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Experimental determination of solubility constants of saponite at elevated temperatures in high ionic strength solutions

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Abstract

Saponite occurs in a wide range of environments from hydrothermal systems on the Earth to surface deposits on Mars. Of practical importance is that Mg-saponite forms when glasses for nuclear waste are altered in Mg-bearing aqueous solutions. In addition, saponite is favorably considered as candidate buffer material for the disposal of high-level nuclear waste and spent nuclear fuel in harsh environments. However, the thermodynamic properties, especially for Mg-saponites, are not well known. Here the author synthesized Mg-saponite (with nitrate cancrinite) following a previously reported procedure and performed solubility experiments at 80 °C to quantify the thermodynamic stability of this tri-octahedral smectite in the presence of nitrate cancrinite. Then, in combination with the equilibrium constant at 80 °C for the dissolution reaction of nitrate cancrinite from the literature, the author determined the solubility constant of saponite at 80 °C based on the solution chemistry for the equilibrium between saponite and nitrate cancrinite, approaching equilibrium from the direction of supersaturation, with an equilibrium constant of –69.24 ± 2.08 (2σ) for dissolution of saponite at 80 °C. Furthermore, the author extrapolated the equilibrium constant at 80 °C to other temperatures (i.e., 50, 60, 70, 90, and 100 °C) using the one-term isocoulombic method. These equilibrium constants are expected to have applications in numerous fields. For instance, according to the extrapolated solubility constant of saponite at 50 and 90 °C, the author calculated the saturation indexes with regard to saponite for the solution chemistry from glass corrosion experiments at 50 and 90 °C from the literature. The results are in close agreement with the experimental data. This example demonstrates that the equilibrium constants determined in this study can be used for reliable modeling of the solution chemistry of glass corrosion experiments.

Keywords: Glass corrosion, Mars, nuclear waste management, buffer materials, hydrothermal alteration of basalt, disposal in salt formations, disposal in clay formations