

## Analysis Of Clay Mineral Transformation In Plambik Village, Central Lombok Using X-Ray Diffraction And Scanning Electron Microscope Methods

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### ABSTRACT

Analysis of minerals and microfabric changes during the burning process of clay in Plambik village, Central Lombok has been carried out using 2 methods: X-Ray Diffraction and Scanning Electron Microscope. A brick of dimension (10 x 20 x 5) mm was prepared which was formed through a gypsum mould. After drying it is burned using an electric furnace with temperatures: 800°C, 1000°C and 1100°C. The results show that using the X-ray diffraction method when burned at 800°C there are quartz minerals with several illite peaks, at 1000°C there is only quartz, the illite peaks disappear while at 1100°C the quartz mineral peaks remain accompanied by the appearance of mullite peaks. Then the effect of the firing temperature on the clay microfabric was analyzed by observing the sample with a Scanning Electron Microscope. This test aims to obtain information about the changes in structure and texture that develop during the combustion process and more specifically on the morphology of the particles and their interaction with the development of the vitrification process and the distribution of the shape and size of the pores. The results showed that before burning, the kaolinite and illite particles were randomly distributed with porous aggregates and the grain size was between 1 – 5 microns. During firing at 800°C some parts have undergone melting and welding phases with the development of secondary porosity. Meanwhile, when burning with a temperature of 1100°C, it appears that the welding and smelting processes that occur are wider and the emergence of mullite crystals from the glass matrix.

### 1. Introduction

One of the obstacles in the development of the ceramics industry in Indonesia is the supply of raw materials whose quality cannot be maintained. The inconsistency of the mineral content of raw materials causes the quality of ceramic products to become non-standard, resulting in Indonesian ceramic products becoming less competitive. This obstacle is caused by the absence of a material refining industry and the lack of ceramics research institutes in Indonesia that have adequate research equipment. One of the materials whose mineral content has been studied is clay from the village of Plambik, located in Central Lombok Regency, West Nusa Tenggara Province. According to data obtained from the Department of Energy and Mineral Resources of West Nusa Tenggara Province, the clay content is estimated to be  $\pm 6,040$  m<sup>3</sup>. (Anon n.d.). So it is necessary to carry out research on the mineral content of the clay for further utilization which in time can be used as raw material for ceramics. Mineral analysis is carried out with the aim of providing a basis and evaluation of the possible use of these materials. Each mineral contained in the clay has a very important role in influencing the physical and chemical properties of ceramics.

The mineral analysis was carried out using an X-Ray Diffractometer and was carried out for 1 week. While the analysis of mineral transformation was carried out in the same place using a Scanning Electron Microscope. While the analysis of mineral transformation aims to obtain information about changes in structure and texture that develop during the combustion process and more specifically on the morphology of particles and their interactions with the development of the vitrification process and the distribution of shape and size of pores.

### 2. Literature review

One of the processes that must be passed from ceramic goods is the combustion process. This process is the last series and is the most important process. One that influences the process of chemical occurrence that occurs during the ceramic firing process is the mineral content of the material is the chemical reactions that occur during combustion can be classified into : chemical reactions that result in processes: dissociation, rearrangement of atoms, decomposition caused by the heat of the material,

chemical reactions caused by impurity materials, a chemical reaction caused by the presence of Oxygen and Carbon Monoxide gases in the furnace atmosphere, re-formation reactions in the form of: recrystallization and recombination, material melting reactions.(Cultrone et al. 2001; El Ouahabi et al. 2015)

### 2.1. X-Ray Diffraction

X-rays are generated by the continuous firing of a metal anode as a target by high-energy electrons from a heated filament in an X-ray tube. The resulting radiation emerges from a thin window which is usually made of beryllium and consists of: a wide map of the continuous radiation (white radiation) produced by electrons from the filament converting their kinetic energy to X-rays upon collision with atoms of the target anode, a number of discrete trajectories of varying intensity which is called the radiation characteristic of the energy released by the rearrangement of electrons in the atomic orbitals of the target anode following the throwing of one or more electrons during the excitation process. These trajectories are called shells K, L, M and so on, the type of trajectory produced is determined by the electron orbitals that play a role in filling up. The scheme that occurs is as follows: the first time the X-rays collimate to produce subparallel rays, the amount of scattering that occurs is controlled by the size of the spreading gap. The beam that spreads will directly hit the sample where the motor rotates at a certain speed in degrees per minute. When the mineral planes in the sample reach a suitable angle, the sample will diffract the X-rays according to Bragg's Law, namely:  $n\lambda = 2d \sin \theta$ . Where n is an integer,  $\lambda$  is the wavelength of the X-ray, d is the lattice gap in the  $^{\circ}$ Angstrom and  $\theta$  is the diffraction angle. The diffracted beam will pass through the receiver and collimator slits and then the scattering slits which are used to reduce the scattering of other X-rays so that the diffracted rays will reach the detector. If a monochromator is used, the light will pass directly from the receiving slit to the monochromator crystal and then to the detector. The signal generated by the X-ray photons on the detector is amplified and then reaches the recording equipment.(Ali, Chiang, and Santos 2021; Moon et al. 2021)

### 2.2. Scanning Electron Microscope

An electron beam generated from a tungsten filament is then accelerated with a potential of up to 50 kV focused by an array of electromagnetic lenses into a fine probe so that when it hits the object of observation it will have a diameter of  $\sim 10$  nm. The probe is used to drive a raster surface that is zig-zagged by two pairs of diverter coils which carry the current from the scan generator. The same current running through the coils of the cathode ray tube will produce on the screen identical objects, but with a bigger picture. Electron probes striking the specimen excite secondary electrons which may be accelerated toward a collector and the resulting signal is amplified and also used to modulate the brightness on the cathode ray tube display. The image produced in the cathode ray tube is an image of secondary electrons on the surface area of the specimen scanned by the primary beam. The magnification is determined by the relative sizes of the two rasters and this can be varied by controlling the magnification of the current flowing in the two diverter coils.

Since the electron beam will be scattered by the residual air molecules, the column is installed in a high vacuum chamber and the specimen is changed by means of an air lock system. The topographical properties of the specimen surface will control the emission of secondary electrons and likewise the arrangement described above provides images of the specimen surface similar to those observed with reflected light microscopy but with a higher potential and greater resolution, depth and field.(Di Remigio, Rocchi, and Zania 2021; Ural 2021)

## 3. Methods

### 3.1 Mineral Analysis

Preparation Stage includes tools and materials : agate mortar, sample holder, drying oven, glycol, sample under study. Ways of working include: dry the sample in the drying oven for 15 minutes at  $60^{\circ}\text{C}$ . Grind the samples with agate mortar. Place the sample that has been ground into the sample holder that has been cleaned with glycol. Tool Operation Stage is X-Ray Diffractometer Rigaku standard type D/max-III C (3.0kW). Ways of working are turn on the cooling machine and monitor and plotter devices then determine the operating parameters, namely as follows: X-ray radiation source :  $\text{CuK}\alpha$ , Operational voltage : 40KV, Operational electric current : 20 mA, Spreader gap : 0.6 mm, Receiver gap : 0.15 mm, Scatter gap : 1 mm, Scan speed :  $2^{\circ}/\text{minute}$ , recording parameter : constant time : 2, paper speed : 20 mm/minute, angle position  $2\theta$  :  $2^{\circ}$  and final position  $2\theta$  :  $60^{\circ}$ . The obtained diffractograms were analyzed for peaks using the Hanawal Standard. So that the mineral content will be obtained in each fraction.

To observe changes in minerals during combustion bricks were prepared using plaster molds with a size of (10 x 20 x 5) mm by means of plastic forming. Then the bricks are dried, after which they are fired in an electric furnace with temperatures:  $800^{\circ}\text{C}$  and  $1100^{\circ}\text{C}$ .

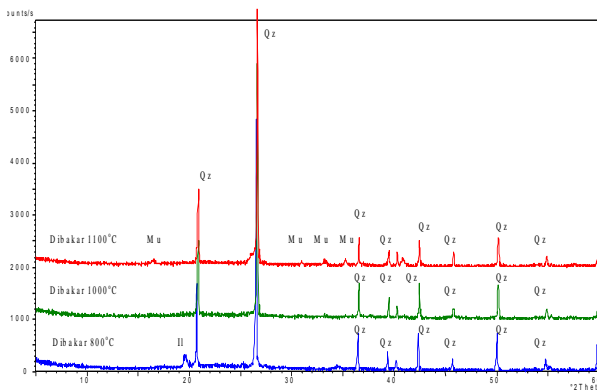
### 3.2 Microstructural Analysis

The sample to be observed has dimensions (2 x 2 x 2) mm. The sample needs to be given a coating by coating it with liquid quick dry silver paint (iso butyl methyl ketone) to make it conductive. The sample is placed in an aluminum stub which is in a sputter coater which is cleaned using alcohol (alcol ethylene denaturato). Before being turned on, the carbon rod in the sputter coater must be sharpened first and put back in an inverted position. After the sample is added, the sputter coater is turned on until the pressure reaches 3 mbar, which is preceded by turning the vacuum valve. Place the sample that has been coated with a conducting layer on the sample holder of the Scanning Electron Microscope. Then we determine the magnitude of the height and angle of observation. The cathode ray tube and monitor are turned on and all processes are computerized (digital). The next process is the digital imaging process. After determining the best image to be taken as a result of the next observation is the process of photography from the image.

When finished the image is then printed and saved. If the resulting image is not good, it is because the coating layer applied is lacking, so the sample is less conductive. The Scanning Electron Microscope used is a type of energy dispersive spectrometer from the Rigaku brand.

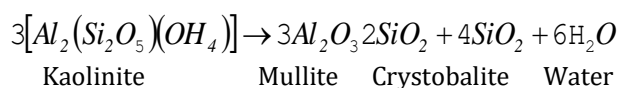
#### 4. Result and Discussion

Observations from mineral analysis of Pambik village clay with temperatures of 800°C, 1000°C and 1100°C are shown in Figure 1. And the results of observations of mineral transformation in Pambik village clay before being fired are shown in Figure 2, while Pambik village clay which has been fired at 800°C is shown in Figure 2. Figure 3 and Pambik Village clay which was fired at 1100°C is shown in Figure 4.



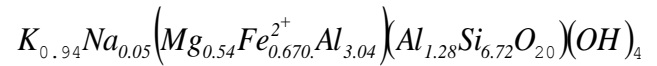
**Figure 1** Results of X-ray diffraction analysis of Pambik village clay fired at 800°C, 1000°C and 1100°C (Remarks Qz = quartz, Mu = Mullite, Il = Illite)

From Figure 1 it can be seen in diffractogram 1 where the sample was burned at 800°C, the dominant mineral is quartz, this is indicated by the many peaks of quartz minerals and there are also peaks of illite minerals which are groups of clay minerals. Whereas in the second diffractogram where the sample was burned at 1000°C the illite mineral peaks disappeared and the quartz mineral peaks were still there with the same intensity as when fired at 800°C. In the third diffractogram where the sample was burned at 1100°C, peaks of mullite mineral appeared, while the peaks of quartz mineral remained with an unchanged intensity. The formation of the mullite mineral at 1100°C comes from the kaolinite mineral which has undergone a phase change during the combustion process, chemically this can be explained by the reaction below:



From the three diffractograms, we can conclude that kaolinite clay is also a type of refractory clay because of its dominant mineral content of quartz. If kaolinite clay is used for building material products, the presence of mullite in the product indicates that the body of the product has been properly fired.

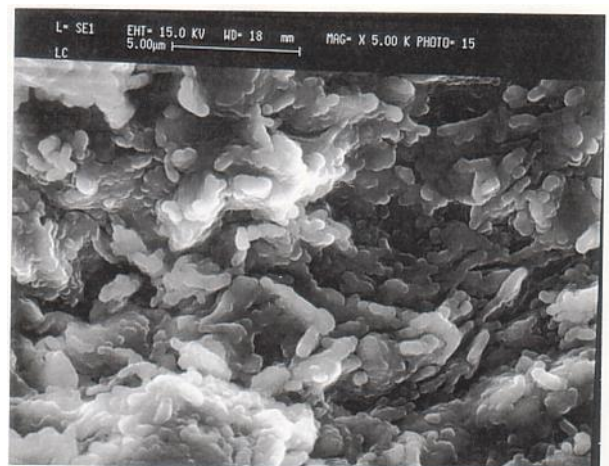
Mullite is tough, chemically resistant and its elongated crystal structure provides great mechanical strength. (Burgos-Montes et al. 2006; Mesin and Anggono n.d.) Illite minerals sometimes provide compositional variations in clay, this is due to the substitution of one ion for another. However, the amount of illite minerals contained in the clay of Pambik Village is not large and will not have a major effect on phase changes during the combustion process. Purely illite has the chemical formula:



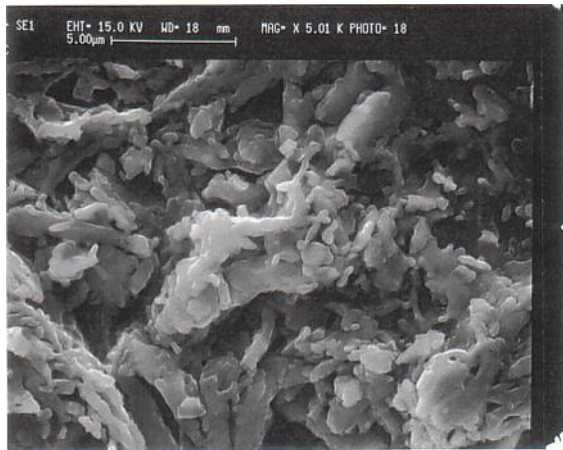
That defines the intensity of the illite peak will appear at 500°C and its intensity will continue to increase until 1150°C the illite peak will disappear or more precisely he defines the illite peak d(001) will disappear if it is burned the temperature is around 925°C while for the illite d(020) peak it will disappear if it is burned at 950°C. Meanwhile, the mullite peaks will begin to appear if the kaolinite clay is fired at 950°C and will continue to increase in intensity until the temperature is above 1300°C. (Húlan, Štubňa, and Jakubec 2019)



**Figure 2.** SEM photo of Clay in Pambik Village before firing



**Figure 3** SEM photo The clay of Pambik village is fired at 800°C



**Figure 4.** SEM Photo of Pambik Village Clay fired at 1100°C

In Figure 2, the minerals kaolinite and illite are randomly distributed with porous aggregates. The magnification applied to this observation is 499 times with a potential of 15 kV. The results of the Scanning Electron Microscope photo on clay from Pambik village which was fired at 800°C showed that a partial melting phase of the clay particles had occurred and this phase was also accompanied by the formation of secondary porosity. The magnification used in this observation is 500 times. The results of SEM photographs of clay from Pambik village which were fired at 1100°C with a magnification of 501 times showed that the melting phase was expanding and was also marked by the appearance of mullite crystals from the glass matrix. The results of microstructural analysis using a Scanning Electron Microscope strengthened the results of the X-ray diffraction analysis that the Pambik village clay is a kaolinite type clay. In practice, this clay can be used for ceramic products with high firing temperatures.

## 5. Conclusions

Based on the analysis using the X-ray diffraction method and the Scanning Electron Microscope it can be concluded as follows: the mineral content contained in the Pambik village clay is: quartz, illite and kaolinite. Where quartz is the most dominant mineral, Pambik village clay is a type of kaolinite clay which is white in color and can be applied to porcelain-type ceramic products. Mullite crystals are formed when clay from Pambik village is burned at 1100°C from the combustion of the mineral kaolinite. The presence of mullite crystals is very useful in providing mechanical strength to products produced using Pambik village clay.

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