ESRF	**Experiment title:** The influence of Pu local environment on the non ideality of $(U,Pu)O_2$ solid solution	**Experiment number**: CH-619
Beamline: BM 20	**Date of experiment**: from: 12/06 to 15/06 and from 07/07 to 11/07	**Date of report**: 30/08/1999
Shifts: 21	**Local contact(s)**: Tobias Reich	*Received at ESRF:*

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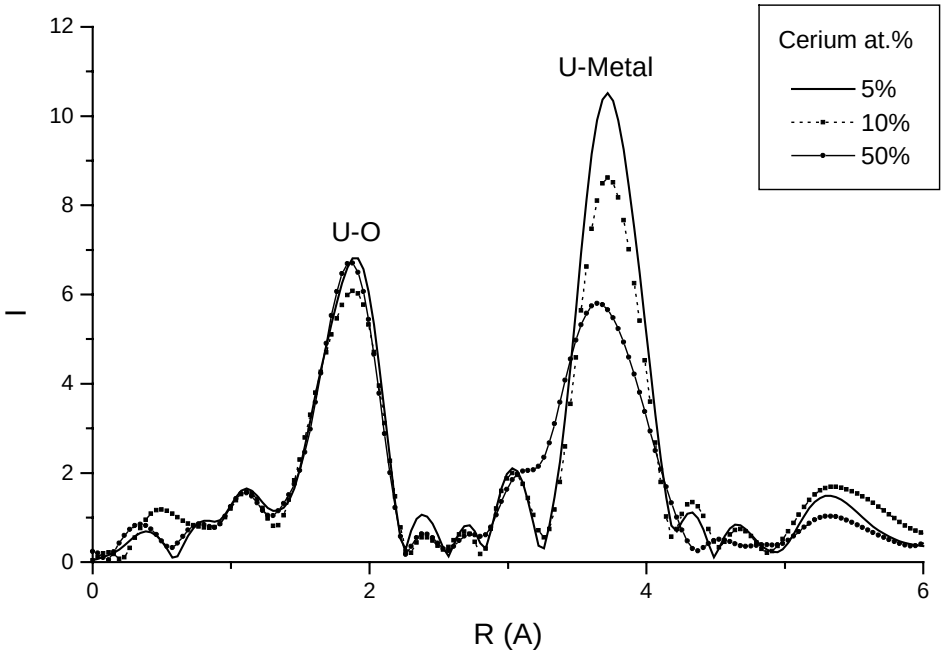
Report:

Some experimental results on unit cell parameter, electrical conductivity, specific heat and thermal conductivity show that $(U,Pu)O_2$ is not an ideal solid solution. M. Beauvy [1] notes that these anomalies occur when Pu concentration is around 3.1 and 12.5 at%. They respectively correspond to one Pu atom for 8 unit cells and one Pu atom for 2 unit cells. He supposed that these thresholds could correspond to different Pu local environments in UO_2 matrix.

Our aim was to see if Pu local environment is connected with Pu concentration in $(U,Pu)O_{2+x}$ matrix. In other words, we wanted to check if any modification occurs when Pu content varies.
However, because of the limitation in plutonium isotopes accepted on the beam line, we studied $(U,Ce)O_{2+x}$ instead of $(U,Pu)O_{2+x}$. $(U,Ce)O_{2+x}$ system is expected to show many similarities to $(U,Pu)O_{2+x}$, cerium is known to be a non active analogue of plutonium [2].

We thus used the 21 shifts allowed by the Review Committee to collect the EXAFS spectra of 5 $(U,Ce)O_{2+x}$ pellets (with Ce amount of 0, 5, 10, 25 and 50 at.%) in fluorescence detection mode.

In order to both probe U and Ce local environment, we worked at U L_{III} and Ce L_I edges (17.17 and 6.55 keV respectively). The following figure shows the Fourier transform moduli obtained at U L_{III} edge for the different cerium concentration.



Due to the high concentration of U and Ce, the amplitude of the XAFS spectra was corrected for self-absorption effects using the procedure of Tröger *et al.* [3]. To analyse our data we use the phase and amplitude calculated by the FEFF 7.02 program [4].

As a first result, the U-O coordination shell shows very low evolution with the amount of cerium. On the opposite, the intensity of the second peak, connected to metal-metal distances, decreases as increasing the cerium concentration.

The determination of metal-metal distance and coordination number is difficult because of the numerous multiple-scattering paths included in this contribution. Furthermore due to the nearness of the experiment we only deduced the parameters of the first coordination shell (U-O) : uranium is coordinated with 8 oxygen atoms at 0.236 nm (the same as in UO_2). This first coordination shell remains the same whatever cerium concentration is.

Due to the complexity and low ratio signal over noise of the Ce L_I edge spectra, we could not have currently quantitative results on cerium environment, but work is still in progress.

References :

[1] M. Beauvy J. Nucl. Mater. 188 (1992) 232-238.
[2] D.J. Lones, J. Roziere, G.C. Allen and P.A. Tempest, Journal de Physique, Colloque C8, Tome 47, (1986) C8-745-C8-748.
[3] L. Tröger, D. Arvanitis and K. Baberschke Phys.Rev. B 46 (1992) 3283-3289.
[4] J. J. Rehr, S. I. Zabinsky, R. C. Albers, Phys.Rev.Lett. 69 (1992) 3397.