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Quality Improvement of Lampang Clay for Porcelain Bodies

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ABSTRACT Lampang clay from Kao Pangka, Thailand was graded into three fractions: particles sizes < 63 microns, < 40 microns, and < 10 microns. Chemical composition, mineral composition, plasticity of moist clay, and rheological properties of clay slip were determined. After firing to various temperatures, fired shrinkage, bending strength, and whiteness were measured. Lampang clay with particles less than 40 microns has the best working quality and was chosen to produce a porcelain body that matured at 1370 °C in a 2 % CO reduction atmosphere with an overall firing time of 4 hours in an industrial porcelain tunnel kiln. The mineralogy, chemical composition, and important physical properties of various porcelain body samples were measured. The best porcelain body was composed of 40 % Lampang clay of < 40 microns, 40 % ground Lampang stone, and 20 % kaolin.

KEYWORDS: Lampang clay, porcelain body, particle size, mineralogy, chemical composition, physical properties.

INTRODUCTION

Lampang, a province in northern Thailand, has many important ceramic raw materials.[1-4] These include deposits of white clay, pottery stone, plastic clay, limestone, and feldspar. These raw materials are mined in different quarries and have been used for many years for the manufacture of ceramic products. The area of Kao Pangka, where there are two mines, is especially interesting. In one mine of approximately 1.5 square kilometers, approximately 325,000 tons of materials have been mined between 1974 and 1995. The second mine of about 0.75 square kilometer began mining in 1965. Today, ceramic industries in Thailand are expanding rapidly. Lampang had approximately 500 factories in 1997. Most factories produce tableware and giftware. Lampang white clay and Lampang pottery stone are increasingly used both in Thailand and in other countries. Knowledge about Lampang white clay and Lampang pottery stone is not sufficient at this time. Especially lacking is knowledge of how properties of these materials are related to grain size. This research work was an attempt to study this grain size problem and to generate some ideas about how to improve clay technology currently being used in Lampang.

MATERIALS AND METHODS

The Lampang clay samples were refined using a size grading method. The materials' mineralogical properties were examined by different methods, as follows : chemical composition by X-ray fluorescence (XRF, Rikagu-Miniflex); atomic absorption spectrophotometry (Shimadzu AA-640-13) and spectrophotometry (Lambda 19). Mineral composition of a ceramic material was determined using X-ray diffraction (XRD). In the present work, semi-quantitative mineralogical phase analysis was performed using a X-ray diffractometer, Phillips, PW 3710. Plasticity behavior as flow stress and maximum deformation, which depends on the moisture content, were examined using the TIRA tester, series 2420. Rheological behavior can be obtained as viscosity and thixotropy. Rheological information, as viscosity and shear stress curves, was obtained after optimal deflocculating with Na_2CO_3. Shear rate and the corresponding changes in other properties were obtained with a rotation-viscometer, VT-550, in accordance with German Standard DIN-53019.[5]

After various firing temperatures in a heated SiC electric kiln, with a heating rate of 5 K/minute and a 30 minute soaking time at the final temperature, shrinkage study followed "total shrinkage" in accordance with German Standard DIN-51066[6] and

German Standard DIN-51045 part 2.[7] Strength study followed "bending strength" in accordance with German Standard DIN EN 100[8] and German Standard DIN-52292[9] using a TIRA tester 2420. Density was measured in accordance with German Standard DIN ISO 5018[10]. Porosity was studied by water adsorption and open porosity was studied in accordance with German Standard, DIN-51056[11]. Whiteness is reported in comparison to a $BaSO_4$ plate with a 464 nm filter. Pressed test pieces with an area of 15 mm^2 were made in an electric kiln using different final temperatures and in a tunnel kiln at a porcelain factory at a temperature of 1370°C. Measurements of whiteness were made with a Lambda 19 spectrophotometer.

Lampang clay after size grading to less than 40 microns was used for making a porcelain body by mixing it with feldspar and MEKA kaolin from Germany and firing at 1370 °C in a 2 % CO reduction atmosphere. Two formulae were used :

Sample 1: 50 % Lampang clay at < 40 microns size, 40 % feldspar at < 100 microns size, and 10 % MEKA kaolin

Sample 2: 40 % Lampang clay at < 40 microns size, 40 % feldspar at < 100 microns size, and 20 % MEKA kaolin

All standard techniques were done as decribed by Ryan.[12]

Results and Discussion

The relationships between particle size and various specific properties of the raw and fired materials were determined.

Sample selection

For this research, representative white clay and pottery stone from Kao Pangka in Lampang were selected from the main production centers of white clay and pottery stone. A representative sample was taken and thoroughly evaluated. This sample was graded in size ranges of < 63 microns, < 40 microns, and < 10 microns, so that any differences due to particle size, including ceramic-technological behavior, could be evaluated.

Sample preparation

Wet sieve screens and an air cyclone were used to make size fractions.[13] The fractions < 63- and < 40-micron were obtained using wet sieve screens. The < 10-micron fraction was obtained using an air cyclone. After separation, the properties of the dry clay were measured. The results of size sorting are shown in Table 1.

About 70 % of the clay has particle sizes < 40 microns. The < 40-micron fraction is of special interest because of its desirable industrial properties.

Chemical composition

Chemical composition is important because of its effect on ceramic behavior. The chemical composition of the three size fractions of Lampang clay was determined by X-ray fluorescence and atomic absorption spectrophotometry and is shown in Table 2 and Figure 1.

The < 63-micron fraction is comparable in composition to that of the raw material. The < 40-microns fraction has less SiO_2 than the original

Table 1. Size sorting Lampang white clay.

63-micron sieve		40-micron sieve		10 microns air separated	
Retained	**Passed**	**Retained**	**Passed**	**Retained**	**Passed**
+ 63 15 %	-63 85 %	+ 40 30 %	- 40 70 %	+ 10 63 %	- 10 37 %
100 %		**100 %**		**100 %**	

Table 2. Chemical composition of Lampang white clay, in percentage.

	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	LOI	Total
U, raw mat.	76.62	15.30	0.60	0.06	0.06	0.14	4.20	0.67	2.75	100.0
C, < 63 µm	75.96	15.46	0.55	0.08	0.04	0.11	4.36	0.54	2.90	100.0
M, < 40 µm	69.52	19.69	0.69	0.11	0.07	0.16	5.34	0.79	3.63	100.0
F, < 10 µm	52.01	31.34	1.09	0.11	0.06	0.27	8.23	1.02	5.87	100.0

LOI: Loss on ignition

materials, more Al_2O_3, and slightly more of the other oxides. The < 10-microns fraction has less SiO_2 and more Al_2O_3 than the other fractions. The amounts of Fe_2O_3, K_2O, and Na_2O in this fraction are larger than in the other two fractions. Significantly, Fe_2O_3, which is an undesirable constituent, is concentrated in the fine fraction, making it possible to improve the raw clay by merely eliminating the finest particles. The amount of SiO_2 decreased with decreasing particle size in the three fractions compared to the amount of SiO_2 in the raw material. The amount of Al_2O_3 is highest in the < 10-microns fraction.

Mineralogical composition

The third feature studied was mineral composition. Mineral composition, as determined by X-ray diffraction patterns for each size fraction, is shown in Table 3 and Figure 2. X-ray diffraction patterns of all size fractions are shown in Figures 3 - 6.

Quartz is more prominent in the size fractions > 10 microns. In the fraction that passed through the 63-microns sieve, two- and three-layer clay minerals are prominent. In contrast to kaolinite, illite increases with decreasing grain size. These differences in mineral constituents reflect the differences in chemical composition, shown in Table 2 . The small amounts of alkali salts correlate with the small amounts of feldspar, as shown in Figure 2. It appears that there has been essentially no incorporation of

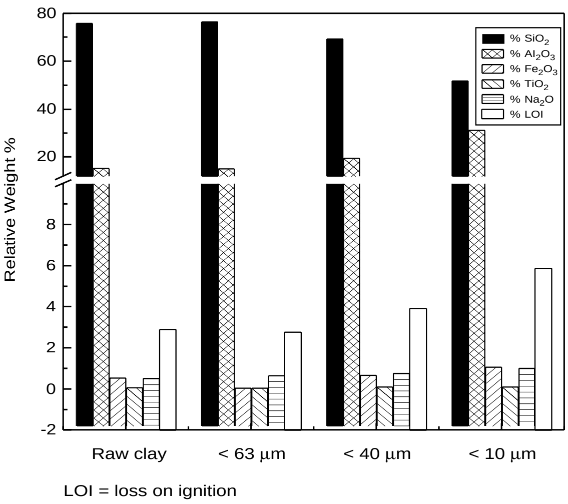

Fig 1. Chemical composition of Lampang white clay.

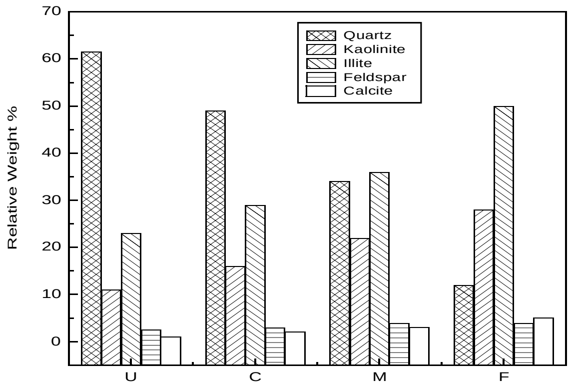

Fig 2. Mineral composition of Lampang white clay.

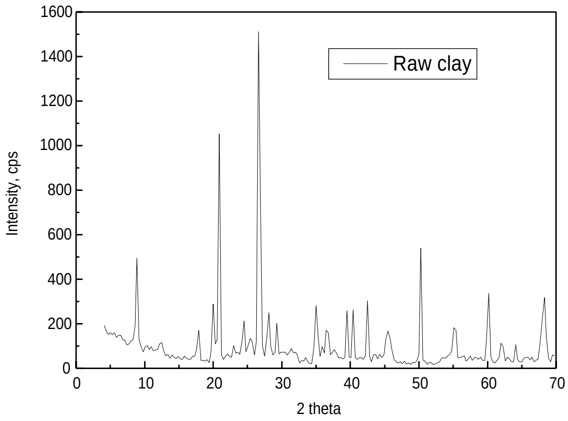

Fig 3. XRD patterns of raw Lampang clay sample.

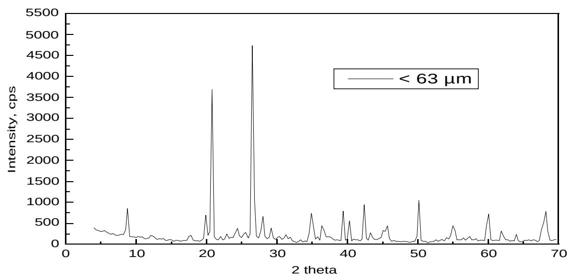

Fig 4. XRD patterns of Lampang clay, < 63 µm sample.

Table 3. Mineral composition of Lampang white clay, in percentage.

	Quartz	Feldspar	Calcite	Illite	Kaolinite	Anatase	Total
U, raw material	61.5	2.5	1.0	23.0	11.0	1.0	100.0
C, sample passing < 63 µm screen	49.0	3.0	2.0	29.0	16.0	1.0	100.0
M, sample passing < 40 µm screen	34.0	4.0	3.0	36.0	22.0	1.0	100.0
F, sample passing < 10 µm screen	12.0	4.0	5.0	50.0	28.0	1.0	100.0

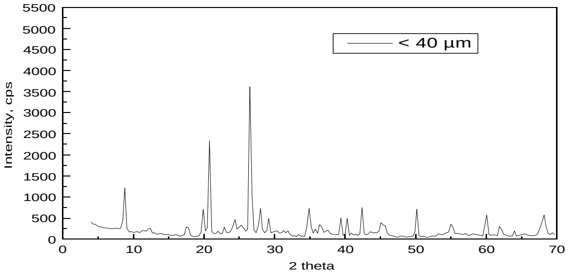

Fig 5. XRD patterns of Lampang clay, < 40 µm sample.

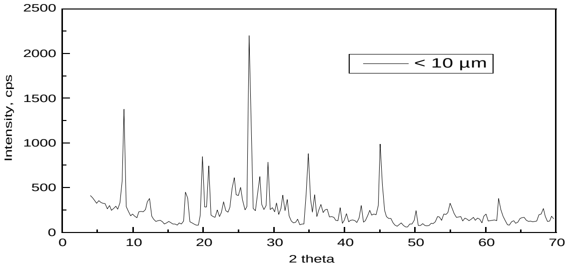

Fig 6. XRD patterns of Lampang clay, < 10 µm sample.

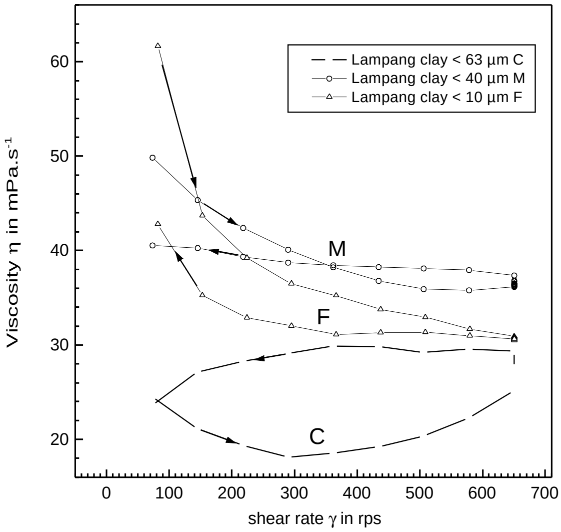

Fig 7. Viscosity curve of Lampang white clay.

alkali ions in the clay mineral lattice. The larger amount of iron ions occurs in the finer grain size fractions. This is conspicuous and probably important.

Rheological property : viscosity

An important rheological characteristic is viscosity. If viscosity is plotted against shear rate, the observed value depends on the history of the sample, that is, hysteresis is demonstrated. The appropriate viscometer has a rotary cylinder and viscosity is indicated by the extent to which the liquid material retards rotation. This retardation depends on the rotation rate of the measuring cylinder and whether the rotation rate is increasing or decreasing. The effect of an increasing rotation rate is not the same as for a decreasing rotation rate. Hysteresis loops are measured and have the largest area for samples that are the most thixotropic. However, thixotropy is not a desirable property if ceramic greenware is to be produced by casting clay slip.

All three size fractions of Lampang white clay have some degree of thixotropic behavior. For the < 63-micron fraction (Figure 7), if the applied shear rate is increased, the viscosity decreases. This is because particles form new arrangements and flow at lower viscosity. When the shear rate is high, the viscosity increases again because the course grains of the quartz retard the rotating viscometer cylinder. If the rotation is greater than 650 revolutions per second, the slurry is allowed to rest for one minute. This causes the viscosity to decrease and it is then necessary to apply reduced shear to reach an equilibrium point. With sedimentation of the quartz, the viscosity increased suddenly from approximately 25 $mPa.s^{-1}$ to 29 $mPa.s^{-1}$ and the slurry then had high thixotropy. After 650 revolutions per second, the viscosity decreased with a decrease in applied shear rate, but there was a large difference between the stirred materials and the materials that had been allowed to rest. The < 63-microns fraction has viscosity behavior that would make it difficult to control in an industrial setting. For the < 40-microns fraction, the behavior is similar, but increasing the applied shear rate to about 360 revolutions per second produced a smaller change in viscosity, that is, there was less thixotropy. This is because the < 40-microns fraction has smaller quartz particles and more clay mineral with plate structure than the < 63-microns fraction. Regarding quality control in the ceramic process, the < 40-microns fraction has a large range of shear rate that must be controlled. For the < 10- microns fraction, a different behavior was encountered. In this fraction, when the applied shear rate for the slip was increased, the viscosity first decreased rapidly and then increased as the shear rate was increased to 650 revolutions per second. The < 40-microns fraction has desirable viscosity and acceptable thixotropy for use in ceramics production.

Plastic behavior of moist clay samples

Plastic behavior is a very important technological property of clay. Plastic behavior of all the Lampang clay samples in this study is shown in Figure 8.

This figure shows that the finest grain size fraction holds more water. For the < 63-microns fraction, the moisture content is least and the sample stress changes very fast with only a small change in moisture content. The < 40-microns fraction has an intermediate moisture content but had the largest stress before breaking. The < 10-microns fraction shows a smaller change in these physical properties with changing moisture content. Figure 8 indicates that the < 40-microns fraction would be the easiest to use in the usual ceramic preparation process. The fine fraction has a higher moisture content than the other two fractions. The maximum deformation of all fractions occurred at a stress of 40 kiloPascals. The < 63-microns fraction had 0.42 maximum deformation at 19.5% moisture. The < 40-microns fraction had 0.85 maximum deformation at 28.7% moisture and the < 10-microns fraction had 0.75 maximum deformation at 41.0% moisture. The < 40-microns fraction has the highest deformation capability and is the easiest control in the usual forming processes in a ceramic factory.

Properties of the fired clay

The most important characteristic for ceramic raw materials is their behavior when fired. Firing behavior is measured in terms of shrinkage due to firing, bending strength, and whiteness after firing.

- Total shrinkage

Fired shrinkage of ceramic materials is used to establish the sintering temperature of clay materials. Shrinkage that results from firing must be known before clay can be used to advantage. The size and shape of the product after firing is strongly dependent on shrinkage in the kiln. Figure 9 shows the fired shrinkage, the shrinkage that occurs in a kiln, for all Lampang clay samples.

Figure 9 shows that all fractions have a similar pattern of shrinkage. In the firing process, with temperatures up to 1300 ˚C, all fractions shrink. The shrinkage from room temperature to 500 ˚C was small and resulted from burning out organic matter and evaporation of water. From 500 ˚C to 700 ˚C, fired shrinkage decreased, because the quartz in the samples changes phase and expanded. From 700 ˚C to 900 ˚C small increases in shrinkage occurred. From 900 ˚C up to 1200 ˚C, shrinkage greatly increased because of sintering and a conversion to a glassy phase. From 1200 ˚C to 1300 ˚C, raw Lampang clay and the < 63-microns and < 40-microns fractions continued to shrink. This means that there will be more shrinkage as the temperature is increased higher than 1300 ˚C. The sintering process was not yet finished at this temperature. For the < 10-microns fraction at 1200 ˚C, shrinkage reached a maximum and after that decreased. This happened because the sintering condition was reached at about 1200 ˚C and after 1200 ˚C the glassy phase expanded as the temperature was further increased.

- Bending strength

Another clay characteristic of interest and importance is bending strength. For industrial purposes, the fired strength must be known. Otherwise, the firing process cannot be controlled and the quality of the product will be variable. The fired strength of all Lampang clay samples is shown in Figure 10.

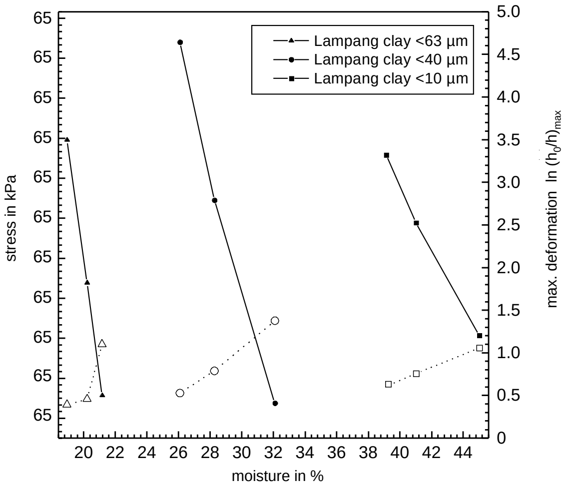

Fig 8. Stress and maximum deformation of moist Lampang clay as a function of water content. The dotted line indicates maximum deformation, the solid line indicates stress.

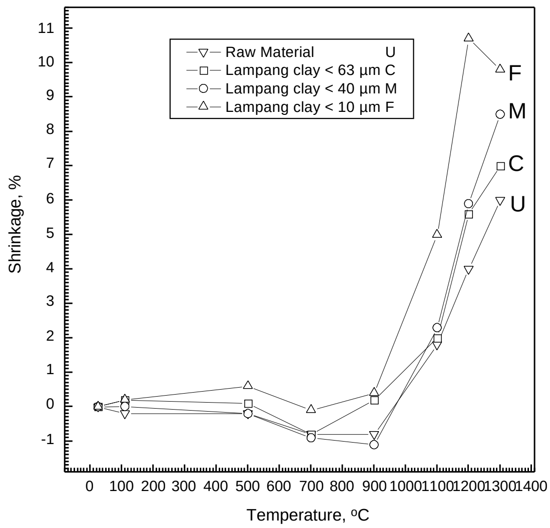

Fig 9. Fired shrinkage of Lampang clay after sieving.

Figure 10 shows that fired bending strength correlates with fired shrinkage. From room temperature to 700 ˚C, fired bending strength of all fractions remained essentially constant. From 700 ˚C to 900 ˚C, there was a small increase in fired bending strength. From 900 ˚C to 1300 ˚C, the < 63-microns and < 40-microns fractions increased in strength to about 33 N/mm^2. The fired bending strength of these two fractions can be increased further by increasing the temperature. However, the < 10-microns fraction reached its highest fired strength, about 44 N/mm^2, at 1200 ˚C. For this fraction, its strength decreases if the temperature is raised further. This behavior correlates with fired shrinkage behavior.

- Whiteness

The most important characteristic of ceramic raw materials, is the degree of whiteness after firing. This whiteness depends on the temperature reached, the atmosphere during firing, and the iron content of the clay. For raw materials that are used for white products, such as porcelain, the degree of whiteness after firing is of critical importance. Even trace amounts of iron degrades whiteness.

Figure 11 shows the whiteness of Lampang clay fractions after firing to different temperatures in an oxidizing atmosphere in an electric kiln. Whiteness achieved by firing to 1385˚C in a reducing atmosphere in a gas kiln is also shown. The whiteness of the test pieces in oxidation and reduction firings correlates well with Fe_2O_3 content. The temperature of approximately 1000˚C caused a brownish color in all fractions because of formation of Fe_3O_4. By reduction to FeO above 1200 ˚C, the whiteness increased again. Use of reduction firing above 1100 ˚C completed the FeO reduction and resulted in a 65 - 70 % improvement in whiteness.

Properties of porcelain body

Lampang clay with particle size less than 40 microns had the best working quality and was chosen to produce a porcelain body.[14]

The mineral composition and chemical composition of two potential porcelain samples are shown in Tables 4 and 5.

After firing at 1370 ˚C in a 2 % CO reducing atmosphere with an overall firing time of 4 hours in an industrial porcelain tunnel kiln, shrinkage, bending strength, bulk density, water absorption, whiteness, and phase composition were determined. Table 6 shows the values of these physical properties.

The refined < 40-microns fraction of Lampang raw clay can be used to produce a porcelain-type white ceramic product. In general, this fraction of Lampang clay can be used to produce a high value porcelain that has a dense body and more than 75 % whiteness. Porcelain bodies can be produced with more than 80 % of their constituents from Lampang. A good mixture for porcelain body has composition 40 % Lampang clay that passes a 40-microns screen, 40 % ground Lampang stone, and 20 % kaolin. This porcelain body sinters at about 1370˚C in a reducing atmosphere. The usual porcelain firing is between about 1100˚C to 1300˚C and produces fired shrinkage of about 12 %. The fired porcelain body has a real density of about 2.45 g/cm^3, bending strength of

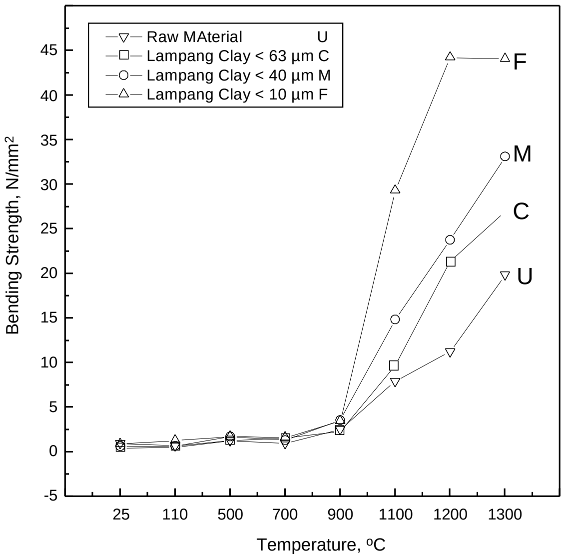

Fig 10.Bending strength of Lampang clay after sieving and firing.

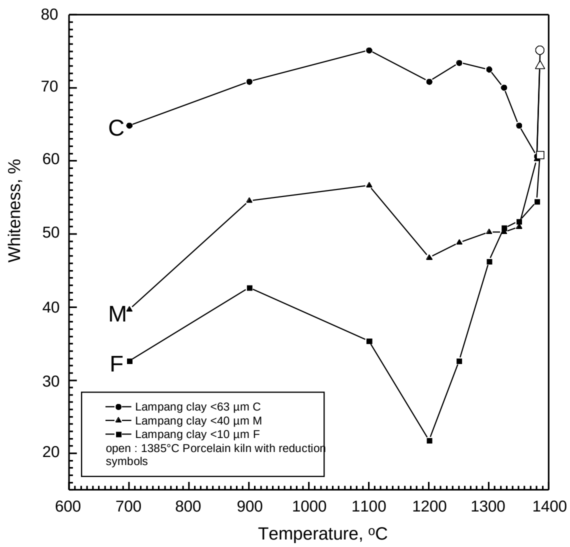

Fig 11.Whiteness of Lampang clay after firing.

more than 60 MPa, and whiteness of more than 75 % compared with a standard sample of $BaSO_4$. This porcelain sample develops a porcelain structure. The fraction of Lampang clay between 40 and 160 microns that is not used to make porcelain bodies can be used to produce floor tiles. A mixture consisting of about 30 % Lampang clay fraction between 40 and 160 microns, about 30 % ground and sieved Lampang stone, and about 40 % Maetan clay can give a product having water adsorption of less than 6 % and a bending strength higher than 25 N/mm^2 if fired to about 1200 ˚C.

CONCLUSIONS

Lampang clay is a quartz-rich clay that has kaolinite, illite, and clay minerals. Most of the quartz has a grain size larger than 10 microns. Technological properties depend on the grain size. Generally, clay minerals have smaller grain sizes than quartz. However, quartz is the larger proportion of the smallest particles. Simultaneously, the illite content increases inversely to that of kaolinite. The shift of quartz to the clay fraction corresponds to changes in Al_2O_3 content. A small increase in the feldspar fraction is correlated with a small increase in the K_2O content in the fraction with smallest grain size. Essentially no incorporation of alkali ions has taken place in the clay mineral lattice. Lampang raw clay is separated into samples according to different particle sizes. About 85 % of the raw material consists of particles with size < 63 microns. About 70 % of the material that passes the 63-microns sieve consists of particles < 40 microns in size. The remaining raw material consists of particles < 10 microns in size. The < 40-microns fraction has an optimal or near optimal behavior. Firing shrinkage, porosity, and firing bending strength seem to be largely independent of particle size. Significant differences only appeared when test pieces were fired above 1200 ˚C. Lampang clay < 40-microns fraction was chosen to produce porcelain bodies due to its mineralogic properties, technological behavior, the whiteness of the sample pieces after firing, and economic reasons. Two specific formulae for making porcelain bodies were chosen as sample 1 and sample 2. Sample 2 was whiter.

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Table 4. Mineral composition of potential porcelain formulae.

Sample number	%Quartz	%Feldspar	%Kaolinite	%Calcite	%Illite	Total
1	40	10	17	3	30	100
2	38	12	22	3	25	100

Table 5. Chemical composition of potential porcelain formulae.

Sample number	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	LOI	Total
1	70.6	19.3	0.6	0.0	0.2	0.2	4.0	1.3	3.8	100.0
2	70.1	19.9	0.5	0.1	0.2	0.2	3.3	1.2	4.5	100.0

LOI = loss on ignition

Table 6. Shrinkage, bending strength, bulk density, water absorption, whiteness and phase composition of two potential formulae in a reduction atmosphere.

Sample number	Shrinkage %	Bending strength N/mm^2	Bulk density g/cm^3	Water absorption %	Whiteness %	Phase composition	
						%Mullite	%Quartz
1	11.8	56.9	2.25	1.4	79.5	14	6
2	12.2	61.5	2.45	0.3	81.4	18	7

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