

Rapid removal of fine particles from mine water using sequential processes of coagulation and flocculation

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Introduction

In South Korea, in total, approximately 2500 mines, including 906 metallic mines, 379 coal mines and 1173 non-metallic mines, have been developed. More than 100,000 tons of acid mine drainage (AMD) per day are currently flowing out from 206 mine adits in 152 coal mines [1,2]. The AMD from coal mines has not only a low pH, but also high concentrations of SO_4^{2-} and heavy metals including Fe, Al and Mn [3]. With these properties, the AMD is contaminating surrounding ground and surface waters, as well as creating yellow boy or white sediments [4]. Basically, AMD treatment utilizes the chemical properties of the main contaminants, such as Fe, Al and Mn. There are principally two different conventional methods to treat AMD: active (or physicochemical) treatment, which requires energy and chemical inputs, and passive (or biological) treatment, which does not require additional inputs of energy or chemicals [5]. For most cases, the attraction of metals for sulphur or carbonate is utilized in passive treatments, whereas the precipitation or adsorption of metal ions is the main mechanism in active treatments. Thus, the pH increase by use of alkaline materials such as limestone or lime turns metal ions of AMD into hydroxide particles. Although these active processes can have advantages, such as the capability to treat a high volume of AMD and reliable treatment efficiency, the produced flocs are usually fine so that they cannot be easily settled down by natural gravity. Thus, a high retention time for the settling basin is required to remove suspended solids, even though this time can vary depending on the treatment

method used for different particle sizes. Hence, in the process of precipitation, the particle size is an important factor because larger particles have more efficiency in terms of precipitation. To enlarge the floc particles, not only should the previously produced sludge be recycled to be used as a core of precipitable particles in the active treatment process, but coagulants or flocculants could be applied in the active treatment process. Since the produced flocs are mostly composed of iron compounds in the process of AMD treatment, they could cause turbidity problems in the effluents when not appropriately treated. In addition, these fine particles have some toxic effects on aquatic fish if they enter rivers [6].

To solve the problems of fine particles, various technologies such as sand filters, microscreens and the flocculation process have been developed. Specifically, alum or ferric chloride has been extensively studied and applied in the flocculation process for wastewater or drinking water treatment. Recently, high molecular weight long-chain polymers have been applied for the flocculation of suspended solids. Through the mutual binding of fine particles into large flocs, coagulants or flocculants as polymeric agents could be used to enhance the precipitation speed of flocs. Compared with inorganic chemicals, these polymers could be used with a lower dosage, reducing the sludge production. The adjustment of pH is also not required for the use of polymers. The most important advantage is that the treatment can be optimized or designed according to the molecular weight and charge densities of the specific polymers used in the removal of fine particles.

In this paper, a sequential process of coagulation and flocculation has been studied in relation to the removal of the fine particles from mine water. The colloidal suspension of fine particles can be destabilized by a coagulant agent and then agglomerated with a flocculant

agent, resulting in separation from water. The objectives of this study were (1) to find out the optimization (mixing speed, types and concentrations of coagulant–flocculant) of the coagulation and flocculation processes for removing the fine particles from mine water, and (2) to elucidate the removal mechanism of fine particles through conducting kinetic tests of turbidity reduction and zeta potential measurement.

Materials and methods

Sampling and characterization of mine water

The water sample was taken from the effluent of an AMD treatment plant, which has a problem with fine floc occurrence, in Korea and delivered to the lab. Then, the sample was exposed to an oxidizing condition for two to three days to ensure that there was no trace of Fe(II) left behind. The pH, oxidation reduction potential (ORP), total dissolved solids (TDS) and the electrical conductivity (EC) of the mine water were measured with a portable pH/ORP and EC/TDS meter (Orion 3 star). To evaluate the effectiveness of the coagulation and flocculation processes on the removal of fine flocs, the total recoverable metals of the samples, obtained before and after treatment, were measured by the EPA method 3005A (acid digestion procedure) [7]. For the analysis of total recoverable metal, 40 mL of water sample was quantified with a mess cylinder and transferred to a conical tube, and 0.8 mL of HNO₃ and 2 mL of HCl were injected. Then, the samples were digested by use of a heating block at 90–95° C, and the volume of the sample was reduced to 8–10 mL. The digested samples were cooled at room temperature and deionized water was added to produce a volume of 40 mL, then filtered with a 0.45 μ m pore membrane filter. Concentrations of

heavy metals of the filtrate were measured by ICP-AES (720-ES, Palo Alto, USA). The alkalinity of the samples, the pH of which was over 4.5, was measured by the following procedure: an indicating agent (Bromcresol Green Methyl Red) was injected and an appropriate concentrated H₂SO₄ solution was added to the sample by a titrator adapted with a cartridge, until the colour of the solution was changed to light pink, and then the volume of the titrated H₂SO₄ solution was measured to calculate the alkalinity. The Fe(II) concentration was measured by the following method: the sample was filtered using a 0.45 pore membrane filter and the ferrouzine iron reagent was added to the filtrate; after three minutes, Fe(II) was measured by UV-Vis (DR 2800, Hach, Loveland, USA) at 510 nm. The physico-chemical properties such as pH, TDS, alkalinity, etc. of the mine water are shown in Table 1. The water had a neutral pH (7.02) and a net high alkalinity (398 mg L⁻¹). The mine water contained 12.8 mg L⁻¹ of iron and 176 NTU of initial turbidity. A particle size analyser (Seishin LMS-300, Tokyo, Japan) was used to analyse the particle size distribution of the fine flocs in the mine water, the results of which are shown in Figure 1a. The median size (d₅₀) and largest floc size were 1.06 and 13.75 μ m, respectively. About 93% of the total mass was less than 3.02 μ m, representing the fine particle size distribution of flocs. An X-ray diffractometer (X'Pert Pro, Phillips, Eindhoven, The Netherlands) was used to obtain the mineralogical composition of the fine flocs (Figure 1b).

Batch jar tests

Jar tests were conducted to optimize not only the type, but also the concentration of the coagulant and flocculant. With different solid contents, charge densities and molecular weights, six different types of acryltype

anionic flocculants and two different types of polyamine-type cationic coagulants were selected from

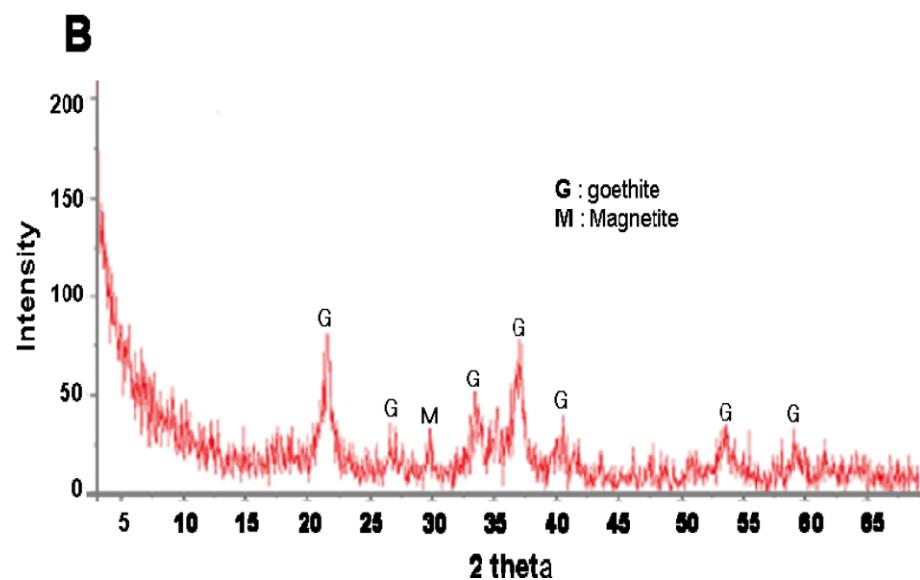
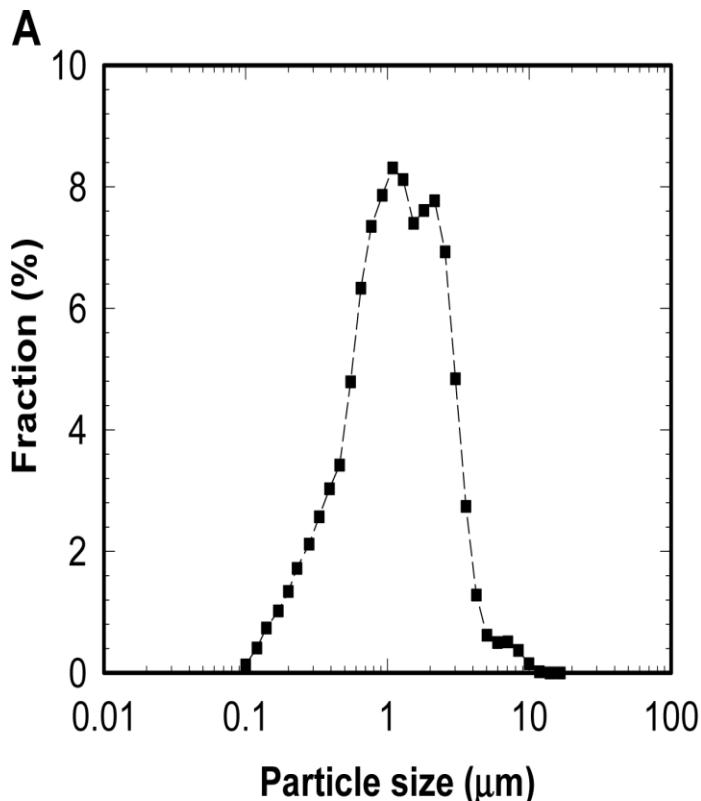


Figure 1. (a) Particle size distribution of fine flocs in mine water, (b) XRD analysis. two companies (Eyang Chemical, Seoul, Korea, and Hansol Chemical, Seoul, Korea). Table 2 shows the physico-chemical properties of these high molecular

weight coagulants and flocculants. Prior to application for treating mine water, the coagulants and flocculants were diluted with deionized water at 1% and 0.5%, respectively. In a 600 mL beaker, 500 mL of water was added, and then a coagulant was added at a predetermined concentration (2–20 mg L⁻¹). This solution was stirred for a minute. Then, a flocculant was also added at a predetermined concentration (4–25 mg L⁻¹) and the suspension was mixed at a predetermined mixing speed (in the range of 100–200 rpm) for 30 min. The following parameters were optimized: (1) type of coagulant and flocculant, (2) mixing speed, (3) coagulant concentration and (4) flocculant concentration. The turbidity was analysed by a turbidimeter (TR-30, K.R.K). To elucidate the flocculation mechanism of the fine flocs, the zeta potential of suspension after applying a coagulant or a flocculant was measured by a zeta potential analyser (ELS-8000, Ostuka). After measuring the turbidity of the suspension kinetically, the following equation (agglomeration equation) was applied to obtain the removal rate [7]. In the agglomeration equation, N usually represents the number of particles in the solution. In this study, however, the removal rate (k_a) was obtained through applying the turbidity instead of the particle numbers, based on an assumption that the turbidity linearly increases as the number of particles increases. The initially turbidity (N_0) and the turbidity at different times (N) were applied.

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