A conventional Pitzer model for {Nd^{3+}–H^+–Na^+–K^+–Ca^{2+}–Cl^-–OH^- }aq at 298 K, 0.1 MPa, and 2 < pH < 13

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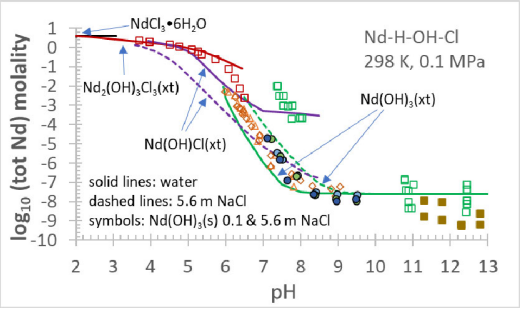
A Pitzer ion-interaction model is presented for the Nd^{3+}–H^+–Na^+–K^+–Ca^{2+}–Cl^-–OH^- system at 298 K and 0.1 MPa. This analog system is used for geochemical modeling of trivalent actinides [An(III)] in evaporite hosted nuclear waste repositories. In contrast to previous geochemical models for the An(III)-H-Na-K-Ca-Cl-OH system, the new model is both parsimonious and accurate.

Our model was fitted to isopiestic, emf and solubility data for the 'ternary' chloride salt systems. The model was then extended to pHs at which solid $Nd(OH)_3$, $Nd(OH)_2Cl$ and a speculative double salt ($Nd(OH)_3$•$NdCl_3$) are stable. The two hydroxychloride salts join the stability fields of $NdCl_3$•$6H_2O$ and $Nd(OH)_3$ to form a continuous model over 2 < pH < 13.

With addition of standard state free energy terms for crystalline (xt) and amorphous (am) $NdOH_3$(s) and $NdOH_2^+$ and $Nd(OH)_3^0$ solute species, the model reproduces the full body of thermodynamic data over an ionic strength range from infinite dilution to the solubility limits of: NaCl, KCl, $CaCl_2$•$6H_2O$, $Nd(OH)_3$(s), the hydroxychloride salts, and for mixtures containing up to 4.5 molal HCl(aq). No Pitzer parameters involving $NdOH^{2+}$, $NdOH_2^+$ and $Nd(OH)_3^0$ were necessary. The model adds 5 'binary' and 3 'ternary' Pitzer ion-interaction parameters to the set in Harvie et al. [1].



[1] Harvie et al. (1984) *Geochim. Cosmochim. Acta* **48**, 723.