

## **Mechanisms for UO<sub>2</sub> alteration to Coffinite: Reconciling the laboratory and Natural Analogue evidences**

JORDI BRUNO<sup>1</sup>, LARA DURO<sup>2</sup> AND LENA Z. EVINS<sup>3</sup>

<sup>1</sup>Amphos 21 Consulting SL, jordi.bruno@amphos21.com

<sup>2</sup>Amphos 21 Consulting SL, lara.duro@amphos21.com

<sup>3</sup>SKB AB, lena.z.evins@skb.se

There is a general consensus now that reducing conditions will prevail in the vicinity of the spent fuel matrix in the KBS 3 repository conditions. Under these conditions, one process that could destabilise the UO<sub>2</sub> spent fuel matrix is its potential alteration to coffinite (USiO<sub>4</sub>).

The potential effect of silicate on the stability of the UO<sub>2</sub> matrix is complex issue. The three main conditions that were identified as potentially causing the formation of coffinite relevant for a KBS-3 repository are i) the likely presence of high concentrations of Si (or silica) from the surrounding clay (bentonite) backfill material or surrounding host rocks; ii) the potential development of an alkaline plume leading to high pH values in the system due to the possible co-location of high intermediate and low activity wastes and iii) the development of highly reducing conditions.

Recently, there has been a substantial effort in the synthesis, characterisation and thermodynamic properties of coffinite reported in the literature. The key conclusion would be that coffinite is metastable with respect to UO<sub>2</sub> (s) + SiO<sub>2</sub>(s) by some 20-26 kJ/mole. This would indicate that coffinite cannot be formed from the mixture of uraninite and silica but still does not explain why this phase is abundant and concurrent to uraninite in so many deposits.

In this presentation we will discuss the potential mechanisms that could give rise to the formation of coffinite and the mineralogical and geochemical observations that could support these potential mechanisms.

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