

Water activity and modeling of ternary solutions of the systems CsCl-MCl-H₂O and Cs₂SO₄-MSO₄-H₂O (M=Na,K,Mg) at 25 to 90 °C

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Cesium-137 is a medium-lifetime radionuclide resulting from fission of nuclear fuel such as uranium-235 and other isotopes.

Description of its behaviour in the vicinity of geological repositories for nuclear waste requires a thermodynamic model that allows the calculation of activity coefficients for the ion Cs⁺ at elevated temperatures relevant for the specific disposal concept (e.g. up to 90°C) and the typical salinity of the formation waters.

Earlier studies by GRS resulted in a set of Pitzer ion interaction parameters for the system Cs, Na, K, Mg, Ca, Cl, SO₄-H₂O at 25°C [1,2].

Additional isopiestic and calorimetric measurements were conducted in binary and ternary aqueous solutions containing cesium at temperatures between 25 and 90°C. The experiments were designed to cover solution concentrations up to the saturation of the individual Na, K, and Mg salts but with restricted Cs content (< 7 mol/kg). Data from the measurements were combined with literature data and the THEREDA Pitzer model for the seawater system [3] in order to provide a model for Cs⁺ applicable at 25 to 90°C. Measurements in ternary systems such as CsCl-NaCl-H₂O and CsCl-MgCl₂-H₂O clearly showed a markable deviation from ideal mixing. This can be interpreted as evidence for significant ion pairing or complex formation. Nevertheless, the developed Pitzer model performs sufficiently well without explicit consideration of additional Cs species.

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[1] Scharge *et al.* (2012) J. Chem. Eng. Data 57, 1637–1647. [2] Scharge *et al.* (2013) J. Chem. Eng. Data 58, 187–188. [3] Moog *et al.* (2015) Appl. Geochem. 55, 72–84