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COMPARATIVE STUDY USING LOW COST ACETIC AND SULFURIC ACIDS ON ROASTING AND PEROXIDE FOR LEACHING PROCESS TO EXTRACT PB FROM GALENA

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Abstract

Extraction of heavy metals from mineral concentrate applying leaching acids is a major issue due to simple, fast, and economical process. This article aims to present the application of acetic acid and sulfuric acid for lead (Pb) extraction from galena concentrate applying roasting (600°C and 60 min.) and peroxide oxidant (0.5 M) for 60 min. leaching process. Nowadays, lead has been broadly applied for batteries, besides for PVC tubes, chemicals, paint color, and alloys for joint. The peroxide increases the percentage of Pb extraction. This study shows the effect of acid concentrations (1.0 M, 1.5 M, and 2.0 M), temperatures (30, 50, and 70°C), and stirring speed (200, 400, and 600 rpm) on Pb extraction from galena. A particle size of galena ≤ 200 mesh after roasting has been used for leaching process. AAS has been used to determine Pb concentration in solution after leaching process. This study shows that acetic acid is a better leaching agent rather than sulfuric acid due to lead sulfate precipitation. Optimization result shows leaching with acetic acid achieved 35.64 ppm Pb extraction using 2.0 M acetic acid, 50°C, and stirring speed of 200 rpm. This study used low-cost acids as leaching agent for Pb recovery that can be viewed as a preliminary breakthrough in heavy metal recovery. The simple leaching technique looks promising for future application on heavy metal separation from mining mineral.

Keywords: Acetic Acid, Galena Concentrate, Lead Extraction, Sulfuric Acid, AAS.

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1. Introduction

Galena is a mineral with its chemical formula as PbS (lead sulfide). Galena ores are usually in association with other minerals like sphalerite, marcasite, pyrite, chalcopyrite, and other hydrothermal minerals[1]. Galena is found as veins in metamorphic deposit due to hydrothermal process. In general, mineralization of galena is related to sedimentation process of upper limestone formation, volcanic rock, granite igneous rock, and alluvial deposit[2]. Exploitation of galena mineral generally uses dump-truck and backhoe heavy equipment to excavate galena after blasting.

Galena is the main source to extract Pb from its concentrate[3]. Fig. 1 shows the galena mineral found in several rocks. Due to its corrosive resistance, up to date, lead has broadly applied for many purposes especially for lead batteries. Besides, lead is used as an isolator for PVC (polyvinyl chloride) tubes, as alloy for joining lines, pigments, lead glass crystal, ammunition, and as protecting material for nuclear reactor. Since galena is found in many places, lead has been used in many industries due to economic reason.

Ongoing with the extraction process of Pb from galena, many previous works reported pyrometallurgy and hydrometallurgy routes[1,4-6,7-8].

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Fig. 1. Galena mineral

Previous investigations concerned with several factors such as environmental problems with dust, SO_2, and volatile lead liquid applying pyrometallurgical process[6], and expensive and toxic leaching agent applied in hydrometallurgical process[4,8-9]. Besides the two routes mentioned above, electrometallurgical route has also been applied to produce metal from its mineral concentrate using electrical energy as the metal ion reduced at the cathode in electrolysis[10]. Mubarok[8] studied the effect of acetic acid concentrations on Pb leaching using shrinkage core model. Dessy[5] and Anugrah[4] applied fluorosilicic acid and peroxide to extract Pb from galena concentrate to study the parameter effects (leaching time, leaching temperature, acid concentration, particle size, and stirring speed) on Pb leaching process. Suryana[1] studied the effect of peroxide and lead oxide cathode from used batteries on Pb leaching. Moreover, Feng[11] conducted separation of galena and pyrite applying serpentine as depressant, while Feng[12] reported separation of galena and chalcopyrite applying highly efficient, non-toxic, biodegradable hydroxyethyl cellulose as depressant and H_2O_2 in flotation process, and Wang[7] separated galena from sphalerite using flotation through chelation with different nitrogen functional groups.

Ongoing with metal recovery from some mineral wastes that induced major issue in mining field, previous studies[13–15] reported some investigations on other metal recovery using leaching acids. Agatzini[13] reported titanium recovery from bauxite mineral applying diluted sulfuric acid at atmospheric pressure, while Barakat[14] reported indium recovery from wire scrap using H_3PO_4 leaching and precipitated as indium phosphate. Other study on tin recovery from copper waste was reported by Nurul[15] using HCl and the result found almost 60% tin dissolved in 2 M HCl at 80ºC and 8h leaching. Considering the pollution effect, toxic and expensive leaching agents, this study attempts to use low cost and less toxic acids (acetic acid and sulfuric acid) separately as comparative study using peroxide (H_2O_2) oxidant with galena roasting treatment prior to leaching process. The work investigates the effects of acid concentrations (1.0 M, 1.5 M, and 2.0 M), leaching temperatures (30, 50, and 70ºC), and stirring speed (200 rpm. 400 rpm, and 600 rpm) on Pb leaching from galena concentrate. The AAS (atomic absorption spectrophotometer) will be used for determination of Pb extraction.

2. Experimental and Procedures

2.1 Material and Apparatus

The chemicals applied in this study were 1.0–2.0 M acetic acid (CH_3COOH), 1.0–2.0 M sulfuric acid (H_2SO_4), and 0.5 M hydrogen peroxide (H_2O_2). This study used beaker glass, measured glass, filter paper, stirrer, analytical balance, hot plate, thermometer, muffle furnace, AAS and XRF equipment.

2.2 Roasting

The galena ores were obtained from a flotation plant of galena in West Java Province, Indonesia. The roasting process of galena ores was conducted at about 500–700ºC for 60 min. until a dark brown sample yielded. The chemical reaction (1) during roasting is as follows:

$$2PbS + 3O_2 \rightarrow 2PbO + 2SO_2 \quad (1)$$

During roasting, the galena was undergone oxidation to form lead oxide and toxic SO_2 gas. The roasting process was done in a chamber provided with an exhauster in order to remove the toxic SO_2 gas resulted from the chemical reaction above to avoid chamber pollution. After roasting and drying, a sieve shaker was used to filter the mineral powder to obtain the desired particle size, i.e., ≤ 200 mesh. Then, the galena concentrates in powder form obtained from rusting process with given particle size was prepared for leaching process to extract Pb. Fig. 2 shows the galena concentrate in powder form.

Fig. 2. Galena concentrate in powder form after roasting process

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2.3 Leaching

The leaching procedure is given as follows: 5g galena concentrate from roasting dissolved in acetic acid. A 0.5 M hydrogen peroxide was added to galena concentrate in acetic acid in a beaker glass. The leaching process of Pb from galena concentrate using acetic acid is a modified process from earlier study[8]. This procedure was repeated for galena concentrate in sulfuric acid. The given acid concentration was made following the simple dilution rule. i.e.

$$V_1 \times C_1 = V_2 \times C_2 \quad (2)$$

where:
V_1 = initial acid volume (ml)
C_1 = initial acid concentration (ppm)
V_2 = final acid volume (ml)
C_2 = final acid concentration (ppm)

During leaching process, the chemical reaction (3) referred to reaction of galena concentrate with acetic acid (CH_3COOH), while the chemical reaction (4) referred to reaction of galena concentrate with sulfuric acid (H_2SO_4). The flow diagram of this comparative study applying acetic acid and sulfuric acid as leaching agent for Pb extraction is shown in Fig. 3.

$$PbO + 2CH_3COOH \rightarrow Pb(CH_3COO)_2 + 2H_2O \quad (3)$$

$$PbO + H_2SO_4 \rightarrow PbSO_4 + H_2O \quad (4)$$

The leaching process was used to examine the effect of acid concentrations (1.0 M, 1.5 M, and 2.0 M), leaching temperatures (30, 50, 70°C), and stirring speed (200, 400, and 600 rpm).

2.4 AAS examination

An Analytik Jena with acetylene/nitrous oxide flame for atomizer of atomic absorption spectrophotometer was used to determine Pb concentrations in leaching solutions using acetic acid and sulfuric acid leaching agents. The lamp current of hollow cathode HCl lamp is set to be 2–20 mA,

2.5 XRF examination

A PANalytical, type Minipal 4 XRF (X-Ray Fluorescence) Spectrophotometer was used to analyze qualitatively and quantitatively chemical composition of galena ore.

3. Results and Discussion

3.1 Pb extraction from galena concentrate

Table 1 shows the data of Pb extract in terms of type of leaching acid, acid concentration, leaching temperature, and stirring speed. The Pb concentrations have been determined by AAS.

Table 1 Pb extraction with respect to leaching acid, temperature, and stirring speed. H_2O_2 oxidant. AAS assay

Acetic acid conc. molar	Temp. °C	Stirring speed rpm	Pb conc. ppm
1 M	50	200	19.66
1.5 M	50	200	29.63
2M	50	200	35.64
1 M	30	200	13.90
1 M	50	200	L8.09
1 M	70	200	23.92
1 M	50	200	19.66
1 M	50	400	25.97
1 M	50	600	30.74
Sulfuric acid conc. molar	**Temp. °C**	**Stirring speed rpm**	**Pb conc. ppm**
1 M	50	200	7.I8
1.5 M	50	200	11.46
2 M	50	200	17.63
1 M	30	200	5.16
1 M	50	200	7.18
1 M	70	200	11.21
1 M	50	200	5.67
1 M	50	400	5.93
1 M	50	600	7.18

3.2 Effect of acid concentration on Pb leaching

As shown in Fig. 4, acetic acid as leaching agent gives more effect on Pb leaching from galena compared to that of sulfuric acid. The AAS assayed Pb concentration in the leaching solution. Pb(II) ion and sulfate ion may form Pb-sulfate deposit or precipitate, therefore, less Pb(II) ion existence in leaching solution. In general, Table 1 shows that increase concentration of both leaching acids from 1.0 M to 2.0 M yielding increased Pb concentration in leaching solution. It is reasonable since the acid may withdraw the Pb from galena matrix to yield salt of Pb in the form of Pb acetate and Pb sulfate and therefore, the Pb in the form of Pb(II) ion. Furthermore, increase of leaching temperature from 50°C to 70°C causes increased Pb extraction for both acids at constant acid concentration and constant stirring speed. It is not surprising since increased temperature causes increased solubility of Pb(II) ion in leaching solution. Moreover, Table 1 shows increased stirring speed from 200 rpm to 600 rpm

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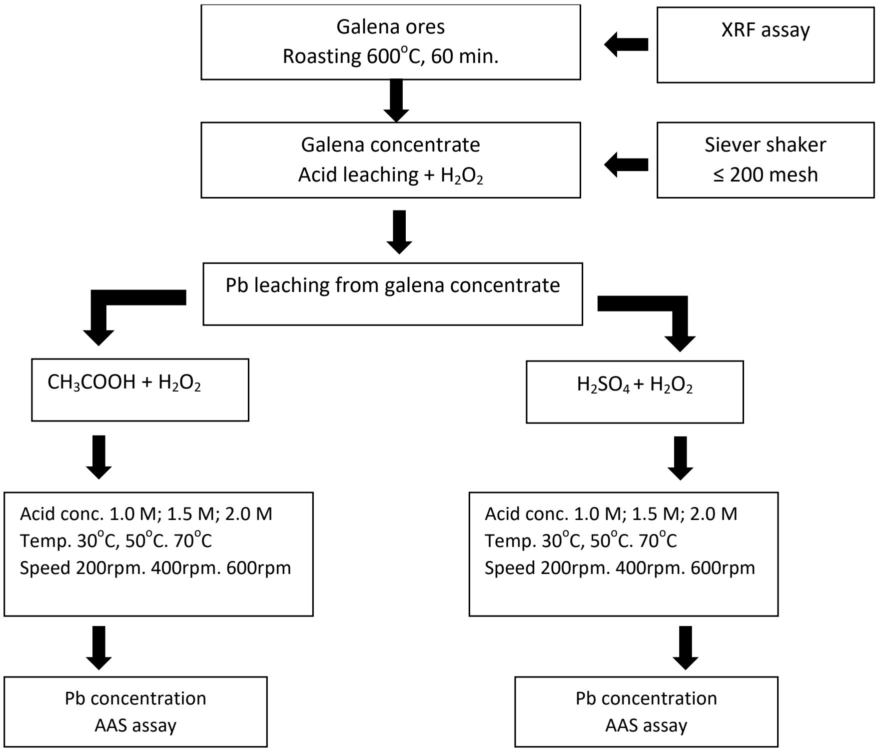

Fig. 3. The flowchart diagram of Pb leaching from galena concentrate in this study

causes increased Pb extraction for both acids, however, the increment is not too significant for sulfuric acid as leaching agent compared to that of acetic acid. It seems that thermal effect (temperature) is more dominant rather than mechanical effect (stirring speed) addressing to Pb extraction from galena matrix.

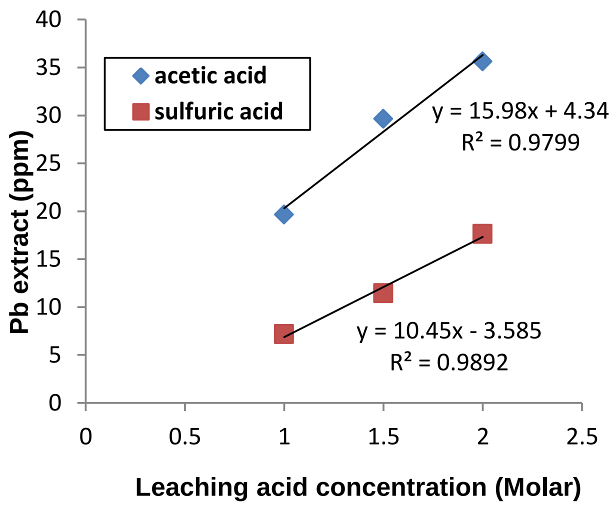

Fig. 4. Pb extraction from galena using acetic and sulfuric acids. 50°C. 200 rpm. H_2O_2 oxidant. AAS assay

The temperature effect (Fig. 5) gives less influence on Pb extraction from galena compared to the effect of acid concentration (Fig. 4). With regard to acetic acid on Pb leaching, an increase of Pb concentration from about 20 ppm to almost 40 ppm as shown by Fig. 4, while an increase of Pb concentration from about 15 ppm to about 25 ppm as shown by Fig. 5. The effect of acid concentration on Pb leaching almost doubled the effect of temperature with respect to acetic acid.

3.3 Effect of leaching temperature on Pb leaching

The Pb leaching effect using acetic acid due to both increase of acid concentration (Fig. 4) and temperature (Fig. 5) is greater than the leaching effect with sulfuric acid. As already mentioned above, the formation of Pb sulfate deposit caused lower concentration of Pb(II) ion in leaching solution applying sulfuric acid leaching.

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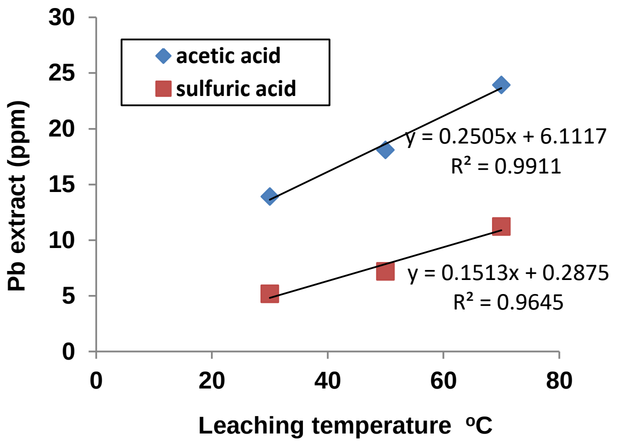

Fig. 5. Pb extraction from galena using acetic and sulfuric acids. Acid concentration 1.0 M, 200 rpm. H_2O_2 oxidant. AAS assay.

As shown by Fig. 4 and Fig. 5, the trend of Pb leaching from galena concentrate at given varied acid concentrations (1.0 M, 1.5 M. and 2.0 M) and given leaching temperature (30ºC, 50ºC, and 70ºC) show almost linearly. The range of linearity (Fig. 4 and Fig. 5) in the range of R^2 of 0.96–0.99, which is moderately linear both for given acid concentration and temperature effects on Pb extraction. The upper limit of acid concentration was made to be 2.0 M on the reason of chemical safety, while the temperature was limited to 70ºC due to energy reason. There is a consistence between concentrations of Pb extract and both effects of acid concentration and temperature applying either acetic acid or sulfuric acid as leaching agent.

3.4 Effect of stirring speed on Pb leaching

This article has also investigated the effect of stirring speed (200 rpm, 400 rpm, and 600 rpm) on Pb leaching applying acetic and sulfuric leaching acids, as shown by Fig. 6. Furthermore, Fig. 6 shows that effect of given varied stirring speed on Pb leaching from galena is not too significant particularly using sulfuric acid as leaching agent. As shown by other effects earlier (acid concentration and leaching temperature), the effect of stirring speed on Pb leaching from galena concentrate shows that acetic acid leaching extracted more Pb(II) ion from galena rather than leaching by sulfuric acid. This phenomenon is due to the formation of Pb sulfate precipitate. According to the rule of solubility, the solubility product (Ksp) of Pb sulfate is much smaller than that one of Pb acetate at given range of temperature. The linearity of stirring speed effect particularly for sulfuric acid leaching shows no significant effect on Pb extraction with varied given stirring speed (200, 400, and 600 rpm).

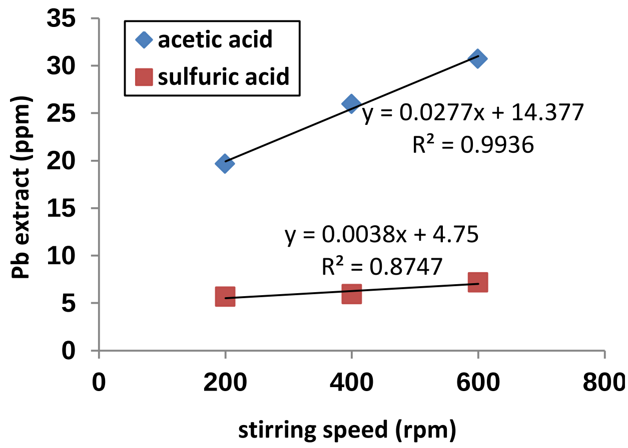

Fig. 6. Pb extraction from galena using acetic and sulfuric acids. Acid concentration 1.0 M, 50ºC. H_2O_2 oxidant. AAS assay.

3.5 XRF examination of galena ore

As mentioned earlier, galena mineral is commonly found with other minerals such as sphalerite, marcasite, pyrite, chalcopyrite[1,7]. Sphalerite is a sulfide mineral with chemical formula as (Zn, Fe) S that is commonly called as zinc blende. Table 2 shows that the result of XRF examination reported Fe (32.8%) and Zn (26.6%) together with Pb (13.9%) as transition elements in galena ores. Marcasite and pyrite, both are sulfide minerals with same chemical formula, i.e., FeS_2, however, they have different crystal structure. Marcasite has orthorhombic structure, while pyrite has cubic crystal structure. In addition, marcasite is less dense and lighter than pyrite, therefore, marcasite is more brittle. Chalcopyrite is a copper iron sulfide mineral (Cu_5FeS_4).

Several studies reported the flotation separation of galena from pyrite using serpentine depressant[11], isolation of galena from chalcopyrite using HEC depressant[12], and separation of galena from sphalerite using nitrogen functional groups[7]. Table 2 reported copper as 3.29% and sulfur as 4.8% related to the existence of chalcopyrite in galena ore. Thus, there are the important minerals in galena ores based on their concentrations. Furthermore, it is apparently that transition elements look dominant in galena ores (Table 2). It should be noted that rare earth metals (lanthanum, europium, and ytterbium) are also detected in galena ore (Table 2). Fig. 6 shows the XRF spectra of galena ore before roasting treatment.

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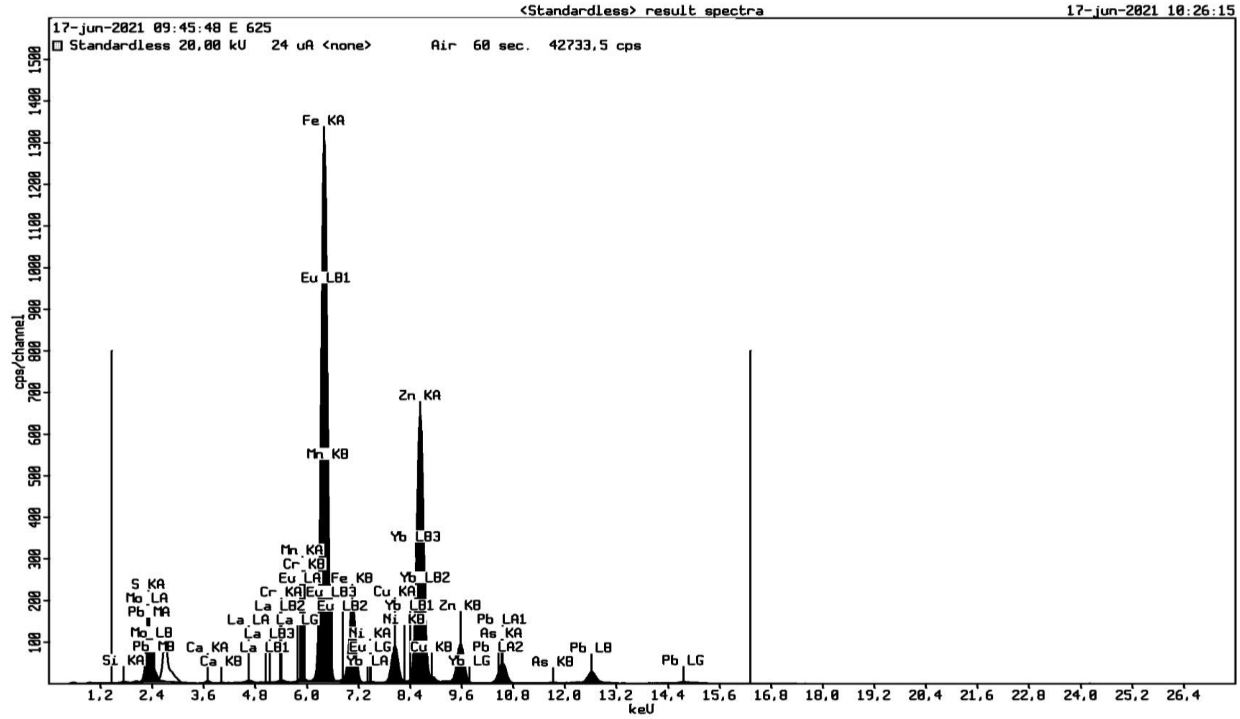

Fig. 6. XRF spectra of galena ore before roasting

Table 2. XRF examination of galena ores from West Java Province, Indonesia.

Chemical element	Content (%)
Si	1.0
S	4.8
Ca	0.28
Cr	0.095
Mn	0.13
Fe	32.8
Pb	13.9
Ni	0.05
Cu	3.29
Zn	26.6
As	0.78
Mo	16.0
La	0.03
Eu	0.3
Yb	0.1

4. Conclusions

Previous study reported Pb leaching from galena concentrate using expensive and toxic leaching agents. However, this study attempts to apply low-cost acids (acetic and sulfuric acids) as leaching agent for Pb extraction from galena concentrate. Further study needs modification on leaching parameters addressing to particle size, selection of oxidant, and leaching time to get results improvement.

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