Seasonal color change of the oxyhydrous precipitates in the Taebaek coal mine drainage, south Korea, and implications for mineralogical and geochemical controls

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Abstract

The seasonal changes in pH, Fe, Al and SO_4^{2-} contents of acid drainage released from coal mine dumps play a major role in precipitation of metal hydroxides in the Taebaek coal field area, southeastern Korea. Precipitates in the creeks underwent a cycle of the color change showing white, reddish brown and brownish yellow, which depends on geochemical factors of the creek waters. White precipitates consist of Al-sulfate (basaluminite and hydrobasaluminite) and reddish brown ones are composed of ferrihydrite and brownish yellow ones are of schwertmannite. Goethite coprecipitates with ferrihydrite and schwertmannite. Ferrihydrite formed at higher values than pH 5.3 and schwertmannite precipitated below pH 4.3, and goethite formed at the intermediate pH range between the two minerals. With the pH being increased from acid to intermediate regions, Fe is present both as schwertmannite and goethite. From the present observation, the most favorable pH that basauluminte can precipitate is in the range of pH 4.45-5.95. SEM examination of precipitates at stream bottom shows that they basically consist of agglomerates of spheroid and rod-shape bacteria. Bacteria species are remarkably different among bottom precipitates and, to a less extent, there are slightly different chemical compositions even within the same bacteria. The speciation and calculation of the mineral saturation index were made using MINTEQA2. In waters associated with yellowish brown precipitates mainly composed of schwertmannite, SO_4 species is mostly free SO_4^{2-} ion with less $AlSO_4^+$, $CaSO_{4(aq)}$, and $MgSO_{4(aq)}$. Ferrous iron is present mostly as free Fe^{2+}, and $FeSO_{4(aq)}$ and ferric iron exists predominantly as $Fe(OH)_2^+$, with less $FeSO_{4(aq)}$, $Fe(OH)_2^-$, $FeSO_4^-$ and Fe^{3+}, respectively. Al exists as free Al^{3+}, $AlOH_2^-$, $(AlSO_4)^+$, and $Al(SO_4)^{2-}$. Fe is generally saturated with respect to hematite, magnetite, and goethite, with nearly saturation with lepidocrocite. Aluminum and sulfate are supersaturated with respect to

predominant alunite and less jubanite, and they approach a saturation state with respect to diaspore, gibbsite, boehmite and gypsum. In the case of waters associated with whitish precipitates mainly composed of basaluminite, Al is present as predominant Al^{3+} and $Al(SO_4)^+$, with less $Al(OH)^{2+}$, $Al(OH)_2^+$ and $Al(SO_4)^{2-}$. According to calculation for the mineral saturation, aluminum and sulfate are greatly supersaturated with respect to basaluminite and alunite. Diaspore is fairly well supersaturated while jubanite, gibbsite, and boehmite are already supersaturated, and gypsum approaches its saturation state. The observation that the only mineral phase we can easily detect in the whitish precipitate is basaluminite suggests that growth rate of alunite is much slower than that of basaluminite. Neutralization of acid mine drainage due to the dilution caused by the dilution effect due to mixing of unpolluted waters prevails over the buffering effect by the dissolution of carbonate or aluminosilicates. The main factors to affect color change are variations in aqueous geochemistry, which are controlled by dilution effect due to rainfall, water mixing from adjacent creeks, and the extent to which water-rock interaction takes place with seasons. pH, Fe, Al and SO_4 contents of the creek water are the most important factors leading to color changes in the precipitates. A geochemical cycle showing color variations in the precipitates provides the potential control on acid mine drainage and can be applied as a reclamation tool in a temperate region with four seasons.