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The relationship of physical and mineralogical properties and geochemical compositions of limestone and its implications to the quality as cement raw material in Gunem District, **Rembang Regency, Central Java**

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Abstract. The increasing development in Indonesia today will require higher portland cement consumption. Good quality cement comes from good quality raw materials. A research was conducted in the Gunem area, Rembang, Central Java which is a limestone quarry of PT Semen Indonesia. The research aims to determine physical properties of the limestone and determine the percentage of CaO and MgO and determine the quality of limestone as raw material for Portland cement. This research uses several methods, which are megascopic analysis, petrographic analysis and XRF (X-Ray Fluorescence) analysis. The results shows that wackestone has the lowest content of CaO and the highest content of MgO, while packstone and grainstone have the highest content of CaO and the lowest content of MgO. Packstone and grainstone are classified as high grade for cement raw material, while wackestone is classified as low grade for cement raw material.

1. Introduction

Limestone is one of the raw materials for the cement manufacture, apart from claystone, gypsum, silica, and iron as additives. In the current development era, cement demand has always increased in line with the pace of development throughout Indonesia [1]. Indonesia has enormous limestone potential, which is spread almost throughout the Indonesian archipelago. In general, the potential of limestone in Indonesia based on its geological map is estimated at around 28.678 billion tons [2].

One of the cement factories in Indonesia is located in Rembang, Central Java, i.e., PT Semen Indonesia Tbk. The cement factory is located in the limestone quarry, which is a member of the Paciran Formation. The Paciran Formation consists of brownish and yellowish white limestone, coral texture and has a layer thickness of ± 25 m [3].

The purpose of this research is to determine the physical characteristics and components of limestone in the limestone quarry based on megascopic and petrographic features. This research also aims to determine the chemical composition of limestone in the limestone quarry based on XRF analysis and to determine the relationship between physical and chemical properties of rock samples to determine the limestone's quality in cement manufacture.

2. Literature Review

2.1. Regional Geology

The research location, which is the limestone quarry of PT Semen Indonesia, is at Tegaldowo Village, in Gunem District, Rembang Regency. The research location is included in the Paciran Formation (Figure 1). The original Paciran Formation was referred to as Karren Limestone [4]. This formation is found only in the northern part of the Rembang Zone. This formation is characterized by coral limestone, which is grayish-white, massive, often dolomitic, consisting of algae, corals, large foraminifera, and other reef-forming organisms. The distinctive feature of this limestone is the disappear of the fossil content of Lepydocyclina, so that it can be distinguished from the limestone underneath this formation [3].

The age of the Paciran Formation is Pliocene - Pleistocene, based on the discovery of *Alveolinella quoyi* which is a fossil that often appears in the Pliocene. The depositional environment of this formation is in shallow seas, near the coast, warm climates, clear, depth <50 m, the littoral zone - sublittoral edge where the environment allows reef growths [4].



Figure 1. Regional geology map.

2.2. Limestone as Cement Raw Material

Cement is a crystalline compound of calcium silicates and other calcium compounds having hydraulic properties [5]. The raw material for artificial cement is a mixture of the following materials: limestone, clay, silica, iron or copper slag, gypsum and trass (feldspar) [6] [7].

Limestone is the main raw material in the manufacture of cement. In the exploration of limestone as a raw material for cement, it is necessary to analyze the distribution of high-quality limestone. The high and low quality of limestone as a raw material for cement is largely influenced by its CaO

content. On the other hand, limestone has different physical characteristics depending on the facies of the limestone where it found. So, it is important to study the relationship between the physical and mineralogical properties and geochemical of limestone [8].

Based on the standards of PT Semen Indonesia Rembang, limestone which meet the requirements for cement raw materials (high grade class) must have > 50% CaO and <3% MgO content [9].

3. Methods

The methods used in this study are field observations, X-Ray Fluorescence (XRF) analysis, and petrographic analysis.

- a. Field observations were carried out to determine the geological conditions of the study area, such as the type of lithology and geological cross section and to collect the rock samples to be analyzed in the laboratory.
- b. Megascopic observations were carried out to determine the physical properties of limestone.
- c. Petrographic analysis was carried out to determine the rock description optically. Thin section samples were prepared to identify mineralogical and petrographical characteristics under polarizing microscope. The rock samples from the research area were polished to make the thin sections. The thin sections were analyzed using polarization microscope. The petrographic analysis was carried out at the Laboratory of Paleontology, Remote Sensing and Optical Geology, at Geological Engineering Department, Universitas Diponegoro.
- d. The geochemical analysis is carried out by using XRF analysis. XRF analysis is carried out to analyze the main elements and trace elements along with their concentrations in the rocks deposits using spectrometric methods. Currently, XRF is the most common method of analysis used in the determination of major elements and traces elements on rock samples. Compared to other geochemical analysis methods, the XRF analysis can detect the lowest elements more sensitive. However, it cannot detect elements lighter than Na, which have an atomic number of 11. The XRF analysis was carried out at Laboratory of Quality Assurance of PT Semen Indonesia Rembang.

4. Result

4.1. Lithology

Based on the field observation in the limestone quarry, three lithologies were found in the study area: wackestone, packstone and grainstone. The distribution of the lithology units can be seen in Figure 2.

4.1.1. Wackestone

This limestone is found in the northwest part of the research area. From the observation of its physical properties, the rock has a moderate level of weathering, white color, massive structure, silt-sized grain texture, and the composition consists of bioclasts (fragments of fine foraminifera test). Based on petrographic observations, the thin section of the rock shows the texture of the rock is mud-supported, grain size <1 mm, with subangular grain shape and poorly sorted. The composition of limestone consists of skeletal grain, including foraminifera and algae (15%), the matrix is microsparite (80%) and calcite cement (5%) (Figure 3) and then the rock can be classified as wackestone [10].

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Figure 2. Geological map of research area.



Figure 3. Outcrop of wackestone (A) and its petrographic thin section (B). Note: Frm (foraminifera), Alg (algae).

4.1.2. Packstone

This limestone is found in almost all parts of the research area. From the observation of its physical properties, this limestone has a yellowish to brownish-white color, has medium to coarse sand grain size (0.25 - 1 mm), and the composition consists of bioclasts (fine shell fragments) and calcite. Based on the petrographic observation, the thin section of the rock shows grain size <1mm, subangular grain shape and poorly sorted. The composition of the limestone consists of skeletal grain of foraminifera (50%), calcite crystal (10%), the matrix is microsparites (30%) and also calcite cement (10%) (Figure 4). Based on the compositions and classification of [10], the rock can be categorized as packstone.



Figure 4. Outcrop of packstone (A) and its petrographic thin section (B). Note: Frm (foraminifera), Alg (algae), Pr (porosity), Ms (microsparite).

4.1.3. Grainstone

This limestone is found in the middle and northeast part of the research area. From the observation of its physical properties, this limestone has a low level of weathering. This limestone has a darker color than packstone and wackestone, which is yellowish-white to reddish-brown. It has a grain size of coarse sand (1-2 mm) and consists of bioclast (foraminifera test). Based on the petrographic observation, the thin section of the rock shows grain size <1mm-2 mm, subangular grain shape and poorly sorted. The composition of the limestone consists of skeletal grain of foraminifera and algae (60%), calcite crystal (20%), the matrix is microsparites (10%) and also calcite cement (10%) (Figure 5). Based on the compositions and classification of [10], the rock can be categorized as grainstone.

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Figure 5. Outcrop of grainstone (A) and its petrographic thin section (B). Note: Frm (foraminifera), Alg (algae), Pr (porosity), Ms (microsparite).

4.2. Geochemical Analysis

The geochemical analysis was carried out by using XRF analysis method to determine the major oxide compounds of limestone. XRF analysis was carried out on nine rock samples of different lithology types. The XRF analysis result shows that each lithology has a different chemical composition (Table 1). There is no LOI data available from Laboratory of Quality Assurance of PT Semen Indonesia Rembang.

Lithology	SiO ₂	Al_2O_3	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	SO_3
	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)
Wackestone	0.56	0.30	0.17	49.20	4.19	0.02	0.09	0.02
Packstone	0.80	0.48	0.24	55.80	0.50	0.02	0.04	0.01
Packstone	1.47	0.76	0.22	55.38	0.68	0.05	0.05	0.03
Packstone	0.38	0.23	0.13	55.56	1.05	0.01	0.05	0.00
Packstone	0.99	0.61	0.25	55.25	0.74	0.04	0.04	0.00
Packstone	1.11	0.67	0.30	55.14	0.68	0.04	0.05	0.00
Packstone	1.26	0.71	0.30	55.38	0.52	0.04	0.04	0.02
Grainstone	2.03	1.24	0.49	54.38	0.57	0.03	0.04	0.07
Grainstone	1.19	0.69	0.27	55.33	0.57	0.04	0.04	0.01

Table 1. Chemical composition of limestone.

Based on the XRF analysis, it shows that wackestone has a lower CaO content of 49.20% and higher MgO content (4.19%), compared to other samples (packstone and grainstone) which have Cao content> 50% and lower MgO content (0.50-1.05%).

4.3. The relationship of physical properties and chemical composistion of limestone

The wackestone has less than 50% CaO content and 4.19% MgO content. The wackestone has white color, very fine grain size and contains less fossil composition. The composition of wackestone is mostly micrite (80%), skeletal grain (15%) and calcite cement (5%). It can be concluded that the fine-grained limestone with low content of skeletal grain and calcite mineral has low content of CaO.

Packstone generally has more than 50% CaO content and 0.5-1.05 % MgO content. The limestone has a yellowish to brownish-white color, and medium to coarse sand grain size (0.25 - 1 mm). The composition of packstone is mostly skeletal grain (50%), calcite crystals (10%), micrite (30%) and calcite cement (10%). Likewise, grainstone has more than 50% CaO content and 0.57% MgO content. The physical properties of this limestone are yellowish-white to reddish-brown, and coarse sand grain size (1-2 mm). The composition of grainstone is mostly skeletal grain (60%), calcite crystals (20%), micrite (10%) and calcite cement (10%). It can be concluded that the coarse-grained limestone with high content of skeletal grain and calcite mineral has high content of CaO.

The above discussion shows that the fine-grained limestone has low CaO content and high MgO content, while the coarse limestone has high CaO content and low MgO content. This is because the fine-grained limestone contains less bioclastic and calcite minerals, while the coarse limestone contains more abundant fossil shells (bioclastic) and calcite minerals. The fossil shells and calcite minerals contain high levels of CaO.

Based on the limestone classification for cement raw material from PT Semen Indonesia Rembang, wackestone is classified as a low grade because it contains low CaO content but high MgO content, which is not suitable for cement raw material [9]. While packstone and grainstone are classified as a high grade because they contain high Cao content and low MgO content. Thus, packstone and grainstone are recommended to be used as cement raw material, while wackestone must be mixed with other limestones with high CaO content to be used as cement raw material.

In comparison, the same result also occurred in PT Indocement Tunggal Prakarsa, Palimanan, Cirebon, West Java, Indonesia. The results in this area showed that grainstone, rudstone, bafflestone and packstone (coarse-grain limestone) tend to have higher CaO content and lower MgO content, while wackestone and mudstone (fine-grain limestone) tend to have lower CaO content and higher MgO content. The high level of CaO and low level of MgO in grain-supported limestones are interpreted to occur because the coarse-grain limestone has a higher grain percentage than the matrix. The CaO content in limestone comes from limestone grains consisting of fragments of organisms (foraminifera, algae, molluscs, corals, etc.) or cement consisting of aragonite or calcite with the chemical formula CaCO₃ [8].

5. Conclusion

- 1. The physical properties of wackestone have fine grain and contains less skeletal grain and calcite mineral, while packstone and grainstone have coarse grain and contain higher skeletal grain and calcite mineral.
- 2. Wackestone has lowest content of CaO and highest content of MgO, while packstone and grainstone have highest content of CaO and lowest content of MgO.
- 3. Packstone and grainstone are classified as a high grade for cement raw material, while wackstone is classified as a low grade for cement raw material.

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