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Combination of CaCO3 and Ca(OH)2 as Agents for Treatment Acid Mine Drainage Poedji Loekitowati Hariani1,\*, Salni Salni2, and Fahma Riyanti1 1,3Department

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Palembang 30139, Indonesia Abstract. Acid mine drainage (AMD) has characteristic very low pH solution and containing metal ions in high concentration. This paper presents the use of CaCO3, Ca(OH)2 and the combination of both to increase the pH and decreased the concentration of Fe and Mn ions for acid mine drainage. The research variables are the effect of reactant dosage, contact time and temperature by batch studies. The AMD before treatment has pH solution of 3.38, Fe and Mn ions concentration of 44.6 and 7.19 mg/L, respectively. The dosage of CaCO3 to increased pH solution about 7.0 was found 2400 mg/L at contact time 60 minutes and temperature 40 0C. The amount of Ca(OH)2 for the neutralization of AMD solution smaller than CaCO3 is 210 mg/L at contact time 45 minutes and temperature 40 0C. The combination dosage of CaCO3 1000 mg/L and Ca(OH)2 90 mg/L can increased the pH of AMD solution to 7.10 and reduction concentration of Fe to 3.53 and Mn to 4.51 mg/L. Therefore, the integrated of CaCO3 and Ca(OH)2 has the potential to be applied to treatment

acid mine drainage. 1 INTRODUCTION Acid mine drainage (AMD) is

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an important problem in the environment and a serious concern in many countries. The AMD contains of heavy metal from the oxidation of sulphidic minerals and producing low of pH solution. The treatment of AMD must be done on the exhaust into the environment because this substance is harmful to aquatic life at low concentration, not

biodegradable and media surroundings. The AMD contains a lot of heavy metal ions such as Cu2+, Fe3+, Mn2+, Zn2+, Cd2+ and Pb2+. The heavy metal ions are not biodegrable and tends to accumulate in the bodies in living organis [1].. The AMD solution usually is orange colour from precipitation of iron oxide and hydroxide [2]. Characteristic of AMD of each region is different depending on typical mine water for the individual deposits [3]. The characteristic of AMD solution is influenced by several factors such as

### bacteria, temperature, starting pH and alternative oxidants like iron or manganese [4]. The

treatment of AMD must be efficient and continual [5]. The method is often used for AMD treatment is oxidation, coagulation/flocculation, neutralization and precipitation of metal ions. The oxidants for the AMD such as Ca(OCI)2, NaClO, CaO2, H2O2, whereas coagulands often to treatment of AMD are

## Al2(SO4)3, FeSO4, Fe2(SO4)3, NaAlO2

[2]. Some materials can be used for the neutralization process such as CaCO3 [3, 6], Mg(OH)2 [7], fly ash [8], NaOH [9]. The use of CaCO3 for the neutralization process of AMD was patented by the US Geological Survey Leetown Science Center [10]. It has been reported that the pH of AMD treatment using CaCO3 (limestone) more economical but slow rate of dissolution \* Corresponding author: pujilukitowati@yahoo.com with effectiveness  $\pm$  30%) [11]. The other research, effect neutralization of AMD with limestone in the reactor during 48 hours produced an armor coating in the bottom [6]. It causes the decline in the effectiveness of the use of CaCO3. Two step reaction process of limestone with sulfuric acid in the AMD as follows: CaCO3 + H2SO4  $\rightarrow$  CaHSO4+ + HCO3- HCO3- + CaHSO4+  $\rightarrow$  CO2 + H2O + CaSO4 (1) (2) At the end of the reaction occurs of precipitated calcium sulfate. The other studies have shown that calcium carbonate to raise the pH by consuming hydrogen ions and adding alkanity to form bicarbonate ions, according the following reaction: [12]

 $CaCO3 + 2H+ \rightarrow Ca2+ + H2O + CO2 CaCO3 + H2CO3 \rightarrow Ca2+ + 2HCO3- (3)$ 

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(4) The metal ions can be precipitate to form hydroxides or oxyhydrosides.

#### Several methods have been developed for treatment of AMD.

In this paper, integrated of limestone (CaCO3) and hydrated lime (Ca(OH)2) was used for neutralization of AMD solution. Effectiveness Ca(OH)2 for AMD treatment three times greater than CaCO3, but the cost of processing using these materials more expensive [11]. Hydrated lime has the ability to raise the pH quickly and precipitate the metal ions as

greater than CaCO3, but the cost of processing using these materials more expensive [11]. Hydrated lime has the ability to raise the pH quickly and precipitate the metal ions as hydroxide such as manganese at pH 9 to 9.5. Reaction of Ca(OH)2 with hydrogen ions or metal ios as follow: [13] Ca(OH)2 + 2H+  $\rightarrow$  Ca2+ + 2H2O (5) Ca(OH)2 + Me2+/Me3+  $\rightarrow$  Me(OH)2/Me(OH)3 + Ca2+ (6) The other research, the use of CaCO3 on AMD treatment can increase the pH solution from 2.9 to over 7 at the contact time of 48 hours [6]. Beside that, the addition of Ca(OH)2 do not only neutralized, but also the OH- anionic ions increased the rate of speciation with reaction to metal ions and increased the

pH of AMD. CaCO3 and Ca(OH)2

can also destabilize the hydrolysis of acid mine drainage so that the value of TSS decrease [14]. At present, the combination of CaCO3 and Ca(OH)2 to treatment of AMD 2 is expected to increase the pH and reduction the heavy metal ions at the faster contact time. Ca(OH)2 has a greater solubility than CaCO3 are 1850 and 14 mg/L, respectively [7]. The parameters of studied are the effect of reactant dosage, contact time and temperature by batch studies. 2 EXPERIMENTAL DETAILS 2.1.1 Materials The reagents such as CaCO3, Ca(OH)2 were analytical grade by Merck and all solution were prepare with deionized water. Acid mine drainage collected from a mine in Tanjung Enim, South Sumatera, Indonesia. 2.1.2 Effect of dosage, contact time and temperature

The effect of dosage was added of CaCO3 and Ca(OH)2

to 1 L of AMD solution at constant stiring (120 rpm) at room temperature for 60 minutes. The amount of CaCO3 was added from 100-2500 mg/L (interval 100 mg) and Ca(OH)2 10-240 mg/L (interval 10 mg). The data of effect contact time obtained from 0-100 minutes with interval 5 minutes. The effect of the temperature was performed using thermostatic water bath at temperature of 30-90 0C. Data is the observed pH of the solution. The pH solution is obtained using pH meter Orion star A2111. 2.1.3 Neutralization of AMD using combination CaCO3 and Ca(OH)2 For this purpose

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1 L of AMD. The amount of CaCO3 1000 mg while the Ca(OH)2 in the range 10-120 mg (interval 10 mg) with contact time and temperature using the latest result of previous studies. Determination of metal ions (Fe and Mn) in AMD before and after treatment were determined using Atomic Absorption Spectroscopy Shimadzu AA 7000 with atomization by N2O-acetilene flame. The wave number for obtaining Fe and Mn ions at 248.3 and 279.8 nm, respectively. 3 RESULT AND DISCUSSION There are two technologies for

treatment of acid mine drainage	(AMD), these are	active treatment	1
and			

### passive treatment processes. The active treatment method is

the addition of chemicals to raise the pH and precipitate the metal ions. The active treatment using chemical reagents. This method is very effective for the treatment of AMD. The passive treatment includes aerobic and anaerobic wetland. This method is natural process. In traditional treatment of AMD, CaCO3 has been used for neutralization of AMD in many countries. There used in anoxic limestone drain and open limestone channel [9]. In this study, the combination of CaCO3 and Ca(OH)2 to treatment of AMD was evaluated. Before integrating of both, investigated the optimum conditions of treatment in each material includes



temperature. Neutralization of AMD using CaCO3 and Ca(OH)2 are presented in Fig 1. and Fig 2. The AMD solution has initial pH of 3.38. The AMD is classified as types of 1 that has pH solution very lower. In the AMD, pyrite is oxidized to soluble iron and sulphuric acid as follows : [15] 2FeS2 + 702 + 2H2O  $\rightarrow$  2Fe2+ + 4SO42- + 4H+ (7) Fe2+ is oxidaxed to

Fe3+ by oxygen and hydrogen and oxidation of sulfure by Thiobacillus and Ferroplasma bacteria. The oxidation process is influenced by pH solution, this reaction occurs fast at low pH (<4). Ferrous hydroxide formed is namely yellow boy. [5,16]  $2Fe2+702+2H20 \rightarrow 4Fe3+4H20$  (8)  $4Fe3+12H20 \rightarrow 4Fe(OH)3+12H+$  (9) The result obtained that the increased dosage of CaCO3 and Ca(OH)2 increased of pH and reached at pH 7 on the addition CaCO3 of 2400 mg while

Ca(OH)2 of 210 mg. Ca (OH)2 has the ability to

increase the pH solution of AMD better than CaCO3. The positive 8 7 7.03 6 5 pH 4 3 2 1 0 0 500 1000 1500 2000 2500 Dosage of CaCO3 (mg/L) correlation that fact with increasing dosage of neutralization agents, more hydrogen ions can be neutralized using CaCO3 and Ca(OH)2. Fig. 1. Effect of dosage CaCO3 for pH solution 8 7 6 7.06 5 pH 4 3 2 1 0 0 50 100 150 200 250

Dosage of Ca(OH)2 (mg/L)

Fig. 2.

Effect of dosage Ca(OH)2 for pH solution The both of

agents have the same acid equivalent is 2 but Ca(OH)2 has an efficiency factor of 0.8 higher than CaCO3 of 0.4 [17]. Efficiency factor is an empirical estimate for neutralizing acidity. Besides than, Ca(OH)2 is a strong alkaline has a neutralization efficiency of 90 % greater than CaCO3 only 30 % [11]. The alkaline necessary to raise the pH solution and produce hydroxide to precipated with metal ions. Table 1 shows some of the materials

used to raise the pH solution of AMD. Compared to the data in

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table, the results of this research requires fewer doses to increase pH of AMD. The type and dosage of materials have effect to the increase of pH solution. Table 1. The materials to treatment of AMD Materials Dosage pH of Acid mine drainage Initial Final pH pH Mg(OH)2 [7] Ba(OH)2.8H2O [7] Fly ash [8] Fly ash [8] Fly ash [8] Limestone [6] Bentonite [18] 0.25 g/L 7.35 g/L AMD:Fly ash (2:1) AMD

# :Fly ash (1:1) AMD :Fly ash (3:1)

- 1 g/L 3.4 8.3 3.4 12 2.7 11.5 2.7 11.5 2.7 9.5 2.9 6.5 2.7 7.5 Figure 3 showed effect contact time of CaCO3 and Ca(OH)2 on the neutralization of AMD. The data obtained that the equilibrium time required for the neutralization was almost 60 minutes for CaCO3 and 45 minutes for Ca(OH)2. The contact time affect the number of hydrogen ions that can be neutralized by chemical agents. The longer the contact time so the more hydrogen ions that can be neutralized. From the result, the neutralization using Ca(OH)2 faster than CaCO3. In this work indicated that an increase in contact time resulted in increased of pH solution. After the equilibrium time, the pH relatively constant at pH 7. It is clear that the pH value dependence of contact time. The longer of the contact time, the more neutralization process. Furthermore, pH relatively constant at about pH 7 for a limited amount of reagents. To compare the other research, the optimal dosage to reach at pH 7 on acid mine water in the locality of Jiří Mine in the Sokolov Region is 150 mg/L of Ca(OH)2 with contact time 30 minutes [3]. The reduction of sulfate from AMD also dependent dosage of CaCO3 and Ca(OH)2. The research indicated that the relationship between sulfate solubilization and pH was direct and linear [19]. 8 7.08 7 6 7.06 5 pH 4 CaCO3 3 Ca(OH)2 2 1 0 0 10 20 30 40 50 60 70 80 90 100 Contact time (minutes) Fig. 3. Effect of contact time for pH solution The effect of temperature for AMD neutralization is presented in Fig 4. On evaluating the result, it was clear that an increase of pH at the temperature 20-40 0C, and then the pH decreased at higher temperature. The solubility indicated the maximum concentration of substance that can be dissolved at solution (ussulaly at room temperature). Beside pH solution, the solubility of compound is also dependent by temperature. The increase of temperature so the greater solubility of compounds. 7.2 7 6.8 pH 6.6 CaCO3 6.4 Ca(OH)2 6.2 0 10 20 30 40 50 60 70 80 90 100 Temperature (0C) Fig.4. Effect of temperature for pH solution The chemical reagents of CaCO3 and Ca(OH)2 have the same pattern of obtained pH 7 at temperature 40 0C. If the temperature is increased, the average kinetics also increases, it destabilized the solid state and thus the dissolve of the precipitate. This causes solubility of hydrogen ions and then the pH solution is decreased. The same result observed the influence of temperature for neutralization of AMD solution using NaOH. The optimum temperature at 20 0C, there was no change in pH at temperature 40 and 60 0C. The pH dropped to 2.2 at temperature 90 0C [9]. The combined of CaCO3 and Ca(OH)2 is expected to reduce the dosage of CaCO3 and improve efficiency process and economical. The concentration of Fe and Mn ions in AMD solution during the addition of 1000 mg/L CaCO3 and variation

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showed in Fig 5. The process at contact time 45 minutes and temperature 40 0C. The AMD have contain of Fe is 44.6 mg/L while the Mn is 7.19 mg/L. Characteristic of AMD solution exceeds of acid mine drainage quality standard. The maximum of Fe is 7 mg/L, Mn is 4 mg/L and pH solution in the range 6-9 [20]. We can see that after neutralization using CaCO3 and Ca(OH)2 was reached Fe and Mn ions of 3.53 and 4.51 mg/L, respectively. 50 45 40 35 pH Value 30 Fe (mg/L) 25 20 Mn (mg/L) 15 3.53 10 7.02 4.51 5 0 0 10 20 30 40 50 60 70 80 90 100 110 120 Concentration of Ca(OH)2 mg/L Fig. 5. Combination of CaCO3 with Ca(OH)2 to treatment of AMD solution The pH solution increased from 3.38 to 7.02. The effectively decrease of Fe and Mn ions were 92.42 and 37.27 %. The effectivity to removal Fe ions from AMD greater than Mn ions. The oxidixed and precipitation of Fe ions about at pH 7.0 while Mn ions at higher pH is 8. The other studies shows that CaCO3 and Ca(OH)2 effectif to reduced TSS by mechanism coagulation and floculation [21]. The study also investigated interaction double layer metal ions on both reagents of the aqua- colloids. The use of

bottom ash, bentonite and fly ash to reduce the concentration of

iron ions obtained at the optimum weight of 3, 4, 4 g in 100 mL of AMD solution, respectively [18]. In this study, has a smaller dosage than the result. Another study shows that adsorption capacity for adsorption of Mn ions in AMD is 6.03 mg/g using bone char [22]. The AMD solution is very complex, there is competition between the metal ions to form precipitate. The metal ions form precipitates depending value of Solubility Product Constant (KSP). The Ksp is the equilibrium constant, indicated that saturated solutions of ionic dissolving in an aqueous. The metal ions have small solubility product constants will be precipitated first. Example, Ksp Fe(OH)3 = 6.10-38 smaller than Ksp Mn(OH)2 = 2.10-13, so Fe ions to form precipitate earlier than Mn ions [23]. 4 CONCLUTIONS The combination of CaCO3 and Ca(OH)2 can be used for neutralization and reduce metal ions on acid mine drainage (AMD) solution. Integrated of both with a dosage of CaCO3 1000 mg/L and Ca(OH)2 90 mg/L can raise the pH solution from 3.38 to 7.02 and decrease of Fe and Mn ions with efficiency 92.42 and 37.27 %, respectively. In this study show that combination of CaCO3 and Ca(OH)2 an effectif to treatment of AMD.

ACKNOWLEDGMENTS This research was supported by The Directorate General of Technology Research and Higher Education

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