ESRF	**Experiment title:** In situ XAS investigations on SIMFUEL samples to study volatile fission products behavior in severe accident conditions	**Experiment number**: CH 5020
Beamline: BM30B	**Date of experiment**: from: 19/07/2017 to: 26/07/2017	**Date of report**: 25/07/2017
Shifts: 18	**Local contact(s)**: Mauro Rovezzi	*Received at ESRF:*

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Report:

The objective of the project is to determine Fission Products (FP) behavior in conditions representative of a Severe Accident (SA) of a nuclear pressurized water reactor. This will help improving the actual understanding of their release mechanisms and confirming or not the assumptions on which some codes for SA scenario predictions are based. Within this frame, several XAS (X-ray Absorption Spectroscopy) experiments have been performed on SIMFUEL samples composed of a UO_2 matrix doped with 12 oxides as FP surrogates (**Table 1**) after thermal treatments [1–4]. However, post-test analyses do not allow the observation of potential intermediate compounds formed at high temperature which is of particular importance to understand volatile FP speciation.

The goal of this experiment was to demonstrate the feasibility of an *in situ* XAS experiment at high temperature under controlled atmosphere on SIMFUEL samples at the ESRF (**Figure 1**). The sample (with an activity of 14 Bq) was maintained on a W-Re5 wire with two crimped nuts as shown in **Figure 2**. It was heated by Joule effect up to 700°C [5–7]. The atmosphere was set inside the furnace by pumping or adding oxygen to a gas mixture composed of He or He+4%H_2. The temperature of the sample was monitored thanks to a thermocouple and a pyrometer. This experiment was set up and performed in collaboration with FAME-UHD beamline and the ESRF SAFETY group.

Ba speciation was studied at two different oxygen potential conditions at room temperature in the sample as fabricated (T_0), 400°C and 700°C. Thermodynamic calculations indicate that Ba would be present in the samples as $BaZrO_3$ in reducing conditions whereas $BaMoO_4$ predominates above -300 $kJ.mol_{O2}^{-1}$[1,8]. To avoid interferences with the signal of other elements present in the sample, High Energy Resolution Fluorescence Detection (HERFD) XANES have been performed at Ba L3 edge (5.247 keV). The Crystal Analyzer Spectrometer (CAS) consisted in two Ge(400) crystals with a Bragg angle of 79° and the fluorescence signal was recorded with a VORTEX Si detector. In order to limit absorption of photons by the air, the glovebox used to ensure the second containment barrier of the sample was filled in with He and a He bag was placed between the glovebox, the crystals and the detector.

The XANES spectra obtained for the sample in its initial state at room temperature and in the two probed atmospheres are plotted on **Figure 3**. Edge energy E_0 of each spectrum are similar confirming that Ba is in oxidation state +II as expected for an alkaline earth element. At first glance, no evolution of Ba speciation is visible between both samples. The shape of the T_0 spectrum is very close to $BaZrO_3$ spectrum as already observed in [4]. The spectra recorded at 700°C are broadened and flattened if compared to the T_0 (due to thermal agitation effects) but still look similar to $BaZrO_3$ spectrum. However, further analysis of the obtained spectra is still under way.

In conclusion, the feasibility of an in-temperature XAS experiment on SIMFUEL samples on FAME-UHD beamline has been demonstrated. Some optimizations of the sample holder still have to be done particularly to be able to reach higher temperatures. A second experiment will be needed in the first semester of 2018 at Cs L3 edge (5.012 keV).

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[1] E. Geiger, Paris-Saclay, CEA Cadarache, 2016.
[2] E. Geiger *et al.*, *J. Nucl. Mater.*, vol. 471, pp. 25–33, Apr. 2016.
[3] E. Geiger *et al.*, *J. Phys. Conf. Ser.*, vol. 712, no. 1, p. 012098, 2016.
[4] C. Le Gall *et al.*, FAME beamline, ESRF, Experimental report 30-02-113, 25/07 2016.
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[6] D. R. Neuville *et al.*, *Rev. Mineral. Geochem.*, vol. 78, no. 1, pp. 779–800, 2014.
[7] C. Le Gall *et al.*, *Proceedings of the 8th ERMSAR*, Warsaw, Poland, 2017.
[8] H. Kleykamp, *J. Nucl. Mater.*, vol. 131, no. 2–3, pp. 221–246, 1985.

Table 1: SIMFUEL composition (concentration in wt%)

Samples Composition (wt%) representative of a 76 GWj/tU irradiated fuel										
Ba	Ce	La	Mo	Nd	Pd	Rh	Ru	Sr	Y	Zr
0.30	0.52	0.30	0.64	1.12	0.40	0.06	0.69	0.13	0.07	0.61

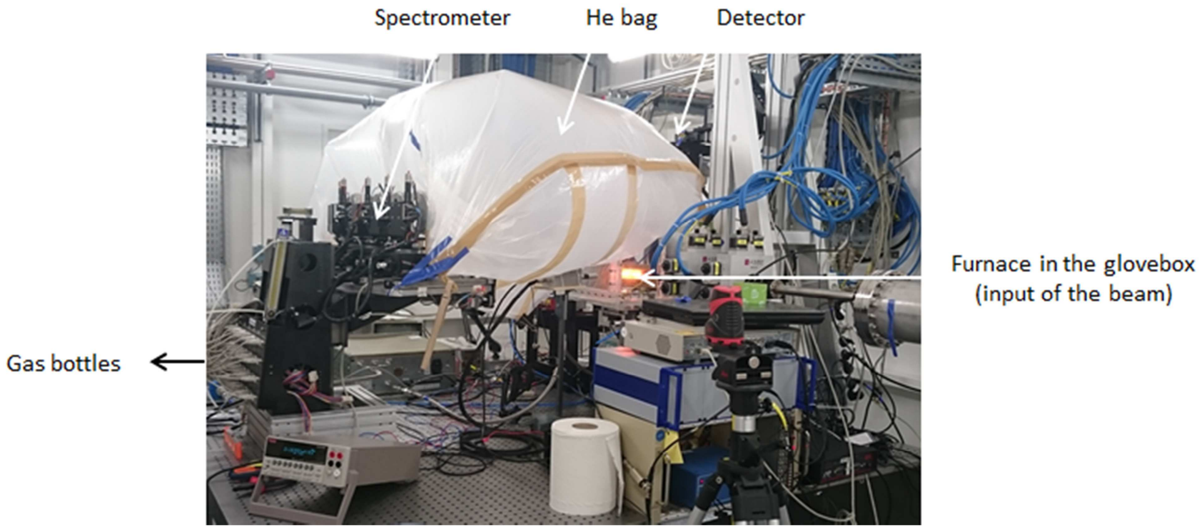

Figure 1: Picture of the in-temperature XAS experimental set-up

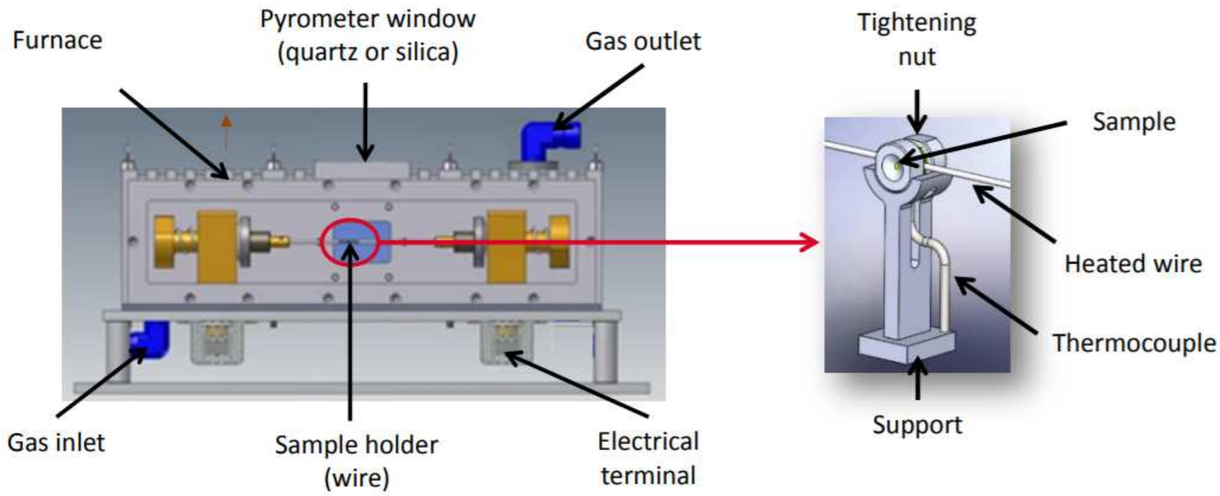

Figure 2: In-temperature XAS apparatus (left) and the sample holder (right) [5,7]

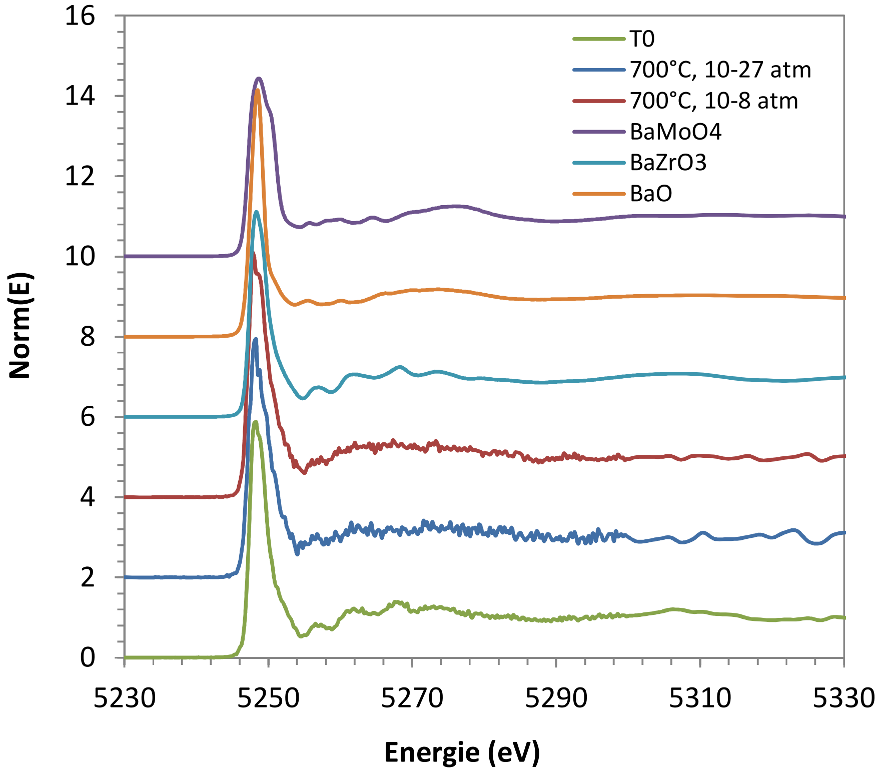

Figure 3: HERFD XANES spectra collected at Ba L3 edge on sample P9C2 at room temperature and as fabricated (T_0), at 400°C and 700°C under different atmospheres, along with several reference samples