

## **Uptake of uranium by carbonate crystallization from reduced and oxidized hydrothermal fluids.**

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Societal importance of uranium (U) cannot be underestimated: it is one of the few remaining sources of clean energy not associated with continuous environmental pollution. Although clean and ecologically-friendly if properly operated, nuclear energy production can, however, be associated with devastating accidents (for example Fukushima Daichi, 2011). Besides such accidents, nuclear energy production poses a serious issue of the disposal of nuclear waste, which needs to be securely stored for extremely long durations exceeding thousands of years. It is currently believed that repositories operating under reducing conditions will have limited U mobility because of the limited solubility of uraninite under reducing conditions. However, this assumption has recently been challenged by new experimental data showing that elevated temperatures drastically increase the stability of  $U^{4+}$  aqueous chloride complexes (Timofeev et al., 2018). Here we provide a solution for immobilization of  $U^{4+}$  and  $U^{6+}$  by abundant carbonate mineral (calcite). We experimentally investigated how much U could be entrapped by calcite growing from chloride solution at temperatures up to 350°C. Oxidation state of U was set by controlling oxygen fugacity via redox buffers (Ni-NiO and  $MoO_2/MoO_3$ ). Solid  $U_3O_8$  was used as a source of U in the calcite growth media. We found the uptake of  $U^{4+}$  by calcite is higher than that of  $U^{6+}$  by up to a factor of 100. Our work provides the foundation necessary for the quantitative assessment of potential risks associated with geological disposal of radioactive waste.

[1] Timofeev A., Migdisov A.A., Williams-Jones A.E., Roback R., Nelson A.T., and Xu H. (2018) *Nat. Commun.* 9, 1469.