

Abstract

U migration was studied in order to better understand to processes of safety relevant elements, particularly U. The rock sequence on the site can be analogous to a potential rock overburden of deep geology repositories.

A multidisciplinary approach was undertaken in order to identify and characterise U mobilisation/immobilisation processes within sedimentary clayey rocks with organic matter enriched interlayers. Both conventional methods and modern sophisticated spectroscopic methods were combined. Sequential extraction, wet chemical method and even spectroscopic methods proved that U prevailed in the U(IV) form in low-oxidised samples. It moved towards more easily releasable fractions with sample ageing (oxidation).

The combination of SE, μ -XRF and μ -XAFS results proved U to be unexpectedly associated with As and P, leading to the presumption that U(VI) from groundwater was reduced to U(IV) on As pyrite.

The evaluation of the hydrogeochemical conditions and isotope analyses then brought the results into broader context: Sedimentary organic matter within the sedimentary layers was microbially oxidised, releasing dissolved organic matter and providing H^+ in order to dissolve sedimentary inorganic carbonates. SO_4^{2-} could be reduced under reducing groundwater conditions, thus causing FeS_2 formation. As got also reduced and precipitates on FeS_2 surfaces. U was reduced on As covered pyrite surfaces to U(IV) and reacted with phosphates (PO_4^{3-}), produced by microbial organic matter oxidation. U phosphates (ningyoite) were thus formed. The $^{234}U/^{238}U$ activity ratio determination proved the fact that U(IV) phases had been stable for more than 1 Ma. This fact strongly supports potential barrier function of similar rock systems (clayey sedimentary rock with organic matter content). On the other hand, it is necessary to stress important role of CO_2 in increased mobility of U(VI) in reducing groundwater conditions.