THE EFFECT OF AQUEOUS BICARBONATE ON THE DISSOLUTION RATE OF AUTUNITE IN SUBSURFACE SOIL OF THE US DOE HANFORD SITE

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Background

Bicarbonate plays a significant role in the uranium geochemical cycle. It is one of the dominant anions that complexes uranium and can yield highly soluble and mobile species. Under advective conditions bicarbonate can decrease the solution saturation state resulting in an increase in the dissolution rate of uranium.

It is important to understand and be able to predict the fate and transport of uranium in groundwater. Autunite is a primary uranium mineral that can impart significant control over aqueous uranium concentrations in natural and anthropogenically altered environments.

Environmental variables that influence autunite dissolution include: bicarbonate concentrations, pH and temperature.

Objective

The purpose of this study is to quantify the effect of aqueous bicarbonate, temperature and pH on the dissolution kinetics of natural Ca meta- autunite I, Ca $[(UO_2)(PO_4)]_2 \cdot 3H_2O$.

Materials and Methods

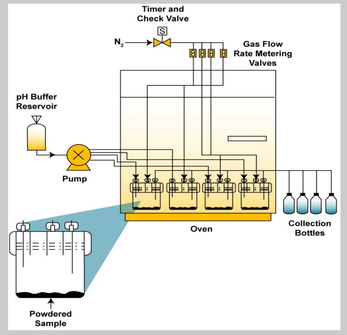
The dissolution kinetics of autunite were determined using single-pass flow-through (SPFT) apparatus

Bicarbonate Concentrations:
0.0005, 0.001, 0.002, 0.003 M

pH:
7, 8, 9, 10, 11

Temperature:
23, 40, 60, 90 °C

Flow Rates:
1, 1.5, 2.5, 3 L/day

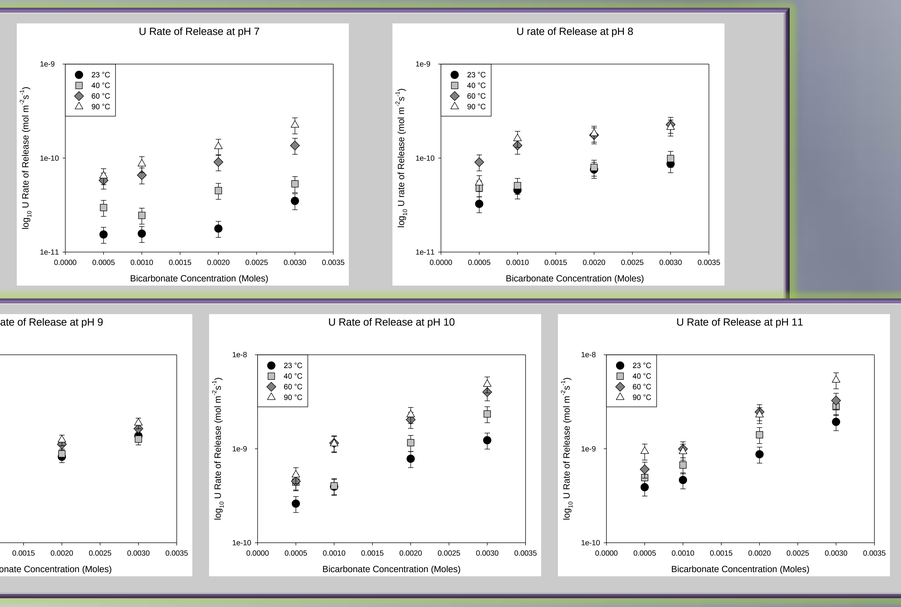


The rates obtained from the SPFT were normalized to the element mass fraction in autunite by the following formula:

$$r_{i,j} = \frac{(C_{i,j} - C_{i,b})q_j}{f_i S_j}$$

(McGrail 1997)

Results



Conclusions

Plots presented above illustrate the release of uranium from Ca-meta-autunite under the bicarbonate concentration range of 0.0005 to 0.003 M and the temperature range of 23° to 90°C across the pH range of 7 to 11.

Consistent with previous studies, autunite minerals have shown to be very stable with respect to temperatures lower than 90°C. Additionally, the rate of uranium release increases as a function of pH.

The release rate of uranium from Ca-meta-autunite is directly correlated to increasing bicarbonate concentration. The presence of bicarbonate increases the chemical affinity within the system resulting in an increase in autunite dissolution as a function of bicarbonate. Moreover, uranium-carbonate complexes are generally negatively charged or neutral, resulting in highly mobile aqueous species in subsurface environments. Thus, understanding the impact of bicarbonate on the release of uranium from autunite is critical to predicting the long-term transport and fate of uranium.

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Reference

[1] D.M. Wellman, et al. Effects of pH, temperature, and aqueous organic material on the dissolution kinetics of meta-autunite minerals, $(Na, Ca)_{2\text{-}1}$ $[(UO_2)(PO_4)]_2$ $3H_2O$, *American Mineralogist* **2006,** 91, 143.