

RESEARCH ARTICLE

The Analysis of Pyrophyllite Quality as a Potential Industrial Raw Material in Argotirto Area, Sumbermanjing Wetan District, Malang Regency, East Java, Indonesia

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Abstract

The Argotirto area, Sumbermanjing Wetan District, Malang Regency, East Java is one of area which had industrial mineral, namely pyrophyllite. Pyrophyllite minerals in this area were generally found in rocks that have been altered by hydrothermal alteration. Pyrophyllite is an industrial mineral that can be used as a raw material in various industries.

This research aims to determine the geological conditions, physical properties, mineralogical and geochemical properties of pyrophyllite in the research area and to determine the quality of pyrophyllite as a potential industrial raw material. The methods used in this research were megascopic analysis, petrographic analysis, X-Ray Diffraction (XRD) analysis, and X-Ray Fluorescence (XRF), and the quality assessment of pyrophyllite using the standard of PT Gunung Bale, SNI No. 15-1023-1989, and SNI No. 15-1325-1989.

The lithology in the study area consists of basaltic lava, andesite breccia, dacitic tuff breccia, dacite intrusion, and limestone. Pyrophyllite in the study area has a quality ranging from low grade to high grade. The EIP sample is classified as high grade quality and can be used as raw materials for the refractory industry, class I refractory materials, and materials for making fine ceramics. The WSP sample is classified as medium grade quality, and can be used as class III refractory materials. The DSP sample is classified as low grade quality, and can be used as a class III refractory material.

Keywords: Pyrophyllite, Sumbermanjing Wetan, XRD, XRF, Pyrophyllite Quality

1. Introduction

Industrial minerals and rocks are the raw materials of economic value that are not classified as metallic minerals, fossil fuels or precious stones (Charalampides et al., 2014). Indonesia is a country which has a lot of industrial minerals potential (Badan Standarisasi Nasional, 2011).

The Argotirto area, Sumbermanjing Wetan District, Malang Regency, East Java is one of area which has abundant potential of industrial mineral resources and rocks. One of the industrial mineral resources is pyrophyllite (Mutrofin et al., 2006). Pyrophyllite minerals in this area were generally found in rocks that have been altered by hydrothermal alteration. Pyrophyllite in Argotirto area had a fairly high silica content reaching 85% (Anggraini, 2008). Pyrophyllite was used as raw material for the ceramic and porcelain industry, as a substitute for talc in the paper industry, and also as a substitute for concrete (Prasetya, 2012).

This study aims to determine the geological conditions, the physical properties, mineralogical and geochemical properties of pyrophyllite in the research area and to determine the quality of pyrophyllite as a potential industrial raw material based on the quality standard of PT Gunung Bale, (Badan Standarisasi Nasional, 1989)

2. Literature Review

2.1. Geological Setting

The research area is located on the geological map of the Turen sheet (Fig. 1) with stratigraphy from oldest to youngest consists of the Mandalika Formation, Tuff Members of the Mandalika Formation, and the Wonosari Formation (Sujanto et al., 1992).

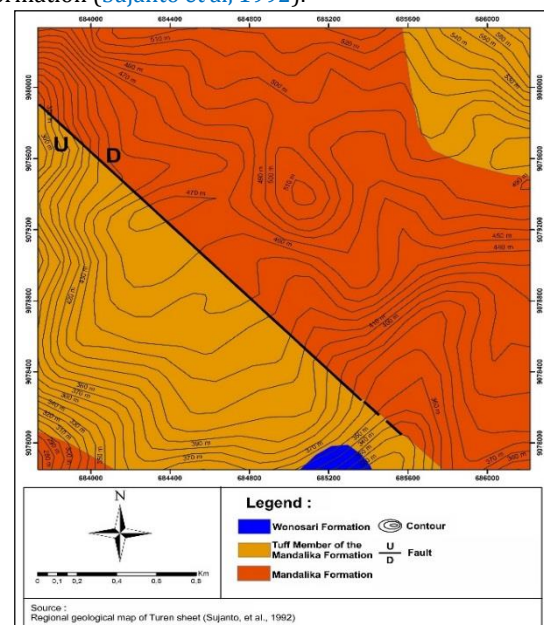


Fig 1. Regional geology map of study area

2.1.1 Mandalika Formation (Tommm)

The Mandalika Formation was composed of volcanic rocks which consisted of andesitic lava, basalt, trachyte, dacite, tuff and andesite breccia. The age of this formation was Late Oligocene – Early Early Miocene.

2.1.2 Member of the Mandalika Tuff Formation Tuff (Tomt)

The Member of Mandalika Tuff Formation consisted of rhyolitic tuff, andesite, dacitic tuff, and pumiceous tuff breccia. The age of this formation was Oligocene – Miocene.

2.1.3 Wonosari Formation (Tmw1)

The Wonosari Formation consisted of limestone, sandy marl with intercalation of claystone. Limestone consisted of sandy limestone, reef limestone, crystalline limestone, and some fossils of foraminifera. The age of this formation was Early Miocene to Middle Miocene.

2.2. Pyrophyllite

Pyrophyllite is a hydrous aluminum silicate with the chemical formula of $Al_2Si_4O_{10}(OH)_2$ and is commonly associated with other minerals such as quartz, mica, kaolinite, epidote and rutile (Ali et al., 2021). The structure of pyrophyllite consists of an octahedral Al-O layer sandwiched between two tetrahedral Si-O layers (Mackenzie, K. J. D., 2009).

Pyrophyllite were usually products of acidic volcanic rocks, such as rhyolite, dacite or metavolcanic rocks which were decomposed and/ or converted by hydrothermal or metasomatic processes, such as in Japan. Pyrophyllite also occurred from hydrothermal alteration of intermediate volcanic rocks and tuffs, such as in South Korea and in The Avalon Zone of Newfoundland, Canada (Öner and Taş, 2013) and (Arbiol et al, 2021). In Indonesia, pyrophyllites were usually formed in association with the Old Andesite Formation with Oligo – Miocene age and had a strong control of the structure and intensity of hydrothermal alteration (Heryono, 1997), (Yuwanto and Solichah, 2015).

Pyrophyllite had wide applications as a substitute for feldspar and silica due to its beneficial technical properties. The uses of pyrophyllite in industry include its application as a refractory material in the refractory industry and as a raw material in the ceramic, fiberglass, and cosmetic industries (Li et al, 2014). Pyrophyllite was also used as filler in the paper, plastic, paint, and pharmaceutical industries, as a soil conditioner in fertilizer applications and as a dusting agent in the rubber and asphalt industries (Ali et al., 2021).

The pyrophyllite in Malang Regency has not been utilized properly, although the potential is quite large (Novianto, 2012). Therefore, a study is needed to determine its characteristics in order to know its use in industry.

3. Methodology

3.1. Research Methods

There were three pyrophyllite samples and five rock samples taken from the quarry in the Sumbermanjing Wetan area. The pyrophyllite samples were then analyzed by using physical/ megascopic, petrographic, XRD and geochemical/ XRF methods, while the rock samples were analyzed using petrographic method.

Megascopic analysis was performed to examine the physical characteristics of the pyrophyllite. Petrographic analysis was carried out to determine the mineral composition of rock and pyrophyllite samples using a polarizing microscope. The determination of rocks used the classification of Travis, 1955. XRD analysis was carried out to determine the type of clay mineral in the pyrophyllite samples using the Table of Key Lines In X-Ray Powder Diffraction Patterns of Mineral (Chen, 1977).

XRF analysis was carried out to analyse the major oxides and trace elements along with their concentrations in the rock 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. It is versatile and can analyse up to 80 elements over a wide range of sensitivities, detecting concentrations from 100% down to a few parts per million (Rollinson, 2014). XRF analysis was carried out to determine the chemical content in rock samples, especially SiO_2 , Al_2O_3 , Fe_2O_3 , TiO_2 , CaO, MgO, K_2O , and Na_2O .

3.2. Pyrophyllite Specifications in Industry

Analysis of pyrophyllite quality as an industrial raw material refers to the quality standard of PT Gunung Bale, SNI No. 15-1023-1989 concerning pyrophyllite for the manufacture of refractory material, and SNI No. 15-1325-1989 concerning pyrophyllite for the manufacture of fine ceramic raw materials.

a. Quality Standard of PT Gunung Bale

Based on the quality standard set by PT Gunung Bale, 2021, pyrophyllite was classified into three quality grades based on the content of its chemical composition (Table 1) and its use in the industrial sector in terms of the content of Al_2O_3 , SiO_2 , and Fe_2O_3 compounds as in Table 2.

Table 1. The quality of pyrophyllite based on quality requirements from PT Gunung Bale (PT Gunung Bale, 2021)

Pyrophyllite grade	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O
High Grade	>60%	>20%	<1%	<1%	<0.1%	<0.1%	<0.1%	<0.1%
Medium Grade	>60%	<20%	<1%	<1%	<1%	<1%	<1%	<1%
Low Grade	>60%	<20%	>1%	<1%	<1%	<1%	<1%	<1%

Table 2. Pyrophyllite quality requirements for the industrial sector (PT Gunung Bale, 2021)

Industrial Sector	Al_2O_3	SiO_2	Fe_2O_3
Fiberglass	22 – 26%	70 – 74%	<0.08%
Refractory	26 – 28%	-	<0.10%

b. SNI No. 15-1023-1989

Badan Standarisasi Nasional, 1989, classified pyrophyllite for the manufacture of refractory material into

three levels of quality, namely class I, class II, and class III which were reviewed based on the chemical composition of SiO_2 , Al_2O_3 , Fe_2O_3 , CaO, and alkali as shown in Table 3.

Table 3. Quality requirements of pyrophyllite for the manufacture of refractory materials based on SNI No. 15-1023-1989 (Badan Standarisasi Nasional, 1989)

Rank	Quality Requirements
I	SiO ₂ maximum 67%
	Al ₂ O ₃ minimum 29%
	Fe ₂ O ₃ maximum 0.2%
	CaO maximum 0.3%
	Alkali maximum 0.2%
II	SiO ₂ maximum 75%
	Al ₂ O ₃ minimum 20%
	Fe ₂ O ₃ maximum 0.4%
	CaO maximum 0.3%
	Alkali maximum 0.7%
III	SiO ₂ maximum 80%
	Al ₂ O ₃ minimum 14%
	Fe ₂ O ₃ maximum 0.5%
	CaO maximum 0.3%
	Alkali maximum 2.5%

Table 4. Quality requirements of pyrophyllite for the manufacture of fine ceramics based on SNI No. 15-1325-1989 (Badan Standarisasi Nasional, 1989)

Requirements	Percentage
SiO ₂ (%) maximum	70%
Fe ₂ O ₃ (%) maximum	0.3%
TiO ₂ (%) maximum	0.5%
Al ₂ O ₃ (%) minimum	20%
CaO (%) maximum	1.5%
K ₂ O+Na ₂ O (%) maximum	0.75%

c. SNI 15-1325-1989

Based on SNI No. 15-1325-1989 (Badan Standarisasi Nasional, 1989) pyrophyllite used as raw material for the manufacture of fine ceramics must meet certain quality requirements, such as maximum of 70% SiO₂, maximum of 0.3% Fe₂O₃, maximum of 0.5% TiO₂, minimum of 20% Al₂O₃, maximum of 1.5% CaO, and maximum of 0.75% K₂O+Na₂O as shown in Table 4.

4. Results

4.1. Lithology of Research Area

There were five kinds of lithology in the research area, which were basalt, andesite breccia, dacitic tuff, dacite and limestone.

4.1.1 Basalt

Based on megascopic observation, the rock had a grey-black color, aphanitic texture and massive structure (Fig. 2). The petrographic observations (Fig. 3) showed the composition of the rock were anorthite plagioclase (45%), quartz (3%), clinopyroxene (15%), hornblende (5%), opaque minerals (2%), sericite (15%), chlorite (3%) and volcanic glass (22%).

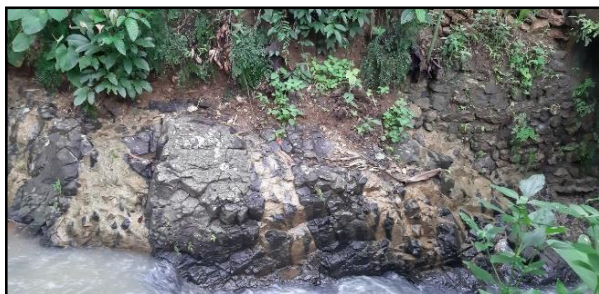


Fig 2. The outcrop of basalt in research area

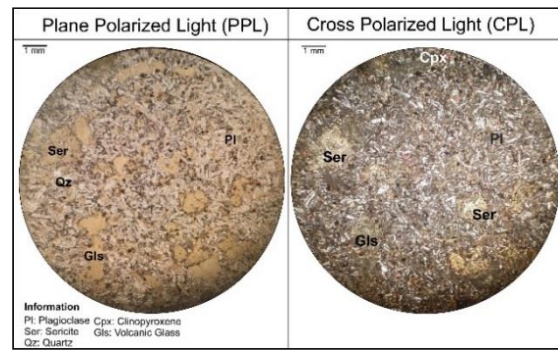


Fig 3. Thin section of basalt

4.1.2 Andesite Breccia

This rock generally were found in weathered condition. Based on megascopic observation the rock had reddish-brown color, massive structure, grain size ranged from cobble (64 - 256 mm) to coarse sand (1/2 - 1 mm), poorly sorted and sub-angular shape (Fig. 4). The rock composition was dominated by andesite fragments and tuffaceous matrix. Based on petrographic observations of the fragment (Fig. 5), the composition of the rock were andesine plagioclase (45%), orthoclase (2%), opaque minerals (1%), volcanic glass (36%) and secondary quartz (16%).

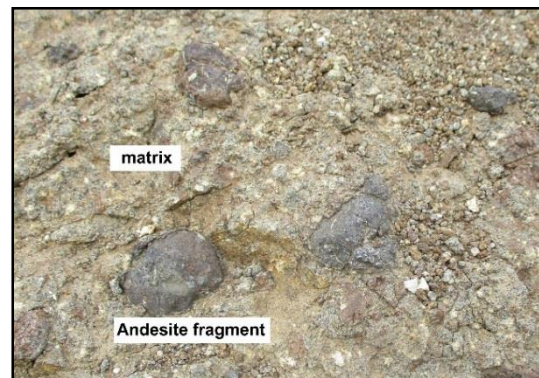


Fig 4. The outcrop of andesite breccia in research area

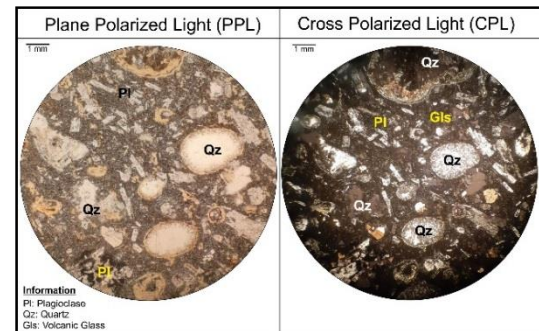


Fig 5. Thin section of andesite fragment of andesite breccia

4.1.3 Dacitic Tuff

This rock generally were found in weathered condition. Based on megascopic observation, the rock had a reddish-brown color, massive structure, grain size ranged from pebble (4 - 64 mm) to fine sand (1/8 - 1/4 mm), poorly sorted and subangular grain shape (Fig. 6). Based on the petrographic observation (Fig. 7), the composition of the rock were secondary quartz (40%), sericite (25%), pyrophyllite (33%) and opaque mineral (2%).

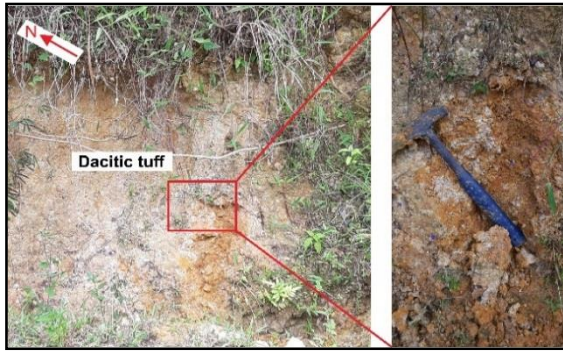


Fig 6. The outcrop of dacitic tuff in research area

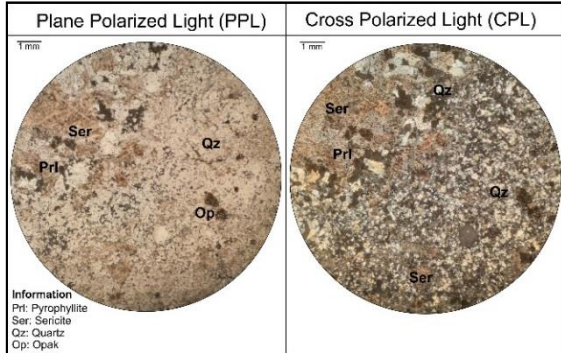


Fig 7. Thin section of dacitic tuff

4.1.5 Limestone

Based on megascopic observation, the rock had brown color with fine grain size, massive structure and moderately sorted (Fig. 10). Based on petrographic observation (Fig. 11), the composition of the rock were foraminifera test (32%), coral (20%), micrite (30%) and calcite cement (15%).



Fig 10. The outcrop of limestone in research area

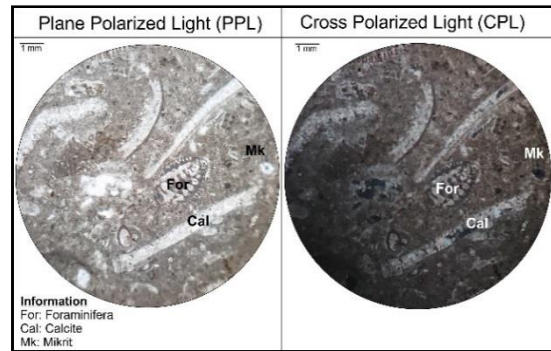


Fig 11. Thin section of limestone

4.1.4 Dacite

Based on megascopic observation, the rock had brown color, porphyritic texture with fine to medium crystal size, massive structure (Fig. 8) Based on petrographic observations (Fig. 9), the composition of the rock were andesine plagioclase 12%, quartz (35%), clinopyroxene (3%), opaque minerals (5%), groundmass (10%) and clay minerals (35%).



Fig 8. The hand specimen of dacite in research area

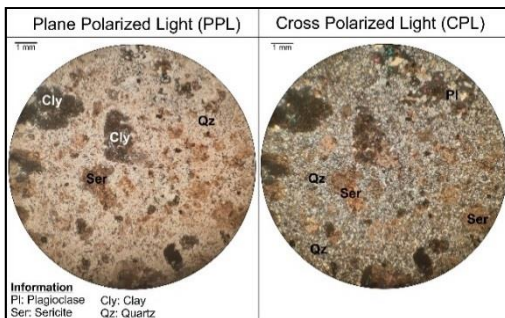


Fig 9. Thin section of dacite

4.2. The Characteristics and Quality of Pyrophyllite

There were three pyrophyllite samples analyzed from the mining area of PT Gunung Bale, namely the EIP sample, WSP sample, and DSP sample.

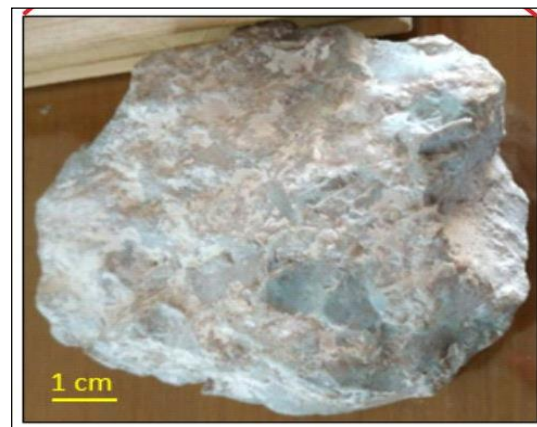


Fig 12. The hand specimen of EIP sample

a. EIP Sample

The physical properties of the EIP sample (Fig. 12) showed greyish-green color, dull luster, massive, hardness about 5.5 Mohs scale and white streaks.

The mineral composition from petrographic observations (Fig. 13) were quartz (13%), clay minerals (29%), opaque minerals (3%), pyrophyllite (40%) and lithic rocks (15%). XRD analysis (Fig. 14) showed the EIP sample consisted of pyrophyllite, K-feldspar, sericite, cristobalite, illite and quartz.

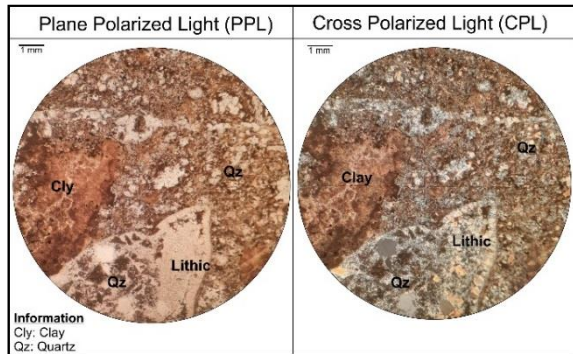


Fig 13. Thin section of ESP sample

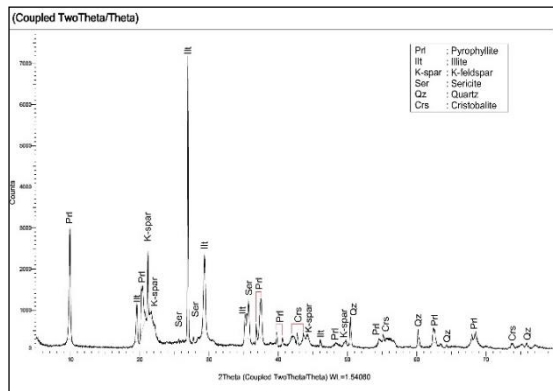


Fig 14. The XRD analysis of ESP sample

The chemical composition of the EIP sample from XRF analysis can be seen in (Table 5), it can be interpreted that this EIP sample belongs to high grade quality based on the quality standards set by PT Gunung Bale (Table 1) because it contains high Al₂O₃ (alumina) or more than 20% .

Table 5. The chemical composition of EIP sample, DSP sample and WSP sample

Chemical Composition	EIP Sample	DSP Sample	WSP Sample
SiO ₂	64.86%	82.40%	78.67%
Al ₂ O ₃	28.07%	14.03%	16.78%
Fe ₂ O ₃	0.06%	1.05%	0.06%
TiO ₂	0.81%	0.31%	0.65%
CaO	0.03%	0.03%	0.04%
MgO	0.05%	0.07%	0.06%
K ₂ O	0.03%	0.02%	0.03%
Na ₂ O	0.22%	0.09%	0.11%

b. DSP Sample

The physical properties of the DSP samples (Fig. 15) showed white color, dull luster, massive, hardness about 2.5 Mohs scale, white streaks and brittle.



Fig 15. The hand specimen of DSP sample

The mineral compositions from petrographic observation were (Fig. 16) quartz (10%), sericite (15%), clay minerals (45%), clinopyroxene (5%) and pyrophyllite (25%). XRD analysis showed the DSP sample consisted of (Fig. 17) pyrophyllite, kaolinite and quartz.

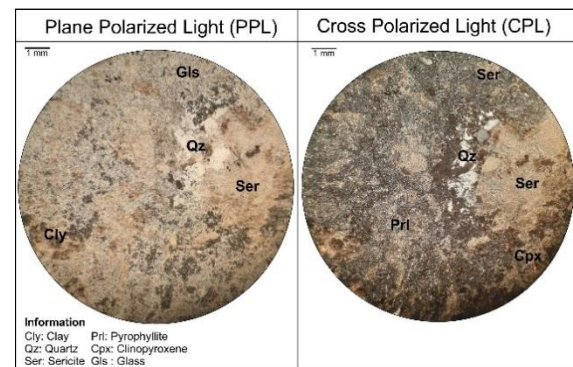


Fig 16. Thin section of DSP sample

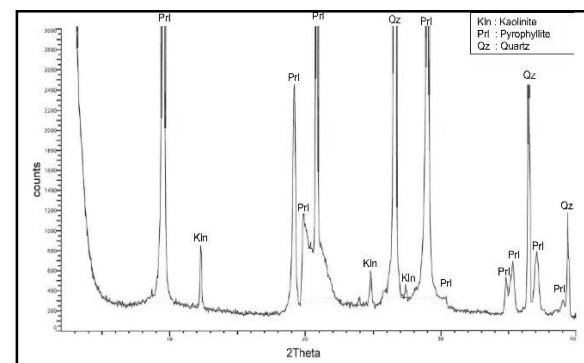


Fig 17. The XRD analysis of DSP sample

The chemical composition of the EIP sample from XRF analysis can be seen in (Table 5), it can be interpreted that this EIP sample belongs to low grade quality based on the quality standards set by PT Gunung Bale (Table 1) because it contains Fe₂O₃ > 1% and Al₂O₃ < 20% .

c. WSP Sample

The physical properties of WSP sample (Fig. 18) showed white color, dull luster, massive, hardness about 3.5 Mohs scale, white streak.

The mineral compositions from petrographic observation were (Fig. 19) quartz (40%), sericite (25%), pyrophyllite (33%) and opaque mineral (2%). XRD analysis

showed the WSP sample consisted of (Fig. 20) pyrophyllite, halloysite, sericite, illite, K-feldspar and quartz.

The chemical composition of the WSP sample from XRF analysis can be seen in (Table 5), it can be interpreted that this EIP sample belongs to medium grade based on the quality standards set by PT Gunung Bale (Table 1) because it contains $Al_2O_3 < 20\%$.

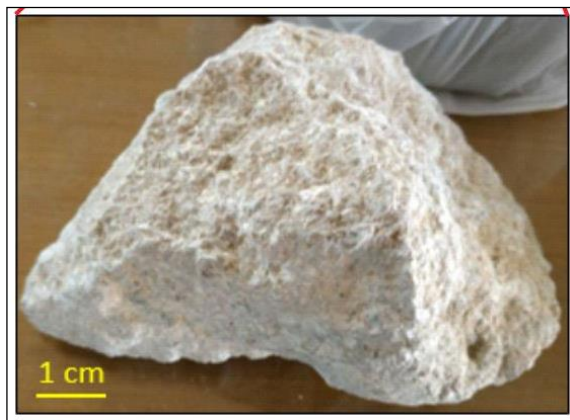


Fig 18. The hand specimen of DSP sample

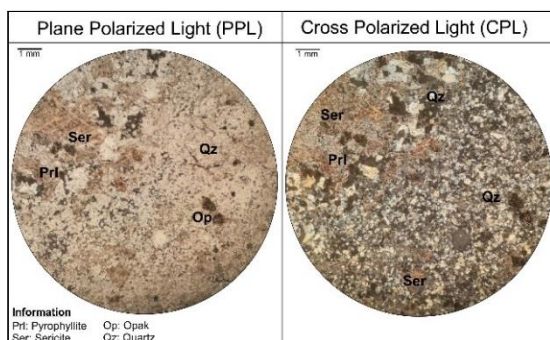


Fig 19. Thin section of WSP sample

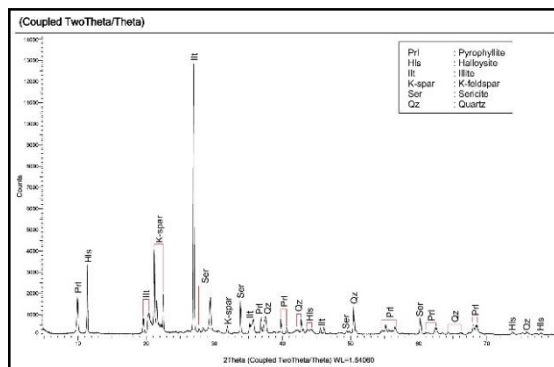


Fig 20. The XRD analysis of WSP sample

4.3. Genesis of Pyrophyllite in Research Area

Based on the results of the analysis which has been carried out including the petrographic analysis and XRD analysis on the three samples (EIP sample, DSP sample and WSP sample), there were some secondary minerals found such as pyrophyllite, sericite, cristobalite, illite, kaolinite and halloysite. While the rock sample which contained pyrophyllite mineral were dacitic tuff.

Pyrophyllite in the research area was interpreted to be formed from volcanic rock, especially dacitic tuff, which underwent hydrothermal alteration. Volcanic rock was the host rock, while hydrothermal fluid came from dacite intrusion. The interaction between the hydrothermal fluid

and the host rock caused high feldspar content in dacitic tuff converted into secondary minerals such as pyrophyllite.

4.4. The Application of Pyrophyllite in Industry

Based on the quality standard requirements of PT Gunung Bale (Table 2), pyrophyllite can be used as raw material in the fiber glass and refractory industries, which were reviewed based on the content of Al_2O_3 , SiO_2 , and Fe_2O_3 of each sample. From the chemical composition, it can be seen that only the EIP sample can meet the quality requirements of PT Gunung Bale to be used as refractory raw material.

Based on Indonesian National Standard (SNI) No. 15-1023-1989 (Table 3) regarding the quality requirements of pyrophyllite as a refractory material, classified it into three levels of quality, namely class I, class II, and class III. Based on the chemical composition of the three pyrophyllite samples in the study area, the EIP sample met the qualifications as class I refractory material by reducing the alkali content slightly (K_2O+Na_2O), while the WSP and DSP samples could meet the qualifications as class III refractory material.

Based on Indonesian National Standard (SNI) No. 15-1325-1989 regarding the requirements for pyrophyllite as a fine ceramic material as shown in Table 4. Based on the chemical composition of the three pyrophyllite samples, only EIP samples can be used as raw materials for the manufacture of fine ceramics.

5. Conclusions

There were five kinds of lithology in the research area, which were basalt, andesite breccia, dacitic tuff, dacite and limestone. There were three pyrophyllite samples analyzed from the mining area of PT Gunung Bale, namely the EIP sample, WSP sample, and DSP sample. Based on the characteristics, the EIP sample can be used as refractory raw material and fine ceramics, while the DSP sample and WSP sample can be used as class III refractory material.

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