

Research on fabrication of non-calcined geopolymer ceramics from kaolin Tung Ba, Vi Xuyen, Ha Giang

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ABSTRACT

The mineral component of kaolin clay in Tung Ba commune analyzed by X-ray diffraction is composed mainly of kaolinite (24–27%), illite (26–30%) and quartz (40–44%). Chemical composition (wt%) of Tung Ba kaolin determined by X-ray fluorescence (XRF) shows SiO₂: 59.92–64.98, Al₂O₃: 18.95–20.82, Fe₂O₃: 2.28–3.95, SO₃: 2.47–4.66, CaO: 0.12–0.36, MgO: 1.72–2.13 and TiO₂ at 0.75–1.08. To fabricate geopolymer ceramic, kaolin clay was transformed into metakaolin with the optimal parameters of calcination: the temperature at 750°C and heating time of 120 min. Obtained metakaolin is mixed with mixtures of NaOH/Na₂SiO₃ (40%wt) with ratios of 0.2, 0.25, 0.33, 0.5 and NaOH molarities are 10M, 12M, 14M. Testing geopolymer ceramics after 28 days had a compressive strength of 40–196 KG/cm², water absorption of 15.25–17.98% and density of 1.54–1.69 g/cm³, totally satisfied the Vietnamese standard for construction bricks and ceramics.

Keywords: Kaolin; metakaolin; geopolymer; ceramic; alkaline activator.

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1. Introduction

“Geopolymer” term was first used by Davidovits in 1976 for inorganic polymeric materials synthesized from aluminosilicates, with the three-dimensional network structure of “Si-Al-O” sequence and Na⁺, K⁺ alkaline cations. This polymer can be solidified under conditions of room temperature and pressure, exists in amorphous to semi-crystalline polymers (Davidovits, 1991, 2011). Geopolymers have a variety of applications in many different fields including treatment of heavy metal pollution or making fire-resistant

materials, refractory materials and new building materials. These materials do not cause environmental pollution. They have high compressive strength, high corrosion resistance, and environmentally sustainable characteristics, without emission of toxic gases during the production process. (Davidovits, 1991; Romisuhani et al., 2015; Chen et al., 2016; Hubadillah et al., 2017). Raw materials for geopolymer production often contain large amounts of SiO₂ and Al₂O₃ existing in the amorphous form such as fly ash, furnace slag, metakaolin... (Davidovits, 1991, 2011; Mohshen et al., 2010).

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Studies on geopolymer production from kaolin have been carried out by Berg since 1964 but without any successful implementation in industries. In 1972, two ceramicists Jean-Paul Latapie and Michel Davidovits confirmed that ceramic tiles could be fabricated from kaolinite at a temperature lower than 450°C, with high mechanical strength and water-resistance (Davidovits, 2011). Later, research mainly focused on the effect of temperature on the physical and mechanical properties and characteristics of geopolymer ceramics such as studies of Kuenzel et al. (2013), Dikko et al. (2015) and Jaya et al. (2016) or on production of high-quality geopolymer ceramics with reduction of fabrication time and more environmentally friendly implemented by Ahmad et al. (2016).

In Vietnam, research on the fabrication of geopolymers is still a new field. Although there have been some research projects of some authors such as Kieu Q.N (2006), Kieu Q.N and Nguyen A.D (2010, 2015), Nguyen et al. (2014) on fabrication of non-calcined geopolymer bricks from red basaltic soils, semi-weathered basalt, and fly ash; Phan et al. (2015) on effect of activator composition on compressive strength of concrete; or Dao et al. (2009, 2011) and Tran et al. (2017) on physical and mechanical properties of fly ash-based geopolymers, studies of kaolin-based geopolymer ceramics be still limited. While the source of kaolin clay is quite abundant, especially low-quality kaolin clay which cannot be enriched to improve the quality and use for producing traditional ceramics, refractory materials, abrasive materials, additives in paint, or making papers,... It is very necessary to study and fabricate products from this source. In this paper, we will present obtained research results: mineral components

and chemical composition of kaolin from Tung Ba, metakaolin and the physical, mechanical properties of geopolymer ceramics. In the result, we assess the ability to manufacture geopolymer ceramics from low-quality kaolin clay.

2. Samples and methods

2.1. Research samples and alkaline activator

2.1.1. Research samples

Samples of kaolin clay used to make geopolymer ceramics were taken from Tung Ba kaolin mine. This mine was located in Nam Ria village, Tung Ba commune, Vi Xuyen district, Ha Giang province (Fig. 1) with the geographical coordinate: 22°51'45"-22°51'54" North latitude, 105°04'00"-105°04'19" East longitude. The study area was mainly covered by Quaternary sediments including clay, dark brown to yellowish-brown, coarse-grained sand. In addition, a minor part was Tung Ba formation (D_{1tb}) with the upper part containing many thick layers of extrusive rocks interbedding with iron ore seams. Tung Ba kaolin mine has a sedimentary origin with a total reserve of TN-3 up to 53.180 tons (TN-2C= 31.188 tons) (According to the evaluation result of Exploration Group 31-General Department of Geology and Mineral Vietnam on an area of 18.000m²).

2.1.2. Alkaline activator

Sodium hydroxide (NaOH) powder with purity of 98% was prepared into different concentrations of 10M, 12M, 14M.

Sodium silicate (NaSiO₃) has chemical composition of SiO₂ (30.1%), Na₂O (9.4%) and H₂O (60.5%) (SiO₂/ Na₂O = 3.2).

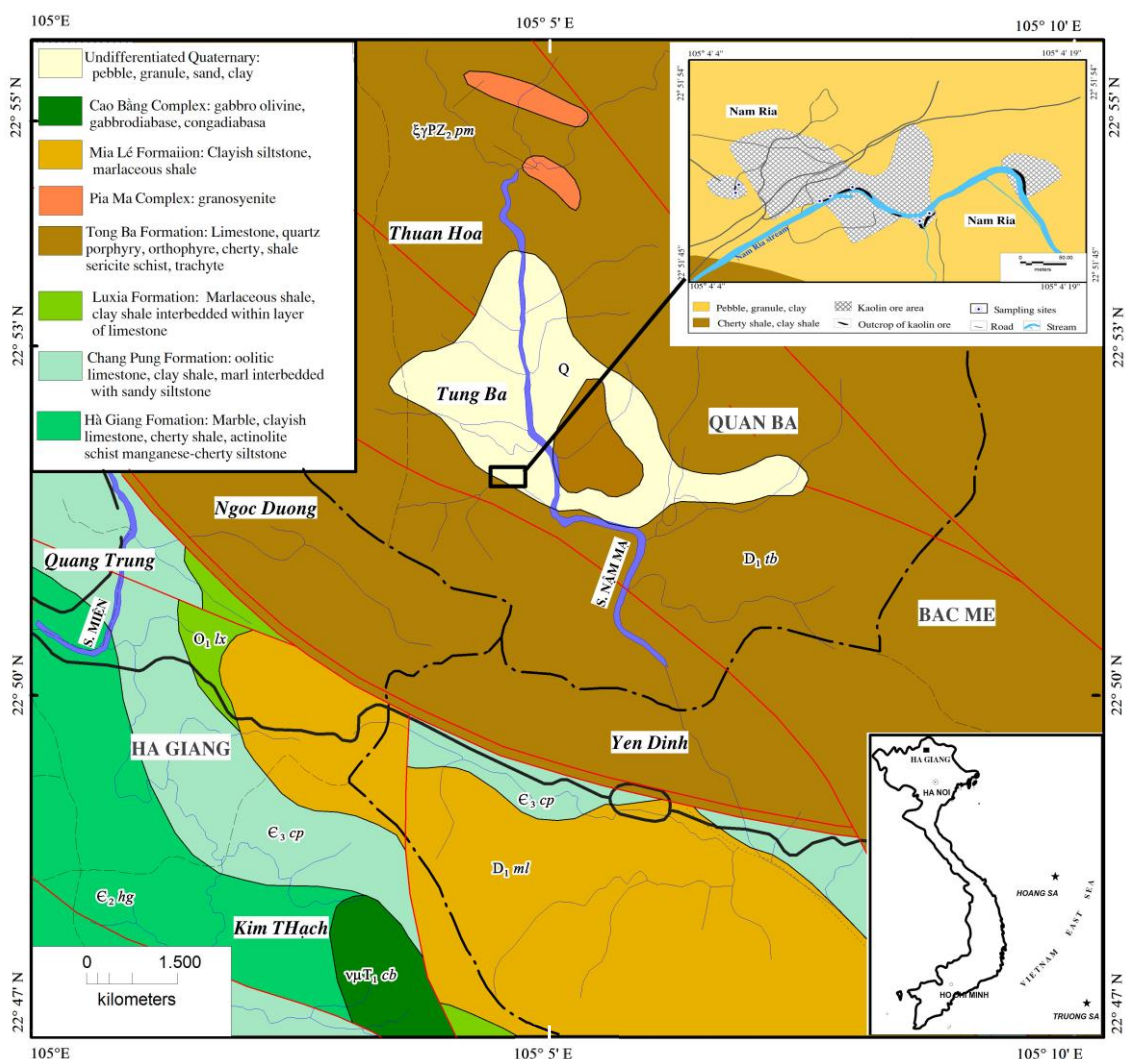


Figure 1. Geological map of the study area (after Geological and mineral resource map of Vietnam on 1:200.000 compiled by Hoang Xuan Tinh-editor). Upper right corner is the schematic map of the distribution of kaolin clay ore body in Tung Ba, Ha Giang and sampling sites (after the database of Exploration Group 31-General Department of Geology and Mineral of Vietnam). Lower right corner shows the regional position

2.2. Methods

X-ray diffraction (XRD) is used in the qualitative and semi-quantitative analysis of a mineral component of kaolin clay, metakaolin obtained from the calcination of kaolin clay at different temperatures and heating times. Research samples were analyzed on PANalytical Empyrean XRD system at

Institute of Geological Sciences-Vietnam Academy of Science and Technology, using $CuK\alpha$ radiation, tube working conditions of 45KV–40mA, an angular range of 5.0–70.0 $^{\circ}2\theta$, and divergence slit of 4 $^{\circ}$.

Differential thermal method (DTA) is the method for identifying mineral phase based on endothermic-exothermic reaction of each

mineral and the change in mass of sample during heating to quantify mineral phases. This method was performed on the STD-PT 1600 system-Center for Geological Experimental Analysis-General Department of Geology and Mineral Vietnam.

X-ray fluorescence (XRF) analysis for chemical composition was carried out on Bruker S4 Pioneer XRF spectrometer at the Institute of Geological Sciences, Vietnam Academy of Science and Technology.

Infrared spectroscopy (IR) method was performed for the analysis of structures of compounds based on information of the vibrational frequencies of molecules. The IR spectra of samples were recorded using a Nordantec-TEA 4000 FTIR spectrometer at the VNU University of Science. The spectra were scanned on a thin film dish of samples mixed with dried KBr (for 1200–400 cm^{-1} range, resolution of 1 cm^{-1}).

Dehydroxylation of kaolin is used for determining the optimal heating parameters of the process in which kaolinite was transformed into metakaolin. Samples of kaolin clay were dried at 105°C until no change in mass, then calcined in the furnace at different temperatures (550°C, 600°C, 650°C, 700°C, 750°C and 800°C) and at different heating times (30, 60, 120, 180 min) to calculate the degree of the dehydroxylation.

Mixing test method for making geopolymers: To fabricate geopolymer ceramics, 150g of metakaolin were mixed with 60ml solution of NaOH/ Na_2SiO_3 at the ratio of 0.5, 0.33, 0.25, 0.2 and the NaOH concentration of 10M, 12M, 14M (Table 5). Water was added to 2.5–5% by weighing the total ingredient. Geopolymer ceramic objects were molded into cylindrical specimens (5×5cm) and rectangular prism specimens (16×11×3cm) (Fig. 6). The molded samples were allowed to mature at room temperature for 28 days. After that, they were tested with physical and mechanical properties.

Test methods to determine the physical, mechanical properties of raw materials and test samples of geopolymer ceramics include moisture content of raw materials (TCVN 250:1986), density (TCVN 250:1986), water absorption ((TCVN 248-1986), water resistance (TCVN 3735:1982), compressive strength (TCVN 246-1986), and flexural strength (TCVN 6415:1998) of the samples. The analysis was performed at the Institute of Geological Sciences, Vietnam Academy of Science and Technology and at the Hanoi University of Mining and Geology.

3. Results

3.1. Tung Ba kaolin clay characterization

Results of XRD and DTA analysis shows that the mineral component (wt%) of kaolin clay in Tung Ba was mainly composed of kaolinite (24–27), illite (26–30), quartz (40–44), minor phases of chlorite (0-1), feldspar (2–4) and goethite (1–2) (Table 1, Figs. 2, 3).

Table 1. Mineral component of Tung Ba kaolin clay

Sample	Mineral component and percentage (%)					
	Illite	Kaolinite	Quartz	Feldspar	Goethite	Chlorite
TB1	28	26	43	2	1	0
TB2	29	27	41	2	1	0
TB3	27	24	44	3	1	1
TB4	30	24	40	3	1	2
TB5	30	26	41	2	1	0
TB6	28	25	43	2	1	1
TB7	29	27	40	3	1	1
TB8	26	25	42	4	2	1

The chemical compositions (wt%) of Tung Ba kaolin clay were SiO_2 at 59.92–64.98, Al_2O_3 : 18.95–20.82, Fe_2O_3 : 2.28–3.95, SO_3 :2.47–4.66, CaO about 0.12–0.36, MgO at 1.72–2.13 and TiO_2 at 0.75–1.08 (Table 2).

As a result, based on the mineral characteristic, kaolin clay in Tung Ba belongs to low-quality kaolin clay.

According to the Vietnamese standard TCVN 6301:1997 on enriched kaolin, comparing to chemical composition, it can be seen that kaolin clay in Tung Ba contains higher concentrations of SiO₂, Fe₂O₃, and CaO and lower contents of Al₂O₃ and TiO₂. Moreover, the improvement of the quality of Tung Ba kaolin clay is difficult because it has a high content of fine-grained quartz. Therefore, the kaolin raw material in Tung Ba

does not reach the requirement of the quality for producing traditional ceramics, porcelain, refractory materials, or papers,... However, the chemical composition of Tung Ba kaolin raw material passes all requirement of the Vietnamese standard TCVN 6301:1997 on clay for manufacturing construction ceramics. Consequently, for initial assessment, kaolin clay in Tung Ba can be used in the fabrication of building ceramic materials.

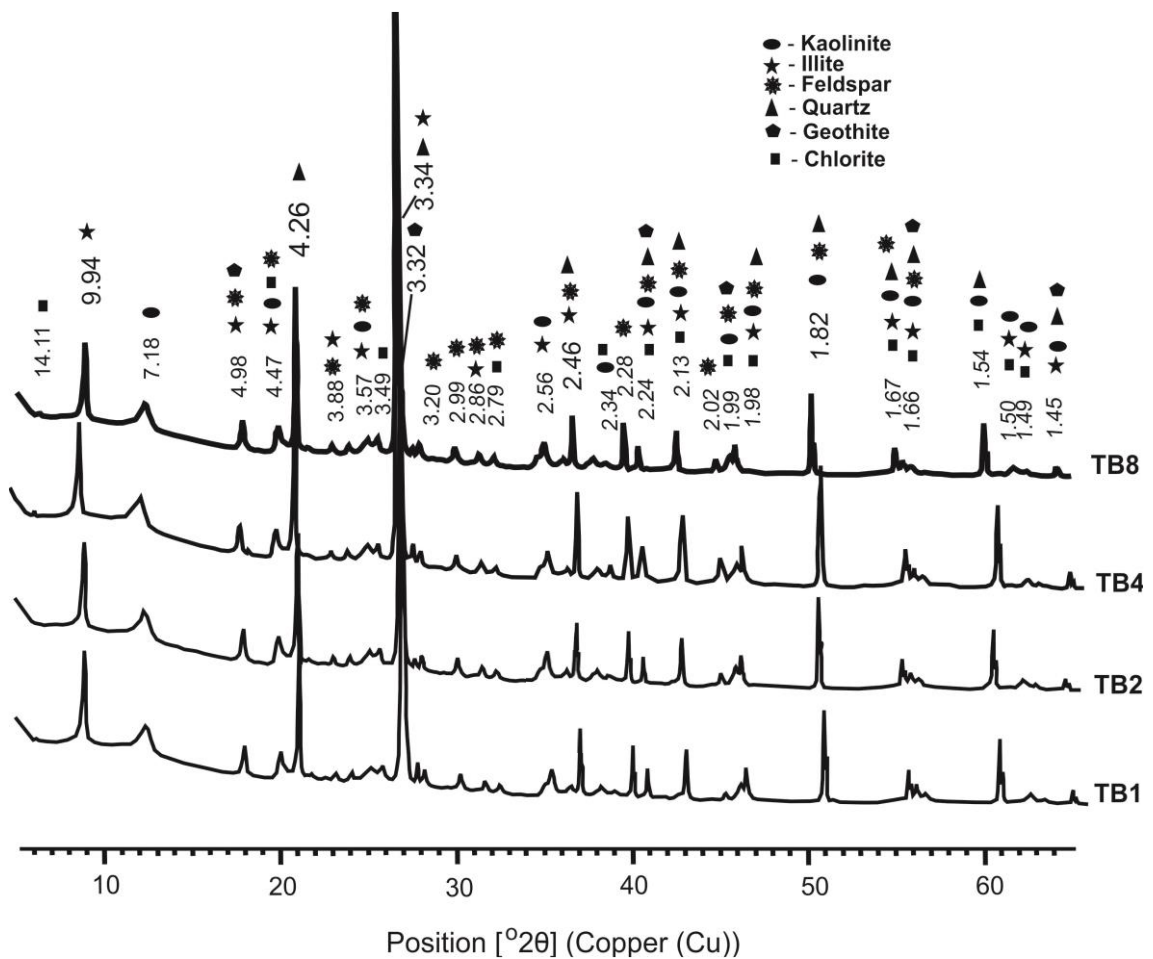


Figure 2. XRD patterns of kaolin clay in Tung Ba

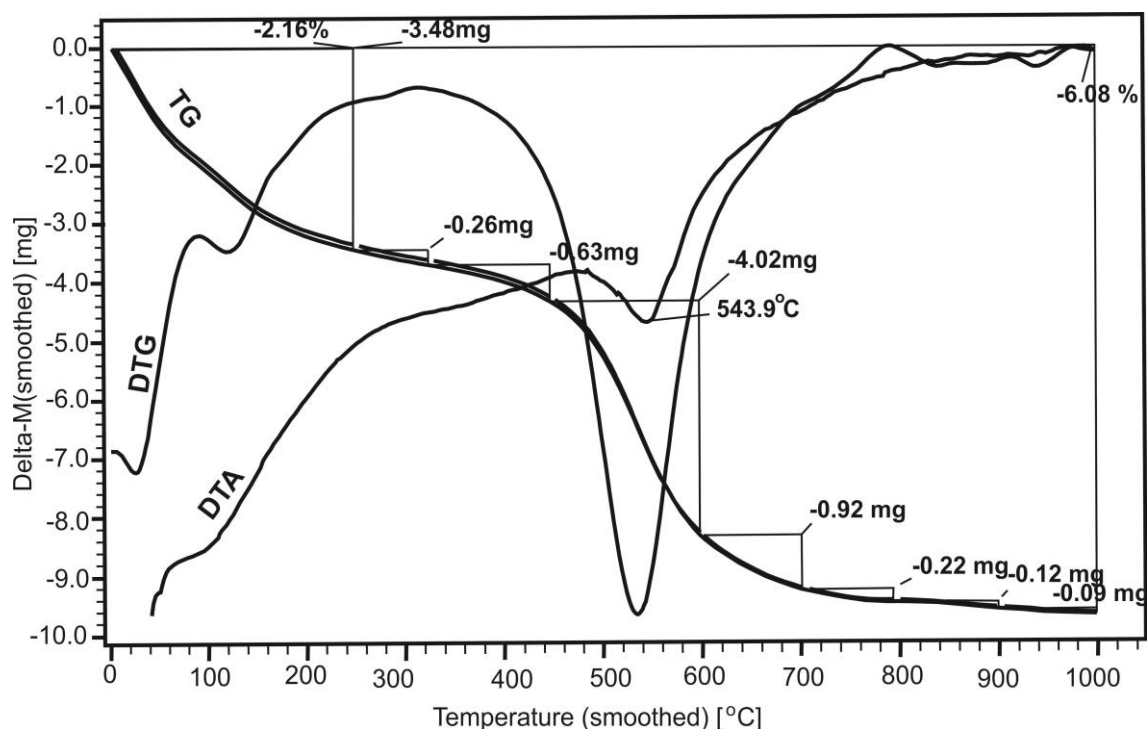


Figure 3. DTA/TG curve of kaolin clay in Tung Ba

Table 2. Chemical composition of kaolin clay in Tung Ba mine and the Vietnamese standards for chemical composition of kaolin

Sample	Chemical composition (%)							
	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	CaO	MgO	TiO ₂	SO ₃	MKN
TB1	62.26	3.95	20.48	0.12	1.72	1.08	2.47	5.67
TB2	61.66	2.50	19.04	0.36	1.94	1.02	4.66	6.22
TB3	64.98	2.28	18.95	0.27	2.13	0.95	2.51	5.61
TB4	60.26	3.15	20.82	0.25	1.89	1.05	4.54	5.67
TB5	59.92	3.67	20.74	0.19	1.82	1.07	4.03	6.03
TB6	64.76	3.32	20.6	0.17	2.02	0.98	2.93	5.6
TB7	62.55	2.90	20.35	0.25	2.07	0.75	3.36	5.64
TB8	63.84	2.46	19.86	0.21	1.90	0.96	2.57	6.16
TCVN 6301: 1997	51–53	1.0–1.7	30–33	0.7–0.9	-	1.4–10	-	-
TCVN 6300: 1997	50–70	2.0–5.0	≥19	-	-	-	<0.5	-

("-" no limit on content (mass%))

3.2. Degree of dehydroxylation of kaolin clay

Mohsen and Mostafa (2010) and Davidovits (2011) showed that kaolinite could be transformed into metakaolin to increase the pozzolanic activity and the use in the production of high-quality geopolymers. Thus, to improve the usability of Tung Ba kaolin clay, it should be heated at an

appropriate temperature and time. The degree of kaolinite transformation was evaluated by calculating the degree of dehydroxylation of kaolinite (Kakali et al., 2001; Shvarzman et al., 2002; Ilić et al., 2010; Kenne et al., 2014.)

The result of the differential thermal analysis (DTA) indicated that the endothermic peak of the dehydroxylation process with a maximum had occurred at 543.9°C (Fig. 3).

Therefore, in order to determine optimal calcination parameters for kaolinite transformation, samples of kaolin clay were heat-treated at temperatures (550°C, 600°C, 650°C, 700°C, 750°C and 800°C) and calcining times (30, 60, 120, 180 min). At different calcination temperatures and times, mass loss values of Tung Ba kaolin clay were recorded in Table 3. At 550°C, mass loss gradually increases from 3.17wt% in the heating time of 30 min to 3.75 in 180 min. At the temperatures of 600°C, 650°C, 700°C, 750°C and 800°C, mass loss also rises when heating time increases. At the same time, when the temperature climbs from 550°C to 800°C, mass loss also grows up, mass loss increases from 3.17% at 550°C to 4.85% at 800°C in 30 min and from 3.75% at 550°C to 5.17% at 800°C in 180 min.

Table 3. Mass loss (%) of Tung Ba kaolin clay at different heating temperatures and times

Heating times(min)	Heating temperatures (°C)					
	550	600	650	700	750	800
	Loss mass (%)					
30	3.17	3.46	3.55	3.77	4.41	4.85
60	3.45	3.64	3.86	3.95	4.87	4.97
120	3.56	3.97	4.05	4.16	5.17	5.15
180	3.75	4.17	4.28	4.38	5.14	5.17

Based on loss mass (M) during calcination at different temperatures and times and the loss on ignition (LOI) value achieved by XRF analysis (M_{max}) in Table 2, the degree of dehydroxylation (D_{tg}) of Tung Ba kaolin clay were calculated by the equation $D_{tg} = M/M_{max}$ (Rahier et al., 2000) and shown in Fig. 4. The lowest degree of dehydroxylation is 0.56 at 550°C for 30 min and the highest value reaches 0.91 at 750°C and 800°C for 120 min and 180 min, respectively. In term of economic factors, to transform kaolinite into metakaolinite completely, the optimal parameters for calcination at 750°C in 120 min were selected.

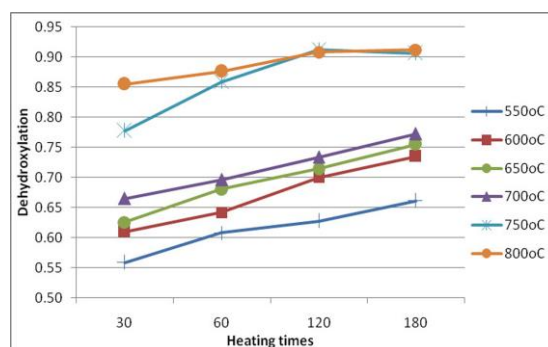


Figure 4. Dependence of the degree of dehydroxylation (D_{tg}) on calcination duration and temperatures

3.3. Metakaolin characterization and pozzolanic activity

Using the obtained results of the degree of dehydroxylation of Tung Ba kaolin clay, metakaolin was fabricated at the heating temperature of 750°C and the time of 120 min. After that, metakaolin was analyzed for mineral component, chemical composition and calculated the amount of calcium oxide for evaluation of pozzolanic activity and test production of geopolymer ceramics.

On mineralogical component, metakaolin fabricating from Tung Ba kaolin clay mainly contained quartz (60–62%), illite (32–35%), few feldspard (5–7%), (Fig. 5). On chemical composition, metakaolin was mainly composed of SiO_2 (66.72%), Al_2O_3 (21.95%), a small content of other oxides such as Fe_2O_3 (4.01%), MgO (1.74%), SO_3 (1.87%), LOI (0.41%).

The analytical result of the amount of lime reacting with metakaolin indicated that the lime absorption of metakaolin produced at 550–800°C in 180 min increased from 151.69 mgCaO/1g of sample to 157.77 mgCaO/1g of the sample. The highest value is 170.35 mgCaO/1g of the sample at 750°C in 120min (Table 4). Consequently, according to the Vietnamese standard TCVN 3735 for pozzolanic additive, metakaolin producing from Tung Ba kaolin clay belongs to the high pozzolanic activity.

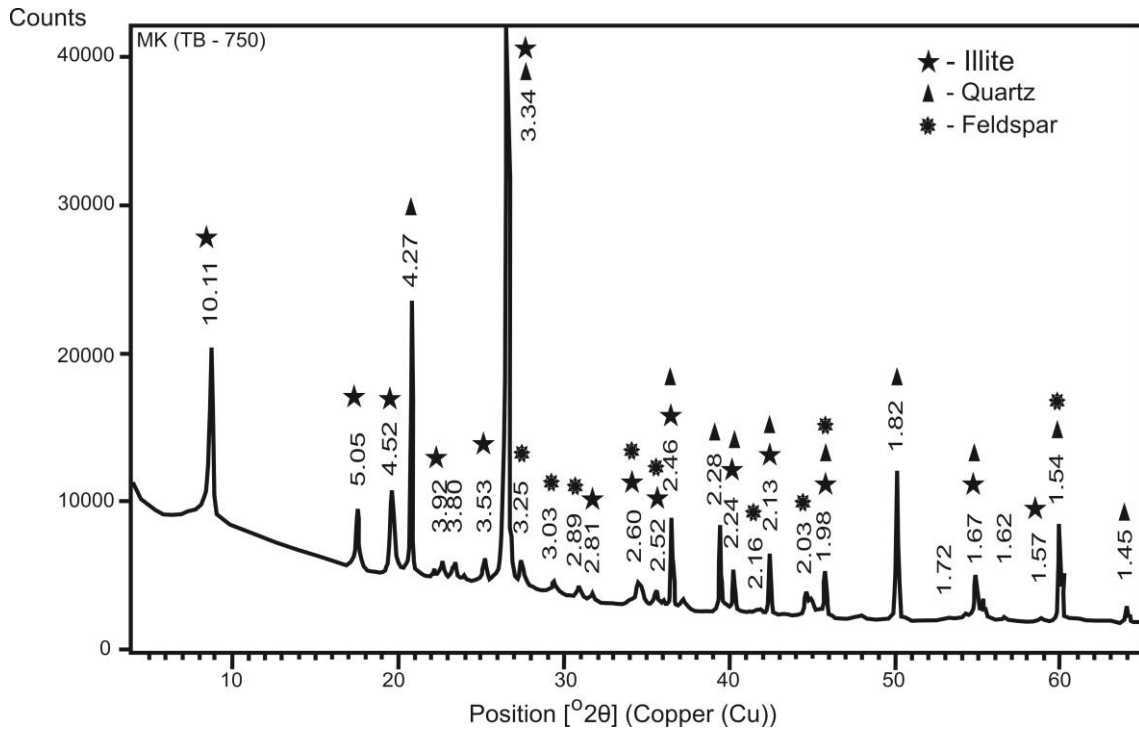


Figure 5. XRD phase of metakaolin (MK) produced from Tung Ba kaolin clay at 750°C for 120 min
 Table 4. Lime absorption of metakaolin produced at different calcination temperatures and times

Calcination Temperature (°C)/time (min)	550/180	600/180	650/180	700/180	750/120	750/180	800/180
Lime absorption (mgCaO/1g of sample)	151.69	154.07	156.25	157.77	170.35	169.21	170.05

3.4. Physical and mechanical properties of geopolymer ceramics

According to the analysis result of metakaolin characterization and its pozzolanic activity in part 3.3, selected material for fabricating geopolymer ceramics is metakaolin which was produced from Tung Ba kaolin clay at 750°C in 120 min. Testing products of geopolymer ceramic were fabricated following mixing ratios in Table 5.

Table 6 represents the physical, mechanical properties of testing geopolymer ceramics. The results show that all geopolymer ceramic samples achieve water resistance. For the NaOH concentration of 10M, the water absorption is 15.25–16.34%, the compressive strength is 40–146KG/cm², the density is 1.55–1.69 g/cm³. At NaOH 12M, the water absorption reaches 17.71–17.98%, the

compressive strength obtains 90–191KG/cm², the density is 1.54–1.60 g/cm³. With the 14M of NaOH solution, the water absorption achieves 17.48–17.92%, the compressive strength is 105–196 KG/cm², the density is 1.54–1.63 g/cm³. The water absorption of geopolymer ceramic samples increases when the NaOH concentration rises. At NaOH 12M and 14M, the values of water absorption are quite high. The compressive strength grows up with the increase of NaOH concentration and ratio of NaOH/Na₂SiO₃ (Fig. 7). The lowest compressive strength is 40 KG/cm² for sample G27 at NaOH 10M, NaOH/Na₂SiO₃ ratio of 0.2. The highest compressive strength is 196 KG/cm² for sample G32 when the NaOH concentration is 14M and the ratio of NaOH/Na₂SiO₃ is 0.5. In addition, for two samples G28 and G32 which is one of the highest values of compressive strength with

191 and 196 KG/cm² respectively, the analysis of flexural strength were performed and the obtained result is 7.84N/mm² (sample G28) and 8.96N/mm² (sample G32).

Table 5. Mixing ratios of Tung Ba metakaolin with NaOH, Na₂SiO₃

Testing sample	NaOH(M)	Mixing ratios (g)				Mol scales		
		Metakaolin	NaOH	Na ₂ SiO ₃	Water	Na ₂ O/SiO ₂	SiO ₂ /Al ₂ O ₃	H ₂ O/Na ₂ O
G24	10	150	20	40	4	0.2834	3.3788	19.2901
G25	10	150	15	45	4	0.2463	3.5512	20.2376
G26	10	150	12	48	4	0.2257	3.6547	19.5079
G27	10	150	10	50	4	0.2127	3.7236	21.3831
G28	12	150	20	40	4	0.3074	3.3788	17.5125
G29	12	150	15	45	4	0.2635	3.5512	18.6958
G30	12	150	12	48	4	0.2391	3.6547	18.2279
G31	12	150	10	50	4	0.2236	3.7236	20.1721
G32	14	150	20	40	4	0.3295	3.3788	16.1069
G33	14	150	15	45	4	0.2792	3.5512	17.4453
G34	14	150	12	48	4	0.2513	3.6547	17.1708
G35	14	150	10	50	4	0.2336	3.7236	19.1584

Table 6. Physical and mechanical properties of geopolymer ceramics

Testing sample	NaOH (M)	NaOH/ Na ₂ SiO ₃ ratio	Water absorption (%)	Compressive strength (KG/cm ²)	Density (g/cm ³)
G24	10	0.5	15.25	146	1.65
G25	10	0.33	15.40	87	1.69
G26	10	0.25	16.17	55	1.56
G27	10	0.2	16.34	40	1.55
G28	12	0.5	17.71	191	1.60
G29	12	0.33	17.95	109	1.58
G30	12	0.25	17.96	98	1.57
G31	12	0.2	17.98	90	1.54
G32	14	0.5	17.92	196	1.63
G33	14	0.33	17.87	112	1.54
G34	14	0.25	17.54	108	1.58
G35	14	0.2	17.48	105	1.60
TCVN 2118–1994 for calcium silicate bricks– Technical requirements			6–18	75–150	≤1.65

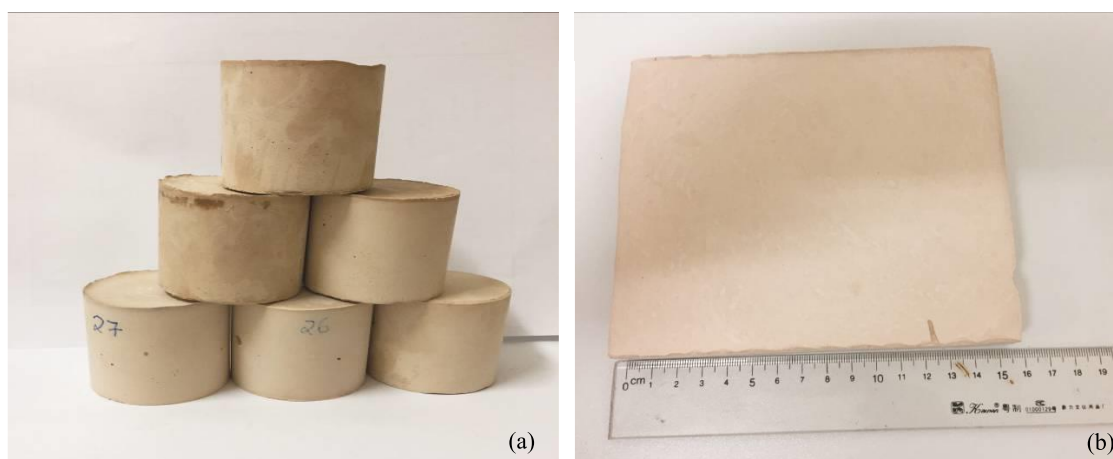


Figure 6. Testing products of geopolymer ceramics

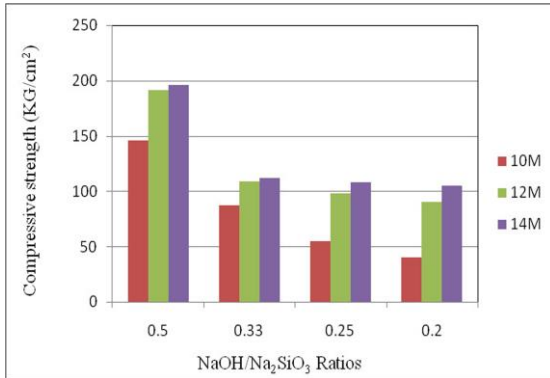


Figure 7. Compressive strength of geopolymer ceramic samples at NaOH 10M, 12M and 14M, at different NaOH/Na₂SiO₃ ratios

4. Discussions

According to Davidovits (2011), to produce geopolymers, raw materials should have high pozzolanic activity, that means they contain a large amount of amorphous SiO₂ and Al₂O₃ content. As a result, for the raw material as kaolin clay, it should be heat

treated to transform into metakaolin which has a strong pozzolanic activity. The calcination process (the dehydroxylation process) makes the release of water in the structure of kaolinite (Al₂O₃.2SiO₂.2H₂O) and destroys its mineral structure, resulting in metakaolinite – an amorphous aluminosilicate (Al₂O₃.2SiO₂). This can be shown in the following equation: Al₂O₃.2SiO₂.2H₂O → Al₂O₃.2SiO₂ + 2H₂O[†] (Ilić et al., 2010).

The transformation of kaolinite in Tung Ba kaolin clay into metakaolin was illustrated by XRD phases (Fig. 8). In the XRD phase of raw material kaolin clay, we can see characteristic peaks clearly (d = 7.18, 12.1 °2θ; d = 4.47, 20.°2θ; d = 2.46, 36 °2θ) while in the XRD phase of metakaolin, these peaks (d = 7.18, 12.1 °2θ; d = 4.47, 20.°2θ) disappear. The peak of d = 2.46, 36 °2θ in the XRD phase of metakaolin still be exist but it coincides with the characteristic peak for quartz.

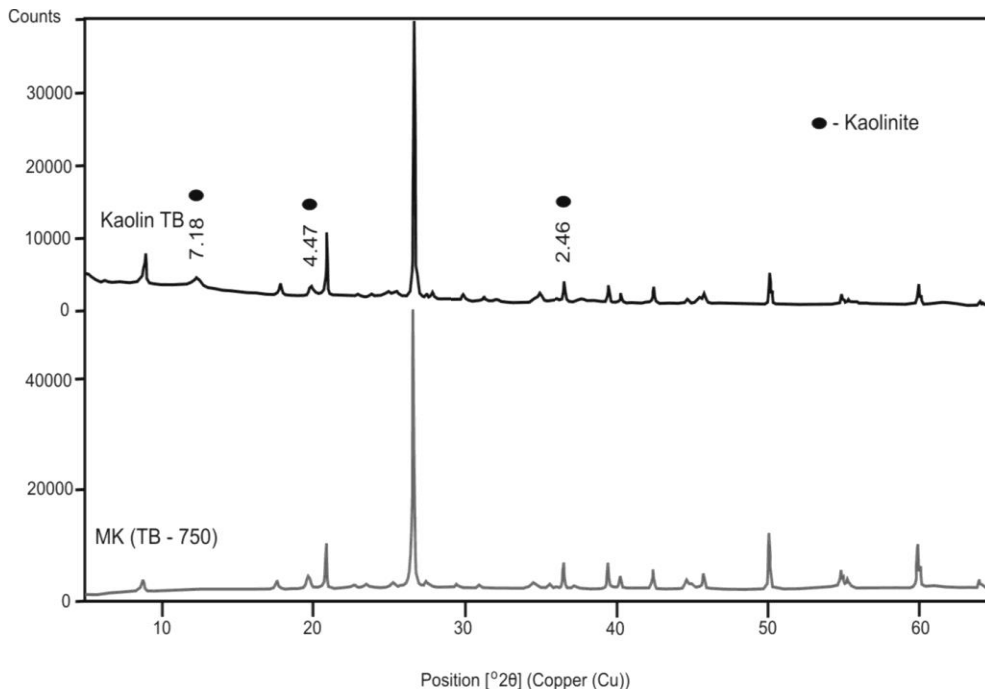
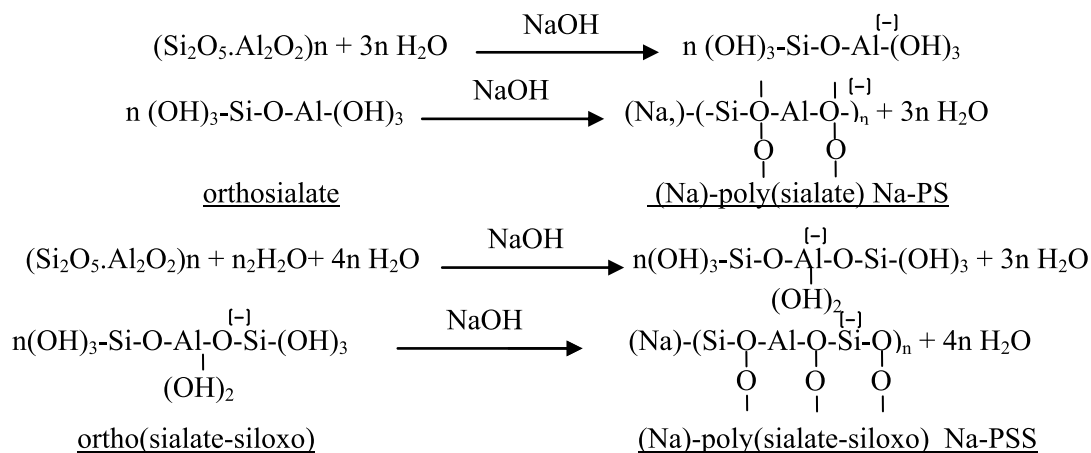


Figure 8. Comparison of XRD phases of Tung Ba kaolin clay (Kaolin TB) and metakaolin MK (TB-750)

Moreover, the destroy of kaolinite structure in Tung Ba kaolin clay to transform into metakaolin was also presented in Fourier transform infrared (FTIR) spectra (Fig. 10). The FTIR spectra of Tung Ba kaolin clay show that the characteristic bands of kaolinite at 10031.43 cm^{-1} and 1113.28 cm^{-1} corresponded to the symmetrical vibration of the Si-O bond, at 1007.32 cm^{-1} expressed for the Si(Al)-O asymmetrical vibration, at 911.94 cm^{-1} assigned to the 6 fold coordinated Al(VI) -OH stretching vibration, at 694.20 cm^{-1} and 469.35 cm^{-1} respectively corresponded to symmetrically stretching vibration and in-plane bending vibration of Si-O bonds. The wavenumber position at 796.22 cm^{-1} indicated the 4 coordinated Al(IV)-O stretching vibration and at 535.80 cm^{-1} illustrated for the Si-O-Al bending vibration. The comparison of the FTIR spectra of kaolin

and metakaolin indicated the destroy of Si-O, Al-OH, Al-O, and Si-O-Al bonds of SiO_4 and AlO_4 tetrahedron of kaolinite structure, replaced by Si=O and Al=O bonds of metakaolin. Evidence for the complete transformation is the disappearance of the Si-O bonds at 1113.28 cm^{-1} , 10031.43 cm^{-1} , 469.35 cm^{-1} , Si(Al)-O, Al-OH and Si-O-Al bonds at 1007.32 cm^{-1} , 911.94 cm^{-1} , 535.80 cm^{-1} respectively, and instead of the bands at 1039.81 cm^{-1} , 1086.10 cm^{-1} , 694.20 cm^{-1} , 478.09 cm^{-1} of the Si-O bond, at 796.83 cm^{-1} of the Al-O bond in amorphous SiO_2 and Al_2O_3 .

When metakaolin was mixed with solution of alkaline activators of NaOH and Na_2SiO_3 , the polymerization occurred to create geopolymer product with polymeric Si-O-Al-O bonds as the following equations (Davidovits, 1991; 2011):



The chemical reaction of geopolymer production from Tung Ba kaolin clay is illustrated by the comparison of XRD phases of NaOH, Na_2SiO_3 , metakaolin and geopolymer. Although the mixed content of NaOH/ Na_2SiO_3 was 40%, it can not be seen the characteristic peaks of NaOH. This proved that NaOH was completely reacted to metakaolin during the formation of polymeric chain (Fig. 9). Moreover, according to Davidovits (2011), the formation of

geopolymer structure in form of Na-poly(sialate) Na-PS type was shown in FTIR spectra. To recognize this, it can be based on the transmittance bands at $1080\text{--}1100\text{ cm}^{-1}$ of kaolinite and metakaolinite. However, after the geopolymerization, this band shifts toward the lower wave number of (Na-PS) geopolymer about approximately $80\text{--}90\text{ cm}^{-1}$. The same shift also can be clearly observed when we compare the FTIR spectra of geopolymer G32 and metakaolin (MK)

produced from Tung Ba kaolin clay. In the FTIR spectrum of metakaolin, the characteristic bands at 1039.81 cm^{-1} and 1086.10 cm^{-1} moved toward the lower wavenumber (1023.87 cm^{-1}) of the FTIR spectrum of geopolymer (Fig. 10). Once again, this confirms the formation of geopolymer from metakaolin which was fabricated from Tung Ba kaolin clay. At different mixing ratios of metakaolin and NaOH, Na_2SiO_3 , almost testing samples of geopolymer ceramics achieved the requirement of the Vietnamese standard TCVN: 2118-1994 and the American standard ASTM-C62 for the physical, mechanical properties of building bricks. The water absorption of geopolymer ceramic products is less than 18%. Most of the density is smaller than 1.65 g/cm^3 and satisfy the requirement of the standard, except for the sample G25 ($d=1.69\text{ g/cm}^3$) which is negligible higher than the standard. The compressive strength of G25 (40 KG/cm^2) and G26 (50 KG/cm^2) are lower than that of the standard because the

content and concentration of mixing NaOH solution are low. The remaining samples of geopolymer ceramics had the compressive strength of $75\text{--}196\text{ KG/cm}^2$, all of them meet the requirement of the standards. In addition, comparing with the compressive strength of geopolymer products fabricating from fly ash ($140\text{--}170\text{ KG/cm}^2$) (Kieu Quy Nam et al., 2010), red basaltic soils (95 KG/cm^2) (Nguyen Anh Duong et al., 2014) or clay soils at 85°C of Davidovits ($61\text{--}184\text{ KG/cm}^2$), the compressive strength of geopolymer ceramics made from metakaolin are similar. Comparison of the values of the flexural strength of two samples G28 and G32 with the Vietnamese standard TCVN:6414-1998 ($> 2.2\text{ N/mm}^2$) for ceramic bricks, it can be seen that their flexural strength are higher than 2.2 N/mm^2 so they achieved the standard for the production of ceramic bricks. According to Davidovits, the flexural strength of these samples satisfy the requirement of the flexural strength of ceramics ($7\text{--}10\text{ N/mm}^2$) applying for decoration.

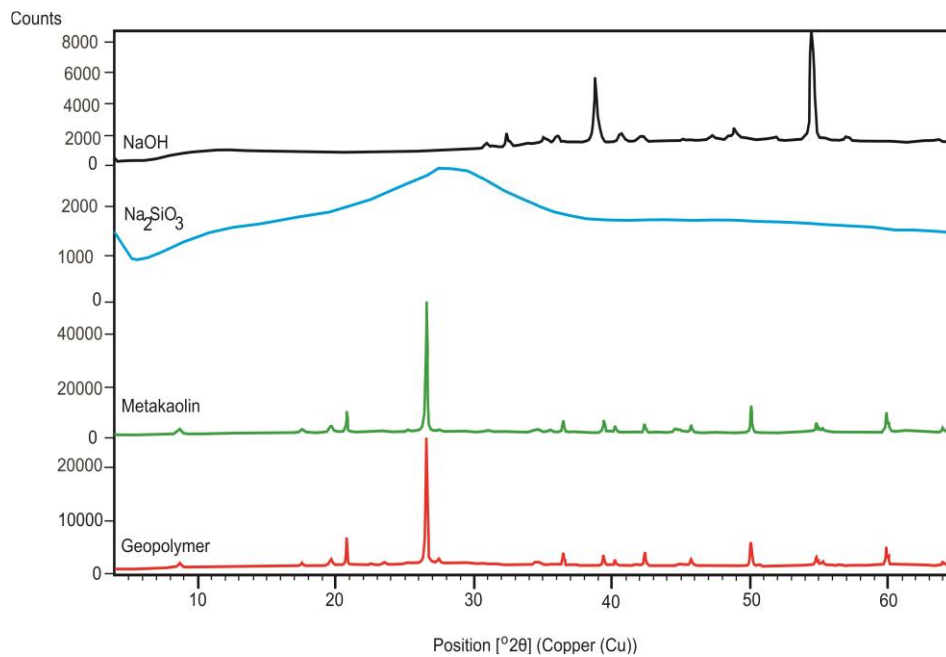


Figure 9. XRD phases of NaOH, Na_2SiO_3 , metakaolin and geopolymer

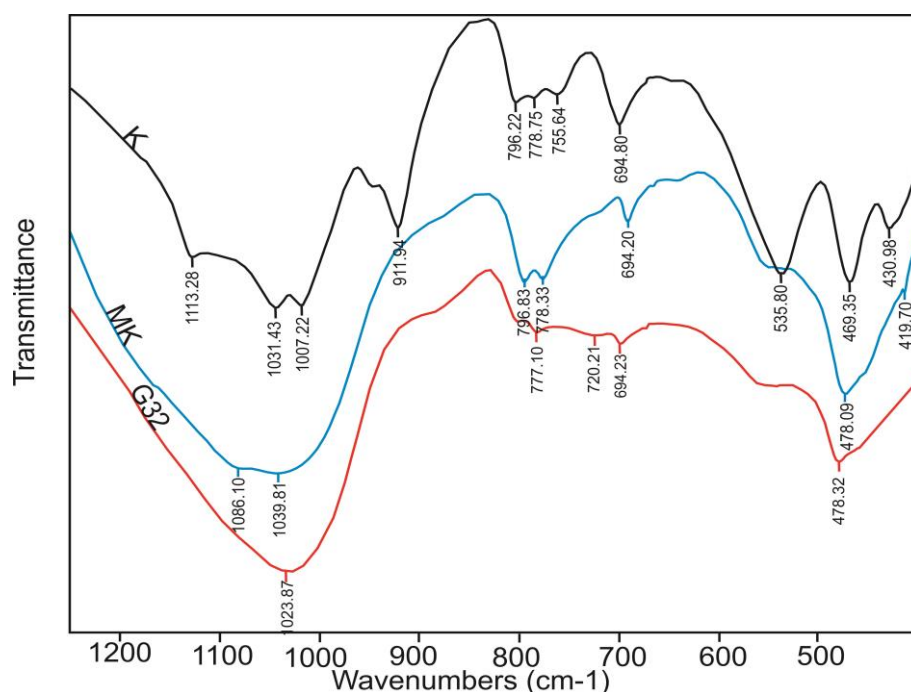


Figure 10. FTIR spectra of Tung Ba kaolin clay (K), metakaolin (MK) and geopolymer

As the result, based on the comparison of the physical, mechanical properties of test samples of geopolymer ceramics with different standards, to produce high quality geopolymer ceramics, metakaolin fabricating from Tung Ba kaolin clay should be mixed with NaOH/Na₂SiO₃ (40%) and water (2.6%) in which the optimal ratio of NaOH/Na₂SiO₃ is 0.5 and the concentration of NaOH solution is 12M or 14M. In general, the products of geopolymer ceramics fabricating from this material can be used in the production of building bricks or ceramic bricks.

5. Conclusions

From the research results on the fabrication of non-calcined geopolymer ceramics from Tung Ba kaolin clay, we can give the conclusion as follow:

Tung Ba kaolin clay belongs to the low quality-kaolin and does not meet the requirement of the Vietnamese standard TCVN: 6301-1997 on enriched kaolin. Therefore, Tung Ba kaolin clay cannot be

used for producing traditional ceramic and porcelain. However, this raw material can be used in geopolymer production.

To produce non-calcined geopolymer ceramics from Tung Ba kaolin clay, it should be the transformation of Tung Ba kaolin clay into metakaolin. The optimal calcination parameter is at the temperature of 750°C and in the time of 120 min. The obtained metakaolin had a high pozzolanic activity with the lime absorption of 170,35 mgCaO/1g.sample.

The testing products of geopolymer ceramics achieved the highest quality at the mixing ratios of metakaolin and NaOH/Na₂SiO₃ (40%), water content (2,6%), in which the optimal ratio of the NaOH/Na₂SiO₃ solution was 0.5 and the NaOH concentration of 12M or 14M. At these ratios, the compressive strength of the products was quite high and obtained the values of 191 KG/cm² and 196 KG/cm², respectively.

Indeed, for all of the low quality-kaolin clay, it can be used in non-calcined geopolymer materials. However, it is necessary to perform testing products in the scale of a pilot and after that, apply and expand for the practical production, thereby effectively using locally available, low-quality raw materials.

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