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Study of Uranium Behavior in Lignite Sediments from Ruprechtov Natural Analogue Site

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Study of uranium oxidation states in natural objects is important to estimate radioecological impact of U and as a natural redox monitor for assessing geological sites considered for long-term highly radioactive waste storage. In this work we studied U oxidation state distribution in the sediment samples by wet chemistry and synchrotron radiation method XAS. Novel wet chemical method for separating U(IV) and U(VI) from solid samples was developed and utilized. Two lignite samples having 172 and 539 ppm of U collected from the sediment layer of the Ruprechtov natural analogue site in Czech Republic were studied by described methods. Redox processes during U extraction has been monitored using 236-U(IV) and 236-U(VI) tracers. Ascorbic acid was used as selective reducing agent for Fe(III) to minimize its oxidizing effect on U(IV). XAS showed for both samples, that bulk of U in the samples is U(IV). Dissolution experiments showed that U(IV) amount varied widely resulting from the strong oxidation caused by Fe3+. Use of ascorbic acid reduced considerably the effect of Fe3+ on U redox balance: about 75% of U after samples'extraction was found in U4+. 234-U/238-U activity ratios (AR) in U(IV) and U(VI) fractions were used as an additional monitor for U redox processes taking place during materials extraction. AR in U(IV) fraction, which is less than 0.5, did not change due to extraction induced oxidation, whereas in U(VI) fraction AR was observed to decrease dramatically, depending on the amount of U(IV) oxidised during the extraction. AR for U(VI) was found below unity after 10 min extraction with pure acid solution but with addition of ascorbic acid AR increased up to 2. Results showed good agreement with lignite samples from other regions of Ruprechtov site.

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