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Article

The Utilization of Lapindo Mud Waste for Aluminium Sulfate Production

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Abstract

The Lapindo mudflow disaster in East Java Province, or also known as LUSI (**LU**mpur "mud"-**SI**doarjo) has become spectacular longest ongoing disaster in recent memory since 2006. The utilization of volcanic Lapindo mud could be the promising solution to prevent further environmental damage. The chemical composition of Lapindo mud contained of 44.1% SiO_2, 23.7% Fe_2O_3, 13% Al_2O_3, 7.02% CaO, 5.35% MoO_3, 2.53% K_2O, 1.84% TiO_2 and 0.7% Na_2O. Aluminium sulfate ($Al_2(SO_4)_3$) or "alum" have been widely used as coagulation compound in water treatment, paper and textiles industry. Aluminium sulfate can be synthesized from aluminium oxide (Al_2O_3) from Lapindo mud with acidic solutions (H_2SO_4). The aim of this work was to synthesize aluminum sulfate from Lapindo mud by using extraction process. The impact of H_2SO_4 concentration and heating time to the production of aluminium sulfate have been investigated. The results showed that the aluminium sulfate can be synthesized from Lapindo mud by using H_2SO_4. Based on XRF analysis, the variation of heating time and H_2SO_4 concentration affect the aluminium sulfate conversion. The increasing of heating time and H_2SO_4 concentration directly enhance the conversion until reach the optimum condition. The optimum condition for aluminium sulfate synthesis from Lapindo mud (75.78% conversion) was found to be 90 min for heating time with H_2SO_4 concentration of 80%.

Keywords: Lapindo mud, aluminum sulfate, heating time, H_2SO_4 concentration

1. Introduction

The Lapindo mudflow disaster in East Java Province, or also known as LUSI (**LU**mpur "mud"-**SI**doarjo) has become spectacular longest ongoing disaster in recent memory since May 29, 2006. The mud volcano has been spewing hot mud and gases in well near Sidoarjo which caused a massive loss of habitable land and environmental damage in the surrounding area [1]. As reported in BPLS data in 2013, the Lapindo mudflow displaced 39700 people, 12 villages in 3 districts (Tanggulangin, Porong and Jabon), 11241 buildings, more than US$600 million property and more than US$2.7 billion damage/management [2].

The location of the Sidoarjo mudflow disaster has an effect on the name attached to the disaster. For the people who believe that the mudflow came from the exploration of Lapindo Brantas Company, they tend to name the disaster become "Lapindo mudflow disaster". On the other hand, for those who think that the mudflow is a natural disaster, they tend to naming it as the Sidoarjo mudflow disaster which is also used by the government [3].

The utilization of volcanic Lapindo mud has been attracted many scientist and could become promising solution to prevent further environmental damage. As reported by Jalil et al., (2010) the chemical composition of Lapindo mud were 53.4% SiO_2, 5.47% FeO, 23.8% Al_2O_3, 5.59% Na_2O, 2.62% MgO, 2.89% Cl_2, 1.63% K_2O and 2.4% CaO [4].

Aluminium sulfate or also known as "alum" is a chemical compound that is indispensable in the several industrial sector. Nowadays, aluminium sulfate is still imported from other country such as Singapore and Australia which have very expensive prices. In the contrary, the high demand of aluminium sulfate in the textile industry, cosmetic industry, paper industry, and batik industry need to be considered [5]. The rapid increasing of Indonesian population causes the increasing of water demand. Therefore, the use of aluminium sulfate also continues to increase. Moreover, the domestic synthesis or production of aluminium sulfate is very important to overcome the shortage of aluminium sulfate. Indonesia have a huge abundance of raw materials for manufacturing process of aluminium sulfate and grouped into 2 namely aluminium sources and sulfate sources.

In 2011, the synthesize of aluminium sulfate from kaolin showed that the optimum condition was 90 min, at 180 ℃ and stirring rate 350 rpm with the conversion of 82%. Time, temperature and ratio of sulphuric acid and kaolin in reaction have a lot effect to conversion value result [6]. Nurcahyo et. al., in 2014 have successfully synthesize aluminium sulfate from clay with the optimum condition at 100 ℃, size of mess was 100 and 40% acid solution. In Soka tile, the conversion was 62%. Meanwhile in Jatiwangi the conversion only 20% [7]. In 2017, the synthesis of aluminium sulfate from cans waste give the optimum condition of 30% KOH concentration with the conversion was 93.2% [8].

Synthesis of Aluminium Sulfate

Aluminium sulfate of known as 'alum' is a chemical compound with the chemical formula $Al_2(SO_4)_3$. The synthesis of aluminium sulfate can be carried out by using aluminium oxide (Al_2O_3) in sulfuric acid solutions (H_2SO_4). The reaction between aluminium oxide and sulfuric acid will produce aluminium sulfate products as shown in Eq. 1. The first step in aluminium sulfate synthesis was dissolve the aluminium oxide with sulfuric acid solution. The solution was heated and filtered with buchner filter under hot condition. The filtrate was left for cooling down until the solid was formed. The obtained solid was further dried in oven [8].

$$Al_2O_3(s)+3H_2SO_4(l)\rightarrow Al_2(SO_4)_3(l)+3H_2O(l) \quad \text{(Eq.1)}$$

Extraction Process

Solid-liquid extraction or also known as 'leaching' is a process of separating a soluble substance (solute) from a mixture with an inert solid using a liquid solvent. The solid-liquid extraction process involve organic and/or inorganic solvents and their blends as extractant solvents in contact with an insoluble solid matrix [9].

Factors affecting extraction process:

1. Temperature

The increasing operating temperature will cause the higher dissolution rate of the solute by the solvent.

2. Size, shape, and solid particles condition.

The smaller size of the reacted particles will increase the contact surface area between the material particles and the solvent, thus causing extraction time will be faster.

3. Extraction Time

The longer extraction time will cause the longer contact time between the solvent and the raw material so as to produce more solute than the material soluble in the solvent.

4. Type of solvent:

A. Solubility, the solvent must have the ability to dissolve the solute as completely as possible.
B. Selectivity, the solvent can only dissolve the desired extract, not the other components of the raw material
C. Chemical activity of solvent, the solvent is a chemical that is stable and inert to other components in the system.

This aim of this study was to synthesize aluminum sulfate from Lapindo mud with

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sulfuric acid as a solvent. The influence of sulfuric acid concentration and heating time reaction to the conversion of aluminium sulfate was investigated in this study.

2. Material and Method

The raw materials used in this experiment was Lapindo mud from Porong district, Sidoarjo, East Jawa, Indonesia. The Lapindo mud was collected precisely located ± 2 km from the center of the Lapindo mudflow. Sulfuric acid (98% H_2SO_4) and aquadest obtained from UD. Eternal Nirvana in Surabaya.

Preliminary Step

The Lapindo mud was washed with aquadest to remove impurities and then dried to remove the water content. After drying, the Lapindo mud is ground using a mortar to speed up the aluminium oxide extraction process. The crushed mud is then pulverized to a size of 100 mesh. The refined Lapindo mud was then analyzed using the X-Ray Fluorescence (XRF) instrument.

Extraction Step

Crushed Lapindo mud with a size of 100 mesh was weighed 100 grams and then put into a three neck flask. Sulfuric acid (200 mL) with a variation concentration of 40%; 50% ; 60% ; 70% and 80% was added into the flask, then heat the mixture while stirring using a spatula periodically every 20 minutes. After the desired heating time variation is reached (70 min, 80 min, 90 min, 100 min, and 110 min), the heating is stopped. The mixture was filtered while in hot condition. The residue was analyzed by the X-Ray Fluorescence (XRF) instrument to determine the chemical content. Filtrate was cooled until a precipitate was formed, the precipitate was then filtered using filter paper, dried and analyzed by using X-Ray Diffraction (XRD) instrument to determine the phase of the solid product [10].

3. Results and Discussion

The Lapindo mud was used as raw materials to synthesize aluminium sulfate. The Lapindo mud was ground with a size of 100 mesh. XRF analysis result of Lapindo mud was tabulated in Table 1. As shown in Table 1, the Lapindo mud has high content of aluminium oxide (Al_2O_3) with percentage of 13%. The results showed that Lapindo mud has the potential to be processed into aluminium sulfate ($Al_2(SO_4)_3$). The content of SiO_2 and Fe_2O_3 also have high concentration in Lapindo mud. This results were in accordance with the similar study reported by Rahmayanti et al., (2020) and Mahardika et. Al., (2017) with the major component of Lapindo mud were SiO_2, Fe_2O_3 and Al_2O_3 [11,12].

Table 1. The chemical composition of Lapindo mud using XRF analysis

No.	Compound	Concentration (%)
1	SiO_2	44,10
2	Fe_2O_3	23,7
3	Al_2O_3	13
4	CaO	7,02
5	MoO_3	5,35
6	K_2O	2,53
7	TiO_2	1,84
8	Na_2O	0,7
9	Cr_2O_3	0,08
10	CuO	0,07

Based on the results of XRF analysis of the product obtained from extraction method, the effect of heating time and sulfuric acid concentration on the conversion of aluminum sulfate ($Al_2(SO_4)_3$) product is summarized in Table 2 and also shown in Figure 1. The result shows that at heating time of 70 min to 90 min with various concentrations of sulfuric acid (H_2SO_4), the conversion aluminum sulfate increased with increasing heating time and sulfuric acid concentration. The increasing of sulfuric acid concentration and heating time, the volume of sulfuric acid in the solvent will increase and be able to dissolve more aluminum oxide in the Lapindo mud. However, due to changes in the concentration of sulfuric acid, the conversion of aluminum sulfate from 90 minutes to 110 minutes decreased because alumina (Al_2O_3) reacted with sulfuric acid, its reaction ability began to decrease the aluminium sulfate conversion.

Similar to the result reported by Ismayanda (2011), the increasing of sulfuric acid concentration causes the conversion of aluminum

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sulfate products become to increase, along with the longer heating time is carried out due to the increasing amount of sulfuric acid that reacts with the existing aluminum, so that large concentrations of sulfuric acid will easily react with the existing aluminum. . However, when the reaction time reaches 90 min, the conversion becomes insignificant, even tends to decrease due to the balance rection. The data optimum data was found to be conversion of 75.78% with the operating conditions of 80% sulfuric acid concentration and 90 minutes heating time [6].

Table 2. The effect of heating time and H_2SO_4 concentration to the $Al_2(SO_4)_3$ conversion

H_2SO_4 Concentration	Heating time	Conversion
(%)	**(min)**	**(%)**
40%	70	50.31
	80	61.14
	90	67.27
	100	62.09
	110	59.67
50%	70	52.99
	80	60.71
	90	70.15
	100	64.44
	110	60.19
60%	70	54.21
	80	65.58
	90	72.34
	100	68.68
	110	65.65
70%	70	57.96
	80	69.47
	90	73.91
	100	71.71
	110	68.59
80%	70	59.24
	80	73.66
	90	75.78
	100	73.39
	110	71.32

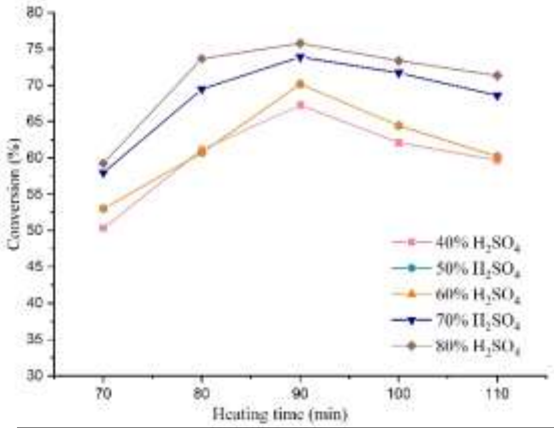

Fig. 1. Correlation between heating time and H_2SO_4 concentration with $Al_2(SO_4)_3$ conversion

The product analysis using X-ray Diffraction (XRD) was used to identify the crystal phase and determine the types of minerals that produce from Lapindo mud extraction. The diffractogram of synthesized product was shown in Figure 2.

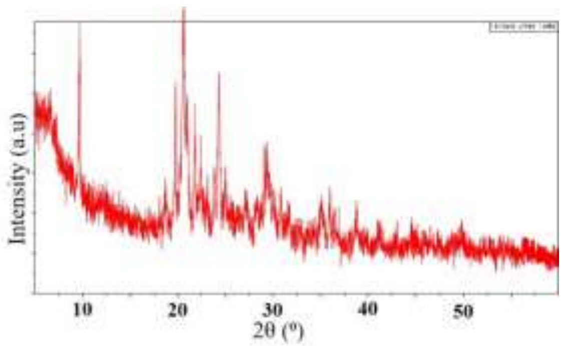

Fig. 2. The diffractogram of product obtained by Lapindo mud extraction

The diffractrogram of synthesized product shows the presence of non-uniform peaks. The highest peak at 2θ= 9.6464°, 20.6242°, 24.2589° correspond with the crystalline phase of aluminum. These results further supported with Fitri's previous study in 2017 which show that aluminum sulfate is appeared in crystalline phase [13]. Then, the diffractrogram was further analyzed to determine any compound content in the synthesized product [13]. The analysis results was shown in Figure 3.

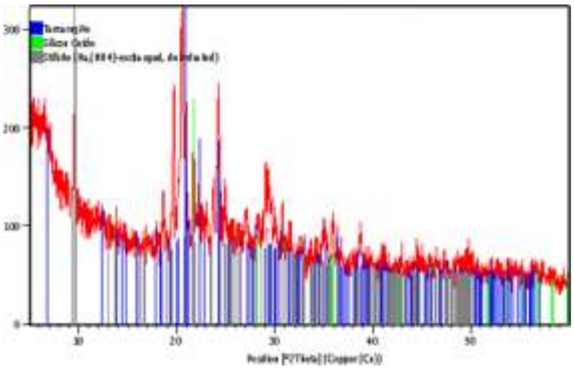

Fig. 3. Analysis using *X-ray Diffraction (X-RD)*

The analysis result from X-ray Diffraction (X-RD) found that the synthesized product was composed of *tamaragite*/Sodium Aluminum Sulfate Hydrate, *stilbite* and silica oxide. The crystal structure from XRD analysis on the synthesized product was tabulated in Table 3.

Table 3. The crystal structure from X-ray Diffraction analysis

Materials	Tamarugite	Silica oxide	Stilbite
Lattice Parameter	a=7.3530	a=4.9984	a=13.5710
	b= 25.2250	b=4.9984	b=18.2640
	c=6.0970	c=7.0242	c=11.2960
Volume (A^3)	1126.21	175.49	-
Crystal size (nm)	90	90	90
Density (g/cm^3)	3.5853	-	1.76
Crystal system	Monoclinic	Tetragonal	Monoclinic

The presence of tamarugite phase or sodium aluminum sulfate hydrate compounds indicates the successfully synthesized of aluminum sulfate from Lapindo mud. According to Irfan and Ramadhani (2014), the possible cause of presence impurities in the form of sodium due to the existence of sodium, silica, iron, and other metals with the high content in raw and supporting materials. Therefore, purification of aluminum sulfate must be carried out so that the metals contained in it can be kept to a minimum [14].

4. Conclusions

Aluminium sulfate have been successfully synthesized from Lapindo mud using sulfuric acid as a solvent. According to XRF analysis data, variations in heating time and sulfuric acid concentration affect the conversion of aluminum sulfate. Increasing the heating time and sulfuric acid concentration will increase the conversion until reach the optimum condition and decrease when the reaction is balanced. The highest aluminum sulfate conversion was obtained at 75.78% with the operating conditions of 80% sulfuric acid concentration and heating time at 90 minutes.

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