

Kinetic Monte Carlo modelling of barite-celestite solid solution growth, dissolution, and radium uptake

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Most of the minerals on Earth occur in form of solid solutions. The study of solid solutions has significant applications in industry as well as in fundamental science: from catalyst design to the prediction of P-T conditions of ore-forming processes. Barite (BaSO₄) is a mineral forming binary and ternary solid solutions with Sr and Ra. These two elements have radioactive isotopes produced by nuclear reactions. The ability of barite to incorporate them into the crystal lattice makes this mineral a promising material to use in nuclear waste repositories.

According to thermodynamic and DFT studies (Vinograd et al., 2018), (Ba_xSr_{1-x})SO₄ systems occur as continuous series of solid solutions. However, in natural samples the concentration of isomorphous components rarely exceeds 0.2 molar fractions (Hanor, 1968). Instead, barite and celestite commonly form oscillatory zoned crystals with different concentrations of solid solutions endmembers (Prieto et al., 1993; L'Heureux and Jamtveit, 2002). Many authors suggested that the oscillatory zonation in minerals is caused by the change of external thermodynamic parameters (e.g., Shore and Fowler, 1996). An alternative concept mentioned by the authors is the possibility of internal self-organization processes. In this study we apply this concept to understand the behavior of (Ba_xSr_{1-x})SO₄ system, and suggest that the kinetic factors may control the oscillatory zonation and limited range of chemical compositions.

The kinetics of the system's behavior is studied by using the Kinetic Monte Carlo (KMC) approach. Previously the model of pure barite dissolution was developed by Kurganskaya et al. (2022). We present here the first KMC model of crystal dissolution, growth, and Ra²⁺ ion adsorption for (Ba_xSr_{1-x})SO₄ system. We discuss how this system evolves over time and what is the role of kinetic factors in spatial organization of its chemical components.

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