ROBL-CRG	**Experiment title:** **EXAFS investigations of uranium species formed by monocellular and polycellular algae**	**Experiment number**: **20-01-656**
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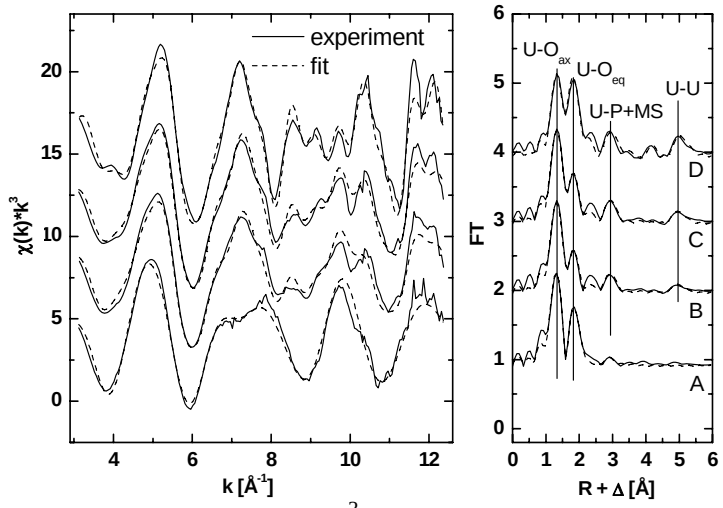
Report

Experimental: The aim of this study is to determine the structural parameters of the formed uranyl complex species in monocellular Chlorella vulgaris cells at pH 3, 5 and 6 and low temperature. The stored algal biomass was re-suspended in 0.9% $NaClO_4$ solution and contacted 72 h with $UO_2(ClO_4)_2$. In each reaction solution the concentration of the algal biomass was 0.75 g dry mass /L. The uranium concentration was $1 \cdot 10^{-4}$ M. The pH values of the solutions were adjusted with $HClO_4$ or NaOH. After separation of the washed algae biomass by centrifugation the fresh samples were put into polyethylene sample holder surrounded with Kapton tape. The U L_{III}-edge spectra were measured in fluorescence mode at 30K using a closed-cycle He-cryostat. The EXAFS spectra were analyzed using the suite of program EXAFSPAK /1/. The theoretical scattering phases and amplitudes were calculated by using x-ray structural data of meta-autunite with the scattering code FEFF8 /2/.

Results and discussion: Figure 1 shows the raw U L_{III}-edge k^3-weighted EXAFS spectra and their corresponding Fourier transforms (FT) of uranium containing algal cells (samples A-C) at 30 K. The corresponding structural parameters are summarized in Table 1. At pH 3 the U-O_{ax} and U-O_{eq} radial distances are similar to those obtained for uranyl hydrate. The spectrum is well reproduced by taking only backscattering on O atoms into account (Fig. 1). At pH 5 and 6 short U-O_{eq} bond distances of 2.28 Å and 2.29 Å and a U-P interaction were found (Tab. 1). The radial U-P distance of 3.57 Å indicates a monodentate binding of uranium on phosphate groups. In addition both samples show a U-U interaction (Fig. 1, Tab. 1). The structural parameters of the samples at pH 5 and 6 are similar to those of meta-autunite (Vo21, Fig. 1, Tab. 1).



meta-autunite, Vo21

U6-47 algae sample pH6

U6-46 algae sample pH5

U6-45 algae sample pH3

Fig. 1. Raw U L_{III}-edge k^3-weighted EXAFS spectra (left) and corresponding Fourier transform (right) of the algae samples and meta-autunite at 30 K.

Tab. 1. EXAFS structural parameters of the uranyl complex species in different algae sample and of a reference sample at 30K

Sample	$U\text{-}O_{ax}$		$U\text{-}O_{eq}$			U-P + MS			U-U		
	R [Å]	$\sigma^2 \cdot 10^3$ [Å^2]	N	R [Å]	$\sigma^2 \cdot 10^3$ [Å^2]	N	R [Å]	$\sigma^2 \cdot 10^3$ [Å^2]	N	R [Å]	$\sigma^2 \cdot 10^3$ [Å^2]
Vo21	1.77	3.9	3.8	2.29	2.0	3.8	3.59	3.0	4	5.27	3.0
U6-47	1.77	2.2	4*	2.28	5.6	4*	3.57	3.8	4*	5.22	7.0
U6-46	1.77	2.3	4*	2.29	7.9	4*	3.57	5.7	4*	5.22	9.0
U6-45	1.77	1.8	4.7	2.39	7.7	-	-	-	-	-	-

N - coordination number with an uncertainty of 25%, N O_{ax} was held constant at N=2, R - radial distance with an uncertainty of ±0.02Å, σ^2 - Debye-Waller factor, * fixed during the shell fitting

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References

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