

## **Environmental factors for the mineral precipitation in the acid mine drainage at Donghae coal mine, Korea**

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### **1. INTRODUCTION**

Many coal and metal mines in Korea have been closed due to economical or environmental reasons during last decade. The Taebaeg coal field, one of the largest coal fields, located in the middle eastern part of the Korean peninsula, has been abandoned since late 1980s. The Donghae coal mine is one of the coal mine in the Taebaeg coal field.

The Donghae mine had been actively mined from early 1960s to early 1990s but the environmental problem has been a sensitive issue soon after the initiation of mining activity. It is located in the high land about 950 meters in elevation in the Hambaeg Mt. The Donghae mine consists of small subsidiary coal mines: Samgyeong, Hanyeong, Namil, Deogyong, Pungsan and Taeyeug. These mines were closed during the period from 1989 to 1991 due to economic and industrial reasons by Coal Industry Promotion Board.

The altitude of the waste dumps in the mine is approximately 1,000 m above sea level. Underground working was done to a depth of 3,000 m. Several hundred square meters of the surface are covered with waste rocks. The AMD produced from the Donghae mine and the leachate through waste dumps enter directly into the local streams.

The geology of the study area is dominated by the Joseon and Pyeongan supergroup metasedimentary rocks of Paleozoic to early Mesozoic period. The Joseon Supergroup consists of the lower Paleozoic sedimentary sequence and the Pyeongan supergroup consists of the predominantly of shale and sandstone with subordinate limestone. The limestone is composed mainly of calcite with minor illite. The shale consists of pyrophyllite, chlorite, quartz with small amount of illite. The sandstone consists mainly of quartz, illite and chlorite. The coal dumps in the mine area are composed of coal, pyrite, quartz and chlorite.

The Sanae creek is approximately 4 km in length and 10-20 m in width, and join the larger Sodo creek. The creek is contaminated with colored mineral precipitates and it has two unpolluted tributaries. Both tributaries join the creek at 2.5km and 2km, respectively, from the outlet of the acid water.

## 2. ANALYTICAL METHODS

The water samples acidified by high-purity nitric acid were analyzed by Perkins-Elmer Optima 3000XL inductively coupled plasma emission spectrometry (ICP-AES) for metal concentration. Anions were analyzed by Dionex 4000i ion chromatography (IC). The mineral identification of stream precipitates and waste dump samples were made using X-ray powder diffraction (XRD). The morphological features of precipitates were studied using a JEOL-JSM-5200LV scanning electron microscope (SEM), equipped with a PV9880 STD energy dispersive X-ray spectrometer.

## 3. RESULTS

### 1) Chemistry of stream water

The Ca, Mg, Fe, Si, Al and SO<sub>4</sub> contents of the creek water decrease downstream from the waste mine dumps or outlets, whereas the pH values increase downstream. On the whole, metal concentrations in creek waters are high in fall whereas low in spring and summer due to dilution effect. The dilution effect of acid water was pronounced during the rainy season.

### 2) Mineralogy of precipitates

X-ray diffraction analyses show that the brownish yellow precipitates consist mainly of poorly crystallized schwertmannite, with traces of silicates including quartz, illite, and pyrophyllite. The reddish brown precipitates consist predominantly of ferrihydrite with a less amount of goethite. The white precipitate consists of low crystalline Al-sulfate with small amounts of gypsum, dolomite and calcite.

### 3) Variations of precipitates with pH

As pH increases within the drains, Fe and Al would likely undergo hydrolysis and precipitate as hydroxide or hydroxysulfate compounds. The precipitates tend to coat the surfaces of pebbles on the stream bottom up to a thickness of approximately 0.1mm. Figure 1 shows the pH ranges within which the mineral species are precipitating on the stream bottom in the study area. Bigam and Schwertmann (1996) observed that schwertmannite was a major component of precipitates and it was the only phase formed at pH 3.0. However, this study shows different stability range for schwertmannite in the Donghae coal mine drainage. The measured pH ranges of precipitation of the brownish yellow and reddish brown precipitates are 3.2-4.5 and 5.3-6.9, respectively.

Al-sulfate is found in the creek where the acid drainage with high dissolved Al and SO<sub>4</sub> is mixed with near-neutral water from tributaries. Al is mostly removed from waters having pH > 5.0, as evidenced from the absence of Al in waters flowing over the whitish precipitates on the bottom of stream. It has been found that even if Al content is relatively high (>20 mg/l), no Al-sulfate precipitates at <pH 4. From the present observations, the most favorable pH at which Al-sulfate can precipitate is in the range

of pH 4.5-6.0.

#### 4) Variations of precipitate with seasons

The key to understanding the spatial variability of SO<sub>4</sub> and the long-term trend of changing EC in mine drainage is to study their changes in the inflow with season. A systematic monitoring for a year demonstrated that concentrations of dissolved metals and SO<sub>4</sub> were highest during fall when flow was low. The highest pH values of the stream were measured in April and May, whereas the lowest pH was recorded in October. The upstream whose summit reaches above 1000m in altitude is characterized typically by

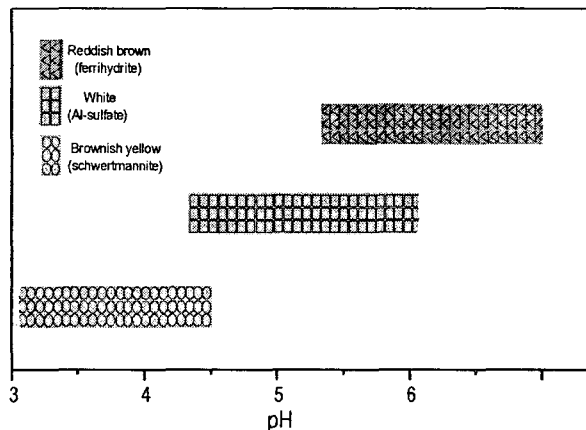


Fig. 1. The pH ranges of stream water within which characteristic mineral precipitates are found on the stream bottom.

freezing temperatures and a deep snow pack during the period from late fall to late spring. In late spring, snow gradually begins to melt down and provides abundant water to the lower part of a creek, and thereby no precipitates resulted from high pH and low dissolved metals. As the pH decreases, the white precipitates are dominated in the downstream in the Sanae creek. If the heavy rainfall in the summer season penetrates into the waste dump, the sulfides present in the coal dump are gradually reacted with water containing acids, resulting in decreasing pH and increasing the dissolved metals of the stream water. The brownish yellow precipitate is formed in the downstream in summer due to inflow of acidic water from pollution source in the upstream of Sanae creek.

#### 5) Variation of precipitate with sampling sites

Bottom sediments at the upstream and downstream exhibit different colors due to different mineralogy with change in the chemistry of stream water. The Fe content is relatively high in waters released from the mine outlet. Its content greatly decreases downward due to precipitation of Fe-hydroxide minerals. Ion contents decrease with distance from the sources of the waste pile or outlets, whereas the pH values increase due to dilution by unpolluted waters from the tributaries.

The brownish yellow materials precipitate in the upstream in July, whereas the whitish and reddish brown materials precipitate in the midstream in the same period. As pH increases downstream, the Al content of water greatly decreases with precipitation of whitish materials. Figure 2 shows X-ray diffraction patterns of precipitates from upstream to downstream in October. The brownish yellow materials consisting chiefly of schwertmannite precipitate in the upstream. The brownish yellow and whitish materials

precipitate in the midstream, whereas the reddish brown, chiefly consisting of ferrihydrite, precipitates in the downstream.

Results of SEM observations show that the brownish yellow material composed of schwertmannite coccus and spirochete with size of 0.5-1 $\mu$ m. The mixture of white and brownish yellow materials with silicate minerals have Fe, Si, Al and S. The reddish brown material consists of small sized (0.2-0.5 $\mu$ m) ferrihydrite cocous and rare rod.

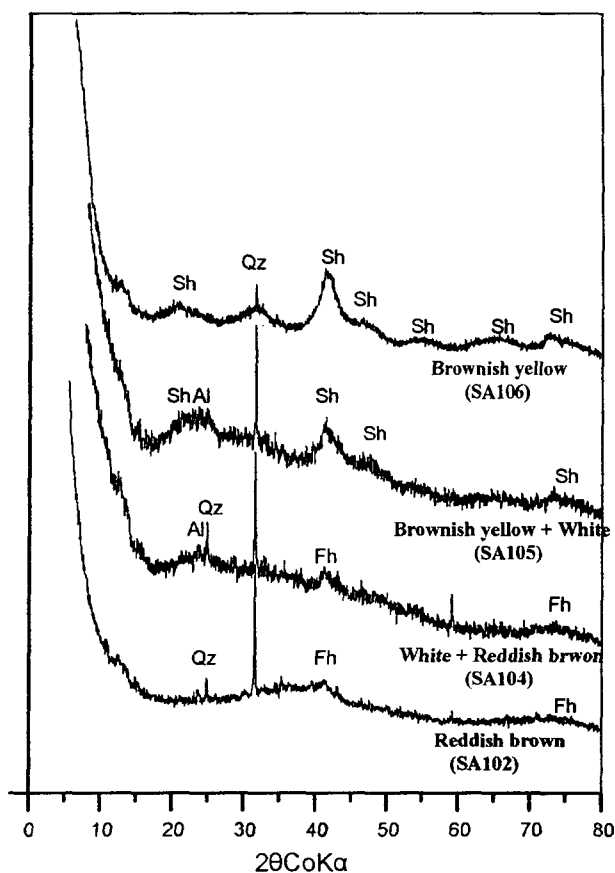


Fig. 2. XRD patterns of precipitates from upstream to downstream in the Sanae creek in October 2000.

(Sh: schwertmannite, Al: Al sulfate, Fh: ferrihydrite, Qz: quartz)