Acid Mine Drainage in Abandoned Mine

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Abstract-Acid mine drainage (AMD) in abandoned mining operations related oxidation of sulfide mineral affording an acidic solution that contains toxic metal ions. Hence acidic water that flow into the stream had potential health risks to both aquatic life and residents in the vicinity of the mine. Study will be conduct to investigate water quality and AMD characteristics which are pH value of the stream or discharge area, mineral composition in the rock and neutralization value of the rock in AMD mining area. Result shows that pH value of water in Kg. Aur, Chini and Sg. Lembing are acidic with value of 2.81, 4.16 and 3.60 respectively. Maximum concentrations of heavy metals in the study area are: Pb (0.2 mg/L), Cd (0.05 mg/L), Zn (5.1 mg/L), Cu (5.2 mg/L), Mn (10.9 mg/L), Cr (0.2 mg/L), Ni (0.2 mg/L), As (0.005 mg/L) and Fe (202.69 mg/L). Prediction of acid formation using acid-base calculations from all samples shows high potential acid production between 22.84-2500.16 kg $CaCO_3$/tonne. The ratio of neutralization (NP) with acid potential (APP) shows a very low value (ratio < 1) Sg. Lembing (0.02), Chini (0.08) and Kg. Aur (0.81). The potential value of this acid production exceeds the neutralization potential value in the mine. It is inferred that AMD is being produced in the Sg Lembing, Cini and Kg Aur. It is recommended that an Environmental Impact Assessment of AMD must be carried out before the mine is reopened.

Keywords- Acid Mine Drainage; Acidic Water; Heavy Metals; Acid-Base Accounting

1. INTRODUCTION

Acid Mine Drainage is quite new environmental problem in Malaysia [22]. Mineral processing of hard rock, metal ores (e.g. Au, Cu, Pb, Zn) and industrial mineral deposits (e.g. phosphate, bauxite) involves the size reduction and separation of the individual minerals. Consequently, the end products of ore or industrial mineral processing are concentrated of the sought-after commodity and a quantity of residue wastes known as tailings [35].

Weathering of tailing contain pyrite mineral (FeS_2) and sulphide minerals can contribute to Acid Mine Drainage AMD [19]. The potential presence of acidic environment when these rocks contain metal sulphide, especially iron disulphide (FeS_2) which is pyrite, and or marcasite, they become oxidized by air and water producing Fe^{2+}, H^+ and SO_4^{2-} ions. When these ions get into the solution, sulphuric acid is produced (AMD). Many researcher use Acid Base Accounting (ABA) as a screening tool, which for many mining situations provides an adequate prediction tool [18]. Since the methods for

ABA were published in [31], ABA soon became the method of choice and began being used as screening tool in other countries as well [15, 36]. tatic tests involve determination of net-acid production potential (NAPP) and/or net acid generation (NAG) values (expressed in either kg H_2SO_4/t as used here or kg $CaCO_3$/t). NAPP is calculated through acid base accounting (ABA) procedures. Positive NAPP values indicate a potential to generate acid. NAG values are obtained through the reaction of pulverised sample with H_2O_2 to accelerate sulphide oxidation with the resulting liquor titrated with NaOH and NAG (kg H_2SO_4/t) values calculated. Its environmental impact, however, can be minimized at three basic levels: through primary prevention of the acid-generating process; secondary control, which involves the deployment of acid drainage migration prevention measures; and tertiary control, or the collection and treatment of effluent [2]. Obejctive of this study is to investigate water quality and AMD characteristics which are pH value of the stream or discharge area, mineral composition in the rock and neutralization value of the rock in AMD mining area. Scope of this study is to analysis samples of seepage water collected in four sites in the Bukit Ibam mining area will be analysis on heavy metal elements (Cd, Pb, Zn, Cu, Mn, Ni, As and Cr). Then to determination of all the in-situ (pH, temperature, conductivity, TDS, salinity) in the water sample. Determination of rock neutralization and toxicity of tailing content for AMD occurrence.

2. MATERIALS AND METHODS

2.1 Mining Site Sampling Procedure

The studies were conducted at four abandoned mining sites (Table 1). These abandoned mining sites were choosing based on potential sulphide minerals contain in these locality. Samples such as water, rock, tailing and soil were collected. Water analysis of in-situ parameters such as pH, dissolved oxygen, potential production, temperature, salinity and conductivity were measured by using Quality meter YSI 656. For water sampling, one liter of water was collected from each sites and preserved by adding ultra-pure acid and then the samples were stored in a cooler box of approximately 4°C before being transported to the laboratory for further analysis. The waste rock samples were collected and put in plastic bag for each site. Meanwhile, tailings and soils were collected from nine sites and for each sampling site, 2-3 kg of the samples were packed and sealed in prewashed plastic bags to maintain its characteristics.

Table 1. The parameter and characteristic of the mining sites.

Mining Sites	GPS coordinate	Type of minerals	Status until 2013	Types of site
Kg. Aur	N 03'08'11.8"/102'57'38.9"	Hematite	Abandoned	Stream
Chini	N03'23'34.9"/E102'56'46.1"	Manganese	Abandoned	Pond
Sungai Lembing	N03'54'35.2"/E103'01'51.1"	Tin	Abandoned	Stream
Kuari JKR Kg. Awah	N03'29'51.8"/E102'32'30.4"	Andesite	Abandoned	Pond

2.2 Analysis of Water from Mining Site

The water analysis procedures are obtained from following steps. Firstly, the AMD solutions were filtered through a membrane of 0.45µm pore to get the clear effluent before tested. Heavy metal elements (Cd, Pb, Zn, Cu, Mn, Ni, As and Cr) and major cation (Ca, Mg, K, Na as well as Fe) were determined by using

Inductively Coupled Plasma Mass Spectrometry (ICPMS) model of Perkin Elmer Elan900. Meanwhile, for major anions (sulfate and chloride) analysis was conducted by filtered the solution through 0.2 µm pore size. Metrohm 850 Professional Ion chromatography with chemical suppression was performed to measure sulfate and chloride anion contents

2.3 Analysis of Rock and Soil

Physical-chemical tests such as pH, neutralization potential, total sulfur of rock and soil were determined using standard method of USEPA [31]. A representative 250 g portion was prepared by splitting, dried and pulverized to less than 0.25mm for the titration test. The pH of the samples obtained by adding 10g of soil and mixed with 25 ml distilled water and then stirred with spatula continuously. The pH value was measured by using Hanna measurement pH instrument with the ratio of water and sample was 1:2.5. The pH meter calibrated with buffer solution at pH:4 and pH:7. Total sulfur in the sample represents the maximum potential of acidity followed by E1915-97 method (ASTM 2000a) (Smith et. al 1974).

2.4 Analysis of Tailings

Toxicity Characteristic Leaching Procedure (TCLP) for these soils was determined using USEPA Method 1311. After sampling, holding time for metal have 180 days until leaching, except for mercury where leaching must start within 28 days. 10 g of soil was mixed with glacial acetic acid (adjusted to pH 4.93 with 1 N NaOH) at 20 L kg^{-1} in polypropylene bottle. Then the bottle was agitated with junior orbital shaker at 200 rpm for 18 h. After 18 h, the liquid was filtered through 0.45 µm membranes, acidified with concentrated nitric acid and stored in amber vials at $4^{o}C$. TCLP tests are completed in a short (18-hour) contact duration. The metal concentrations such as As, Ba, Cd, Cr (VI), Pb, Se, Ag, Hg were determined by inductively coupled plasma mass spectroscopy (ICP-MS).

2.5 Prediction of AMD

Static tests are geochemical analyses of sulfide waste which are used to predict the potential of a waste sample to produce acid. A significant part of the static test is Acid Base Accounting (ABA), which refers to the numerical data used to predict acid generation. The three components of the ABA are: (i) determination of acid production; (ii) determination of acid consumption; and (iii) calculation of net acid production or consumption using the data from (i) and (ii). Here, (i) Determination of acid production from tailings samples deals with the measurement of total sulfur, which is the main goal of research. Acid production potential (APP) based on the assumption that two moles of acid will be produced for each mole of sulfur present in the mine waste. The total sulfur content in percent is multiplied by 31.25 to yield the APP in units of tons acidity as $CaCO_3$ per 1000 tons rock (or equivalently, kg $CaCO_3$/metric ton). Neutralization potential (NP) test was performed to determine the quantity of acid-consuming minerals in a sample [31]. The NP test started with “a fizz test” which was done by adding one or two drops of 25 % HCl solution to about 0.5g of sample and observing the degree of effervescence. The fizz test was used to rank sample as none, slightly, moderate and strong. The appropriate amount and concentration of HCl was added to 2.0g of sample in a flask and placed on a hot plate until the sample was just beginning to boil. The flask was taken off the heat and swirled intermittently until no more effervescence was observed. More distilled water was added to make a total volume of 125 ml. After heated for 1 minute, the sample was left to cool down above room temperature before titration. The sample is then titrated with 1.0 N sulfuric acid until a pH of 7.0 is reached. The pH 7 was selected since this pH represents the amount of acid that a mine waste could neutralize while maintaining drainage pH in a range that meets quality standards. Prediction of acid is determined by the Net Neutralization Potential (NNP), which is the difference between these values (NNP = NP – APP). Typically, this difference is initially assessed by acid– base accounting (ABA) to determine the net acid production potential (NAPP) of a sample. ABA test was developed by [29] and subsequently modified by [31] to evaluate the acid producing capacity of coal mine wastes. [31] indicated that waste would produce acid if and only if NNP was less than -5 kg $CaCO_3$/ ton.

3. RESULTS

3.1 Characterization of AMD Samples

Table 2 indicates the results of pH, conductivity (EC), potential production (Eh), temperature (T), total dissolved solid (TDS), dissolved oxygen (DO) and salinity. Figure 1 show AMD is in acidic phases at pH 2.81- 4.16 for Kampung Aur, Sungai Lembing and Chini respectively. The highest reading of D.O was recorded at Kg. Besul at 10.06 mg/L and the lowest value recorded at Kg. Aur (7.45 mg/L). The value of D.O for water pollution index in clean water class V is more than 1 mg/L and all mining sites were considered safe due to the value of dissolved oxygen. The potential production (Eh) showed a high value were recorded at Kg. Aur, Chini and Sungai Lembing (153.0 mV, 195.8 mV and 244.0 mV) respectively while electrical conductivity was recorded between 438.0 µs/cm to 1699.67µs/cm in some places respectively. Table 3 illustrates the average of concentration of heavy metals such as Pb, Cd, Zn, Cu, Mn, Cr, Ni and As obtained from all mining sites and calculated concentration is shown in Figure 1. The concentration of lead, Pb(II) found very high at Kg. Aur upper (0.215 mg/L) while the concentration of cadmium and zinc were highest recorded at Sungai Lembing (0.047 mg/L and 5.07 mg/L respectively). AMD was strongly acidic and contained significant levels of metal ions, especially Fe.

The acidic characteristic of the AMD results from the percolation of water through sulfide minerals generally pyrite, which oxidizes and dissociates when in contact with air and water. The concentration of Fe was found very high at Kg Aur (bottom part) of 202.69 mg/L, Kg Aur (upper part) of 129.43 mg/L, while the lowest content of Fe found at Batu Malim of 0.05 mg/L. There were no significant differences between mine waters for the concentration of kalium while other metals (Pb, Zn, Cr, Cd, Cu, Ni and As) show significantly differences in terms of concentration. The lead and chromium exceeded for concentration of standard WQA 2000 in Kg. Aur, Sungai Lembing, Selinsing and Chini and the concentration of cadmium in Sungai Lembing exceeded the standard too. The content of heavy metal at Kg. Aur and Sungai Lembing displays very low concentration of Cu but high content of Mn. The mean values for Mn was higher at Chini with concentration of 36.91 mg/L. The anion values of sulfate were found maximum at lower part of Kg Aur (5180.86 mg/L) while very minimum at Cheroh (20.22 mg/L). Meanwhile for chloride was found very high at lower part of Kg Aur (48.56 mg/L) but the concentration at Sungai Lembing was very low (1.21 mg/L).

3.2 AMD Prediction

Table 4 lists AMD prediction results using acid-base accounting method for each mine sites. The values of pH identified the host rock of each mining sites. Kg. Aur has pH value 2.34 which host rock is hematite and Chini with pH value of 2.00 which host rock is manganese. Only five mining sites have sulfur content which are Chini, and Sungai Lembing with total values of APP of 924.27 kg $CaCO_3$/ton and 2500.16 kg $CaCO_3$/ton respectively. Analyses of rock samples from other sites which are JKR Kg. Awah not detected any sulfur content. From total sulfur analysis, the highest value of total sulfur content recorded at Sungai Lembing which are 2500.16 kg $CaCO_3$/ton. The amount of neutralizing bases, including carbonates, present in overburden materials is found by treating a sample with a known excess of standardized hydrochloric acid [29]. The values of neutralizing potential (NP), obtained from the titration test in decreasing order, Chini > Kg Aur > Sungai Lembing were calculated as 337.43 kg $CaCO_3$/ton , 41.18 kg $CaCO_3$/ton and 7.32 kg $CaCO_3$/ton respectively. Meanwhile the classification of acid-bases for rock walls were found at low values of NP/APP at Kg. Aur, Chini and Sungai Lembing of 0.81, 0.37 and 0.003 respectively..

3.3 Toxicity of Tailing and Soil

The toxicity characteristic leaching procedure (TCLP) test is used to determine the mobility of toxic contaminants present in waste materials and to define hazardous wastes under the Resource Conservation and Recovery Act (RCRA), 1984). The results from TCLP experiment showed that the concentration of As leached from sediment in the Kg. Aur bottom samples after an 18h extraction time was 7.955 mg/L, which is exceeding the current maximum US EPA TCLP value for

As. This would be characterized as a hazardous waste. The comparison of the sample between sediment in the Kampung Aur (bottom part) and tailing in the Sungai Lembing show different result of TCLP.

Table 5 identified heavy metals content with the highest concentration of Pb (17.42 mg/L) at Sungai Lembing tailing and the lowest concentration found at Kg. Aur (bottom part), which is 1.21 mg/L. The concentration of Cd found in Sungai Lembing tailing was 1.3 mg/L and soil in Sungai Lembing showed below the limitation value of concentration.

Table 2: In situ water quality parameters of mine water.

Mining Sites	Temp. (^oC)	pH	DO (mg/L)	Eh (mV)	Salinity	Conductivity (μs/cm)	TDS (g/L)
Kg. Aur	29.9	2.81	7.45	244.0	1.46	1699.67	1.57
Chini	29.7	4.16	8.05	195.8	0.20	312.00	0.27
Sungai Lembing	29.6	3.60	8.36	153	0.09	135.33	0.12
Kuari JKR Kg. Awah	33.3	7.89	9.27	-37.4	0.16	260.00	0.22

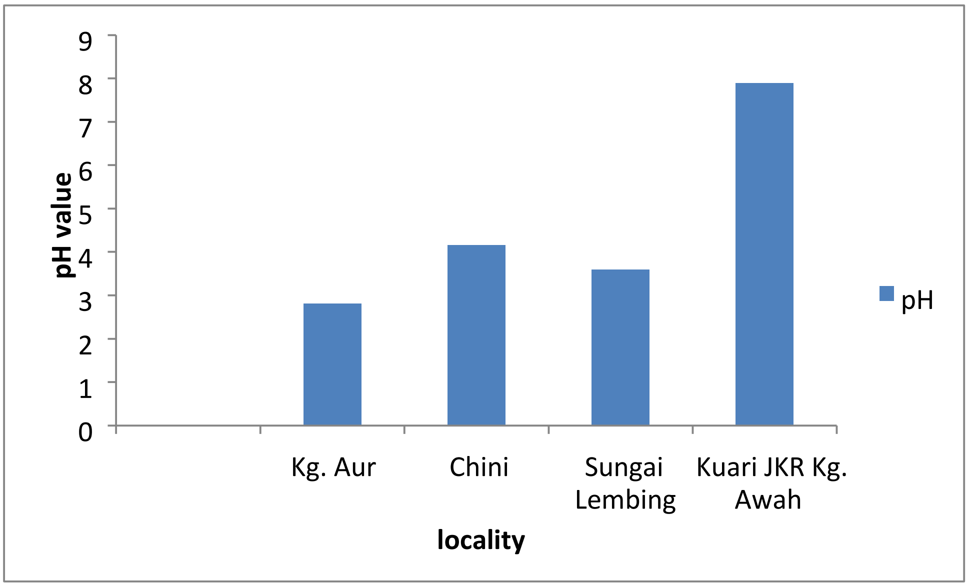

Figure 1: pH value show AMD is in acidic phases at pH lower than 7.

Table 3: The concentration values of heavy metals in mine water as compared to EQA and WQS standard.

Locality/ Symbol	1	3	5	8	Environment quality act	Water quality standard
Element (mg/L)/ Locality	Kg Aur Upper	Sungai Lembing	Chini	Kuari Kg Awah	EQA 1974	WQS 2000
Pb Average n=9	0.215 (0.199-0.22)	0.102 (0.098-0.103)	0.040 (0.037-0.047)	0.000 (-)	0.1	0.01
Cd Average n=9	0.022 (0.012-0.025)	0.047 (0.044-0.05)	0.018 (0.015-0.019)	0.000 (-)	-	0.03
Zn Average n=9	1.005 (0.998-1.00)	5.070 (4.77-5.554)	1.685 (0.999-1.037)	0.000 (-)	2.0	0.3
Cu Average n=9	1.185 (0.988-1.100)	5.232 (4.978-5.55)	0.154 (0.138-0.197)	0.003 (0.001-0.005)	0.2	-
Mn Average n=9	2.157 (1.922-2.00)	10.945 (10.14-11.70)	36.914 (33.58-39.24)	0.006 (0.001-0.011)	-	0.1
Cr Average n=9	0.026 (0.015-0.029)	0.0182 (0.013-0.027)	0.009 (0.006-0.012)	0.010 (0.005-0.015)	0.05	0.05
Ni Average n=9	0.036 (0.026-0.037)	0.126 (0.120-0.131)	0.025 (0.022-0.027)	0.002 (0.002-0.003)	0.2	-
As Average n=9	0.003 (0.001-0.004)	0.002 (0.002-0.002)	0.003 (0.002-0.003)	0.001 (0.0005-0.001)	0.05	0.01
FeAverage n=9	129.433 (115.20-130.00)	1.203 (0.974-1.532)	0.269 (0.188-0.984)	0.142 (0.118-0.165)	1.0	0.3
SO_4^{2-}	3179.6	75.67	48.94	65.86	0.5	250
Cl^-	21.52	1.21	2.36	2.38	2.	250

Table 4: The AMD prediction results of Acid-Base Accounting method for each mine sites .

Locality	Total of APP (kg $CaCO_3$ /ton)	Total of NP (kg $CaCO_3$ /ton)	NPP (NP-APP)	NP/APP	pH	Samples
Kg. Aur	50.79	41.18	-9.61	0.81	2.34	Hematite
Chini	924.27	337.43	-586.84	0.37	2.00	Manganese
Sungai Lembing	2500.16	7.32	-2492.84	0.003	2.90	Tailings

Table 5: The concentration of As, Cd, Cr and Pb for TCLP test.

TCLP	Sungai Lembing soil (mg/L)	Sungai Lembing Tailing(mg/L)	Kg. Aur bottom (mg/L)	USEPA (mg/L)
Arsenic	1.86	0.814	7.955	5
Cadmium	0.319	1.3	0.096	1
Chromium	0.213	0.35	0.223	5
Lead	3.864	17.417	1.207	5

4. DISCUSSION

Based on the result in table 2, the pH values recorded very low at Kg Aur, Chini and Sungai Lembing. Meanwhile, Kg. Aur, Chini and Sungai Lembing have the highest value of Eh showed solution that accept more proton and undergoes an oxidation process. According to [27], the low pH of discharge mine water results in the further dissolution of minerals and release of toxic metals, when it allowed getting discharge into other water bodies. The value of pH can be as low as 2 and continues to be an important water pollution problem in mining industry around the world [27, 32].

The conductivity of the water in the studied was found high. This high conductivity in the water is an indication of its effect on the water quality. Changes in conductivity were not always coincident with changes in Eh and pH, indicating that conductivity may be a more sensitive tool for locating specific zones focus on sulfate reducing bacteria activity [21]. TDS indicates the general nature of water quality or salinity. Based on TDS classification, it can be said all water in the mining area except at Batu Malim where brackish water. Maximum value of sulfate ions at lower part of Kg. Aur exceeds WHO (2000) limited set at 250 mg/L. For example, as a result of weathering of oxidized sulfide scarlet precipitation can be found up to a thousand meters from the AMD polluted river [16]. An excess of chloride ion in water is usually taken as an index of pollution. The high chloride content of water may have originated from natural sources such as rainfall and the dissolution of fluid inclusions.

Fe, Na, K, Ca and Mg were the major cations for the current study and showed significant differences between all samples. Due to the lower pH, the higher oxidation process, hence value of Fe will be higher. Fe was found highest in areas with low pH and found low in areas with high pH. [13] stated due to the low pH, the solubility of the toxic metals contained in could be attributed to the oxidation of H_2S and Fe(II). Calcium and sulfate concentration decreased as the precipitation proceeded. Precipitation using alkaline reagents for this research is the most widely used treatment method for removing metals as hydroxides [4, 19]. The decrease in the chloride concentration probably could be ascribed due to the formation of some metal chloride. Thus, water reservoir in Chini has high value of Mn compared to others. Acidic water facilitates the movement of Mn that easily soluble in acidic condition. In another place, pH values closer to 9.0, manganese ions precipitate as manganese hydroxide, $Mn(OH)_2$ allowing its removal [9]. The concentration of Ni and As were found safe for all water samples.

Prediction of AMD was calculated from APP and result recorded from table 5. The value of NP/APP is below than 1, interpreted as high potential for acid production. Kg. Aur, Chini, Selinsing and Sungai Lembing have been identified as having high acid generation potential (50.79 – 2500.16 kg $CaCO_3$/ton) compare to neutralization potential. ABA considers two factors, total S and NP, assumed to represent FeS_2 and $CaCO_3$ [31]. Total S can consist sulfide, sulfate, and organic sulfur components, and acid can be produced by each of these. Pyrite generally is the acid producer and [8] have shown that the dissolution of iron and aluminium-sulfate minerals and the subsequent hydrolisis of iron and aluminium can produce substantial quantities of acidity. [14] have suggested that some forms of organic S also may be acid-producing. Although there were many pyrite minerals in the mining site of Bukit Ibam, AMD did not occur as they were naturally neutralized by calcite minerals. Previous studies in Penjom showed that drainage will remain neutral to basic if the rate of acid consumption equals or exceeds the rate of acid production [23]. [30] evaluated if the value of NNP is less than zero, the acid-producing potential of the rock exceeds its neutralization potential and if mined, therefore would be expected to produce AMD. Four localities except Bukit Ibam showed negative values of NPP, then the potential exist for the waste to form acid as suggested by [34]. The values of ABA for Bukit Ibam have lower risk for the formation of acid mine drainage. [6] suggested if the ratio is greater than 3:1, experience indicates that there is lower risk for acid mine to develop, meanwhile samples with a ratio of 1:1 or less are potentially to generate acid.

From TCLP analysis, hazardous waste was identified at Sungai Lembing and Kg. Aur. This is due to the fact that As element in Sungai Lembing had high concentration that exceed the limitation. As in tailing at Sungai Lembing and Pb in Kg. Aur were mobile in characteristic. Exceed value concentration of TCLP element showed element was mobility and of great danger to environment. Hence, labeled as hazardous waste. The proportion of metals leached by the TCLP depends on a sludge's neutralization potential. A sludge with small neutralization potential is incapable of neutralizing all the acid added, and the resultant low pH will cause a substantial proportion of the metals present to be leached [10].

The immobilization of heavy metal ions from aqueous solutions is quite a complicated process, consisting of ion exchange and adsorption and is likely to be accompanied by precipitation of metal hydroxide complexes on active sites of the particle surface [26]. The removal of heavy metals from AMD using different types of adsorbent materials. All sorbents produced similar trends of removal the heavy metals with an abrupt decrease within 8 days. LFS is the best material for absorption of Zn, Mn and Cu in the contaminated water followed by bentonite, zeolite, and active carbon. Various researchers including [25, 28] have identified steel slag as a suitable candidate to remediate waters contaminated by acid mine drainage (AMD), since it has been shown to have a significant acid neutralizing potential that can be exploited to precipitate out a majority of dissolved metals by increasing solution pH. The Acid Neutralization Potential of steel slag was determined to be approximately 83% as calcium carbonate ($CaCO_3$) [25]. Meanwhile, bentonit was the best to adsorb more Ni, Fe and Cd. The concentration of heavy metal was decreased greatly due to owning of precipitation, co-precipitation and huge absorbency. Therefore, the utilization of LFS can achieve the purpose to neutralize the high acidity, minimize the release of heavy metal and improve the water quality of drainage.

5. CONCLUSION

Analysis was done at the mine which has a watershed. Water is an important indicator to determine the level of acidity of mine drainage. Overall, Kg Aur, Cini and Sungai Lembing found very acidic with low pH and contain high concentration of heavy metals. Therefore, the acidity and neutralization test were applied for waste rock, tailings or soil at the mining site. Acid-base accounting showed the possibility of field sites to form and produce acid especially for Sungai Lembing with the highest value of APP. TCLP element was made to test the nature of mobility and thus labeled as hazardous waste. Sungai Lembing and Kg. Aur have hazardous waste of Pb and As elements respectively. The AMD levels are identified from low pH and sulfate ions or high of heavy metal. Supported by this static test, results showed that the potential mine suffered of excessive acid production. Overall, AMD identified from all these tests at Kg Aur, Sungai Lembing, Selinsing and Chini. The most dangerous among others is Sungai Lembing and if no action is made, it can affect surrounding area with high generation of acid potential. There are known treatment methods which can be applied before the AMD situation can worsen and consequently source river pollution. By conducting continuous tank experiment tests for about 30 days using synthetic solution prepared, it was found that by product material such as LFS and natural material such as bentonite can effectively adsorb and remove various heavy metals simultaneously. Besides, these adsorbents can also efficiently neutralize the acidic drainage due to its high alkalinity production by calcite dissolution.

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