

The hydrothermal solubility of monazite-(Ce) and xenotime-(Y)

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CePO₄ and YPO₄ are major components in monazite and xenotime, respectively, which are common hydrothermal phases in REE mineral deposits. Both minerals also occur as secondary minerals in iron-oxide-apatite deposits [1,2], and as accessory phases in high-grade metamorphic rocks where they display varying degrees of metasomatism. Studying the cause of their compositional variations using thermodynamic modeling may provide geochemical signals for interpreting P-T-x of crustal fluid-rock interaction. The thermodynamic properties of monazite and xenotime have been determined using several calorimetric methods [3], but only a few solubility studies have been undertaken, which test the reliability of both the calorimetric data and thermodynamic properties of associated REE aqueous species [4]. Combining available calorimetric data with the REE aqueous species from Haas et al. [5], implemented in the Supcrt92 database [6], yields several orders of magnitude differences when compared with our solubility measurements. To reconcile these discrepancies, we have used the GEMS code package [7,8] and GEMSFITS [9] for parameter optimization, and re-evaluated the standard Gibbs energies for aqueous REE species, while maintaining consistency with available calorimetric measurement of the REE phosphates. This study points to a need to revise the thermodynamic properties of the REE hydroxyl species, which will have an impact on the calculated solubilities of the REE phosphates and our understanding of the mobility of REE in hydrothermal fluids. Our new experimental data will be implemented in the MINES thermodynamic database (<http://tdb.mines.edu>) [10] for modeling the chemistry of crustal fluid-rock equilibria.

[1] Harlov et al. (2016), *Econ. Geol.* 111, 1963-1984; [2] Hofstra et al. (2016), *Econ. Geol.* 111, 1985-2016; [3] Navrotsky et al. (2015), *J. Chem. Thermodyn.* 88, 126-141; [4] Gysi et al. (2015), *Chem. Geol.* 83-95; [5] Haas et al. (1995) *Geochim. Cosmochim. Acta* 59, 4329-4350; [6] Johnson, Oelkers, Helgeson (1992), *Comp. Geosci.* 7, 899-947; [7] Wagner et al. (2012), *Can. Miner.* 50, 1173-1195. [8] Kulik et al. (2013), *Computat. Geosci.* 17, 1-24; [9] Miron et al. (2015), *Appl. Geochem.* 55, 28-45; [10] Gysi (2017), *Pure and Appl. Chem.* 89.