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Geochemistry of waste rock at dumping area

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Geochemical characteristics and the increase in the total reactive surface area of the sulphide-bearing rock due to weathering processes at dumping area, are expected to enhance the oxidation of sulphides, leading to the generation of acid mine drainage (AMD). The study of the changes of geochemical characteristics of sulphide-bearing rocks showed that the oxidation uniformly occurred at the surface layer of 2- and 10-year-old waste rock dumps. The changes of mineralogical composition were also investigated, particularly for the cases where significant formation of clay minerals occurred. The study showed that the formation of clay minerals after the weathering processes took place at the dumping area, suggested that the generation of AMD would be minimised because the interior of the waste rock dump provided a barrier to oxygen and water.

Keywords: acid mine drainage; sulphide-bearing rock; oxidation; clay mineral

1. Introduction

Acid mine drainage (AMD) has been acknowledged by some ore mining and most coal mining operations as one of their main environmental concerns. Many studies have been conducted concerning AMD. The studies covered the identification of the sources [1,2], the processes and the influence factors [3–6], prevention methods [7,8] and the treatment of AMD [9].

Considering the fact that the dump area is a large stockpile of waste materials and it is always exposed to the local climatic conditions, this area can therefore be deemed as one of the sources of AMD. The influence of the local climate will furthermore affect the physical, chemical and biological factors of the dump [10]. The factor of rock chemistry or geochemistry plays an important role in determining the process of AMD generation in relation to the existence of potentially acid-forming and also acid-buffering or neutralising materials.

Rock weathering, whether physical or chemical, is one of the processes that occur at the waste rock that is affected by climatic conditions, especially rainfall and temperature. Physical weathering process is generally the forerunner of chemical

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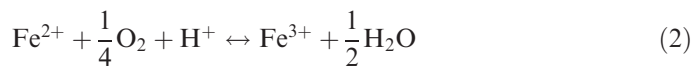
weathering, and their main contributions are to loosen rock masses, reduce particle size and increase the available surface for chemical reactions [11].

Physically, as a result of rock weathering process, hard rock formations will turn into so-called soft rock formations [12,13], and it is also shown by grain size changes in the weathered material [14], which then affect the total reactive surface area. High rainfall intensity, high temperature and high intensity of ultraviolet rays certainly cause intensive rock weathering, particularly chemical weathering [2,13]. Water reacts with rocks and minerals and thereby releases chemical elements to continue the weathering process, or they can be washed out by drainage [14].

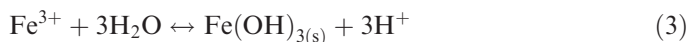
Geochemical characteristics and the increase of the total reactive surface area of sulphide-bearing rock, which is generally found in the waste rock, will accelerate the oxidation rate to form AMD [15,16]. In general, pyrite (FeS_2) that is commonly found in waste rock and typical of many oxidation processes during weathering [17] is oxidised in accordance with the following reaction [18]:



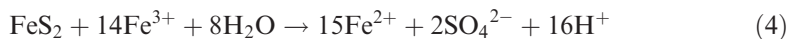
If the surrounding environment is sufficiently oxidising, much of the ferrous iron will oxidise to ferric ion:



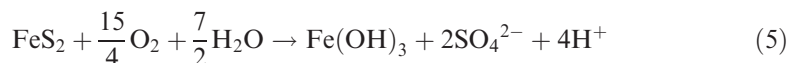
At pH values above 2.3–3.5, the ferric iron will precipitate as $\text{Fe}(\text{OH})_3$, leaving little Fe^{3+} in solution whereas lowering the pH at the same time:



Any Fe^{3+} that does not precipitate from solution may be used to oxidise additional pyrite:



Acid generation that produces iron which eventually precipitates as $\text{Fe}(\text{OH})_3$ may be represented by a combination of reactions:



The pH resulting from the reaction is important, because it influences the solubility of SiO_2 and Al_2O_3 , and the type of clay mineral that may form [11].

Taking into account the physical factors of the cover systems, the change of grain size will also influence permeability of the layer. Decrease of permeability is caused by filling the inter-rock space with fine particles resulting from the weathering. This

degradation of permeability then will control the rate of water infiltration and oxygen diffusion/advection onto and within the dump [10]. Oxygen and water are essential for AMD generation, as shown in Equation (1). Decreasing the rate of such reactions will then minimise the occurrence of AMD.

This article will describe the results of previous and current studies of geochemical and physical characteristic changes on the surface layer of dumps that are affected by the local, tropical climatic conditions.

2. Site description

PT. Kaltim Prima Coal (KPC) is the biggest open pit coal mine company in Indonesia, located in Sengata – East Kalimantan, Indonesia. The location has tropical climate conditions with high average seasonal rainfall varying from 1.6 to 2.5 m annually, and an average temperature varying from 26 to 32°C. As the end of 2007, KPC had cleared 11,398 ha of land for mining activities including supporting facilities, and has rehabilitated 2,842 ha of the area.

The sedimentary coal sequence in the KPC mine area has a variety of sedimentary rock types and includes horizons that are known to be pyretic and those that contain acid producing materials. Pyrite in the KPC mine sequence primarily occurs in the overburden just above (approximately 2 m) and below the coal seams.

According to geochemical studies, one of which is related to Net Acid Generation (NAG) test value, the waste rocks at KPC are classified as Non-Acid Forming (NAF) and Potentially Acid Forming (PAF).

AMD management at KPC is conducted with particular focus on prevention efforts using dry cover systems. The types of the systems used are based on the result of computer modelling and known as DC01 (1 m compacted clay + 2 m loose NAF over PAF), DC02 (2 m compacted NAF + 2 m loose NAF over PAF) and DC03 (10–20 m loose NAF over PAF) (Rehabilitation Specification, PT. Kaltim Prima Coal, 2000).

3. Previous studies

To ensure the effectiveness of each type of cover system, KPC with the assistance of a consultant installed five instruments for measuring oxygen concentration and temperature up to 16–21 m depth, and six lysimeters at 1, 2 and 3 m depth. For comparison purposes, the measurements were also conducted at a dumping location without a cover system that was constructed with PAF material and located at Sangatta North.

The results of the measurements made at Sangatta North in November 1999 concluded the following [19]:

- Pore gas oxygen concentration profiles indicated that diffusion is the dominant oxygen transport mechanism.
- Oxygen was consumed within 5 m, and the concentration dropped quickly along the depth of the probe holes following installation. This suggests that the material in the dump generally has a high oxidation rate.
- Temperature was elevated at depth. Given that very little oxygen is present at depth, these elevated temperatures are likely to be remnant heat from oxidation processes that have occurred in the past.

Hereinafter, the results of the measurement of January–July 2000 concluded the following:

- The water infiltration rate was 22.8% and the water flow rates through the dump were expected to be between 3 and 6.9 m/year.
- Field measurements revealed that oxygen penetration took place at depths of between 2.6 and 7.7 m.

The next measurement from February 2001 until February 2003 gave the same conclusion as the previous measurement [20].

Having learned from these previous studies that the oxygen penetration could reach 2.6–7.7 m depth and that the water infiltration rate was 3–6.9 m/year, it is therefore important to investigate or study the geochemical characteristic changes at the surface layer.

It is also worthwhile to note that the results of previous studies concluded that the mechanical structure of the 10 cm surface is crucial in determining the infiltration rate. This can be explained by the fact that with a slight increase in fines combined with compaction on the surface, rock permeability decreases and this leads to a difference in the infiltration rate. In addition, rock weathering generally converts the dump materials into clay that influences the mechanical properties of the materials and this is due to a quick expand during water absorption [13].

4. Current studies

An investigation was conducted at Sangatta North with 0–2 m waste rock depth in January 2008. The dump was 10 years old (10Y-Sangatta North), and another one for comparison purposes was also conducted at Pit J, which was 2 years old (2Y-Pit J, Figure 1). The samples were taken for each 20 cm interval depth.

This study was particularly intended to know the pattern of geochemical characteristic changes and the potency of clay formation in material, as an indicator of chemical and physical weathering occurrences. Understanding these behaviours will help to investigate the occurrence of AMD at the surface layer.

5. Methodology

5.1. Field sampling

Two holes of $2 \times 2 \times 2$ m were excavated each at flat areas of Pit J and Sangatta North dump, and 5 kg of rock samples were obtained from each layer, and subsequently divided into four quadrants. One quadrant of samples was afterward dried, crushed and pulverised under $75 \mu\text{m}$ in diameter for the following examinations.

5.2. Laboratory analysis

Geochemical analysis of rock samples was conducted by doing static tests. A static test is intended to predict whether a rock sample has acid-producing and/or acid-neutralising capacities [21] using most widely accepted methods such as Paste pH, Paste EC, NAG test and ANC test.

Beside the static tests, mineralogy study with a Rigaku RINT 2000 X-ray diffractometer and elemental compositions analysis with a Rigaku RIX 3100 X-ray fluorescence spectrometer were also carried out.

5.2.1. Net acid generation test

An NAG test was conducted based on the procedure compiled by KPC from the Environmental Geochemistry International Pty. Ltd. procedure, where a 2.5 g rock sample was reacted with 250 mL of 7.5% hydrogen peroxide (H_2O_2) overnight and then followed by heating for 30 minutes.

Having mixed this rock sample with sodium hydroxide (NaOH), the NAG pH is then measured and used as one criteria in determination of the NAG type of the samples. A rock sample with NAG pH less than 4 is classified as PAF and NAG pH greater than 4 is classified as NAF (*NAG Test Manual*, PT. Kaltim Prima Coal, 2003).

5.2.2. Paste pH and Paste EC

Paste pH and Paste EC are used to show the existence of current results of sulphide mineral oxidation on the rock sample. The paste is prepared by soaking the rock sample for 16 h in a water to rock sample ratio of 2:1 [22].

5.2.3. Acid Neutralising Capacity

Acid neutralising capacity (ANC) is used to determine the capacity of rock to neutralise the acidity formed, based on the procedure developed by Sobek *et al.* (1978) [24] and modified by Skousen *et al.* (1997) [25]. Two grams of crushed rock samples are reacted by heating in the presence of known concentrations of hydrochloric acid (HCl), which is determined by the Fizz test (Table 1).

The solution is then titrated with a known concentration of NaOH, based on the table, with addition of H_2O_2 at near to pH 5. The ANC then is determined as kg H_2SO_4 /ton of rock, based on the quantities and concentration of HCl and NaOH used.

6. Results and discussion

6.1. Geochemistry

The comparative study between Paste pH and NAG pH provides an indication of relative reactivity of sulphide mineral oxidation on the rock. Final pH value of rock

Table 1. Fizz ratings and associated quantities and concentrations of HCl to be used in the ANC determination.

| Reaction | Fizz rating | HCl | | NaOH molarity (M) |
|-------------|-------------|--------------|-----------|-------------------|
| | | Molarity (M) | Vol. (ml) | |
| No | 0 | 0.5 | 4 | 0.1 |
| Slight | 1 | 0.5 | 8 | 0.1 |
| Moderate | 2 | 0.5 | 20 | 0.5 |
| Strong | 3 | 0.5 | 40 | 0.5 |
| Very strong | 4 | 1 | 40 | 0.5 |

sample (NAG pH) is determined after all sulphide available is oxidised using H_2O_2 , whereas Paste pH shows the current pH value as the result of reaction with water. Samples containing reactive sulphide minerals (such as framboidal pyrite) will apparently be the most responsive to Paste pH test [22].

All the samples obtained from Pit J had a Paste pH greater than 4 and six samples of those had a NAG pH less than 4 (Figure 2). The differences of Paste pH and NAG pH indicated that the samples still had a considerable amount of neutralising minerals and that they were still in the initial stage of AMD generation. This result was different for the samples obtained from Sangatta North where five samples had a Paste pH less than 4, and seven samples had a NAG pH less than 4.

NAG pH and Paste pH values in each dump showed that, in general, there was no observation of NAG pH and Paste pH changes with depth, except for the samples of Pit J from 20–40 cm depth and Sangatta North from 00–20 cm, 140–160 cm and 180–200 cm depth. This result indicated the uniformity of weathering process at each dump.

The differences between NAG pH and Paste pH values at each dump were generally consistent. Except for those fractions, it can be stated that Pit J had higher gap of pH value compared with Sangatta North. This condition is attributed to the fact that the oxidation process at Sangatta North (10 years old) had advanced further than at Pit J (2 years old).

The different oxidation levels can also be seen from the comparison between Paste pH and Paste EC (Figure 3). Paste EC estimates the total amount of dissolved ions in the water, and it is dependent on the elemental composition of the rock sample. Paste pH from Pit J showed a neutral condition and had a good correlation with Paste EC. It indicated that the alkalinity of the rock samples was still present. During the weathering processes, the alkalinity reacts with hydrogen and sulphate ions that are produced from pyrite oxidation, which results in a decrease of the available ions.

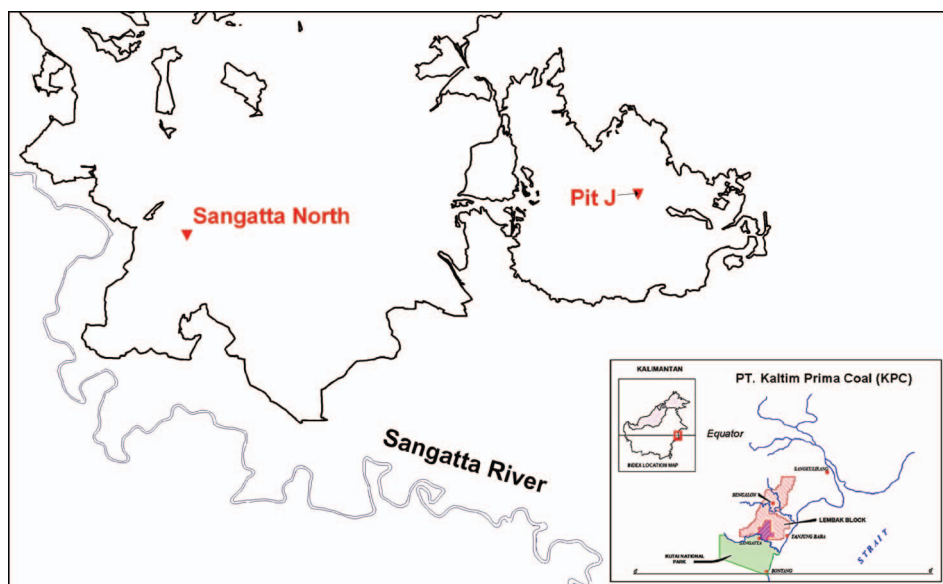


Figure 1. Map of the study area.

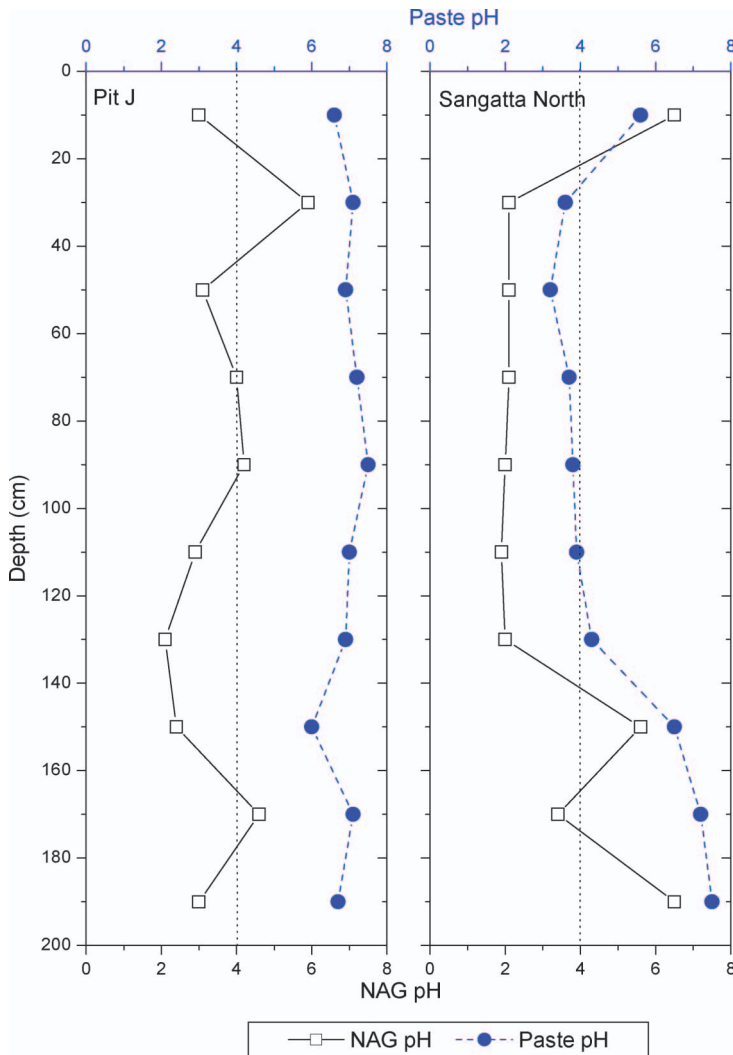


Figure 2. NAG pH and Paste pH values versus depth in Pit J and Sangatta North.

On the contrary, Sangatta North samples showed high Paste EC that varies from 2–5.9 ms/cm, and low Paste pH due to lack of alkalinity especially for the samples at 20–140 cm depth which had no neutralising capacity as shown with ANC values (Table 2).

Considering NAG pH and NAPP values (Table 2), the classification of the rock samples showed that three samples obtained from Pit J at 60 – 80 cm, 80 – 100 cm and 160 – 180 cm depth, and one samples obtained from Sangatta North at 180 – 200 cm, depth were NAF. Samples obtained from Pit J at 20 – 40 cm and from Sangatta North at 00 – 20 cm and 140 – 160 cm depth were classified as “uncertain (UC)” since there was a conflict between NAG pH and NAPP values. The other samples were classified as PAF.

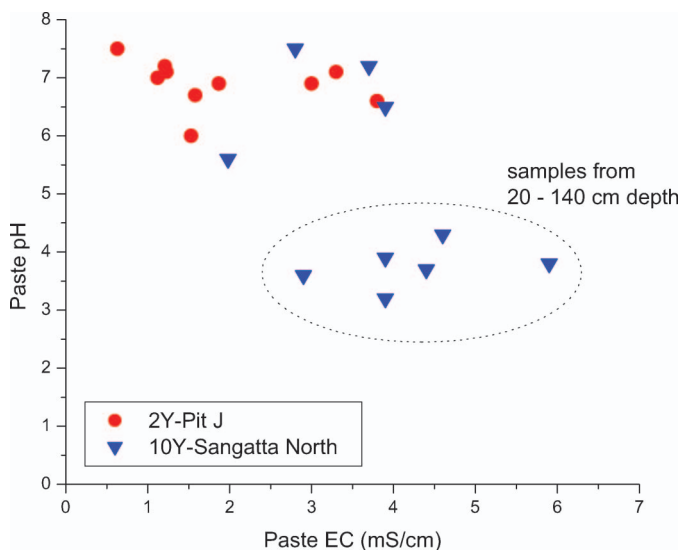


Figure 3. Paste pH versus Paste EC.

Table 2. Static test results of the samples.

| Sample | Depth (cm) | NAG pH | MPA | ANC | NAPP | Type |
|----------------------|------------|--------|-------|--------|-------|------|
| 2Y – Pit J | 00–20 | 3.0 | 20.49 | 9.56 | 10.93 | PAF |
| | 20–40 | 5.9 | 16.01 | 15.20 | 0.81 | UC |
| | 40–60 | 3.1 | 17.70 | 11.74 | 5.97 | PAF |
| | 60–80 | 4.0 | 6.63 | 8.68 | -2.05 | NAF |
| | 80–100 | 4.2 | 6.58 | 8.44 | -1.86 | NAF |
| | 100–120 | 2.9 | 14.23 | 11.97 | 2.26 | PAF |
| | 120–140 | 2.1 | 34.72 | 18.22 | 16.50 | PAF |
| | 140–160 | 2.4 | 27.96 | 12.21 | 15.74 | PAF |
| | 160–180 | 4.6 | 7.48 | 11.14 | -3.66 | NAF |
| 180–200 | 3.0 | 20.11 | 15.01 | 5.10 | PAF | |
| 10Y – Sangatta North | 00–20 | 6.5 | 28.61 | 25.91 | 2.70 | UC |
| | 20–40 | 2.1 | 62.48 | -8.70 | 71.19 | PAF |
| | 40–60 | 2.1 | 72.07 | -14.23 | 86.30 | PAF |
| | 60–80 | 2.1 | 70.47 | -9.30 | 79.77 | PAF |
| | 80–100 | 2.0 | 88.40 | -6.60 | 95.00 | PAF |
| | 100–120 | 1.9 | 69.51 | -10.26 | 79.77 | PAF |
| | 120–140 | 2.0 | 48.55 | -5.74 | 54.29 | PAF |
| | 140–160 | 5.6 | 15.64 | 9.16 | 6.48 | UC |
| | 160–180 | 3.4 | 20.90 | 12.34 | 8.56 | PAF |
| 180–200 | 6.5 | 14.51 | 18.14 | -3.63 | NAF | |

Calculation:

Maximum Potential Acid (MPA) = 30.6 × % Total Sulfur (S).

Net Acid Potential Production (NAPP) = MPA – ANC.

MPA, ANC, NAPP in kg H₂SO₄/ton.

Classification:

NAF: NAG pH ≥ 4; NAPP ≤ 0.

PAF: NAG pH < 4; NAPP > 0.

However, taking into consideration the Paste EC values of those samples and comparing them to the values obtained from each dump, the samples appeared to have a capacity to form acidity. The exception was the sample obtained from Sangatta North from 00–20 cm depth, which had low Paste EC. Consequently, the samples from Pit J and Sangatta North generally could be stated as PAF material and still have the possibility to further oxidise though they are 2 and 10 years old. This uncompleted oxidation was presumably caused by an inadequate presence of oxygen and water as a consequence of natural compaction of the surface layer.

6.2. Mineralogy

Mineralogical examination was conducted for the samples at 00–20 cm, 40–60 cm and 180–200 cm depths for each dump. The minerals such as quartz (SiO_2), kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) and illite ($(\text{K},\text{H}_3\text{O})\text{Al}_2\text{Si}_3\text{AlO}_{10}(\text{OH})_2$) are commonly involved in mine site samples. There was also illite-montmorillonite ($\text{KAl}_4(\text{Si},\text{Al})_8\text{O}_{10}(\text{OH})_4 \cdot 4\text{H}_2\text{O}$) and siderite (FeCO_3).

Clay minerals such as kaolinite and montmorillonite are secondary minerals that are produced by the weathering process of primary silicate minerals such as albite, anorthite, K-feldspar, augite and biotite [26]. Given the location of the mine in an equatorial region, weathering process had occurred before mining activities started and the rocks are currently at intermediate and advanced weathering stages, based on the classification from Mitchell (1976) [11].

The study also indicated that there were differences of secondary mineral composition resulting from pyrite oxidation. At the surface of the Pit J dump, 00–20 cm depth, the secondary mineral formed was melanterite ($\text{Fe}^{\text{II}}\text{SO}_4 \cdot 7\text{H}_2\text{O}$), and this result was consistent with previous study where melanterite is the dominant mineral to be precipitated during the initial stages of sulphide oxidation [27]. This was not found at Sangatta North. However, less soluble secondary minerals such as jarosite ($\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$) was found at the samples of Pit J and Sangatta North, which is commonly formed as the results of pyrite (FeS_2) oxidation.

The focus of the mineralogy was the elemental determination of rock samples. It was conducted based on the premise that chemical weathering depends on the reactions of silica and alumina with water. Silica and alumina, as the major constituents of rocks, are combined as aluminosilicate minerals such as muscovite and K-feldspar. Silica is less soluble at all pH values, whereas alumina is only soluble below pH 4 and above pH 8.5. Various stages of desilication indicate the weathering process and the various rates of desilication indicate the influence of differences in drainage [14]. Nearly all silicate weathering reactions also consume acid and increase the pH [26].

Molecular proportion analysis for SiO_2 at Sangatta North indicated an occurrence of desilication process where the molecular proportion tended to increase gradually with depth (Figure 4). However, this condition was not found on the samples obtained from Pit J.

Al_2O_3 also tended to increase with depth at Sangatta North. Because the solubility of alumina is strongly influenced by pH, Paste pH values could explain this condition. Paste pH of Sangatta North varied from 3.2 to 7.5, and that of Pit J varied from 6–7.5. Because alumina is soluble only below pH 4 and above pH 8, it was an indication that alumina dissolution occurred only at Sangatta North where lower values appeared near to the surface.

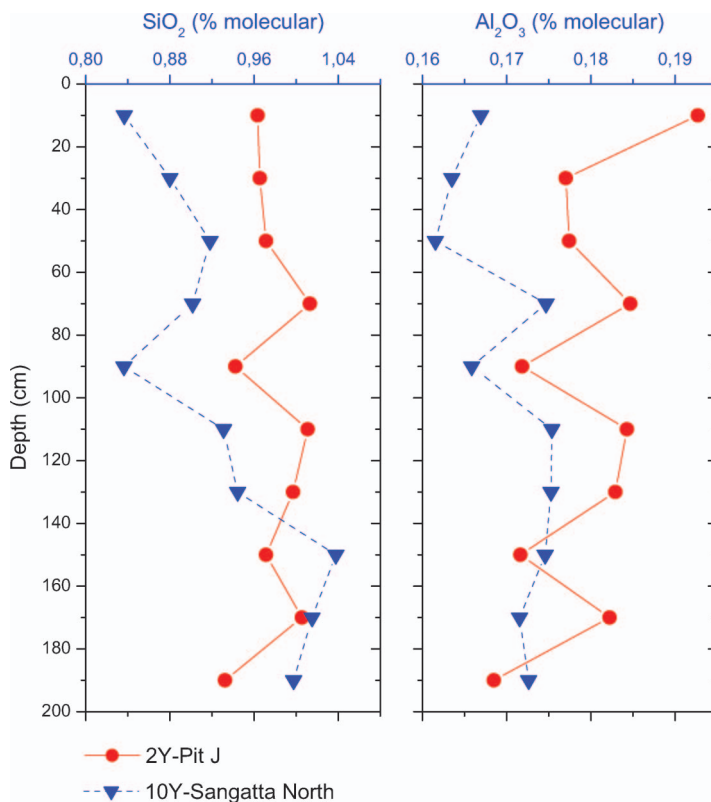


Figure 4. Vertical profiles of SiO₂ and Al₂O₃ at 2Y-Pit J and 10Y-Sangatta North.

The abundance of clay minerals (illite, kaolinite and montmorillonite) at Pit J and Sangatta North indicated that the waste rocks had weathered formerly, and at the dump sites the weathering processes continued and accelerated under the influence of climatic conditions.

In addition, the result of Atterberg Limits analysis for fine-grained samples, according to ASTM D 4318-00 standard, showed that the samples obtained from Pit J and Sangatta North were classified as 'CL', which means 'inorganic clays of low to medium plasticity, gravelly clays, sandy clays, silty clays, lean clays', based on the criteria of the Unified Soil Classification System (Figure 5).

Considering the expansion characteristic of clay minerals due to water absorption, the effect would be a decrease in the permeability of the dump and thus the potency of pyrite oxidation. In this case, where PAF material was placed on the top of the dump, it can be concluded that rock weathering can decrease the potency of AMD generation by creating clay minerals that can act as a barrier against oxygen and water. However, during the formation of clay minerals, the oxidation of pyrite will still occur and should be taken into consideration.

Furthermore, it is important to investigate in more detail the potency of clay formation, including the rate of clay formation, as a weathering product and its physical potency as a water and oxygen barrier that is useful to prevent AMD generation.

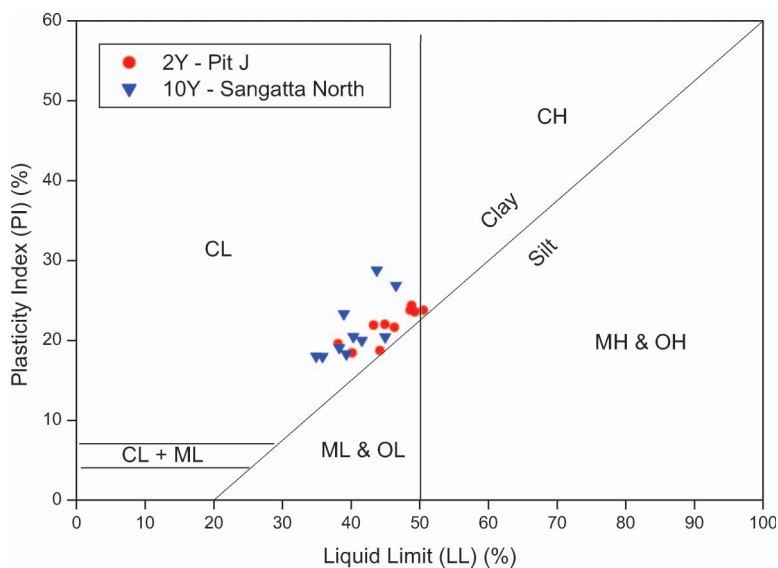


Figure 5. Plasticity Chart for rock samples.

7. Conclusion

The effects of climate factors on the rock weathering and pyrite oxidation processes were investigated in this study. Oxygen and water, as two important factors in pyrite oxidation, were shown to be available above 2 m depth of dump, and they influenced the geochemical and physical characteristic changes.

In general, the uniformity of oxidation results from each dump showed the differences in oxidation periods between Pit J dump (2 years old), which was still in the initial stage of AMD generation, and Sangatta North dump (10 years old), which was more advanced.

Secondary minerals formed by the rock weathering processes showed that the dumps have a potency to form clay minerals as a barrier against oxygen and water to decrease AMD generation.

Acknowledgements

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