Fundamental Study of Effect of Waste Rock Condition on Control of Acid Mine Drainage at Open Pit Coal Mine in Tropical Regions

Hideki Shimada^a*, Takashi Sasaoka^a, Sugeng Wahyudi^a, Kikuo Matsui^a, Ginting J. Kusuma^b and Rudy S. Gautama^b

^aKyushu University, Fukuoka, Japan ^bInstitutTeknologi Bandung, Bandung, Indonesia * shimada@mine.kyushu-u.ac.jp

Abstract

Acid Mine Drainage (AMD) problem is considered as one of the serious problems in mining operation in the world. This problem is generated due to sulfide minerals in waste rocks. Sulfide minerals are very commonly included in any types of crustal rocks, resulting in AMD and a negative impact on the ecosystem. In this research, some experiments were conducted with the rocks sampled in the open pit coal mine in Indonesia to understand the effects of the rock condition on acidic water generation. The result indicated that wetting and drying process affected the disintegration of rocks caused by both physical and chemical factors in the rock sample. Hence, AMD generation is promoted due to both factors while waste rocks are stored in tailings in the current system. It suggests that the system for waste rock management should be improved to prevent AMD generation.

1. Introduction

Mining operation often results in environmental problems. Acid Mine Drainage (AMD) is one of them and considered as serious problem due to the bad influence on the nature. Acidic water is generated when rocks containing sulfide minerals are exposed to oxygen and water. Pyrite is oxidized by oxygen, and the main oxidants are oxygen and ferric ion as shown in Eq. (1) and Eq. (4) in the reaction. Ferrous ion is initially dissolved from pyrite Eq. (1) and ferrous ion is oxidized to ferric ion by oxygen Eq. (2). The pH decreases with the promotion of the reaction above. Oxidation of pyrite is, moreover, accelerated by ferric ion Eq. (4) because ferric ion is strong oxidant. Depending on pH, ferric ion is precipitated as ferric hydroxide Eq. (3). This reaction does not happen at low-pH.

$$FeS_2 + \frac{7}{2}O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+$$
 (1)

$$Fe^{2+} + \frac{1}{4}O_2 + H^+ \to Fe^{3+} + \frac{1}{2}H_2O$$
 (2)

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+$$
(3)

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+$$
 (4)

Acidic water might cause negative impact on the nature in the surrounding area since it is low-pH and usually contains high concentrations of heavy metals. Therefore, effective control method and proper management of AMD should be considered for reducing the overall impact of mining operation on the environment.

Acidic water is often generated in pit while waste rocks are stored in tailings in mining area. The active pit where AMD is generated is shown in Fig.1. Waste rocks weather due to heavy rain and high temperature while being stored, resulting in AMD problems. Some researchers noted that rock weathering affects AMD generation (Gleisner, 2005; Cotter and Brigden; 2006; Dold et al., 2013). Rocks are easily weathered under acid conditions, and most of the elements are released (Jennings et al., 2008). Several studies about the relationship between rock weathering and AMD have been performed in the past; however, further studies are required since mining operation, property of the waste rock, and conditions on the site are different depending on mine site.



Fig.1. AMD generation in the active pit where waste rocks are temporarily stored before backfilling.

As stated above, rock conditions which is dependent on the management of waste rocks can affect AMD generation. Waste rocks, therefore, should be treated properly to prevent the AMD generation. This study was conducted to investigate the effects of the rock conditions on AMD. Based on the results, the current system in mining activities was reviewed to prevent acidic water.

2. Methods

The rocks which were covered with others were obtained in the active pit and they were not exposed to air. Some rocks were exposed to air, leading to acidic water in the area. The sample was characterized by physical and geochemical analysis. Additionally, leaching test was performed to investigate the long-term behavior of acidic water generation (Ginting et al., 2012). Base on the results, slaking test was performed and sequential extraction was carried out with the sample before and after the leaching test to understand the effects of rock condition on acidic water generation.

2.1 Sample Analysis

Waste rocks were classified by conducting the Acid Base Accounting (ABA) test and the Atterberg limits test (Casagrande, 1932; Leon and Edgardo, 2012). In ABA test, Maximum Potential Acidity (MPA) is obtained by calculating the sulfur content in the samples, and the Acid Neutralizing Capacity (ANC) is determined as kg H₂SO₄/ton of sample after HCl and NaOH are titrated (Sobek et al., 1978). Based on the values, Net Acid Producing Potential (NAPP=MPA-ANC) can be calculated (Weber et al., 2004). The sample has a potential of causing AMD when NAPP is higher than 20 (kg H₂SO₄/ton) (Chotpantarat, 2011). In the Atterberg limits test, rock samples could be categorized into rock types based on the liquid limit and the plastic limit of them which are obtained by observing the change of physical property of them under different water content (Casagrande, 1932; Japanese Industrial Standard). Furthermore, the rock sample was analyzed by X-ray diffraction (XRD) to ascertain the constituent minerals associated with AMD under the following conditions: radiation, Cu K-ALPHA1, 40 kV, 26 mA; scan speed, 2.000°/minute; scan step, 0.050°; angle range, 2.5-65 deg/20. In this study, these tests were performed in order to assess the potential to cause acidic water and physical properties of the rock sample.

2.2 Leaching Test to Examine the Change of Water Quality

Leaching test was performed considering the results in the sample analysis in order to investigate the long-term behavior of acidic water generation from the samples. The samples were ground into 1-2 mm of the particle size and packed into the plastic column which inner diameter was 51.6 mm and the height was 156 mm. 200 mL of deionized water was poured into the column and the leachate was sampled from the bottom of the column as in Fig. 2. The quality of the water sample was measured by an electric conductivity meter and a pH meter.

The process of pouring deionized water into the column and measurement of water quality had been conducted for 120 days to evaluate the long-term behavior of acidic water generation. The samples had been dried for 24 hours at 25°C, which is annual average temperature in Indonesia, after pouring water and measuring water quality. The cycle had been repeated until 10 times. After the 10 times of pouring water and the drying process, the samples were completely dried for 2 weeks at 25°C to keep the samples in contact with a large quantity of oxygen. The sequence of the procedure was defined as 1st step. The step had been repeated until 5th step, and the wetting and drying process could promote rock weathering.



Fig. 2.Schematic view of the leaching test.

2.3 Slaking Test

Slaking generally indicates the phenomenon in which sedimentary rocks become sandiness or muddy sediment in the case that they undergo a wetting and drying process. There are two mechanisms of slaking; advance of destructive effect of micro-crack caused by shrinkage-swelling behavior, fracture phenomenon by increase in pore-air pressure (Taylor, 1988). In this study, Slaking test was conducted to evaluate the effects of physical factors on acidic water generation. Slaking characteristics is usually evaluated based on the change of amount of rock samples with wetting and drying process (ISRM, 1981; Taylor, 1988). In this study, the slaking test was performed following the method which was modified in 2003 (Imam et al., 2003). The method included 5 steps of wetting drying process.

2.4 Sequential Extraction by Acids

Sequence extraction was performed to investigate the change of rock conditions before and after the wetting and drying process following the modified method (Huerta-Diaz and Morse, 1990; Sasaki et al., 2002). In the method, three kinds of acid were used to extract elements from the rock sample; HCl, HF, and HNO₃ (Sasaki et al., 2002). Generally, HCl can extract the elements which easily dissolve in water like precipitation and soluble salts, and HNO₃ can extract elements which are not easily dissolved in water like sulfide minerals.

Twenty mL of HCl (1M) was added to 2.5 g of dried samples in a teflon vessel, and the mixture was shaken for 16 hours at 30°C, then centrifuged at 3000 rpm for 20 minutes and decanted. The residue was washed with 10 mL of deionized water at three times, and the supernatant water was mixed with the wash water and analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

Subsequently, 30 mL of HF (46%) was added to the residue in a teflon vessel, and the mixture was shaken for 1 hour at 30°C, then centrifuged at 3000 rpm for 20 minutes and decanted. The supernatant water was mixed to the following supernatant and wash water. 30 mL of HF (46%) was added again to the residue in a teflon vessel, then the mixture was shaken for 16 hours at 30°C. 5 g of boric acid, furthermore, was added to the mixture, and shaken for 8 hours at 30°C, and the mixture was centrifuged at 3000 rpm for 20 minutes and decanted. The residue was washed with 10 mL of boiling deionized water at three times. All supernatants water and wash water were mixed and analyzed by ICP-AES.

Ten mL of concentrated HNO_3 was added to the residue in a teflon vessel, and the mixture was shaken for 2 hours at 30°C, then centrifuged at 3000 rpm for 20 minutes and decanted. The residue was washed with 15 mL of deionized water, and the supernatant water was mixed with the wash water and analyzed by ICP-AES.

3. Results

Fig.3 shows the XRD patterns for the rock sample. Pyrite which is a kind of sulfide minerals and leads to AMD was detected in the patterns in the rock sample, thereby resulting in acidic water generation. Moreover, the patterns of kaolinite which composes a clay mineral were detected in the patterns of the rock sample. The rock which contains clay minerals can indicate the disintegration caused by slaking; therefore, the rock sample may be broken down by slaking through wetting and drying process.

Table 1 shows the results of both the ABA test and the Atterberg limits test. Rocks could be classified by using the plasticity chart based on the values on the Table 1: WL is liquid limit, WP is plastic limit, and IP is plasticity index. According to the classification by calculating IP based on WL and WP, and plotting them on the plasticity chart, the rock sample was classified into clay or silt. Net Acid Producing Potential (NAPP) indicates the potential of rocks to cause acidic water. The result suggests that the rock sample have a potential to cause strong acidic water since the rocks which could cause strong acidic water generally indicate around 100-200 (kg H_2SO_4 /ton) of NAPP value in the mine (Chotpantarat, 2011).



Fig. 3.XRD patterns for the rock sample.



W _L (%)	W _P (%)	I _P (%)	NAPP (kg H ₂ SO ₄ /ton)
42.8	30.7	12.1	384.0

Fig.4 and Fig.5 shows the change of water quality at each step in the leaching test. The EC decreased with the each cycle of pouring water; however, it increased after the drying process for 2 weeks. It indicates that large quantities of the elements in the samples dissolved in water after the dry process for 2 weeks since EC is related to the amount of dissolved elements in water. Moreover, the pH decreased after the drying process for 2 weeks, indicating oxygen supply affected acidic water generation. The water quality furthermore, was not improved over time, and many elements still dissolved in water after 90 days when oxygen was supplied to the sample again. These results suggested that the supply of oxygen significantly affected AMD generation, and acidic water may continue to be generated in active pit while waste rocks are stored and exposed to oxygen in the current system in the mining operation.

The form change of the rock sample after wetting and drying process in the slaking test is shown in Fig.6. Some parts of the rock sample were broken down after a cycle of the wetting and drying process as shown in Fig.6B. Besides, most parts of the sample were broken down in accordance with the layers like coming off after the 5 cycles of the wetting and drying process as shown in Fig.6C. The expanding pressure caused by clay minerals like smectite containing water, which is slaking phenomenon, lead to the disintegration of the rock sample. These results suggested that supply of oxygen to the rock sample significantly affected the disintegration by slaking. Moreover, in 2003 Sadisun concluded that the disintegration of rocks by slaking could be classified into 3 types; constant,

accelerated, and decelerated (Imam et al., 2003). Based on the result, the change of the amount of the sample with wetting and drying process in this study was classified into "constant", indicating that the rock sample can continue to be broken down with a certain amount of small particles. The disintegration of rocks leads to the increase of surface area of rocks, resulting in the promotion of acidic water generation.



Fig. 4.Change of EC in the leachate from the rock sample.



Fig. 5.Change of pH in the leachate from the rock sample.



(A) Before the process

(B) After 1 cycle of the process



(C) After 5 cycles of the process

Fig. 6.Form change of the rock sample after wetting and drying process.

The result of sequential extraction at each step is shown in Fig.7. The amount of elements which were extracted by acid was described in Fig.7 by focusing on the iron since it was high content in the sample and existed as sulfide minerals. Two types of sample were used in the extraction; the sample which was not weathered and the sample which was weathered through wetting and drying process. The amount of Iron, which was extracted by HNO₃, in the sample before the weathering process was larger than that in the sample after. On the other hand, the amount of iron, which was extracted by HCl, in the sample after the wetting and drying process, was larger than that in the sample before. These results suggested that the form of elements were changed after the weathering process, indicating that iron existed as sulfide minerals before weathering, and they existed as the products which easily dissolve in water after the process since HCl can extract products which easily dissolve in water. Similarly, the amount of sulfur in the rock sample which was extracted by HCl was high in the sample after the wetting and drying process compared to the amount before the process. In addition to the experiment, the surface conditions of the sample before and after pouring water were observed with the Scanning Electron Microscope (SEM) as shown in Fig.8. Some products were attached to the surface before pouring water and after wetting and drying process as shown in Fig.8A, whereas they were not observed after pouring water as shown in Fig.8B. Hence, some products formed on the surface of rocks after weathering process were dissolved in water after pouring deionized water in the leaching test.



Fig. 7.The amount of extracted elements at each step in both samples.



(A) Before pouring water

(B) After pouring water

Fig. 8.Surface conditions of the rock samples.

4. Discussion and Conclusions

For these results described above, waste rocks weather while being stored in tailings and exposed to air, leading to disintegration by slaking and AMD generation since the supply of oxygen to rocks affected both slaking phenomena and AMD generation in this study. The increase of surface area of rocks could be caused by the disintegration of rocks, and it results in the promotion of AMD generation. Meanwhile, rocks can be easily broken down in the case that some parts of them are low in intensity by dissolving products formed on the surface of rocks with the chemical reaction of AMD. In the mining area, rock weathering by the both factors is generated, resulting in AMD generation. These situations are caused by the system in which waste rocks are stored separately for the order of

the backfilled materials. AMD is not generated in the case that waste rocks are not stored in tailings and they are backfilled in a short time. The current system, therefore, should be improved or the waste rocks should be treated properly in order to prevent AMD in the area; waste rocks which can cause acidic water problem should be backfilled in a short time after the excavation, or they should be covered with impermeable sheet or layer composed of clay while being stored in tailings.

The results of the sample analysis indicated that the rock sample had a potential to cause strong acidic water and they were easily broken down by slaking. These characteristics lead to the promotion of chemical reaction of AMD in the mining area while waste rocks are stored and weathered by the climate. The results can be summarized as follows:

- AMD continue to be generated for a long term while oxygen is provided to waste rocks in the mining area.
- Oxygen supply significantly affected the disintegration of the rock sample by slaking and AMD generation.
- AMD generation could be promoted by both factors; the increase of surface area of rocks caused by slake-disintegration, and the deterioration of strength in some parts of rocks caused by dissolution of products formed on the surface of rock after weathering process.
- The current system for treating waste rocks should be improved to prevent AMD generation: waste rock associated with AMD should be backfilled in a short time after the excavation, or they should be covered with impermeable sheet or layer composed of clay while being stored in tailings.

References

Casagrande, A., 1932, Research on the Atterberg limits of soils, Public roads, 13(3), 12-130.

Chotpantarat, S., 2011, A review of static tests and recent studies, *American Journal of Applied Science*, 8(4), 400-406.

Cotter, J.,Brigden, K., 2006, Acid mine drainage: the case of the Lafayette mine, Philippines, GRL-TN, 09-2006.

Dold, B., Toril, G.E., Aguilera, A., Pamo, L.E., Cisternas, E.M., Bucchi, F., Amils, R., 2013, Acid rock drainage and rock weathering in Antarctica: important sources for iron cycling in the southern ocean, *Environ. Sci. Technology*, 47, 6129-6136.

Ginting, J.K., Hideki, S., Candra, N., Akihiro, H., Takashi, S., Kikuo, M., Rudy, S.G., Budi, S., 2012, Study on co-placement of coal combustion ash-coal waste rock for minimizing acid mine drainage generation: a preliminary result of field column test experiment, *Proc. of International Symposium on Earth Science and Technology 2012*, Bandung Indonesia, 257-262.

Gleisner, M., 2005, Quantification of mineral weathering rates in sulfidic mine railings under water-saturated conditions, *Doctor thesis*, Department of Geology and Geochemistry Stockholm University, SE-106 91 Stockholm, Sweden.

Huerta-Diaz, M.D., Morse, J.W., 1990, Quantitative method for determination of trace metal concentration in sedimentary pyrite, *Marine Chemistry*, 29, 119-144.

Imam, A.S., Shimada, H., Ichinose, M., Matsui, K., 2003, Further developments in procedures to determine durability characteristics of argillaceous rocks using a static slaking index test, *Proc. of International Workshop on Earth Science and Technology*, Fukuoka, Japan, 179-186.

ISRM, Rock characterization testing and monitoring, ISRM suggested methods, Brown, E.T., 1981, Pergamon Press, Oxford, United Kingdom, 211.

Jennings, S.R., Neuman, D.R., Blicker, P.S., 2008, Acid mine drainage and effects on fish health and ecology: a review, *Reclamation Research Group Publication*, Bozeman, Montana, United States, 5-10. JIS (Japanese Industrial Standards), liquid limits and plastic limits test, code, A-1205.

Leon, A.L., Edgardo, 2012, Acid Metalliferous/Mine Drainage (AMD) and Management, Roper Bar Project Area, Western Desert Resources Ltd, *Report held ta the Western Desert Resources*, Darwin, Australia, 6-13.

Sasaki, K., Haga, T., Hirajima, T., Kurosawa, K., 2002, Distribution and transition of heavy metals in mine tailing dumps, *Materials Transactions*, 43, 2778-2783.

Sobek, A.A., Schuller, W.A., Freeman, J.R., Smith, R.M., 1978, Field and laboratory methods applicable to overburdens and mine soils, West Virginia University, *EPA*, 600, 2-78.

Taylor, R.K., 1988, Coal measure mudrocks: composition, classification and weathering process, *Geology*, 21, 85-99.

Weber, P.A., Stewart, W.A., Skinner, W.M., Weisener, C.G., Thomas, J.E., 2004, Geochemical effects of oxidation products and framboidal pyrite oxidation in acid mine drainage prediction techniques, *Geochemistry*, 19, 1953-1974.