

The first Kinetic Monte Carlo model focused on the dissolution of ternary solid solution: a case study on (Ba,Sr,Ra)SO₄ system

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Barite is the main mineral of barium in the Earth's crust being the subject of interest for industrial as well as for academical scientists. The ability of barite to form solid solutions with the radioactive isotopes of Sr and Ra makes this mineral a promising backfilling material for construction of nuclear waste repositories. The studies on dissolution of Ra-Sr-bearing barite are important to model the situation of water injection into the repository, constructed with use of mineral matrices.

The kinetical processes on mineral-water interface are studied by experimental, e.g., AFM and VSI as well as theoretical techniques such as DFT and MD. The AFM and VSI can provide a detailed information on the surface topography at nano- and micrometer scale, however these methods cannot be used to get the information on molecular scale reactions. Such methods as DFT or MD can be utilized for extraction the mechanisms of molecular reactions, however are limited on the system size and time trajectory possible to model. The Kinetic Monte Carlo method can easily close this gap, cause it requires the rates of molecular reactions as an input, however provides an information on macroscopic behavior of the system.

The KMC modelling of the solid solution-water interface is a challenging task due to high complexity of system. In our previous studies we found that the implementation the bond lengths parameters into the parameterization procedure significantly improves the parameterization. In the current study we provide the first KMC model of the dissolution of (Ba,Sr,Ra)SO₄ ternary solid solution, considering also the binary solid solutions. For our model we use the data from the DFT geometry optimization to correctly introduce the effect of the bonds distortion and the strain field.

