

## **Evolution of uranium and thorium minerals**

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### **ABSTRACT**

The origins and near-surface distributions of the ~250 known uranium and/or thorium minerals elucidate principles of mineral evolution. This history can be divided into four phases. The first, from ~4.5 to 3.5 Ga, involved successive concentrations of uranium and thorium from their initial uniform trace distribution into magmatic-related fluids from which the first U<sup>4+</sup> and Th<sup>4+</sup> minerals, uraninite (ideally UO<sub>2</sub>), thorianite (ThO<sub>2</sub>), and coffinite (USiO<sub>4</sub>), precipitated in the crust. The second period, from ~3.5 to 2.2 Ga, saw the formation of large low-grade concentrations of detrital uraninite (containing several wt% Th) in the Witwatersrand-type quartz-pebble conglomerates deposited in a highly anoxic fluvial environment. Abiotic alteration of uraninite and coffinite, including radiolysis and auto-oxidation caused by radioactive decay and the formation of helium from alpha particles, may have resulted in the formation of a limited suite of uranyl oxide-hydroxides.

Earth's third phase of uranium mineral evolution, during which most known U minerals first precipitated from reactions of soluble uranyl (U<sup>6+</sup>O<sub>2</sub>)<sup>2+</sup> complexes, followed the Great Oxidation Event (GOE) at ~2.2 Ga and thus was mediated indirectly by biologic activity. Most uraninite deposited during this phase was low in Th and precipitated from saline and oxidizing hydrothermal solutions (100 to 300 °C) transporting (UO<sub>2</sub>)<sup>2+</sup>-chloride complexes. Examples include the unconformity- and vein-type U deposits (Australia and Canada) and the unique Oklo natural nuclear reactors in Gabon. The onset of hydrothermal transport of (UO<sub>2</sub>)<sup>2+</sup> complexes in the upper crust may reflect the availability of CaSO<sub>4</sub>-bearing evaporites after the GOE. During this phase, most uranyl minerals would have been able to form in the O<sub>2</sub>-bearing near-surface environment for the first time through weathering processes. The fourth phase of uranium mineralization began ~400 million years ago, as the rise of land plants led to non-marine organic-rich sediments that promoted new sandstone-type ore deposits.

The modes of accumulation and even the compositions of uraninite, as well as the multiple oxidation states of U (4+, 5+, and 6+), are a sensitive indicator of global redox conditions. In contrast, the behavior of thorium, which has only a single oxidation state (4+) that has a very low solubility in the absence of aqueous F-complexes, cannot reflect changing redox conditions. Geochemical concentration of Th relative to U at high temperatures is therefore limited to special magmatic-related environments, where U<sup>4+</sup> is preferentially removed by chloride or carbonate complexes, and at low temperatures by mineral surface reactions.

The near-surface mineralogy of uranium and thorium provide a measure of a planet's geotectonic and geobiological history. In the absence of extensive magmatic-related fluid reworking of the crust and upper mantle, uranium and thorium will not become sufficiently concentrated to form their own minerals or ore deposits. Furthermore, in the absence of surface oxidation, all but a handful of the known uranium minerals are unlikely to have formed.

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