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LIMESTONE AS MATERIAL TO REDUCE SULPHATE AND ARSENIC CONTENT IN ACID MINE DRAINAGE

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Abstract. In this paper, limestone was used to reduce sulphate and arsenic content in acid mine drainage (AMD). The sample of acid mine drainage was collected from tailing tin mine pond located in Perak. The sulphate content in acid mine drainage was analysed before and after treatment with limestone by using DR 2800 spectrophotometer. ICP-OES was used to analyse water sample before and after treatment for arsenic content analysis. pH multi parameter was used to determine pH value, oxidation reduction potential and electrical conductivity (EC) of water sample before and after treatment. Limestone used was collected from a quarry in Simpang Pulai, Ipoh. The size of limestone used was less than 2 mm. The experiment was carried out by using experimental column. After treatment with limestone the result showed that the pH value increased from 2.4 to 6.5, whilst sulphate and arsenic content decreased. The best parameter was 500 g limestone with 75 minutes retention time.

Keywords Limestone; sulphate; arsenic; acid mine drainage; experimental column

1.0 INTRODUCTION

Acid mine drainage (AMD) is caused when sulphide minerals had exposed to oxygen and water [1] and assisted by sulphate oxidising bacteria (SOB). Acid mine drainage can be categorized as low pH and high concentration of toxic elements such as arsenic, copper, cadmium etc. The reaction is shown in equation 1.0:

$$2FeS_{2(s)} + 7O_{2(g)} + 2H_2O_{(l)} \longrightarrow FeSO_{4(s)} + 2H_2SO_{4(aq)} \quad (1.0)$$

Limestone rock is a sedimentary deposition of calcium carbonate caused by combination of dissolved calcium ions and carbon dioxide. Limestone deposits cover about 10% on the land surface of earth and can be found around the world [2] especially in Malaysia. Limestone is a versatile material that can be used in many fields such as in construction, agricultural, environmental, and industrial material.

Chemical property of limestone gives pH values in water between 8.0 and 9.0 [2] and is suitable to be used in acid mine drainage treatment despite encrustation by iron and aluminium hydroxides [3]. A major anion, sulphate that present in natural waters and industrial effluents either in acid mine drainage or neutral mine drainage can be adsorbed by limestone because calcium ion on the solid surface can bind anion [4]. Sulphate is only mildly hazardous anion compared to other toxic elements [4] such as arsenic, cadmium, copper etc. However, sulphate can cause laxative effect and can affect the taste of water at concentration above 600 mg/L. Therefore, certain countries especially WHO and Europe countries have set maximum values of sulphate content in mine drainages and industrial effluents around 250 mg/L to 500 mg/L [4]. The reaction between limestone and sulphuric acid is shown in equation 1.1.

$$CaCO_{3(s)} + H_2SO_{4(aq)} \longrightarrow CaSO_{4(s)} + CO_{2(g)} + H_2O_{(l)} \quad (1.1)$$

Another toxic element that present in acid mine drainage is arsenic. Arsenic is an element with a name derived from the Greek word known *arsenikon* meaning potent [4]. The arsenic presents in an environment with different oxidation states such as As (V), As (III), As (0) and As (-III). Arsenic is very difficult to vanish and it can only be transformed into insoluble compounds combined with other elements especially iron [5]. Arsenite, As (III) and arsenate, As (V) are the most toxic compounds compared to other arsenic oxidation states and both are the most abundance in water [6].

Other parameters studied besides pH were ORP and EC. Oxidation reduction potential (ORP) is a measure of the water cleanliness and an indicator the level of breakdown contaminants. Acidic water has positive ORP and alkaline water has negative ORP and is an antioxidant. Electrical conductivity (EC) is a measurement to determine the capability of water to pass electrical flow and its related to concentration of ions in water. These conductive ions exist from dissolved salts and non organic materials such as alkalis, sulphides, chlorides and carbonate compounds.

2.0 EXPERIMENTAL

2.1 Materials

The materials used in this study were limestone from quarry in Simpang Pulai, Ipoh and water sample from a tin mine pond tailing in Perak.

2.2 Method

2.2.1 Sampling

50 kg limestone was collected from a quarry in Simpang Pulai, Ipoh. Limestone was dried in a tray before sieving by using 2 mm sieve size. After sieving with 2 mm sieve the size of limestone less than 2 mm was used for acid mine drainage treatment.

Water samples from tailing pond of an active tin mine located in Perak were collected. In situ pH, ORP, EC and temperature values were recorded from this pond. pH values recorded were less than 4. The water samples were kept in 15

L jerry can and another two 2 L bottles samples were preserved in ice box before brought back to laboratory. The pond is a detention reservoir which is the overflow water from retention pond. The water of this pond was pumped from the nearest river and the discharge water from the tin processing plant.

2.2.2 Experimental Column

The experiments were carried out using experimental column as shown in Figure 2.0.

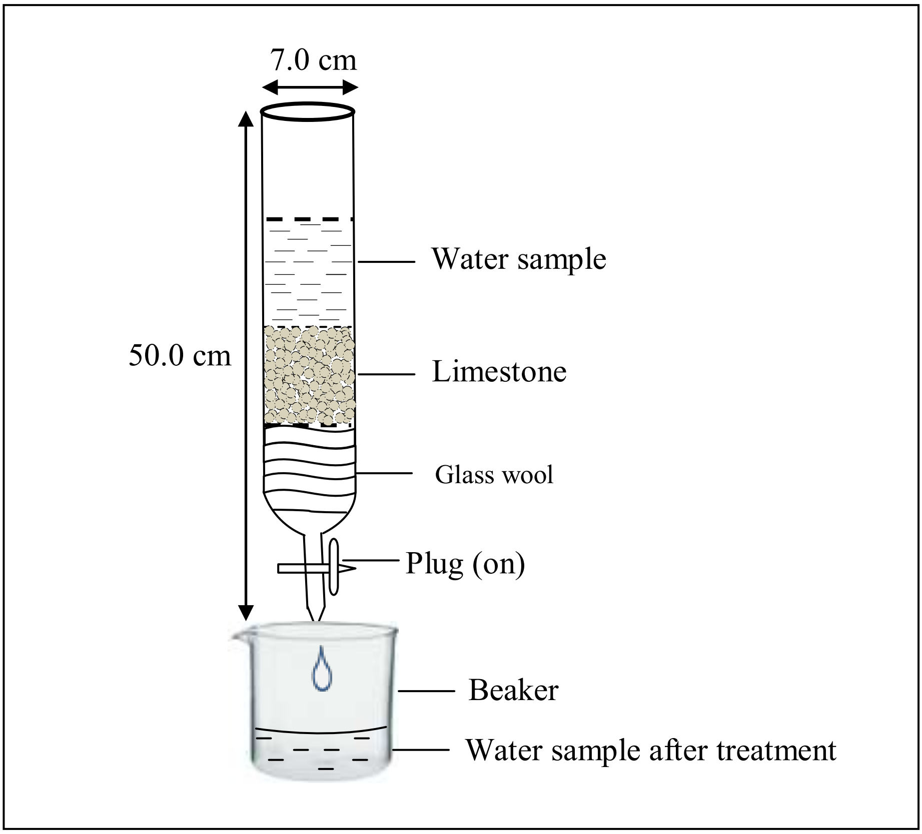

Figure 2.0: Experimental column

10 g glass wool was packed into the column. The limestone with size less than 2 mm and different weights 200 g, 300 g and 500 g were packed into the column one at a time. 500 ml water sampel was poured into the column. The retention times used for every weight of limestone were 15 minutes, 30 minutes, 45 minutes, 60 minutes and 75 minutes. pH, ORP and EC values of water sample before and after treatment were recorded. The experiment was carried out at room temperature. Water sample was analysed before and after treatment to detect sulphate and arsenic.

2.2.3 Instrumentations

ICP-OES (Optima 5300 DV, Perkin Elmer, USA) was used to detect arsenic, Portable spectrophotometer (DR2800, Hach, USA) was used to detect sulphate content in water samples and Portable multi parameter meter (A329, Thermo Scientific, Indonesia).

3.0 RESULTS AND DISCUSSION

Table 3.0 shows the values in situ parameters of pH, EC, ORP and temperature in the studied pond. The results showed that the values of pH, EC and ORP were 2.48, 699.2 µs/cm and 681.8 mV respectively. The pH value 2.4 indicated that the AMD problem occurred in this pond because any pH value around 2 to 4 can be assumed as AMD problem [7].

Table 3.0: In situ reading of pH, EC and ORP.

pH				EC (µs/cm)				ORP (mV)				Temperature (°C)			
1	2	3	mean	1	2	3	mean	1	2	3	mean	1	2	3	mean
2.49	2.47	2.48	2.48	2425	2338	2399	2387	699.2	696.9	649.4	681.8	36.1	36.1	36.1	36.1

Table 3.1, Table 3.2 and Table 3.3 show the result of experimental column carried out by using different weights of limestone 200 g, 300 g and 500 g respectively. The size of limestone used in this study was less than 2 mm and

retention times used in this study were 15 minutes, 30 minutes, 45 minutes, 60 minutes and 75 minutes.

Table 3.1(a): Reading of pH value

Retention Time (min)	Weight of Limestone (g)	Before pH				After pH			
		1	2	3	Mean	1	2	3	Mean
15	200	2.56	2.56	2.55	2.56	5.94	5.95	5.95	5.95
30	200	2.58	2.60	2.58	2.59	5.93	5.91	5.91	5.92
45	200	2.56	2.55	2.54	2.55	6.00	6.01	6.02	6.01
60	200	2.58	2.59	2.60	2.59	6.08	6.08	6.09	6.08
75	200	2.57	2.56	2.55	2.56	5.80	5.79	5.81	5.80

Table 3.1(b): Reading of EC value

Retention Time (min)	Weight of Limestone (g)	Before EC (μs/cm)				After EC (μs/cm)			
		1	2	3	Mean	1	2	3	Mean
15	200	2684	2682	2679	2682	2449	2451	2452	2451
30	200	2707	2708	2709	2708	2456	2457	2456	2456
45	200	2702	2703	2704	2703	2473	2469	2471	2471
60	200	2674	2674	2674	2674	2438	2438	2438	2438
75	200	2689	2688	2688	2688	2433	2433	2444	2433

Table 3.1(c): Reading of ORP value

Retention Time (min)	Weight of Limestone (g)	Before ORP (mV)				After ORP (mV)			
		1	2	3	Mean	1	2	3	Mean
15	200	780.2	779.1	779.2	779.5	455.4	455.3	454.4	455.0
30	200	733.4	732.7	731.9	732.7	343.4	343.2	341.8	342.8
45	200	786.7	785.9	785.2	785.9	483.6	481.5	482.5	482.5
60	200	741.3	742.1	742.2	741.9	359.2	358.7	356.9	358.3
75	200	775.5	774.3	774.0	774.6	418.5	418.3	417.8	418.2

Table 3.2(a): Reading of pH value

Retention Time (min)	Weight of Limestone (g)	Before pH				After pH			
		1	2	3	Mean	1	2	3	Mean
15	300	2.49	2.50	2.51	2.50	6.12	6.12	6.13	6.12
30	300	2.46	2.47	2.46	2.46	6.21	6.20	6.22	6.21
45	300	2.48	2.48	2.48	2.48	5.99	6.00	6.01	6.00
60	300	2.48	2.49	2.47	2.48	5.87	5.87	5.85	5.86
75	300	2.48	2.47	2.47	2.47	6.07	6.08	6.09	6.08

Table 3.2(b): Reading of EC value

Retention Time (min)	Weight of Limestone (g)	Before EC (µs/cm)				After EC (µs/cm)			
		1	2	3	Mean	1	2	3	Mean
15	300	2753	2754	2753	2753	2515	2514	2513	2514
30	300	2760	2762	2763	2762	2532	2533	2535	2533
45	300	2708	2708	2708	2708	2396	2395	2394	2395
60	300	2750	2749	2752	2750	2438	2437	2436	2437
75	300	2751	2749	2748	2749	2463	2462	2463	2463

Table 3.2(c): Reading of ORP value

Retention Time (min)	Weight of Limestone (g)	Before ORP (mV)				After ORP (mV)			
		1	2	3	Mean	1	2	3	Mean
15	300	806.0	806.1	806.6	806.2	485.3	484.2	484.1	484.5
30	300	804.5	805.4	805.9	805.3	477.6	477.5	477.8	477.6
45	300	805.7	803.3	803.1	804.0	487.9	486.3	479.5	484.6
60	300	808.9	807.4	806.9	807.7	490.8	489.2	487.9	489.3
75	300	808.2	809.1	808.8	808.7	470.0	469.9	469.3	469.7

Table 3.3(a): Reading of pH value

Retention Time (min)	Weight of Limestone (g)	Before pH				After pH			
		1	2	3	Mean	1	2	3	Mean
15	500	2.50	2.49	2.48	2.49	6.46	6.47	6.48	6.47
30	500	2.50	2.48	2.49	2.49	6.34	6.35	6.35	6.35
45	500	2.45	2.45	2.46	2.45	6.42	6.41	6.42	6.42
60	500	2.48	2.47	2.48	2.48	6.35	6.36	6.36	6.36
75	500	2.47	2.48	2.49	2.48	6.59	6.59	6.58	6.59

Table 3.3(b): Reading of EC value

Retention Time (min)	Weight of Limestone (g)	Before EC (µs/cm)				After EC (µs/cm)			
		1	2	3	Mean	1	2	3	Mean
15	500	2788	2789	2794	2790	2495	2495	2495	2495
30	500	2788	2789	2790	2789	2511	2512	2512	2512
45	500	2797	2796	2797	2797	2538	2537	2538	2538
60	500	2782	2781	2782	2782	2544	2545	2546	2545
75	500	2744	2745	2746	2745	2537	2536	2536	2536

Table 3.3(c): Reading of ORP value

Retention Time (min)	Weight of Limestone (g)	Before ORP (mV)				After ORP (mV)			
		1	2	3	Mean	1	2	3	Mean
15	500	810.0	811.5	813.3	811.6	482.3	480.6	480.5	481.1
30	500	815.6	815.4	815.5	815.5	447.3	446.9	446.0	446.7
45	500	816.6	817.3	815.7	816.5	470.2	470.3	471.3	470.6
60	500	810.9	811.1	811.2	811.1	449.9	450.0	451.2	450.4
75	500	811.9	813.3	812.2	812.5	457.2	456.4	455.6	456.4

Tables 3.1, 3.2 and 3.3 show that reaction had occurred between water sample and limestone. There were demonstrated by the increase in the pH values and decrease in electric conductivity (EC) and oxidation reduction potential (ORP) The highest pH value achieved after treatment was 6.59 by using 500 g limestone with retention time 75 minutes.

Table 3.4 and Table 3.5 show the result of sulphate content and arsenic content respectively in water sample before and after treatment by using experimental column.

Table 3.4: Sulphate content in water samples before and after treatment

Retention Time (min)	Weight of Limestone (g)	Before Sulphate (mg/L)				After Sulphate (mg/L)			
		1	2	3	mean	1	2	3	mean
15	200	1400	1400	1300	1367	1390	1304	1252	1315
30	200	1400	1400	1300	1367	1287	1313	1364	1321
45	200	1400	1400	1300	1367	1322	1397	1273	1331
60	200	1400	1400	1300	1367	1340	1362	1267	1323
75	200	1400	1400	1300	1367	1447	1371	1445	1421
15	300	1400	1400	1300	1367	1240	1383	1358	1327
30	300	1400	1400	1300	1367	1378	1368	1385	1377
45	300	1400	1400	1300	1367	1355	1364	1328	1349
60	300	1400	1400	1300	1367	1352	1332	1351	1345
75	300	1400	1400	1300	1367	1371	1335	1261	1322
15	500	1400	1400	1300	1367	800	1400	1200	1133
30	500	1400	1400	1300	1367	1100	1000	900	1000
45	500	1400	1400	1300	1367	1000	900	1100	1000
60	500	1400	1400	1300	1367	1100	1200	1300	1200
75	500	1400	1400	1300	1367	1385	1378	1355	1373

The results from Table 3.4 show that the content of sulphate decreased slightly after treatment with limestone. The use of 500 g limestone seems more effective compared to others especially with retention times of 30 minutes and 45 minutes. The content of sulphate in these retention times had decreased from 1400 mg/L to 1000 mg/L after treatment.

Table 3.5: Arsenic content in water samples before and after treatment

Retention Time (min)	Weight of Limestone (g)	Before Arsenic (mg/L)				After Arsenic (mg/L)			
		1	2	3	mean	1	2	3	mean
15	200	2.368	2.405	2.527	2.433	-0.002	0.001	0.000	0.001
30	200	2.368	2.405	2.527	2.433	0.003	-0.002	0.003	0.003
45	200	2.368	2.405	2.527	2.433	0.000	0.004	-0.003	0.004
60	200	2.368	2.405	2.527	2.433	0.002	0.000	-0.001	0.002
75	200	2.368	2.405	2.527	2.433	0.011	0.005	0.008	0.024
15	300	2.368	2.405	2.527	2.433	0.006	0.003	0.001	0.003
30	300	2.368	2.405	2.527	2.433	0.007	0.005	0.006	0.006
45	300	2.368	2.405	2.527	2.433	0.005	0.009	0.010	0.008
60	300	2.368	2.405	2.527	2.433	0.007	0.005	0.002	0.005
75	300	2.368	2.405	2.527	2.433	0.004	0.008	0.003	0.005
15	500	2.368	2.405	2.527	2.433	0.007	0.006	0.011	0.008
30	500	2.368	2.405	2.527	2.433	0.008	0.004	0.010	0.007
45	500	2.368	2.405	2.527	2.433	0.011	0.009	0.010	0.010
60	500	2.368	2.405	2.527	2.433	0.006	0.008	0.009	0.008
75	500	2.368	2.405	2.527	2.433	0.010	0.015	0.013	0.013

Table 3.5 shows that limestone was capable to reduce arsenic content in water sample after treatment in all experiments that had been carried out.

4.0 CONCLUSIONS

The results show that limestone can reduce sulphate content in water sample but the concentrate value is much more than international standard (WHO standard 500 mg/L ; Europe standard 250 mg/L). However, limestone can reduce arsenic content in water sample effectively based on the ICP-OES analysis result. Overall result indicated that arsenic content in water sample after treatment had complied with Environmental Quality Act 1974 (Standard A 0.05 mg/L ; Standard B 0.10 mg/L). The highest pH value recorded in this experiment after treatment was 6.59.

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