Locally available Clays of Bangladesh as a Replacement of imported Clays for Ceramic Industries

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Abstract

In Bangladesh, generally imported clays are used as the main ingredient in ceramicware industries. Though locally available clays can be refined and used instead of imported ones to minimize the high manufacturing cost. In this research, 3 locally available clays were investigated both mechanically and spectrally. The XRF analysis of local non-refined clays in contrast to imported clays has indicated the presence of excess SiO_2 content (about 35%) in the form of free silica as well as TiO_2 and iron oxide but the Al_2O_3 content are a presence in lower amount (around 8.7%). In the ceramic body, a high amount of silica content shows low plasticity which creates a crack and iron oxide could adversely affect the translucency of ceramic ware. After the refining process from those local clays, the amount of SiO_2 content was reduced up to 40.2% along with 5.5% increment in Al_2O_3 content. From the mechanical analysis of those clays, hardness, impact and compressive strength show very good results compared with imported clays. Copyright © VBRI Press.

Keywords: Clay refining, material analysis, ceramics, Bangladesh.

Introduction

In recent decades of Bangladesh is known as a developing country, at the recent time demand and use of low-cost ceramicware and tiles (both floor and wall) is increasing greatly. But the costs of these products are comparatively high, due to the use of the imported clay raw materials. Consequently, the utilization of locally accessible raw materials for the production value of ceramicwares is significant because such a step will make a good economic figure in the economic condition of the country. The main reasons behind not using local clay are the unavailability of proper clay refining methodology in the country.

Keeping this view in mind, clays of three different areas (Modhupur, Bijoypur, and Sherpur) from Bangladesh are used to prepare the samples at three different temperature (1050ºC, 1100ºC and 1150ºC). This non-refined treated clay sample, after being characterized (chemical and mechanical properties) compared with the properties of refined clay samples at 1100ºC. Refined clays had shown better physical and mechanical properties, it can be used for the production of ceramic ware. Using the data of the different physical, compositional and mechanical test, the investigation is aimed to use these refined local clay materials in ceramic ware and quality tiles by replacing imported clay in order to reduce cost.

Literature review

Adnan Mousharraf, Md. Sazzad Hossain and Md. Fakhrul Islam were concentrated on Potentiality of Locally accessible Clay as Raw Material for Traditional Ceramic Manufacturing Industries [**1**]. Most of the whiteware manufacturing industries in Bangladesh, imported clay as the major raw material from abroad. The main points are the unavailable technology and the lack of proper clay refining methods in the country and in some part the unsuitable chemical and structural composition of the locally available naturally occurring clays. Moreover, particle size identification has been a major issue in the processing of naturally available clay. These two vital features affect the strength and plasticity of clay processing technology.

Research on the physical substance of Patia clay (Chittagong), Bangladesh is done by A. H. Dewan, S. Mustafi, M. Ahsan and M. S. Ullah [**2**]. The microstructural and physical characteristics of Patia clay of individual areas (Hydgaon-1, Hydgaon-2 and Kanchannagar) were examined. Patia clays were resulted in good by repeated sedimentation technique.

The chemical configurations of raw materials (Which is unwashed) and washed clay was destined. The physical substance of fired Patia clay was determined. Chemical configurations of Patia clay (unwashed and washed) collected from the individual area of Patia were brought to pass using X-ray Fluorescence Spectrometer (XRF) (PANalytical XRF, Model PW-2404 X-Ray Spectrometer) and the fired Patia clay sample (unwashed and washed) was put down to numerous tests.

The firing shrinkage was determined by Eliche-Quesada *et al.*,2018 in an environment-friendly process. The beneficiation technique of the clay was obtained on the principle of washing and pass through in a slow stream of water [**3**]. The ratio of silica in the unwashed clay is a high and low percentage of Alumina is present in it. And the higher concentration of silica and a lower concentration of alumina of the clays are not fit for the manufacturing of quality ceramic wares. The raw clays are appropriate for manufacturing regular pottery. On the contrary, the washed clays contain a lower percentage of silica and a higher percentage of alumina comparable to unwashed clays. Both the washed and unwashed clays contain a small amount of MgO, CaO, K_2O and Na_2O which acts as a change factor in a ceramic substance during firing because of the lessening of free silica washing of raw material of clay.

Research on the Physio-Chemical Parameters of Ceramic Tiles Produced from Locally Available Raw Materials is completed by S. A. Jahan, S. Parveen, S. Ahmed and M. Moniruz Zaman [**4**]. Due to the lessening demand of low-cost tiles in Bangladesh, using the locally available raw materials, there are five different batches of tiles have been prepared and their physical properties (bending strength, bulk modulus, firing shrinkage, water absorption, etc.), as well as chemical properties, were studied in order to assess the quality of the products. The research showed that quality tiles can be manufactured by using different ratios of the local raw materials (quartz, zircon, clay, and feldspar) along with the molasses (a waste product from sugar industry) as a binder. Especially tiles example C and D fulfills the properties of value tiles which reflect that among the five-combination researched, the combination of raw materials for these two assortments can be streamlined for the production of quality tiles [**5-7**].

Experimental

Methodology

The structure and the size of the locally available clay particle were determined by using X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD) to unboxing the effective potential of these clays for the refining purpose. Researches have been brought to pass out so far on clays from the several regions across the country in order to distinguish between the compositional variation among different types of clays. This kind of analysis is typically performed with the X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD) technique. The local material is much higher in Fe_2O_3, SiO_2 and TiO_2 and undesired amount of Al_2O_3 content. In a typical manner, most of the Silica content stay in the free form as Quartz. Oddment of the Silica content is allied with Alumina in bonded form, which forms phases like Kaolinite, Halloysite, etc. Also, impurities like Fe_2O_3 and TiO_2 are also higher in amount in most of the local raw clay types. Bijoypur Clay (White Clay) has contained a higher SiO_2 content, and yet Al_2O_3 content contains low impurities in it. Thus, this clay brings out a great potential which can be turned into the suitable raw materials for the conventional ceramic manufacturing technique. The local clays consist a percentage about 65-73% of Silica, 7-9% of Iron (III) oxide, 22-27% of Alumina and 1% of Titanium oxide which helps to meet the challenge of using local clay because of the higher content of Silica. X-Ray Fluorescence (XRF) analysis showed the appearance of SiO_2 content in the form of Silicate, TiO_2, and Fe_2O_3 all of which represent a challenge in the refining process. Also, the amount of Al_2O_3 is not up to the minimum level in most composition. By utilizing this information dependent on the insights on molecule sizes of raw materials of various starting points, this examination was planned to reveal the structure and properties of the locally available raw material of clay that can be anticipated for further refining to make appropriate as raw material for whiteware industries.

Material refining

X-Ray Fluorescence (XRF) analysis showed the appearance of SiO_2 content in the form of Silicate, TiO_2, and Fe_2O_3 all of which represent a challenge in the refining process. The amount of Al_2O_3 in composition must not be up to the minimum level. The X-Ray Fluorescence (XRF) analysis shows the percentile of chemical components of the raw materials. **Table 1** shows the X-Ray Fluorescence (XRF) data of the 3 samples along with imported clays [**1**].

Table 1. XRF analysis of local and imported clays.

Composition (%)	Modhupur	Sherpur	Bijoypur	China	Ball
SiO_2	69.5	70.4	72.8	50.4	48.3
Al_2O_3	24	22.5	22.2	32.8	33.1
Fe_2O_3	1.8	1.9	1.78	1.85	1.33
TiO_2	1.33	1.4	1.32	0.6	3.57
Others	3.37	3.8	1.9	14.35	13.7

The intramolecular structure of local clay and imported clay are examined by using X-Ray Fluorescence (XRF). The X-Ray Fluorescence (XRF) analysis has indicated the abundance of SiO_2 content as free silica, TiO_2, and Iron (III) Oxide and the presence of the Al_2O_3 content in a lower amount. In the ceramic body, a high amount of silica content shows low plasticity which creates a crack and iron oxide could adversely affect the translucency of ceramic ware.

Primary refining

Firstly, raw material clay was mixed with pure water to form a semiliquid mixture called slurry. After that, the slurry was kept in a pot for 24 hours. After that clay particles were settled down in the bottom of the pot and some clay impurities were floating upon the top surface of the slurry.

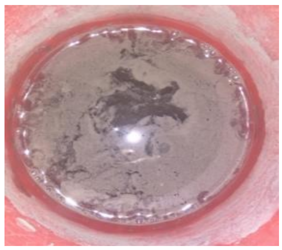

Fig. 1. Primary refining.

Fig. 2. (a) Sodium silicate deflocculant and (b) Poly-acrylamide flocculant.

Addition of deflocculant and flocculant chemicals

Appropriate deflocculant, for example, sodium silicate was added to the slip and the slurry was constantly fomented. For deflocculating reason, around 3.5 - 3.75 kg of sodium silicate was included per ton of clay. At the point when the clay particles become deflocculated, the dispersed clay slip was additionally diluted to roughly 10% to 20% solid substance by including pure water. Poly-acrylamide type anionic or non-ionic flocculants were added to the slip at an extent of 3.5 - 3.75 kg/ton of clay. **Fig. 2** demonstrates the picture of used Deflocculant and Flocculant chemicals.

Before adding Polyacrylamide, pH (puissance of Hydrogen ions) range should be kept between 8.0 - 9.5; for removing the impurities like Fe_2O_3 and TiO_2. At this pH range, the contamination particles got specifically flocculated with the poly-acrylamide polymer to form substantial flocs; which rapidly settle at the bottom and the suspended clay was evacuated either by centrifugal or gravitational forces. Then again, for the expulsion of fine SiO_2 particles, alkali like Ammonium hydroxide (NH_4OH) should be added at a proportion of 0.5 kg/ton of clay; to maintain pH level. At this pH level, Clay particles got specifically flocculated with the polyacrylamide polymer to form substantial flocs and settle at the bottom. The suspended Quartz particles were then expelled either by centrifugal or gravitational forces. At last, the prepared clay was dried to accomplish the process for supplying clay into the market.

Experimental procedure

Having acquired the knowledge about the chemical constituents of the raw materials, compositions of five different batches of tiles body were formulated as shown in **Table 2**.

Table 2. Chemical composition of experimental samples.

Different Clays	Feldspar (% wt)	Quartz (% wt)	Local Clay (% wt)	Ball Clay (% wt)	China Clay (% wt)	Water (% wt)
Modhupur	30	20	40	10	0	80
Bijoypur	30	20	40	10	0	80
Sherpur	30	20	40	10	0	80
Re. Bijoypur	30	20	40	10	0	80
Standard	30	20	0	30	20	80

From the above extents of the distinctive ingredients, 500 gm of each batch composition was arranged and exposed to ball milling for 1 day, which was then trailed by drying on a hot plate. It was then ground very well so as to accomplish fine powder with particle size under 200 meshes. Particle size is a significant factor since the littler molecule estimate is especially alluring to acquire a quality item. A few circle samples were set up at a weight of 8-10 ton using a hand squeezing machine. Such pressure accomplished the sample with a compact shape which was first dried at 110ºC. Followed by 1050ºC of firing temperature, 1100ºC and 1150ºC in a furnace to get the completed terminated fired products. An outline of the previously mentioned planning methodology appears in **Fig. 3**.

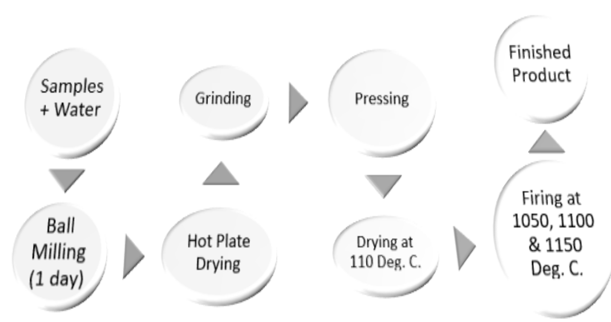

Fig. 3. Flow diagram of tile samples preparation from locally available raw materials.

From the refined firing sample density, firing shrinkage, water absorption, hardness, compressive strength & impact testing were carried out.

Results and discussion

The Identification of compositional variation needed a lot of researches to be performed in the various region across the country. The leading methods associated with these analyses are the X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD) methods. We can see the

XRF analysis of some clay types that are available locally in Table 1 and in reference to that, a comparison between local clay and imported material (standard compositions) reflects that the indigenous material is much higher in SiO_2, Fe_2O_3, and TiO_2 content, whereas, falling short in Al_2O_3 content. Generally, most of the Silica is found in free form as Quartz. The remaining can be found associated with Alumina in bonded form, which results in phases like Kaolinite, Halloysite, etc. Moreover, impure percentages like Fe_2O_3 and TiO_2 are also high in most of the local clay types. The X-Ray Diffraction pattern of three types of clays can be seen in **Fig. 4**. **Fig. 4(a)** shows the pattern for standard Bijoypur Clay and the existence of Quartz and Kaolinite in the composition. **Fig. 4(b)** and **Fig. 4(c)** represents the pattern for locally available Modhupur Clay and imported China Clay, respectively. From the XRD patterns, we can come into conclusion that Quartz is higher in comparison to kaolinite in local clays, but the china clay would show otherwise. So, we must take substantial refining action if we want to make the indigenous material suitable for industrial manufacturing.

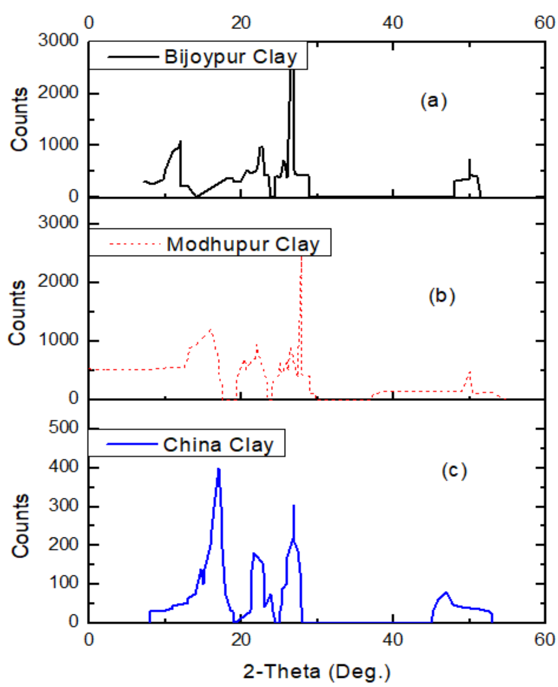

Fig. 4. XRD analysis of Bijoypur Clay (a) [1], Modhupur Clay (b) [1] and China Clay (c) [7].

For a comparative analysis total 5 types of clays are taken under consideration. From those 5 types of clays, 3 (Bijoypur, Modhupur & Sherpur) types were experimented both mechanically and chemically. From the chemical tests before refining it showed about 35% excess SiO_2 content in contrast to imported china and ball clays but after refining the SiO_2 content was reduced up to 42.01% and Al_2O_3 content was increased up to 5.5% from the 3 samples. **Fig. 5 (a)** to **(d)**, shows the comparative and percentile analysis of refined and non-refined clay samples.

From **Fig. 5(c)**, it can be seen that Modhupur (Green Dashed) and Sherpur (Blue Dashed) after refining shows almost same compositional change with respect to percentile point from the non-refined ones. But in **Fig. 5(d)**, from the compositional percentile change, the refined Modhupur clay (Green Dashed) sample shows the best likely compositions with the imported clays. So, the refined Modhupur clay has the best quality to be used instead of conventional imported clays.

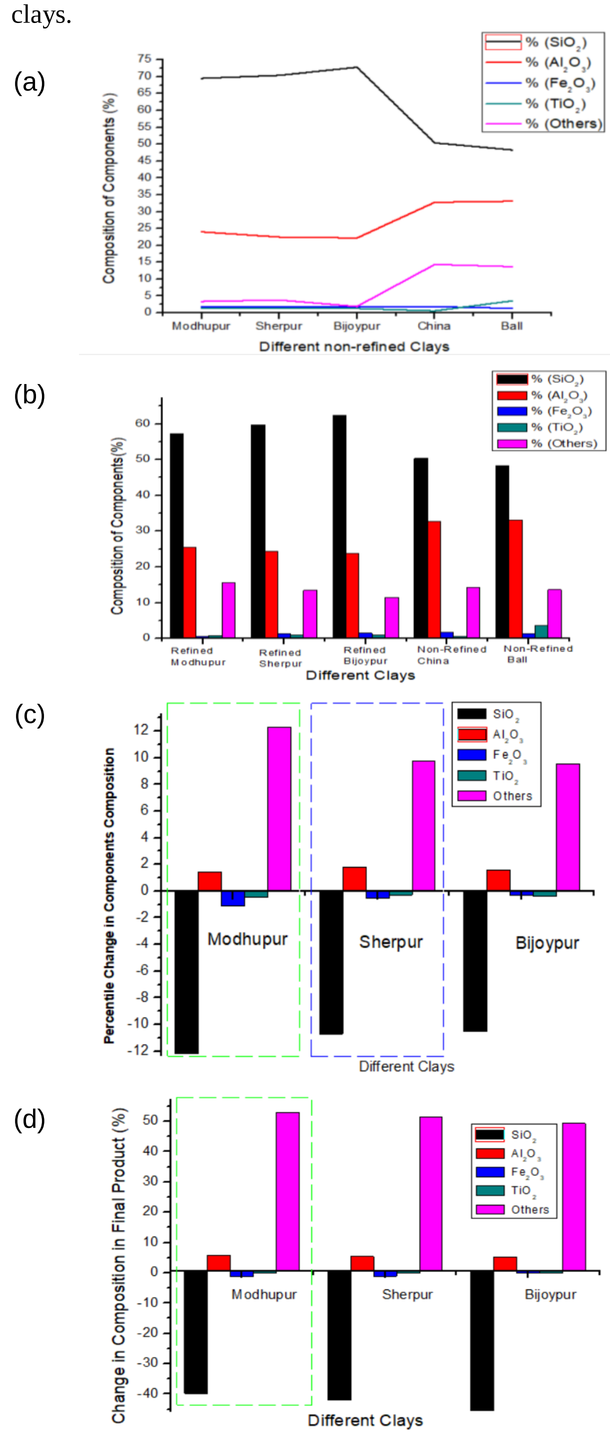

Fig. 5. (a) Non-refined Samples composition, (b) Refined local clay samples in contrast to imported china and ball clay, (c) Percentile point change in new refined local clay samples and (d) Change in composition in refined local clay samples from the non-refined samples.

Conclusion

The main motive of this thesis was to reduce the cost for producing ceramic ware and to ensure the proper utilization of local clay resources. The compositions of locally available clays were determined by using X-Ray Fluorescence (XRF). Due to high percentages of impurity content in local clay, these clays were refined. The samples were made by the traditional method of tiles production using hydraulic press machine. To fulfill the objectives of the experiment three types of non-refined local clay (Modhupur, Bijoypur and Sherpur) sample of three different temperatures (1050^{o}C, 1100^{o}C and 1150^{o}C) were used. Then some basic physical and mechanical properties such as firing shrinkage, water absorption, density, impact strength, compressive strength, and micro hardness had been tested. Various properties of Modhupur clay at 1100^{o}C was similar or higher than standard clay sample but both Bijoypur and Sherpur non-refined clay shown lower properties than both standard and Modhupur non-refined clay sample. So 1100^{o}C was the optimum temperature for Modhupur local clay. At 1150^{o}C all non-refined local clay shown good chemical properties which was comparable with standard clay, but their volume shrinkage was high for what we could not use them for prepare ceramic tiles.

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Author's contributions

Conceived the plan: M. Akanda, A. Mallik; Performed the expeirments: M. Akanda, M. Ali; Data analysis: A. Mallik, M. Akanda and A. B. Karim; Wrote the paper: A. Mallik, M. Akanda, M. Ali. Authors have no competing financial interests.

References

1. Adnan, M.; Hossain, M. S.; Islam, M. F.; *J. Chem. Eng.*, **2011**, *26*, 34.
2. Dewan, A. H.; Mustafi, S.; Ahsan, M.; Ullah, M. S.; *Bang. J. Sci. Ind. Res.*, **2014**, *49*, 255.
3. Eliche-Quesada, D.; Sandalio-Pérez, J. A.; Martínez-Martínez, S.; Pérez-Villarejo, L.; Sánchez-Soto, P. J.; *Cer. Int.*, **2018**, *44*, 4400.
4. Jahan, S. A.; Parveen, S.; Ahmed, S.; Zaman, M. M.; *Bang. J. Sci. Ind. Res.*, **2008**, *43*, 77.
5. Fahlman, B. D.; What Is "Materials Chemistry"? Mat. Chem. Springer, Dordrecht, **2018**, 1.
6. Mirsayar, M. M.; *Theo. App. Frac. Mech.*, **2018**, *93*, 170.
7. Chung, Y.; Hyojeon K; Taek-Seung, K.; Young, M. K.; Seoktae, K.; *Sep. Puri. Technol.* **2019**, *219*, 216.

