

The Chemistry of Fe in the Characteristics, Formation Mechanism, and Prevention of Silica Scale from Acidic Geothermal Water

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論文題名 : The Chemistry of Fe in the Characteristics, Formation Mechanism, and Prevention of Silica Scale from Acidic Geothermal Water
(酸性地熱熱水から生成するシリカスケールの特徴、機構および防止における鉄の化学)

論 文 内 容 の 要 旨

Scaling is currently one of the main obstacles in the development of geothermal power plants over the world. The most common scales concerned are siliceous scales formed inside of the surface facilities such as separators, reinjection pipelines, and even within the geologic formation surrounding the reinjection wells. This type of scaling commonly occurs at a low temperature, low pressure, and near neutral pH condition. Therefore, the most widely applied mitigation for this issue is either high temperature reinjection of geothermal fluid and/or pH modification through injection of sulfuric acid. Each has its own merit and demerit. In Dieng geothermal power plant, Central Jawa, Indonesia, the geothermal water is acidified by injection of sulfuric acid to control the formation of silica scale, however the silica scale still persists. Considering few research has been done on the silica scaling formation at acidic pH, this study aims to elucidate the formation mechanism of silica scale at acidic pH. Moreover, this study also serves as foundation for future utilization of acidic geothermal fluid in order to boost the carbon-neutral energy production for a sustainable development. The results of scale characterization, investigation on the formation mechanism of silica scale, and its prevention method are documented in a 5-chapters dissertation.

Chapter I introduces the general nature related to this research. This includes the general introduction to the scaling problem in geothermal power plant, introduction to the Dieng geothermal power plant, general background of the research, objectives, and its significance.

Chapter II describes the diverse characteristics of scales collected at the Dieng geothermal power plant and its relations to the change in the properties of the geothermal water. The characterization of the scale includes physical characteristics, chemical composition, and the texture of the scale, whereas the characterization of the geothermal water was based on the chemical composition and chemical state of dissolved species, in addition to pressure, temperature, and fugacity of gasses. The scales can be generally categorized into three types. (1) Scale in the two-phase pipeline composed of amorphous silica, a mixture of Al, K, and Fe rich silicate, and sulfide minerals. (2) Scale in the brine pipeline (after separator) mainly composed of amorphous silica with small amount of Al, K rich silicate and sulfide minerals. (3) Scale in the open canal mostly composed of amorphous silica containing small particles of hydrous ferric oxide. The study shows that the characteristic of the scales is directly correlated to the change of the physicochemical properties of the geothermal water, which in turn control the saturation index of certain minerals at each location.

Chapter III explains the role of Fe on the polymerization and precipitation of silica under atmospheric pressure and acidic pH conditions. The study was carried out through series of characterization of geothermal water and plates immersion experiment at Dieng geothermal power plant, as well as characterization and mixing of acidic and weakly alkaline geothermal water at Hatchobaru geothermal power plant, Japan. The experiments at Dieng show that under pH 5.2, small particles (<450 nm) of hydrous ferric oxide combining silicic acid precipitated on the surface of metal plates. The particles then slowly interacted with monosilicic acid in geothermal water and resulted in the growth of silica scale with low Si/Fe atomic ratio (<50). Under pH 6.5, copolymerization of Fe and silicic acid rapidly occurred in the geothermal water, forming large (10–50 μm) polysilicic acid particles containing Fe, while the small particle of hydrous ferric oxide was also formed. Both then precipitated, resulting in a precipitate with high Si/Fe atomic ratio (>150). The experiments at Hatchobaru show that Fe in the acidic geothermal water existed as colloidal hydrous ferric oxide and hydrolyzed species of Fe^{3+} under atmospheric condition. Under strongly acidic pH condition, the hydrous ferric oxide bound with monosilicic acid and then accumulated, resulting in a precipitate with low Si/Fe atomic ratio. Under near neutral and weakly alkaline pH conditions, polymerization of silicic acid rapidly occurred, and the hydrolyzed species of Fe^{3+} combined to the surface of polysilicic acid particles. The polysilicic acid particles aggregated through the reaction with the hydrous ferric oxide, resulting in a precipitate with high Si/Fe atomic ratio. In both locations, Fe clearly initiates the formation of silica scale at acidic pH and atmospheric condition.

Chapter IV describes the potential methods of the prevention of silica scaling through removal and deactivation of Fe in geothermal water. For the removal of Fe, activated carbon was used in on-site experiments at Dieng, Hatchobaru, and Yamagawa geothermal power plants under strongly acidic to weakly alkaline pH conditions. Fe was effectively adsorbed on the surface of activated carbon at pH 5 to 6, even though the adsorption rate was slow. Simultaneously, the polymerization of silicic acid was also prevented. This effect was more pronounced at near neutral to acidic pH conditions. For the deactivation of Fe, tetrasodium ethylenediaminetetraacetate ($\text{EDTA}\cdot 4\text{Na}$) was used to adjust the pH of acidic geothermal water (pH ~ 3) to pH 5. Sodium hydroxide (NaOH) was also used as a comparison. From series of polymerization and adsorption experiments in laboratory and at Hatchobaru geothermal power plant, Fe remained in aqueous geothermal water throughout the experiments when $\text{EDTA}\cdot 4\text{Na}$ was used. However, a Ferron-reactive analysis suggested that Fe was present as a non-reactive complex. On the contrary, Fe concentration was decreased during the experiments when NaOH was used. Similarly, the concentrations of monosilicic acid and total silicic acid slightly decreased when $\text{EDTA}\cdot 4\text{Na}$ was used, but significantly decreased when NaOH was used. The results show an effective trapping of Fe(III) by EDTA leading to the prevention of its accelerating effect on the polymerization of silicic acid.

Chapter V presents summary of the findings and conclusions of this research which emphasizes the important role of Fe in the formation of silica scale. Furthermore, implications and recommendations for future study are also presented in this final chapter.