

Uranium Mobilization and Enrichment in Sedimentary Basin Environments of the Pale-Mesoproterozoic: Insights from Geochemical Modelling

Evgeniy Bastrakov,* Roger Skirrow, and David Huston

Geoscience Australia, GPO Box 378, ACT 2601, Australia

*E-mail, Evgeniy.Bastrakov@ga.gov.au

Up to 90% of Australia's uranium resources occur in deposits of Pale-Mesoproterozoic (~1.9-1.5 Ga) age, including hematite granitic breccias at Olympic Dam in South Australia and unconformity-related deposits in the Northern Territory. Published fluid inclusion data for unconformity-related uranium deposits suggest that uranium was transported by low- to moderate temperature (<250°C), Na-Ca-Mg brines of seawater evaporation origin.

Secular changes in geochemical behavior of uranium through Earth history are well known. The most prominent changes are attributed to stepped oxygenation of the Earth's atmosphere. This process resulted in oxidation of U(IV) to U(VI) forming highly soluble aqueous uranyl complexes. The oxygenation is thought to have occurred as two stepwise increases in atmospheric oxygen at the beginning and end of the Proterozoic, at ~2.3 and ~0.6 Ga. High aqueous mobility of uranium after the second oxygenation event is globally recorded by elevated concentrations of uranium in organic-rich shales.

Large-scale processes of crustal enrichment of uranium in the Proterozoic rocks pre-dating the second oxygenation events can be explained by a number of endogenic factors, including high paleogeothermal gradients and large volumes of uranium-enriched granitic rocks emplaced at shallow crustal levels. Other decisive factors leading to the formation of the giant uranium deposits may be of exogenic origin. One would be a unique combination of moderately elevated levels of atmospheric oxygen and high levels of atmospheric CO₂, with the latter exceeding present day levels at least by ~1.5 orders of magnitude. Under these conditions, for a wide range of surficial waters and groundwaters, uranium aqueous speciation would be dominated by carbonate uranyl complexes (e.g., UO₂CO₃), with uranyl concentrations proportional to CO₂ pressures. Another exogenic factor is Paleoclimatic conditions favourable to the formation of evaporative basins suggested as sources of uraniferous fluids.

In the present study, we examine these two exogenic factors quantitatively, modelling solubility of uranium in natural waters and progressively evaporated seawater at boundary conditions characteristic of Pale-Mesoproterozoic atmosphere ($\log f\text{CO}_2 > \square 2$, $\log f\text{O}_2 \sim \square 1.4$).

The modelling indicates that Pale-Mesoproterozoic environment could be especially favorable for mobilization of uranium in weathering profiles due to elevated content of atmospheric CO₂. Evaporation of seawater is indeed a chemically feasible process that might have determined the initial (Na-Mg) composition of brines associated with uranium mineral systems. The range of the Na-Ca-Mg brine compositions reported in the literature can be explained by "sampling" of spatially separate parts of the same "brine factory" characterized by different degrees of seawater evaporation and the extent of the subsequent brine interaction with Ca-rich basin and basement lithologies via Na-Ca and Mg-Ca exchange reactions.