

## Formation rate of iron colloids at pH 2-3

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Various types of iron colloids are widely distributed in the earth's surface conditions, and their formation process, adsorption characteristic, and the material transport mediated by iron colloids are receiving attention. Iron colloids are often formed through several reactions including the dissolution of  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  from minerals, oxidation from  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$ , hydrolysis, and crystallization. In the present study, the reaction rate was evaluated focusing on the process in which iron colloids are formed from dissolved  $\text{Fe}^{3+}$ .

A solution of  $\text{Fe}^{3+}$  100 ppm was prepared by dissolving  $\text{FeCl}_3$  into pure water. This solution was reacted at 15, 25, 35, 45, and 55 °C. At 25 °C, pH of the solution was  $\sim 2.7$  at the start of the reaction and decreased to  $\sim 2.2$  as reaction time passed. Such change in pH is known to occur as a result of the following reactions: hydration of  $\text{Fe}^{3+}$  (release of  $\text{H}^+$ )  $\rightarrow$  formation of dissolved  $\text{Fe}(\text{OH})_3 \rightarrow$  formation of solid  $\text{Fe}(\text{OH})_3$  (Grundl and Delwiche, 1993). Therefore, information of the formation rate of iron colloids can be obtained by monitoring the pH of the solution. After the initial period of the reaction in which the rate of the decrease of dissolved  $\text{Fe}(\text{OH})_3$  concentration was slow presumably due to nucleation, the concentration of dissolved  $\text{Fe}(\text{OH})_3$  decreased in a manner like first-order reaction, and the reaction behavior deviated from the first-order reaction at the later stage. By assuming first-order reaction, rate constants of  $3.3\text{E-}5 - 1.1\text{E-}2 \text{ s}^{-1}$  were obtained at 15-55 °C, and good linearity was confirmed in an Arrhenius plot of these rate constants.

In addition to the above experiments in which time variation of the total amount of solid is considered, the time variation of the grain size of iron colloids was evaluated. A solution of  $\text{Fe}^{3+}$  100 ppm was prepared and the change of grain size at 25 °C was continuously measured using a dynamic light scattering apparatus (Zetasizer  $\mu\text{V}$ , Malvern). As a result, enough scattering intensity began to be detected after the mean diameter of iron colloids grew to 10 nm, then the grain size increased. The increase of the grain size almost stopped after 8 hours, and mean diameter at this stage was approximately 30 – 40 nm. By evaluating the number of grains from the total amount of solid and mean grain size, the grain number was estimated to decrease with time.