Nuclearized Raman microscope coupled with a hot-stage: new tool to study (U, Pu)O_{2-x} fuel microstructure

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In the frame of the development of uranium-plutonium mixed oxide fuels for Sodium-cooled Fast Reactors (SFRs), characterizing nuclear materials by various techniques is paramount. These fast neutron reactors imply the use of a (U,Pu)O_{2-x} ceramic fuel with a Pu/(U+Pu) content between 19 and 30 mol.%. Furthermore, the physico-chemical and microstructural properties of such fuels, such as chemical homogeneity, oxygen stoichiometry (O/(U+Pu) ratio) and crystallographic structure, have to meet precise criteria for being introduced in the reactor core. As shown in recent studies by Talip *et al.* (2018) and Elorrietta *et al.* (2017), Raman microscopy is a promising tool for characterizing the physico-chemical properties such as, among many others, the cation distribution homogeneity, the grain size, the crystal defects that are of main interest for the production of nuclear fuels.

The development of a new *in situ* Raman device dedicated to handling transplutonium-bearing materials is currently in progress in the new hot-laboratory L26 located at the ATALANTE facility (CEA Marcoule, France) in the framework of the TARRA project[4]. This new experimental set-up installed in a glovebox is complementary to the Raman set-up existing at the C19 hotcell also at the ATALANTE facility and mainly dedicated to study irradiated fuels (Jérou et al., 2010).

This new experimental set-up designed by Optique H. Peter company consists in a confocal optical microscope coupled with an iHR-320 Raman spectrometer supplied by HORIBA company. Moreover, a new nuclearized Raman micro-furnace will allow us performing *in situ* Raman measurements up to 1800°C under controlled atmosphere.

Experimental set-up

Confocal microscope. The confocal optical microscope designed by the Optique H. Peter company and shown in Figure P33a, is installed inside a nitrogen-filled glovebox (Cf figure P34). The glovebox stage has been reinforced in order to increase its rigidity to minimize vibrations induced by the laboratory/facility ventilation. The microscope is fixed on an optical table which is fixed on the glovebox floor. The microscope is equipped with two turrets which can be manually exchanged: the first one equipped with Olympus© objectives (x5 to x100 magnification) and dedicated to optical

[4] More details about the TARRA project and the L26 laboratory can be found in O. Dugne *et al.* abstract entitled « TARRA Project: transfer of MOX R & D between 2 CEA sites » presented during the same conference.

observation and Raman mapping on polished samples; the second one dedicated to *in situ* Raman experiment Mitutoyo© objectives (x5 up to x50 magnification) with long working distance in order to accommodate the micro-furnace geometry. The turret dedicated to optical observation is also equipped with a micro-indenter device in order to perform Vickers hardening tests. On each turret, the change of objective is motorized. The sample displacements (x, y, z and rotation) are performed using independent motorized stages.

Raman spectrometer

The spectrometer is an iHR-320 supplied by the HORIBA© company located outside the glovebox. The spectrometer is equipped with 2 lasers (532 nm and 660 nm) with adjustable power by the labspec software developed by HORIBA©. Each laser is connected to a HORIBA Raman superhead mounted on the microscope (Cf Figure P33a) via an optimized optical fibber (10 µm diameter). The Raman signal is transmitted to the spectrometer using a 100 µm diameter optical fibber. Thanks to the microscope motorized stages, Raman mapping/imaging at micrometre scale of polished sintered samples can be easily performed.

Raman micro-furnace

The nuclearized version of Raman micro-furnace initially developed by Montagnac *et al.* (2013) is shown in Figure P33b. The furnace is a confined 1 mm diameter heating-wire (Pt with 20% Ir alloy) flattened in the middle where the sample will be placed. The wire is connected to a power supply and the temperature is monitored/measured using an optical pyrometer from the Lumasense© company. The latter is installed on a dedicated x,y,z stage in order to compensate sample displacements due to the wire thermal expansion. This heating system has low thermal inertia and it is possible to change the temperature between room temperature up to temperatures >1700 °C in few seconds (Neuville et al., 2014). The micro-furnace is water-cooled using a close circuit coupled to a heat exchanger. Thanks to gas inlet and outlet, a continuous flushing of a controlled atmosphere around the sample during a heat treatment can be performed. Furthermore, a set-up controlling, imposing and measuring the variations in the oxygen partial pressure is also available.

As illustrated by the 3 figures, the whole experimental set-up is already installed at L26 laboratory. As describe here, the whole system is optimized to minimize as much as possible sample and equipment handling: once the sample is installed (except a turret change), the experiments/observations are performed/conducted remotely in an office located near the hot laboratory The first radioactive experiments on $(U,Pu)O_{2-x}$ samples are scheduled in November 2018.

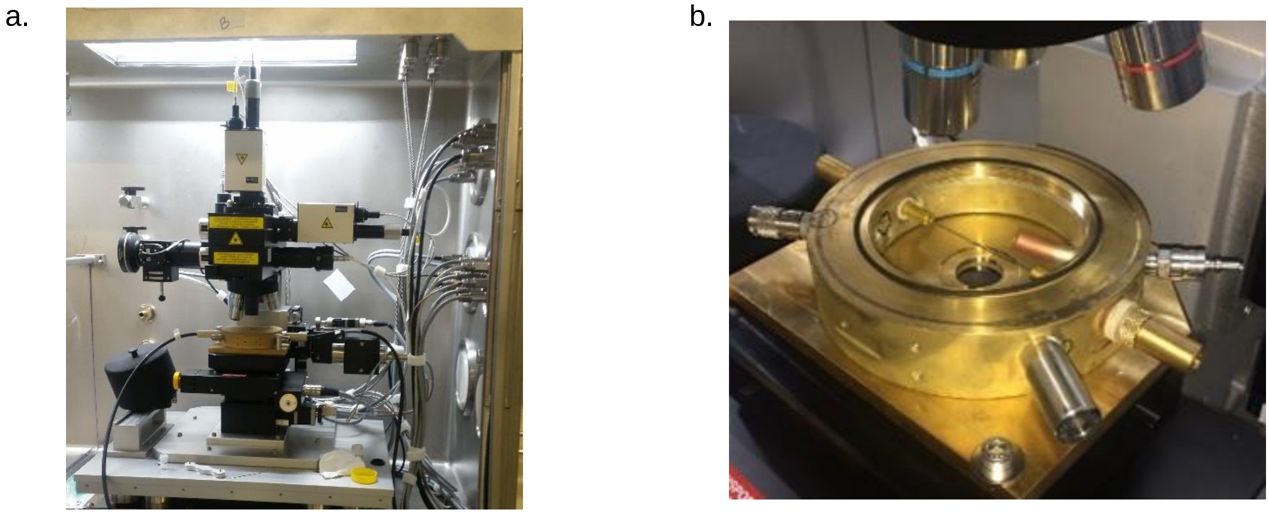

Figure P33: Detailed views of (a) the Raman microscope installed inside the glovebox; (b) the nuclearized Raman micro furnace installed on microscope motorized stage.

Figure P34**:** View of the new glovebox dedicated to Raman microscope installed in the L26 laboratory (ATALANTE Facility, CEA Marcoule, France).

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