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THERMODYNAMICS OF ZEOLITES: A MODEL OF POLYHEDRAL CONTRIBUTIONS BASED ON A VARIETY OF INDEPENDENT EXPERIMENTAL DATA

ARNAULT LASSIN[1], SYLVAIN GUIGNOT[1], PHILIPPE BLANC[1] AND BENOIT MADÉ[2]

[1]BRGM
[2]Andra

Presenting Author: a.lassin@brgm.fr

Zeolite minerals are common aluminosilicate minerals in the environment and widespread in commercial and industrial applications. Either natural or synthetic, they are characterized by a great variety of chemical compositions, and by specific ion-exchange and hydration properties. In the context of deep underground nuclear waste repositories in clayey rock formations, they are expected to form at the interface between cement barriers and the clay-rock, with potential impacts on the local flow and transport properties. Because repository performance is assessed over long periods of time (100 000 years, usually), their geochemical stability is a relevant issue. Numerous efforts were made to determine the thermodynamic properties of zeolites both experimentally and theoretically. Large sets of experimental data, including calorimetric and solubility data are now available, which makes it possible to develop thermodynamic models with the aim to show predictive capabilities.

In the present work, a polyhedral contribution approach is developed for calculating standard formation properties of zeolites, namely Gibbs energy, enthalpy and entropy. This stepwise development relies first on previous critical selections of published data, regarding mostly calorimetric data (Blanc et al., 2015). In a second step, recently published solubility data are considered for implementation in the critical selection. The polyhedral method is then applied, allowing sorting out the thermodynamic datasets, depending on their contribution to the overall model performance.

A series of different optimization strategies were tested and their results compared. Finally, an optimized set of thermodynamic properties could be obtained for the constitutive polyhedra of an ensemble of 49 zeolite minerals. Except for 5 zeolites, the average deviations on standard formation properties are less than 1% for Gibbs free energy and enthalpy, and less than 6% for entropy.

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