

The non-ideal mixing thermodynamic effects on REE formation and fractionation in natural hydrothermal systems

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REE ores are commonly associated with enrichment in U and Th, which cause separation issues for HREE from LREE, and radioactive waste streams during their mining and processing. Thus, it is important to be able to predict conditions under which An-depleted ores are formed. U and Th are often getting incorporated in REE-bearing minerals as solid solutions, and, thus, this ability largely depends on thermodynamic stability and phase equilibria of the latter. However, our understanding of the mixing thermodynamics of REE and U/Th in mineral phases is limited. Conventionally, the mixing of various REE is assumed to be ideal and described by the random distribution or regular solution models. Although being a convenient model used for geochemical modeling, its application in REE hydrothermal systems may yield incorrect or inaccurate geochemical prediction. In this talk, we will present two showcases to emphasize the importance of non-ideal mixing and its impacts on REE mineralization and fractionation in solid phases. In the first case, we examined the thermodynamics of LREE binary hydroxylbastnäsite solid solutions and HREE binary xenotime solid solutions, which have positive enthalpies of mixing and lead to miscibility gaps at low temperatures. In the second case, we showed that the mixing of U and Th in uranothorite (isostructural to xenotime) has a strong negative enthalpic effect and can only be described by a subregular solid solution model. The consequence of the non-ideal mixing is an extended phase boundary in terms of temperature and oxygen fugacity, where pure U endmember (coffinite) is unstable. These two cases highlight the importance of correctly accounting for the non-ideal mixing effects in accurate prediction of phase boundary and mineral behavior under hydrothermal systems, which will enable further development of new exploration techniques permitting the identification and localization of REE-fractionated or Th- and U-depleted REE ore deposits.

