

Re-evaluation of equilibrium relationships involving U^{6+}/U^{4+} and Fe^{3+}/Fe^{2+} in hydrothermal fluids and their implications for U mineralization

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Uranium (U) and iron (Fe) are generally considered to have contrasting aqueous geochemical behavior, with U dissolution being favored in oxidizing fluids as oxidized U (U^{6+}) and Fe in reducing fluids as reduced Fe (Fe^{2+}) species. However, high concentrations of both U and Fe have been reported for the ore fluids of uranium deposits, suggesting that the two elements can be co-transported in the same fluid. Geochemical modeling in this study indicates that high concentrations of U (>10 ppm) and Fe (>1000 ppm) can be dissolved in acidic aqueous fluids, for a wide range of $\log fO_2$ above and below the MH, at temperatures from 150 to 250 °C. There are four different combinations of dominant dissolved U and Fe species depending on the pH and redox conditions: U^{6+} and Fe^{3+} species at relatively high fO_2 , U^{4+} and Fe^{2+} species at relatively low fO_2 , and U^{6+} and Fe^{2+} species or U^{4+} and Fe^{3+} species at intermediate fO_2 conditions. Because the most important U mineral in hydrothermal uranium deposits is uraninite, in which U is present as U^{4+} , a necessary condition for U mineralization is that U is either transported as U^{4+} species or, if transported as U^{6+} species, precipitation of uraninite is induced by a reducing agent. The recognition that U^{6+} and Fe^{2+} species can be co-transported in the same fluid indicates that uraninite can be precipitated without the need to invoke a reducing agent external to the ore fluid. The driver of uranium mineralization in this case is an increase in pH due to fluid-rock interaction that leads to the alteration of the rocks by clay minerals and associated coupled sorption-reduction. These findings provide for a more extensive range of conditions for U transport and deposition than previously imagined and require that interpretation of the conditions of uranium mineralization consider both oxygen fugacity and pH. This study underscores the potential for uranium deposits to occur in geological settings where reducing agents are either absent or insufficiently abundant to account for the observed mineralization, and the possibility for the transport of U in environments that might otherwise be considered unfavorable.