

ThEREDA Database Project:

Extensions of the Pitzer Database with Respect to Phosphate, Alkaline Earth Metal Sulphates, Heavy Metals and Fission Products – First Results

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ABSTRACT

In an effort to extend the database for fission products, an extensive screening of the literature was done for Cs, Rb, and Sr. The data for Cs were amended by own isopiestic measurements in the GRS, leading to an almost complete set of Pitzer parameters for interactions with Na, K, Mg, Ca - Cl, SO₄ for 298.15 K and 1 atm [1, 2]. For Sr and (to a lesser extent) for Rb data gaps remain.

As a first step to model the solubility of low soluble phosphates in brines, the literature was screened for suitable data, essentially water vapor pressures and solubilities. Missing data were amended by own isopiestic measurements [3, 4].

For the solubility of alkaline earth sulfates at elevated temperatures, data sets were prepared specifically with regard to mixed NaCl-CaCl₂-solutions. For Sr and Ba credit was taken from Monnin (1999) [5]. Experimental solubilities can be reproduced up to 100°C. For higher temperatures considerable deviations occur. It is hypothesized that this is due to the limited temperature up to which the Pitzer parameters in ThEREDA for the systems of oceanic salts is valid (about 100-120°C). As the extension of the database for these Pitzer parameters up to 200°C is presently underway, these results may change in the near future.

For binary RaCl₂ interactions at 298.15K we adopted Pitzer parameters as estimated in Rosenberg 2011 [6]. The temperature dependence of BaCl₂-interactions as given in [5] was projected on top so as to give the well founded values at 298.15K as given by Rosenberg et al. [6]. Temperature dependent RaSO₄ interactions were adopted from Paige et al (1998) [7]. It was further assumed that BaSO₄ and RaSO₄ form an ideal solid solution at low values for x(RaSO₄) [6].

For lead we adopted a set of internally consistent Pitzer parameters, solubility constants, and formations constants of chloro- and sulfato-complexes from Hagemann [8]. Temperature functions for the formation of chloro-complexes [9] were projected on top such as to yield the values from Hagemann at 298.15K.

No solubility data for Anglesite at elevated temperature were found. Thus, standard enthalpy of formation, standard entropy and heat capacity data were used to calculate a Gibbs energy function of temperature, which was subsequently adjusted to give the precisely known solubilities at 298.15K.

Finally, a short view is given on the database for cementitious systems. Considered test cases cover low-saline systems mainly, but an example for solubility of cement phases in brine is given also. The presentation gives an account on intermediate results.