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Selection of thermodynamic properties for zeolite minerals

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In the context of deep underground nuclear waste repositories, safety has to be assessed over long periods of time (100 000 years, usually). At the European level, repository concepts may use various types of physical barriers to prevent the release of toxic elements into the geosphere. Among those materials, clay minerals do represent an important contribution, for the engineered barrier made of bentonite, or for the surrounding geological host rock, which may include a large amount of clays. Gaucher et al. (2004) showed that the contact with cement based materials, used for repository underground facilities could induce mineralogical transformations in both sides of the cement / clay interface, producing zeolite minerals. Those 3D framework silicates are stabilized in alkaline environments and were found in many experiments where clay minerals were put in contact with alkaline solution (Gaucher and Blanc, 2006).

Assessing the stability of those minerals over long periods of time is thus of importance for risk assessment calculations. Thermodynamic properties are essential for such applications and within the framework of the Thermochimie project (Giffaut et al., 2014), much efforts have been devoted to filling in gaps in thermodynamic databases in general and for zeolites in particular (Blanc et al., 2015). The current selection establishes the consistency within the selections for clay, zeolites and cement phases (Blanc et al., 2015). However, since then several studies have been published, aiming to investigate the thermodynamic properties of such minerals, like the work proposed recently by Ma and Lothenbach (2020a) and Ma and Lothenbach (2020b), for example. Such studies allow completing the Thermochimie database for the properties estimated in the previous selection, or for the minerals which can not be included, given the lack of data in the literature. Furthermore, the determinations carried out by Ma and Lothenbach (2020a) and Ma and Lothenbach (2020b) enlighten the necessity to distinguish zeolites which have been equilibrated with porewaters over thousands of years and phases freshly precipitated, presenting higher solubilities. This work presents and illustrates the guidelines followed for the new selection of zeolites thermodynamic data. Illustrations are provided, along with comparisons with recent determinations.

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