

## Experimental study of Ca-uranyl-carbonate surface complexation on *Bacillus subtilis* bacterial cells

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We measured the effect of aqueous Ca and carbonate on the adsorption of U onto gram-positive bacterial cells. Gorman-Lewis *et al.* [1] observed enhanced U adsorption to *Bacillus subtilis* in the presence of Ca and dissolved CO<sub>2</sub> at pH>6 and attributed the enhanced binding to the presence of a Ca-uranyl-carbonate bacterial surface complex. However, they only collected data at one Ca:U:bacteria molal ratio. In order to more rigorously test for the presence of this complex and to better constrain its thermodynamic stability, we conducted batch U adsorption experiments at a fixed pH and U:bacteria molal ratio but as a function of Ca concentration.

Each experimental system contained  $2.1 \times 10^{-5}$  M total aqueous U and 0.625 g/L *Bacillus subtilis* in a 0.1 M NaClO<sub>4</sub> electrolyte to buffer ionic strength. The experimental solutions were equilibrated with atmospheric CO<sub>2</sub>, and the pH of each experiment was controlled to remain fixed at either pH 8 or 9. Ca concentrations in solution were adjusted to range from 0.5 mM. We measured the extent of U adsorption, and the results were modeled using a non-electrostatic discrete site model of the reactivity of the bacterial cell wall functional groups.

The experimental results show that U adsorption remains constant in the presence of increasing concentrations of Ca. Speciation calculations based on the stability constants for U-bacterial surface complexes from Gorman-Lewis *et al.* but neglecting the proposed Ca-U-carbonate bacterial surface complex, predict that under our experimental conditions the Ca-U-carbonate aqueous complex should become the dominant U reservoir as Ca increases, causing a corresponding decrease in adsorption of U onto the bacteria. We do not observe this decrease, suggesting that either: 1) the enhanced adsorption relative to the predicted level is caused by the formation of a Ca-U-carbonate bacterial surface complex or 2) the stability constant for the aqueous Ca-U-carbonate complex is incorrect. We use the experimental data to constrain the stability constant for the Ca-U-carbonate bacterial surface complex and note that this complex could control U distributions in bacteria-bearing geologic systems.

[1] Gorman-Lewis *et al.* (2005) *Environ. Sci. Technol.* **39**, 4906-4912.

## Lithium isotope compositions of chondrules and CAIs from Allende and various ordinary chondrites

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Variation in Li isotope compositions has been observed within and among members of ordinary chondrites (OC) and carbonaceous chondrites (CC), indicating isotope heterogeneities in the early solar nebula [1]. The hosts for such heterogeneities may be preserved within components of chondrites of low petrologic grade, e.g., in Calcium Aluminium-rich inclusions (CAIs), chondrules and matrix. In order to unravel the reason for the observed differences in the Li isotope compositions of bulk chondrites we have studied several primitive chondrites. Semarkona (LL3.0), Bishunpur (LL3.15) and Allende (CV3) were chosen because of their low petrologic type and low degree of hydrous or thermal alteration, compared with more equilibrated OCs such as Saratov LL4, Bjurböle L/LL4, Bremervörde H/L3.9.

Bulk compositions of Allende and OCs vary between  $\delta^7\text{Li} = +3.2\%$  at 1.1 µg/g (Bishunpur) and  $\delta^7\text{Li} = +1\%$  at 1.3 µg/g (Bremervörde) and fall in the range reported earlier for Allende and other OCs [1]. The majority of chondrules and CAIs have a  $\delta^7\text{Li}$  varying between -5 to +4‰ at 0.1 to 0.8 µg/g. Only two chondrules, 1 from Bishunpur and 1 from Bjurböle have relatively high Li concentrations of 2.7 and 3 µg/g, respectively. No correlation between Li concentration and  $\delta^7\text{Li}$  is apparent for chondrules or for CAIs.

If compared to the bulk composition average  $\delta^7\text{Li}$  of chondrules in Allende and OCs are similar or lighter and Li abundances are consistently lower. Various processes can be responsible, such as fractionation during condensation, melting and evaporation during chondrule formation, and/or precursor materials affected by pre-solar and early solar nucleosynthetic processes. We conclude that (1) chondrules from OCs and Allende are similar in their Li isotopes and Li compositions, implying that there are no significant differences in their source regions. (2) Higher Li abundances and heavier  $\delta^7\text{Li}$  in chondrites compared to their chondrules and CAIs require the presence of at least one other component, possibly residing in the matrix. (3) The lighter  $\delta^7\text{Li}$  bulk in OCs [1] are likely to be controlled by the proportions of CAI's and chondrules in the bulk chondrite.

[1] Seitz *et al.* (2007) *EPSL* **260**, 582-596.