Determination of Mineral Contents in Meukek Marble Using XRD and SEM-EDS Analysis

I. Lindawati 1 M Mursal2 and A Afdhal3
1Department of Mechanical Engineering, Polytechnic of South Aceh, Indonesia
2Department of Physics Department, Syiah Kuala University, Indonesia
3Department of Industrial Engineering Department, Polytechnic of South Aceh, Indonesia

Email: lindawati203@gmail.com

Abstract. Marble is a metamorphic rock that has been widely used in many applications. In general, it is composed of recrystallized carbonate minerals. The aim of this study was to investigate the minerals present in the marble sample from Meukek area, South Aceh. The mineral identification was conducted by using the X-Ray Diffractometer (XRD). The morphological and elementary analysis was carried out by using Scanning Electron Microscopy supported with Energy Dispersed Spectroscopy (SEM-EDS). Based on the XRD pattern data analysis, it was revealed that the marble sample from Meukek area in South Aceh mainly consists of calcium carbonate (CaCO3). The SEM-EDS analysis indicated the presence of 55.479% Oxygen (O), 0.249% Magnesium (Mg), 1.197% Aluminium (Al), 0.716% Silicon (Si), 0.202% Sulphur (S), 1.388% Potassium (K) and 40.769% Calcium (Ca) elements in Meukek marble. The highest element contents were shown by Oxygen (O) and Calcium (Ca). It confirmed that calcite is the major mineral phase composed in Meukek marble.

Keywords: meukek marble, x-ray diffraction, scanning electron microscopy and energy dispersed spectroscopy

1. Introduction
Meukek is one of areas in South Aceh that is well known for its marble deposits. The marble was primarily used for sea-wave barrier before. The abundance of marble in the area offers the potential for economic growth for the community [1]. The marble has been produced into many valuable products. The properties of marble were highly influenced by the mineral composition [2-3]. Principally, marbles are commonly composed of mineral calcite and dolomite.

However, the information related to mineral content of marble from Meukek was still questioned. Consequently, mineral content of marble from Meukek should be determined. In recent years, numerous studies have been conducted in order to identify mineral composition of marble from an area [4-12]. Marble from various areas were analysed by using X-ray diffraction (XRD). X-Ray diffraction allowed the identification of both major and minor mineralogical components [13-14]. These previous researches reported that marbles from various areas are commonly consist of calcite and dolomite.

Hence, the objective of this study is to determine mineral composed in the marble from Meukek, South Aceh. The mineral content was determined by using X-Ray Diffraction (XRD) analysis. In order to confirm the mineral content, the elements presence over the marble should be determine using Scanning Electron Microscopy and Energy Dispersed Spectroscopy (SEM-EDS) analysis. The results of this research will be useful as reference in subsequent studies.
2. Materials and methods

2.1 Sample Preparation
The marble from Meukek area in South Aceh were selected as raw materials. The samples of marbles from Meukek were collected from Marble Production Unit in Tapaktuan, South Aceh, Indonesia. The samples used for XRD and SEM-EDS analysis were in marble powder form. Marble stones were cut into small pieces using a marble stone cutter. Then, they were crushed and mashed using mortar until became powder that is ready to be analysed. Marble stone from Meukek has a grey colour as shown in Figure 1a and Figure 1b.

![Figure 1a. Marble Stone](image1a.png)  ![Figure 1b. Marble Powder](image1b.png)

2.2 Mineral Phase Identification
In order to determine the mineral content, the sample of marble should be characterized by X-ray diffraction (XRD). Mineral characterization with X-Ray diffraction (XRD) was carried out at the Materials Laboratory. X-ray Diffraction analyses was conducted by using powdered sample the Shimadzu XDR-6100. The sample was radiated with Cu radiation at the wavelength of 1.54060 Å. Besides mineral composition, XRD allowed to determine the crystal structure. The particle size can be calculated by using the following Scherrer equation [10].

$$\tau = \frac{K\lambda}{\beta \cos \theta}$$

Where $\tau$ is the average crystal size (nm), K is Scherrer constant (0.9), $\lambda$ is X-ray (Å) wavelength, $\theta$ is diffraction angle (°), and $\beta$ (FWHM) is maximum half width (radians).

2.3 Morphological Surface
Scanning Electron Microscopy (SEM) testing was carried out in order to observe the surface morphology of the marble sample. The elemental content in the sample was detected by using Energy Dispersive X-Ray Spectroscopy (EDS). The result showed in term of certain peaks that represent a number of elements contained in marble sample. The SEM-EDS analysis was performed at the Material Engineering Laboratory.

3. Results and Discussion

3.1 Mineral Phase
Basically, X-ray diffraction (XRD) has been used to detect the mineral and structure crystal contained in the material. The parameters are resulted in form of diffraction angle ($2\theta$), distance between two fields ($d$), full width at half maximum value (FWHM) and intensity ($I$). The mineral contents in marble were
determined based on the mineral diffraction peaks resulted from the relationship between diffraction angle (2θ) and intensity (I). Table 1 shows the data obtained from X-Ray Diffraction analyses on marble sample from Meukek, South Aceh. The table presents the three highest X-Ray Diffraction peaks found at 2θ = 29.4789°; 26.5781°; and 48.597°. The highest peak appeared at 2θ = 29.4789° with Intensity 1928. According to mineral database equipped in X-Ray Diffractometer, the mineral content identified at that diffraction angle are calcite (CaCO3). Hence, the major mineral phase composed in marble stone from Meukek, South Aceh are Calcite. This finding is in line with the theory that Calcite is the main minerals that form limestone or marble. Marble with large number of calcite content tends to appear in white colour. The presence of some impurities such as oxide, silicate, quartz and graphite may affect the texture and colour of marble in such area. Marble which composes of graphite will pose grey colour. Regarding marble shows a grey colour with some orange and red fibres inside, it was assumed that the Meukek marble consists of calcite and graphite minerals.

Table 1. XRD data

| No | 2θ (°) | d (Å) | FWHM (°) | Intensity (a.u.) |
|----|-------|------|----------|-----------------|
| 1. | 29.4789 | 3.02762 | 0.15680 | 1928 |
| 2. | 26.5781 | 3.35136 | 0.13790 | 501 |
| 3. | 48.5972 | 1.87197 | 0.17650 | 318 |

3.2 Crystalline Size
The size of the crystal of mineral is determined by widening full width at half maximum (FWHM) of the X-ray diffraction peaks. The crystalline sizes, which were calculated by using the Scherrer equation (1), are tabulated in Table 2. The crystalline size counted from each mineral is 10.19, 11.48 and 9.58 nm. It can be seen that the peak width is inversely proportional to the size of the crystal. The larger the size of the crystal, the smaller the width of the diffraction peak appeared and vice versa.

Table 2. Crystalline size of Composed Mineral in Marble from Meukek

| No | FWHM (°) | θ (°) | Crystalline Size (nm) |
|----|----------|------|----------------------|
| 1. | 0.15680 | 14.73 | 10.19 |
| 2. | 0.13790 | 13.29 | 11.48 |
| 3. | 0.17650 | 24.29 | 9.58 |

3.3 Morphological Surface
The presences of certain elements over marble surface were then confirmed by SEM-EDS analysis. The morphological surface of marble from Meukek area are shown in Figure 2a and Figure 2b. The morphology of the surface was obtained by capturing and processing of secondary electrons emitted by the test sample. The picture shows surface morphology resulted from 500 times and 1000 times magnification in each marble stone sample from Meukek. The greater the magnification given, the clearer the crystal formed seen. It can be seen that there are beam-shaped and angled but irregular particles. This is due to impurity factors that allow new phases to emerge. SEM analysis was carried out in line with Energy Dispersive X-Ray Spectroscopy (EDS). Characterization with EDS was conducted to determine the elemental content of a tested sample.
Figure 3 shows the results of Energy Dispersive X-Ray Spectroscopy (EDS) characterization for marble stones from Meukek. The spectrum of EDS analysis indicates the presence of some elements in marble from Meukek area. The elements present are Oxygen (O), Magnesium (Mg), Aluminium (Al), Silicon (Si), Sulphur (S), Potassium (K), and Calcium (Ca) on Meukek marble. The percentage of mass of each element is shown in table. The highest element contents were shown by Oxygen which is equal to 55.479 % and then followed by Calcium which is equal to 40.769 %.

| Element  | Mass (%) | Atom (%) |
|----------|----------|----------|
| Oxygen   | 55.479   | 75.273   |
| Magnesium| 0.249    | 0.222    |
| Aluminium| 1.197    | 0.963    |
| Silicon  | 0.716    | 0.554    |
| Sulphur  | 0.202    | 0.137    |
| Potassium| 1.388    | 0.771    |
| Calcium  | 40.769   | 22.081   |

Figure 3. EDS Spectrum for Marble from Meukek

4. Conclusion
In this study, the mineral composition of South Aceh marble was investigated. XRD analysis reveals that marble stone from Meukek has dominantly consisted of calcite (CaCO3). A grey appearance related to the presence of some impurities such as graphite. The SEM-EDS analysis indicated the presence of 55.479% Oxygen (O), 0.249% Magnesium (Mg), 1.197% Aluminium (Al), 0.716% Silicon (Si), 0.202% Sulphur (S), 1.388% Potassium (K) and 40.769% Calcium (Ca) elements in Meukek marble. The highest element contents were shown by Oxygen (O) and Calcium (Ca). It confirmed that calcite is the major mineral phase composed in Meukek marble.
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