Evolution of Uranium Minerals at Olympic Dam, South Australia

Edeltraud Macmillan,[1,2*] Nigel J. Cook,[3] Cristiana L. Ciobanu,[3] Kathy Ehrig,[2] Vadim S. Kamenetsky,[4] Jay Thompson,[4] and Allan Pring[5]

[1]School of Physical Sciences, The University of Adelaide, Adelaide, SA 5000, Australia
[2]BHP Billiton Olympic Dam, Adelaide, SA 5000. Australia
[3]School of Chemical Engineering, The University of Adelaide, Adelaide, SA 5000, Australia
[4]School of Physical Sciences, University of Tasmania, Hobart, TAS 7001, Australia
[5]School of Chemical and Physical Sciences, Flinders University, Bedford Park, SA 5042, Australia

*E-mail, edeltraud.macmillan@bhpbilliton.com

The Olympic Dam (OD) iron-oxide copper-gold (IOCG)-U-Ag deposit, South Australia, is the world's largest economic uranium resource, but the genesis of U mineralization in the deposit remains poorly constrained. The three main U minerals at OD are uraninite, coffinite, and brannerite. Prior studies of U minerals focused on limited sampling of higher-grade, massive, or vein uraninite specimens. The current microbeam analytical study addresses a broader range of samples at the micron- to nanoscale.

Classification of uraninite here is based on detailed mineralogical and microanalytical investigation. The first category ("primary") occurs as single grains (10–50 µm), with the simplest cubic euhedral morphology, that are oscillatory-zoned and have the highest Pb content. Electron diffraction (ED) and TEM-EDS carried out on a FIB-prepared TEM-foil cut through one of the smallest grains show uraninite is defect free. The high Pb, REE + Y (REY) contents measured are locked within the oscillatory zonation pattern, inferring these elements are lattice-bound. A second category ("zoned") is defined by coarser, subeuhedral grains, with internal zonation patterns defined by variation in Pb and REY, showing morphological changes (e.g., square domains within a prismatic outline) from core to margin. EBSD mapping confirms zonation in a single grain with no differences in orientation from core to margin. This infers that the zonation is intrinsic to either a single growth process, or involves grain-scale element redistribution via mechanisms such as chemical-gradient diffusion or coupled-dissolution re-precipitation reaction (CDRR). A third category ("cob-web") includes a variety of grains that are still coarser (up to several hundred µm), have variable hexagonal to octagonal morphologies, varying degrees of rounding, and feature rhythmic intergrowths with sulfides+fluorite from core to margin. Greater complexity includes different orientation domains within single grains (EBSD and ED), as well as heterogeneity in terms of fields of inclusions (sulfides, fluorite, REE minerals) with sizes down to nanoscale. Compositions for this category vary with respect to Pb content (<than the primary) and other minor elements (Ca, ΣREY). ED and TEM-imaging on FIB-prepared foils on grains from this category confirm they are uraninite, and place constraints on REY measurements, i.e., fields of nanoscale REY mineral inclusions account, partially, for the REY-contents (EPMA data); EBSD mapping and ED show epitaxial relationships between uraninite and Cu-(Fe)-sulfides, backing-up CDRR-replacement. The fourth ("massive") category comprises uraninite occurring as µm-sized grains to massive varieties forming coarser aggregates/vein-filling. This category has the lowest Pb and variable REY contents albeit lower than previous categories.

Coffinite and brannerite, displaying intergrowths with uraninite, sulfides, and gangue, increase in abundance in samples containing "massive" uraninite. Based on the above, there are at least two uraninite generations: early (primary through cob-web) and late (massive). EPMA Pb/U data are used as a proxy for chemical age and confirm that uraninite is multistage. Coffinite and brannerite are tied to the late generation. However, the complexity of mineral growth, replacement, and recrystallization, and formation of discrete REYminerals, observed throughout the cob-web uraninite, indicates multiple cycles of U-remobilization and reprecipitation, which contributed to the observed distributions of U-minerals at OD.