

 ROBL-CRG	Experiment title: Local structure of U(IV) and U(VI) in natural and synthetic minerals	Experiment number: 20-01-642
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Report:

Uranium is very immobile in the environment in its tetravalent oxidation state. However, processes like mining and possibly also oxidation of depleted uranium ammunition cause the release of hexavalent uranium (uranyl), which is forming a vast range of aqueous, soluble complexes, and hence regarded as highly mobile. In addition, there is also a large range of relatively poorly soluble minerals containing uranyl, which may also reduce the mobility of uranium in the environment. A recent investigation (Catalano and Brown, 2004; Catalano et al., 2004) demonstrated that these uranyl minerals are difficult to distinguish by EXAFS spectroscopy, since they commonly form layered structures (silicates, vanadates, carbonates, hydroxides, phosphates, etc.). However, this study was based on a relatively limited number of mineral samples, was performed at room temperature and preferential orientation (texture effects) cannot be completely excluded. Hence, we followed up this study using a large range of mineral samples, which Prof. Vochten from Antwerp University and Dr. Massanek from the Mineral Collection of the technical University Freiberg have provided. These samples comprise both synthetic and natural samples. We have used this collection to build a spectral data base for EXAFS speciation of U in environmental samples. Up to now, we have measured 40 samples to measure the samples at both room temperature and 20 K, and to avoid texture effects by using the magic angle geometry. Examples are shown in Figs. 1 and 2. We have also collected XRD, FTIR and TRLFS data of these samples to verify their identity and provide additional chemical and structural information.

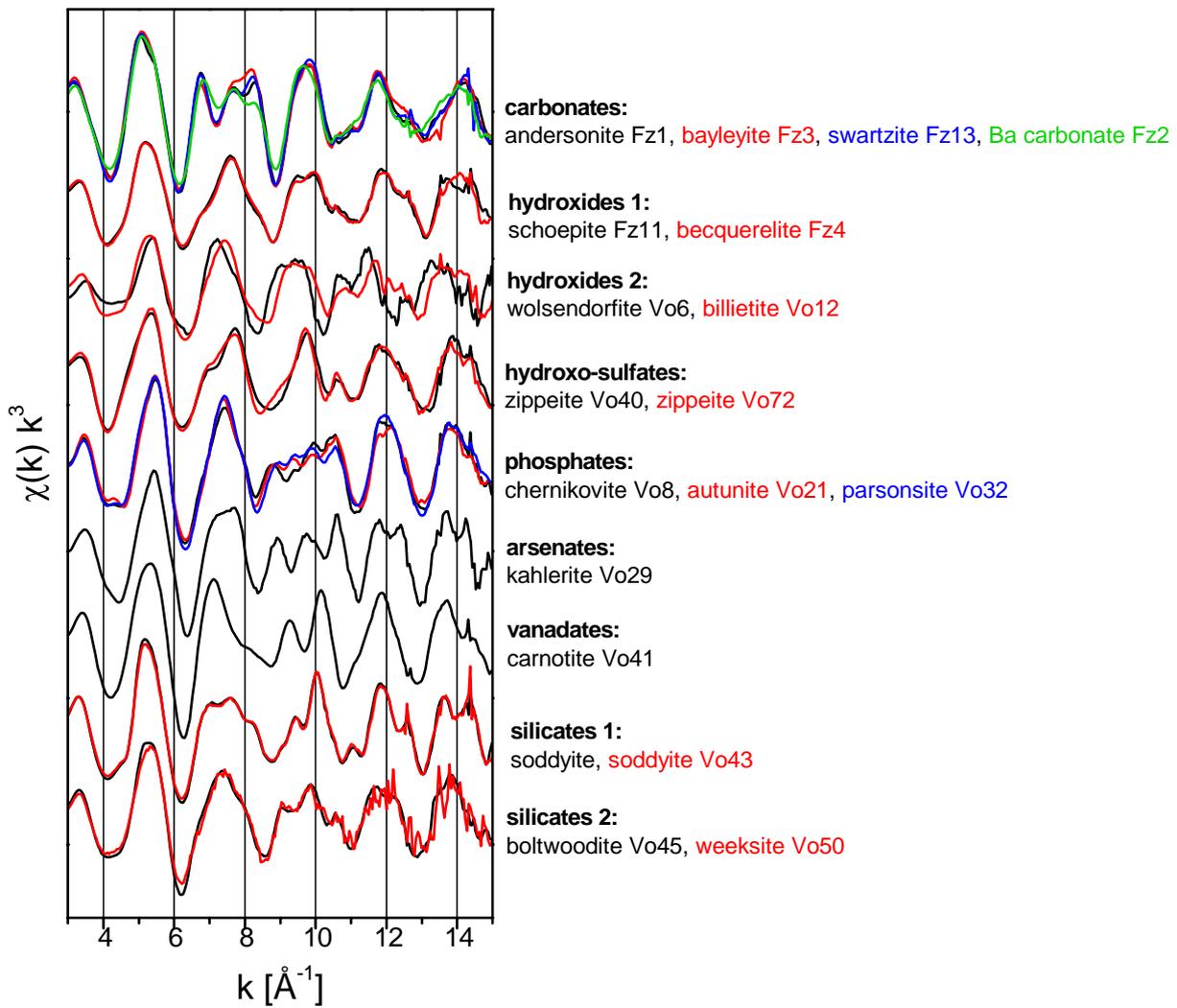


Fig. 1. Comparison of U-L₃ EXAFS spectra of different uranyl minerals. The spectra have been measured at room temperature and at the magic angle to avoid orientation effects.

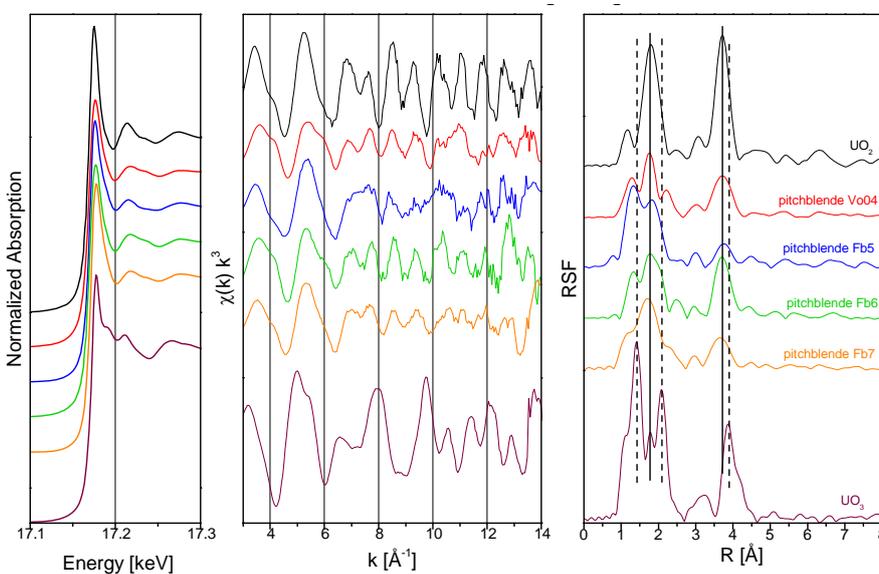


Fig. 2. Comparison of U-L₃ EXAFS spectra of natural pitchblende samples in comparison to a freshly precipitated UO₂ and to UO₃. The spectra have been measured at room temperature and at the magic angle to avoid orientation effects.