Experimental report

25/07/2023

Proposal: 6-06-520 **Council:** 10/2022

Title: Local order in ThO2: what are the differences with UO2?

Research area: Materials

This proposal is a continuation of 6-06-471

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Samples: CeO2
ThO2

Instrument	Requested days	Allocated days	From	To
D4	3	6	20/06/2023	23/06/2023

Abstract:

We aim at measuring ThO and ThTh nearest-neighbor distances in ThO2 as a function of temperature using PDF-analysis in order to compare with our previous results on UO and UU distances in UO2. The nearest-neighbor UO distance in UO2, as defined by the peak in the UO distance distribution seen in the PDF(r), decreases with increasing temperature whereas the UU distance follows the unit cell thermal expansion. A comparison with ThO2 will test whether this unusual behavior is a consequence of electron-lattice interactions from the two 5f electrons in UO2, or due soley to MO2 phonons in the fluorite structure, since the Th atom has no 5f electrons in ThO2. Other than this result, neutron diffraction data on ThO2 PDF at high temperature are lacking in literature, and are needed for proper modeling of the structure and dynamics of actinide dioxides.

Experimental report for 6-06-520 experiment at D4 from 20 to 23rd of June 2023

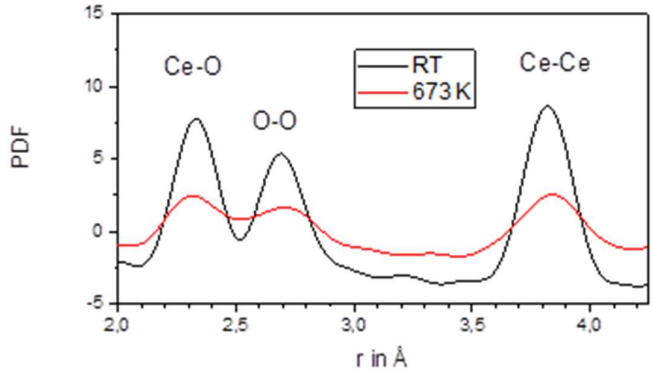
Before the experiment, we contacted SPR service in order to define how to handle our ThO_2 samples, and it was decided that a person from SPR would supervise our ThO_2 manipulation.

After the experiment started, we learnt a supplementary safety procedure was required to handle ThO_2 samples, preventing the planned project.

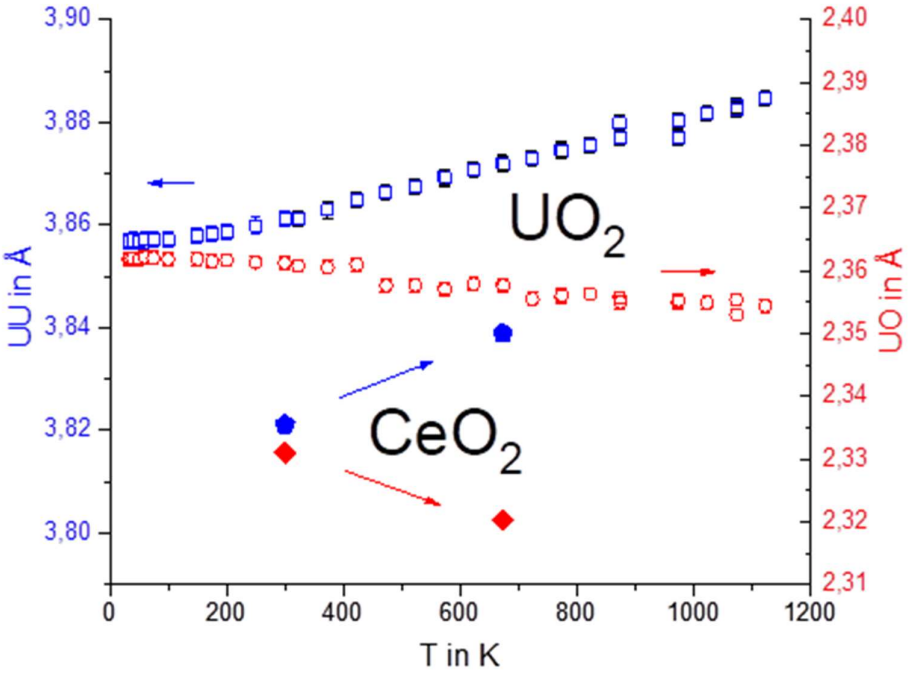
ILL staff was cooperative, but there was no way to solve the problem in time.

Predefined transparent procedures issued by the safety office, covering manipulation of active samples, should be agreed with prior to user arrival to avoid damaging loss of allocated beam time.

When it became clear that no experiment would be performed on ThO_2, we asked user office the authorization to perform measurements on a CeO_2 sample, which we brought with in case. We managed to measure CeO_2 PDF at two different temperatures: room temperature and around 400°C. The corresponding PDF are shown on the figure below.



We extracted the positions of the CeO and Ce-Ce peaks from this figure, and compared their evolution as a function of temperature to the one we previously measured on UO_2. The results seem in qualitative agreement, but additional experimental data are required for these preliminary results to be published.


3,90
3,88
3,86
3,84
3,82
3,80
UU in Å
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2,39
2,38
2,37
2,36
2,35
2,34
2,33
2,32
2,31
UO in Å
0
200
400
600
800
1000
1200
T in K
UO2
CeO2