

## The study of acid leaching and electrodeposition method to enhance Pb recovery from galena concentrate

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### Abstract

Galena is a potential source of lead (Pb), which has many used for lead batteries. Indonesia is a country with potential sources of galena mineral. This investigation showed Pb extraction of 0.274 % using acetic acid leaching method with 1.0 M acetic acid at 50°C from galena concentrate. This study showed remarkable enhancement of Pb extraction until 8.96 % applying electrodeposition method using 1.0 M acetic acid at 50°C. Varied acetic acid concentrations were applied for 1.0 M, 3.0 M, 5.0 M, and 7.0 M and varied temperatures were applied in this study for 50 °C, 65 °C, and 80 °C. The percentage of Pb extraction showed a consistency with the reduction of % Pb extraction with increasing acid concentrations and increasing temperatures. This findings is assumed to be related to exothermic PbS dissolution shifted to decreasing Pb(II) ion concentration and ion pair formation retarding Pb dissolution. The percentage of Pb from leaching process was determined by AAS (Atomic Absorption Spectrophotometer), while the percentage of Pb by electrodeposition method was determined from quantity of Pb deposit on cathode of an electrochemical cell. This investigation shows a significant breakthrough for enhancement of Pb recovery from galena mineral.

**Keywords:** Acetic Acid; Electrodeposition; Galena; Leaching; Pb

### 1 Introduction

Galena mineral has many found in Indonesia particularly in Aceh, North and West Sumatera, West and East Java. This study took galena ores as samples from Bogor, West Java. Galena as potential source of lead (Pb) is a mineral with its chemical formula as PbS (lead sulfide). In nature, galena is found in association with sphalerite mainly containing zinc sulfide (ZnS), marcasite and pyrite with chemical formula as FeS (iron sulfide), and chalcopyrite containing copper sulfide. Marcasite and pyrite are differed in crystal structures as marcasite with orthorhombic and pyrite with its cubic structure. Therefore, galena ores contains mainly lead, zinc, iron, and copper as major metals. Separation of galena from other minerals such as sphalerite, marcasite and chalcopyrite is commonly used differential flotation technique [1 – 3]. Galena is often found as veins in metamorphic deposits undergone hydrothermal processes related to upper limestone sedimentation, volcanic and granite igneous rocks, as well as alluvial deposit [4 – 6]. Fig. 1 shows galena ores, its color strongly affected by its association with other minerals, for example, if blue greenish an indication rich of copper, brown reddish attributed to iron rich content, and greyish indicated zinc and lead content.

As galena significant source for lead (Pb), many applications for Pb particularly for battery industry are used in automotive vehicles. Other applications of Pb as for pigment in painting, Pb alloy for joints, ballast in sailing boat, lead crystal glass, ammunition, and as additive in PVC isolator, as well as protecting layer in a reactor. Due to useful applications of Pb in industry, several methods of Pb recovery from galena and other minerals have been reported.

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**Figure 1** Galena mineral

Pb extraction from galena concentrate using several leaching acids in the existence of oxidant in hydrometallurgy route were reported [2, 6 – 9]. There are several routes corresponding with Pb recovery from minerals such as pyrometallurgy, hydrometallurgy, and electrometallurgy. Pyrometallurgy process yielded pollution effect due to toxic sulfur gas emission and high thermal energy, therefore, in the last decades many Pb recovery used combined routes of pyrometallurgy and hydrometallurgy, which is friendlier environmental [2 – 6].

This study attempts to apply electrodeposition method after Pb leaching from galena concentrate using acetic acid at several concentrations (1.0 M, 3.0 M, 5.0 M, and 7.0 M) at different temperatures (50 °C, 65 °C, 80 °C, and 95 °C). The concentrations of Pb leaching from galena concentrate are determined by AAS method. The percentage of Pb from the electrodeposition method is determined based on Pb deposit on the cathode.

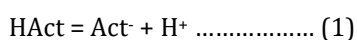
## 2 Experimental Method

Initially, a pyrometallurgical method was used by roasting galena ores in a furnace at moderately high temperature (600 °C) for about 60 minutes [10]. The pyrometallurgical route may yield toxic sulfur gases and not friendly environmental [7 – 8, 11]. Therefore, this study applied combination method using hydrometallurgical route, which did not cause air pollution, i.e. acid leaching to extract Pb from galena concentrate. After galena roasting, the ores were screened by a screen shaker to obtain galena ores with desired particle size of  $\leq 200$  mesh.

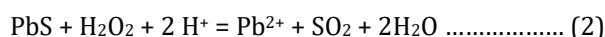
### 2.1 Pb leaching from galena concentrate

In the leaching method, the concentrations of acetic acid were varied (1.0 M, 3.0 M, 5.0 M, and 7.0 M) and the leaching process was conducted at different temperatures (50 °C, 65 °C, 80 °C, and 95 °C). All concentrations of Pb extraction were determined by AAS (Atomic Absorption Spectrophotometry) method. The leaching process used a beaker glass provided with a magnetic stirrer setting at 200 rpm and a hot plate to set the given temperature. Galena ores with given particle size ( $\leq 200$  mesh) in acetic solution with given concentration was placed in a beaker glass. A peroxide ( $\text{H}_2\text{O}_2$ ) was added in the leaching process to increase Pb extraction from galena concentrate.

The concept of chemical reactions in aqueous medium is described as follows:



Reaction (1) is the dissociation reaction of acetic acid (HAct) in aqueous solution.

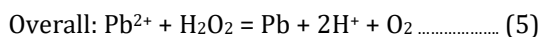
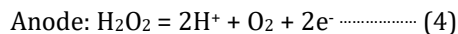
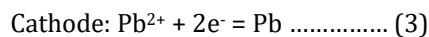


Supply of ion  $\text{H}^+$  due to dissociation of HAct in reaction (1) and the contribution of peroxide ( $\text{H}_2\text{O}_2$ ) in reaction (2) causes the reaction (2) shifted to right side, thus, the PbS (galena) may dissolve in stepwise. Hence, higher concentration of acetic acid and more contribution of strong oxidant ( $\text{H}_2\text{O}_2$ ) may increase solubility of galena to yield Pb(II) ion. The production of  $\text{SO}_2$  gas in reaction (2) is an advantage in order to dissolve galena to form Pb(II) ion. Many previous studies reported the usage of peroxide ( $\text{H}_2\text{O}_2$ ) in hydrometallurgical route for metallic recovery [6 – 8, 11].

## 2.2 Electrodeposition route for Pb precipitation in electrochemical cell

The electrodeposition route is an electrometallurgical route, which is also used in metal recovery [12 – 14]. Brar et al. [12] overviewed integrated bioleaching and redox process for copper recovery from waste resources, while Verbruggen et al. [13] reported arsenic codeposition from acidic copper sulfate solution using linear sweep voltammetry, and Whitworth et al. [14] reviewed minerals processing, metal extraction, and metal recovery from mining wastes due to high economic demand.

This study applied an electrochemical cell consisted of zinc cathode and carbon anode. The acetic acid containing Pb yielded from acid leaching was used as aqueous electrolyte. The electrochemical reaction is described as follows:



The cathode reaction is corresponding with reduction of Pb(II) ion to form Pb deposited at zinc cathode. The quantity of Pb precipitate at cathode is an indication of Pb recovery from galena concentrate. The existence of strong oxidant (H<sub>2</sub>O<sub>2</sub>) may enhance Pb extraction from galena. This matter is in agreement with previous reports [6 – 8, 11].

In nowadays, electrochemical studies have broad applications since it gives a lot of benefits including Al waste beneficiation to yield energy in batteries [15 – 16]. In the electrodeposition process the cathode weights were measured before and after electrodeposition process taken for 60 minutes for each given temperature (50°C, 65°C, and 80°C) and each acetic acid concentration (1.0 M, 3.0 M, 5.0 M, and 7.0 M). 12 mA current was used in this investigation.

## 2.3 AAS examination

An Analytic Jena with acetylene / nitrous oxide flame for atomizer of Atomic Absorption Spectrophotometer was used to determine Pb concentrations in leaching solutions using acetic acid leaching agents. The lamp current of hollow cathode HCl lamp is set to be 2 – 20 mA. In this study, the AAS examination was used to analyze Pb in leaching and electrodeposition processes.

## 2.4 XRF examination

A PANalytical, type Minipal 4 XRF (X-Ray Fluorescence) Spectrophotometer was used for sample characterization. A screen shaker was used to get galena sample with given particle size ( $\leq 200$  mesh) after roasting galena ores in an oven for 600°C about an hour, The galena ores was obtained from Bogor Residence in West Java Province and stored in Galena Maju Karya Mandiri company. 20g sample with  $\leq 200$  mesh was used for XRF examination. The XRF examination in laboratory was applied to determine concentrations of several representative elements, such as iron, copper, zinc, molybdenum, and lead.

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## 3 Results and discussion

### 3.1 XRF characterization

The XRF spectrometry only analyzed representative elements (Fe, Cu, Zn, Mo, and Pb) as already mentioned above. In fact, the galena sample contains many other minor elements that can be viewed as impurities. As seen in Table 1, although iron and zinc are the mayor elements and lead at the third position, however, galena ore is the potential source of lead in nature. Since galena is the main source of lead, Tan et al. [17] extracted Pb from concentrate mixture of galena and sphalerite using redox reaction. Since this study used XRF for elemental analyzes, Hu et al. [18] analyzed Pb in small quantity in Pb – Zn carbonate in a mixture of galena and sphalerite applying ICP – MS (Inductive Coupled Plasma – Mass Spectrometry).

**Table 1** Concentration of elements in galena sample by XRF.

No	Elements	Concentration (%)
1	Fe	32.8
2	Cu	3.29
3	Zn	26.6
4	Mo	16.0
5	Pb	13.9

### 3.2 Pb determination in acid leaching method

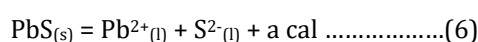
Since the galena sample for AAS examination taken as 20 g and the Pb content in galena was examined by XRF as 13.9 % (Table 1), therefore, the galena sample used for AAS was obtained to be 13.9/100. 20 g = 2.78 g. The atomic weight of Pb (lead) and S (sulfur) from Periodic Table are recorded as 207.2  $\approx$  207 amu and 32.065  $\approx$  32 amu, respectively [19]. Thus, the molecular weight of PbS is found as 239 amu (round off). Thus the weight of Pb in PbS is found to be (207/239) 2.78 g = 2.41 g [19].

The volume used for AAS was taken for 100 ml. At 50°C and 1.0 M acetic acid, the AAS result for Pb using acid leaching method was measured as 65.95 mg/l. Thus, the Pb extraction in leaching method was found to be (100/1000) (65.95/1000) (100/2.41) = 0.274 % using 1.0 M acid and 50°C. Using the same calculation procedure, the results of other Pb extraction obtained from acid leaching with varied temperatures (50°C, 65°C, and 80°C) and varied acetic acid concentration (1.0 M, 3.0 M, 5.0 M, and 7.0 M) that are shown in Table 2.

**Table 2** AAS result of Pb concentrations (mg/l) and Pb extractions from galena (%). Acetic acid leaching method at varied acid concentrations (1.0 M, 3.0 M, 5.0 M, and 7.0 M) and temperatures (50°C, 65°C, and 80°C).

Acetic acid concentration	Leaching temperature (°C)	[Pb] mg/l AAS	Pb extract (%)
1.0 M	50	65.95	0.274
	65	7.64	0.032
	80	6.46	0.027
3.0 M	50	37.14	0.154
	65	5.87	0.024
	80	4.68	0.019
5.0 M	50	26.90	0.112
	65	4.65	0.019
	80	3.96	0.016
7.0 M	50	11.24	0.046
	65	2.93	0.012
	80	2.30	0.010

Table 2 shows that the Pb extraction decreased consistently with increasing temperatures (50°C, 65°C, and 80°C) at each acetic acid concentration (1.0 M, 3.0 M, 5.0 M, and 7.0 M). The solubility reaction of galena is an exothermic reaction (6) as shown below:



The equilibrium reaction of PbS is an exothermic reaction. According to Le Chatelier's principle, if the temperature of system increased, the equilibrium reaction (6) will shift to the left side where system consumes more energy resulting

less concentration of Pb(II) ion in the solution [20]. Hence, it is reasonable with the phenomena shown in Table 2 that increased temperature yielding decreased Pb(II) ion concentration. In addition, increased temperature may cause acetic acid volatility resulting less acetic acid in solution, however, this effect is not too significant. This finding also shows that Pb extraction reduced consistently with increased acid concentration at same temperature. Higher acid concentrations resulting the tendency of ion pair formation of acetate ion and hydrogen ion retarding Pb (II) ion dissolution.

### 3.3 Pb determination in electrodeposition process

The electrochemical cell was applied for electrodeposition of lead at cathode. The electrochemical reactions are shown above by reactions (3) to (5) occurred in electrochemical cell with zinc cathode and carbon anode.

The electrodeposition process took 60 minutes and the result was the increased weight of cathode due to lead deposition on cathode surface. Table 3 shows the electrodeposition results in terms of increased weight of cathode due to lead deposition at varied acetic acid concentrations (1.0 M, 3.0 M, 5.0 M, and 7.0 M) and varied temperatures (50°C, 65°C, and 80°C). The difference of cathode weight was obtained as final cathode weight due to lead deposit minus initial cathode weight before lead deposition due to electrodeposition process as shown in Table 3. For simplicity, this investigation has assumed that the electrodeposition process caused only lead precipitation on cathode surface and other impurities were neglected.

**Table 3** Different weight of cathode (g) due to lead deposit on cathode surface Electrodeposition method at varied concentrations (1.0 M, 3.0 M, 5.0 M, and 7.0 M) and temperatures (50°C, 65°C, and 80°C) for 60 minutes.

Acetic acid concentration	Temp (°C)	Initial cathode weight (g)	Final cathode weight (g)	Difference cathode weight (g)
1.0 M	50	9.510	9.726	0.216
	65	10.340	10.363	0.023
	80	9.990	10.010	0.020
3.0 M	50	9.730	9.845	0.115
	65	10.293	10.311	0.018
	80	9.810	9.824	0.014
5.0 M	50	9.712	9.792	0.080
	65	10.250	10.263	0.013
	80	10.200	10.211	0.011
7.0 M	50	9.640	9.693	0.053
	65	10.200	10.208	0.008
	80	10.150	10.156	0.006

The difference of cathode weight ( $\Delta$ ) is the lead deposit on cathode surface due to electrodeposition process. As already mentioned above, for 20 g sample used in this investigation, the PbS was found as 13.9 % (data Table 1 related to XRF analysis), i.e. 2.78 g. As not repeating the statement in Pb determination using leaching method, the weight of Pb in PbS is found to be  $(32/239) 2.78 \text{ g} = 2.41 \text{ g}$  [19]. The % of Pb extraction obtained from electrodeposition method is based on the difference of cathode weight (the fifth column in Table 3) defined as Pb deposit on cathode surface referred to initial Pb in galena, i.e. 2.41 g. For example,  $\Delta$  weight 0.216 g for acid concentration of 1.0 M and at 50°C shown in Table 3, the percentage of Pb extraction is found to be  $(0.216/2.41) 100\% = 8.96\%$ . Using the same calculation procedure, the results are shown in Table 4.

Table 5 shows the comparison of percentage of lead extraction between the acid leaching and the electrodeposition method at given varied acid concentrations and temperatures. Apparently, that the electrodeposition method elevated percentage of Pb extraction at overall given acid concentrations and temperatures. The electrodeposition method involves with reduction oxidation process, while the acid leaching is only related to acid penetration breaking the Pb (lead) atoms and S (sulfur) atoms in PbS matrix. The acetic acid penetration in PbS (galena) matrix is apparently not as

strong as the Pb(II) ion reduction to produce Pb in the cathode. The electrical effect in the electrodeposition method influenced strongly enough to dissolve PbS in acetic acid solution to form Pb(II) positive ion and sulfur negative ion. Addressing to % of Pb extraction, a similar trend with the acid leaching method is also shown for the electrodeposition method applying same values of varied acid concentrations and temperatures. The reason of the trend is also the same, which is related to equilibrium shift of exothermic PbS dissolution followed the Le Chatelier's Principle and ion pair formation hindering dissolution of PbS in acetic acid solution as the significant effect [20].

**Table 4** Percentage of Pb extraction using electrodeposition method at varied concentrations (1.0 M, 3.0 M, 5.0 M, and 7.0 M) and temperatures (50°C, 65°C, and 80°C) for 60 minutes. 12mA.

Acetic acid concentration	Temp (°C)	Difference cathode weight (g)	% Pb extract
1.0 M	50	0.216	8.96
	65	0.023	0.95
	80	0.020	0.83
3.0 M	50	0.115	4.77
	65	0.018	0.75
	80	0.014	0.58
5.0 M	50	0.080	3.32
	65	0.013	0.54
	80	0.011	0.46
7.0 M	50	0.053	2.20
	65	0.008	0.33
	80	0.006	0.25

**Table 5** Percentage of Pb extraction of acid leaching and electrodeposition method at varied acid concentrations (1.0 M, 3.0 M, 5.0 M, and 7.0 M) and temperatures (50°C, 65°C, and 80°C).

Acetic acid concentration	Temp (°C)	% Pb extract acid leaching	% Pb extract electrodeposit
1.0 M	50	0.274	8.96
	65	0.032	0.95
	80	0.027	0.83
3.0 M	50	0.154	4.77
	65	0.024	0.75
	80	0.019	0.58
5.0 M	50	0.112	3.32
	65	0.019	0.54
	80	0.016	0.46
7.0 M	50	0.046	2.20
	65	0.012	0.33
	80	0.010	0.25

## 4 Conclusion

The electrodeposition method has increased remarkably the percentage of Pb extraction from galena concentrate compared to that of acid leaching method commonly used for metal extraction from minerals in mining field. The electrical current in the electrochemical cell has shown strong effect on PbS dissolution in acetic acid solution at given acid concentrations and temperatures. This study can be developed for future application in relation to metal recovery from mineral ores.

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## Compliance with ethical standards

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### *Disclosure of conflict of interest*

This statement confirms that there is no any conflict of interest between authors corresponding to this manuscript.

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