintering process since the individual clay particles are still existent.

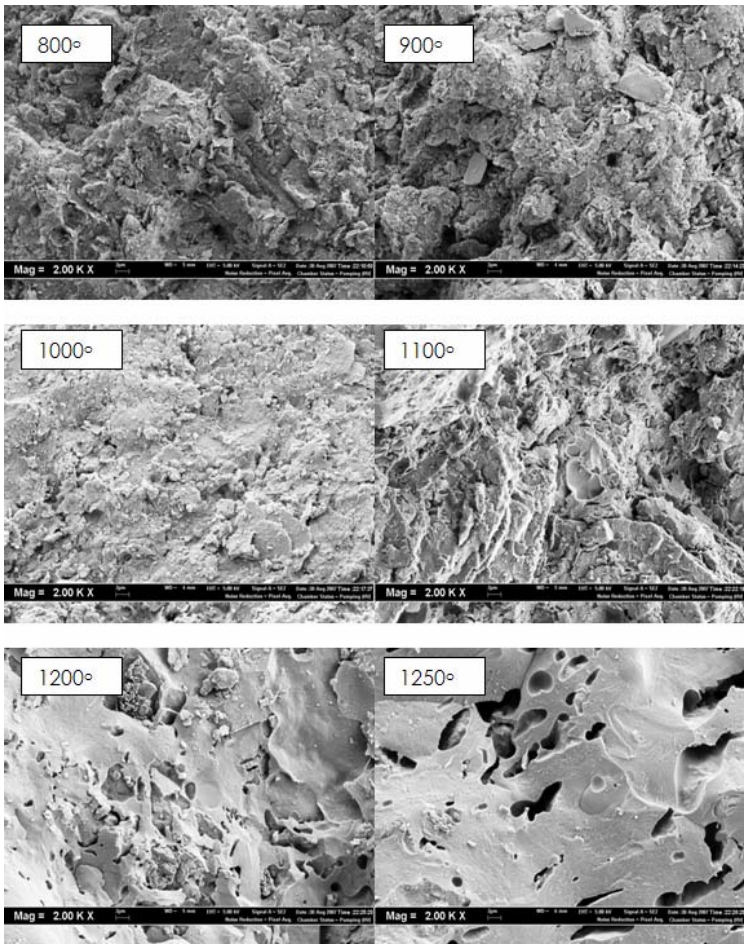

Fig. 6. SEM micrographs for the clay fired at different temperatures

This behaviour is consistent with that found by [11], who showed that the brick structure formed at lower temperatures (840-960 °C) remained essentially the same until temperatures

of over 1080°C are reached. The porosity of brick shows an increment of 1.4% and 0.1% from 800°C to 900°C and 900°C to 1000°C, respectively (Fig. 7). The increasing in porosity was the result of diffusion at relatively low temperature without significant shrinkage. The shrinkage value for temperature 800°C, 900°C and 1000°C is 0.31%, 0.50% and 1.04%, respectively. The surface also looks rough and a bit dusty. The bricks that were sintered until 1000°C are considered as having a porous structure since their water absorption rates are higher than 25%, as shown in Fig. 8.

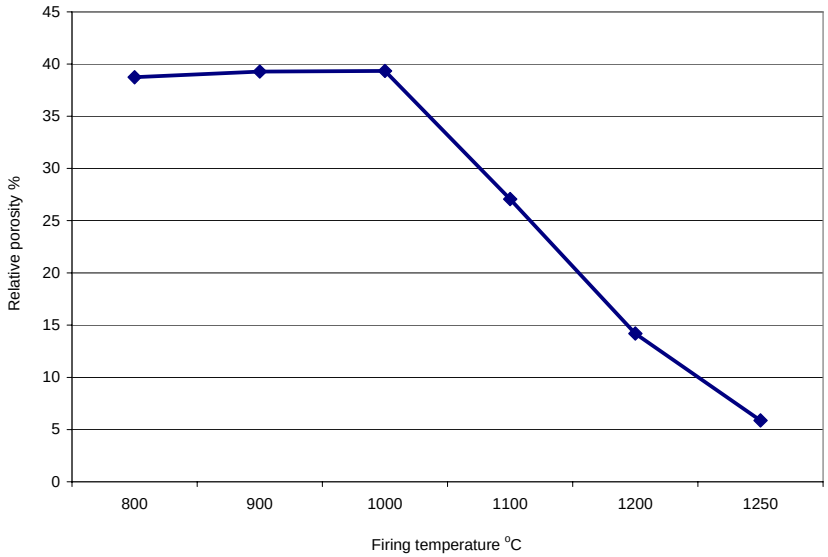

Fig. 7. Effect of firing temperature of the clay on the porosity

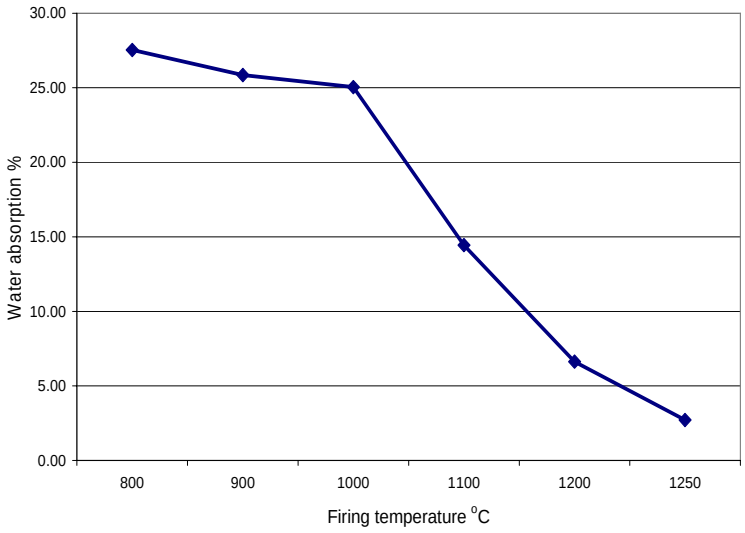

Fig. 8. Effect of firing temperature of the clay on the water absorption

Between 1000°C to 1100°C, the solid state sintering becomes very significant since the clay body had been fully sintered. Very few pores can be seen in the microstructure. Brick porosity value reduces significantly from 39.33% to 27.06% and it was 31% reduction. As agreed by [12] and [13], the purpose of the solid state sintering process is to develop atomic bonding between particles by a diffusion mechanism. This diffusion followed by grain growth will create a dense structure with significant shrinkage. The shrinkage value increases 74%

causing the reduction in volume for brick sintered from temperature 1000^0C to 1100^0C. A progressive gain in strength can be observed on brick sintered at 1100^0C where the compressive strength increased from 25.4N/mm^2 to 71.8 N/mm^2. This is also the temperature at which vitrification was first detected by SEM [14].

Starting from 1100^0C, the liquid phase sintering becomes a very important sintering mechanism. [15] emphasized that the liquid phase sintering was existent if there is a liquid phase that coexists with particulate solids during the sintering process. During this process, the reduction of pores becomes more significant as the compacted structure starts to increase its performances, such as strength and water permeability. The fired-clay brick sintered at 1100^oC begins to diffuse and shrink as the liquid phase starts to form and fill up the pores, creating smaller pores. The brick shrunk 37% when sintered from 1100^0C to 1200^0C causing the porosity to reduce 47.5%. The effect of firing also causes the water absorption value to reduce 42% lower than the value for brick sintered at 1000^oC.

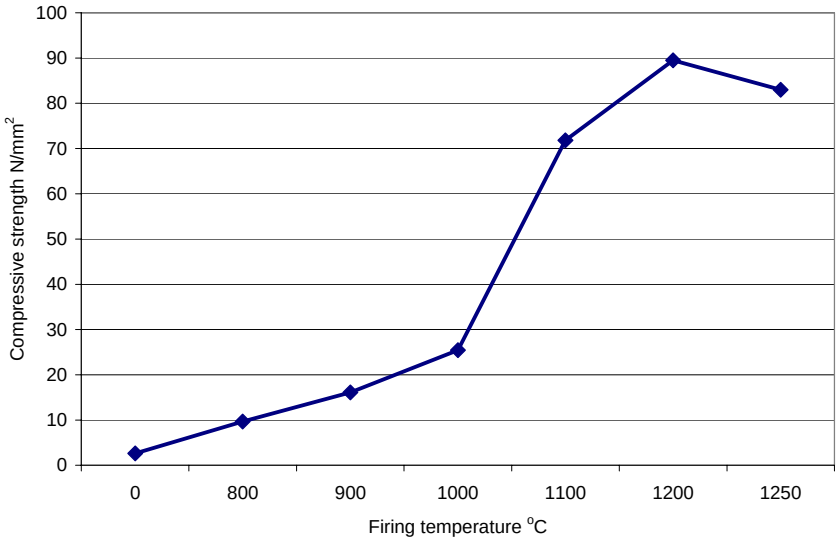

Fig. 9. Effect of firing temperature of the clay on the compressive strength

The internal surface of pores in bricks sintered at 1200^0C and 1250^0C has a "glazed" view (Fig. 6). The sintering process reached the optimum temperature at 1200^0C, whereby its microstructure contains minimum pores with porosity value 14.2 % and produces the highest strength, 89.5 MPa, as shown in Fig. 9. However, at 1250^0C, the microstructure shows larger pore sizes and lower porosity value which is 5.87% with brittle fracture behaviour. The brick becomes more brittle due to a larger portion of glassy phase in the microstructure. Therefore, the strength of the sample becomes lower (83 MPa). Even though the porosity value is lower than the brick sintered at 1200^0C, this only effect on the water absorption properties where the value of water absorption for brick sintered at 1200^0C and 1250^0C was 6.63% and 2.71%, respectively.

4. Conclusions

Firing has a positive influence on the microstructure of brick promoting a dense structure with low permeability. At temperatures of 1000^0C or above, the technical quality and durability of bricks is generally superior. It displays a high in compressive strength, lower porosity and water absorption value. The findings indicate that the physical and mechanical

properties of bricks can be controlled to a significant extent by varying the firing temperature. The best firing temperature for fired-clay bricks with good performance of mechanical properties was discovered to be 1200^0C. There is no doubt about its potential in the construction industry, not only as filler in walling systems, but also as load bearing structures. However, due to economic reasons, the firing temperature can be reduced to between 1050°C to 1100°C. The fired-clay brick can still achieve strength around 40-70 N/mm^2, porosity below 29% and obtain water absorption value below 25%.

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References

1. G. Cultrone, E. Sebastian, M. J. De la Torre, Constr. Build. Mater. 19 (2005) 39.
2. R. A. Livingston, P. E. Stutman and I. Schuman, Conservation of Historic Brick Structure, 11 (1998), 105.
3. A. Bremner, Origin and effect of cryptoflorescence of FEB. PhD thesis, Leeds University, London, 2002.
4. F. M. Khalaf, A. S. DeVenny, J. Mater. Civil Eng. 14 (2002), 334.
5. M. S. Tite and Y. Maniatis, Nature 257 (1975) 122.
6. W. E. Worrall, Clays and Ceramic Raw Materials. 2nd Edition, Elsevier Applied Science Publishers LTD, England, 1986.
7. B. Velde, Introduction to Clay Minerals. 1st Edition, Chapmen & Hall, London, 1992.
8. Y. P. Khanna, Thermal Characterization of Material, in Guide to Materials Characterization and Chemical Analysis. 2nd Edition, Sibilia, J.P. (Ed.), Wiley-VCH: New York, 1996, 261-285.
9. M. M. Jordan, M. A. Montero, S. Meseguer, T. Sanfeliu, Appl. Clay Sci. 42 (2008) 266.
10. P. Chindraprasirt, K. Pimraksa, Powder Technol. 182 (2008) 33.
11. M.S. Tite, Y. Maniatis, Trans. Brit. Ceram. Soc. 74 (1975) 19.
12. M. W. Barsoum, Fundamental of Ceramics. McGraw-Hill, Singapore, 1997.
13. R. Warren, Ceramic – Matrix Composites. Blackie USA Chapman & Hall, New York, 1992.
14. G. Cultrone, C. Rodriguez-Navarro, E. Sebastian Pardo, O. Cazalla, M. J. De la Torre Lopez, Eur. J. Mineral. 13 (2001) 624.
15. R. M. German, Liquid Phase Sintering. Plenum Press, New York, 1985.

Садржај: *Фокус ове студије је понашање жарене глинене цигле из области око Берауса у Малезији која је позната по индустрији цигла. Температура жарења је била између 800 и 1250^{o}C и време потапања је било фиксирано на 1 час. Проучени су ефекти температуре жарења на промену фазе, микроструктуте, компресивну јачину,*

абсорпцију воде и порозност цигле. Резултати тестова показују да је оптимална температура жарења била 1200°C. Процена порозности се значајно смањује од 39.33% до 5.87% када се синтерује од 1000 до 1250°C. Цигле синтероване на 1200°C су имале највећу јачину од 89.5 N/mm^2. Ефекат температуре жарења је значајно побољашао микроструктуру са гледишта порозности и квалитета физичких својстава жарених глинених цигли.

Кључне речи: *Жарена глинена цигла, микроструктура, порозност, фазна анализа, компресивна јачина, абсорпција воде.*