A new device for X-ray Diffraction analyses of irradiated materials

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Abstract

A new X-Ray Diffraction (XRD) equipment is being implemented in the LECA (Cadarache) hot laboratory. The device will be dedicated to structural characterization on irradiated fuels, as PWR fuels, transmutation targets and innovative fuels.
The paper will present the specific design that was decided in order to reduce the number of components in contaminated volume and to make servicing easier:

- Inside a glove box, radioactive samples are mounted on a 3 axis (x, y,z) and rotation (ϕ) sample holder, which allows a well defined positioning of the primary X-ray beam on the area of interest.
- X-ray goniometer, including primary optic (Bragg-Brentano set-up or microdiffraction set-up) and secondary optic (graphite monochromator and point detector) is located outside the glove box but inside the shielded cell.
- Incident and diffracted beam are transmitted through the glove-box walls thanks to a beryllium foil (with a low X-ray attenuation coefficient) that prevents contamination release outside the glove box.

The analytical performances of this new equipment will be illustrated on some model samples:

- micro diffraction capabilities will be detailed on heterogeneous material
- strain and stress analyses on fresh uranium oxide pellets

Keywords:
Irradiated materials, hot cells, X-Ray Diffraction, microanalyses

1. Introduction

The old XRD system available in CEA Cadarache from the early eighty was dedicated to structural analyses on irradiated fuels samples.
Nuclear fuels specificities and structural micro-analytical needs was not covered by the existing device, its replacement has been initiated with three major goals:

- improved analytical performance
- optimized characterization of heterogeneous sample
- optimized-servicing design.

2. Experimental device

A standard XRD goniometer (Type XRD 3001) from GE-Inspection Technologies Company, has been selected for this project (Fig. 1a)

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2.1. Specifications

Specifications of the device have been defined keeping in mind analytical needs on irradiated materials: to avoid gamma radiation effects on X-ray detection, a point detector coupled to a graphite monochromator is used.
Analyzed materials may be heterogeneous, due to their processing (MOX fuel, transmutation target), or after irradiation (HBS effect at fuel pellet periphery). For that reason, local analyses are needed.

- With that purpose a specific focusing device has been designed (Kb mirrors). The X-ray beam dimension (diameter 100 µm) allows structural analyses of these heterogeneous objects (Fig. 1b).
- Thanks to a 4-axes motorized sample holder, local X-ray measurements (point, line-scan, mapping) are possible (Fig. 1a).
- A high magnification digital video camera will be used to locate analyzed zone on heterogeneous samples (Fig. 1a).

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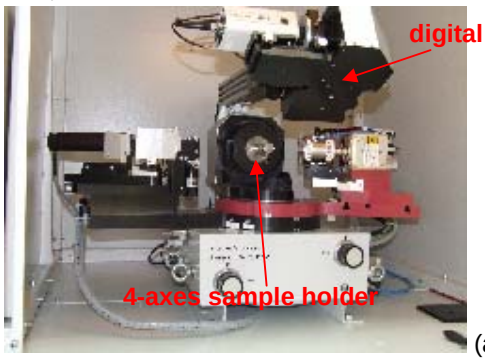


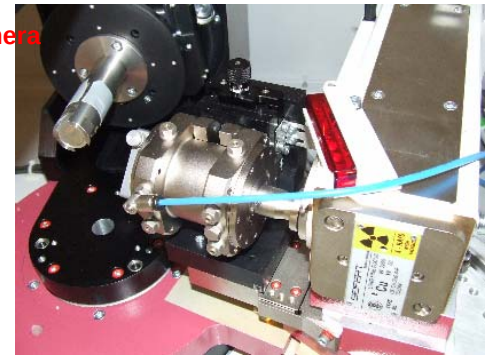
(a)

(b)

Figure. 1 : Standard X-ray goniometer (a) ; detail of the X-ray focusing device (b)

2.2. Device modification

The device modification is in progress. A specific design will be used to avoid contamination of the goniometer, still allowing an easy servicing :

- The analytical system will be implanted in a shielded cell (Fig. 2),
- The 4-axes sample holder will be located inside a glove box on an up/down translation device,
- The goniometer (X-ray tube and X-ray detector), will be outside the glove box (in the lower part of the hot cell) but still inside the hot cell,
- Thanks to a beryllium window located in the lower part of the glove-box, incident and diffracted X-ray beam will be transmitted through the glove-box walls (Fig. 3),
- To reduce gamma irradiation effects on diffraction patterns an additional shielding will be added around the X-ray detector and the lower part of the glove box (close to the beryllium window).

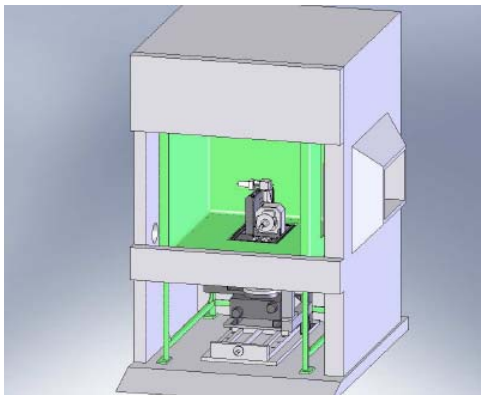


Figure. 2 : View of the system after modification, inside the hot-cell

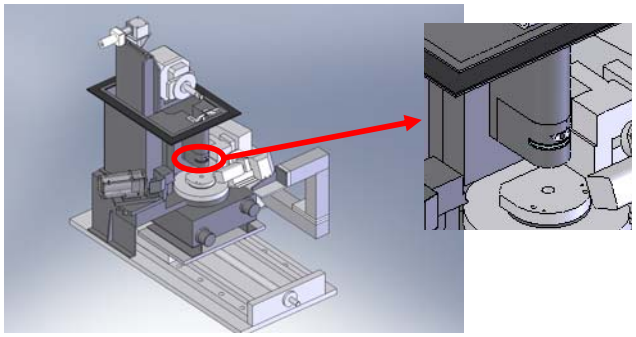


Figure. 3 : details of the goniometer-sample holder interface

3. Analytical performances

Analytical performances are illustrated below from two types of experiments achieved on inert samples representative of irradiated fuels. These experiments were performed on the standard device before its modification.
On the first example, we will demonstrate the improvement of X-ray measurements when using a micro-beam on heterogeneous samples. Then we will illustrate analytical capabilities of the system to quantify mechanical behavior of an irradiated fuel.

3.1. Illustration on heterogeneous samples:

High Temperature Reactor (HTR) is a promising concept especially for safety, co-generation and rentability characteristics. The basic fuel component of HTR Fuel is a kernel of fissile fuel surrounded by a PyC/SiC multilayer coating; the diameter of each particle is about 1 mm. These particles are then coated in a graphite matrix to obtain the final object so-called "compact". The characterization of these fuels involves microanalyses techniques as SEM (Scanning Electron Microscope), EPMA (Electron Probe Micro-Analysis) and SIMS (Secondary Ion Mass Spectrometry). Thanks to X-Ray diffraction device, structural analyses may be carried out.
To perform experiments, ZrO_2 has replaced UO_2 kernel. The other components (SiC/PyC layers) were remaining identical to nuclear material.
A Bragg-Brentano measurement has been made on a sample cross section (Fig. 4b): the surface irradiated by the X-ray beam is around 10 mm^2. Several fuel particles but mostly graphite matrix will contribute to XRD pattern (Fig. 4a).
When using a local beam, specific area in the fuel may be selected. As illustrated in Fig. 4d, the structural analysis of a single fuel particle may be performed using µ-XRD.

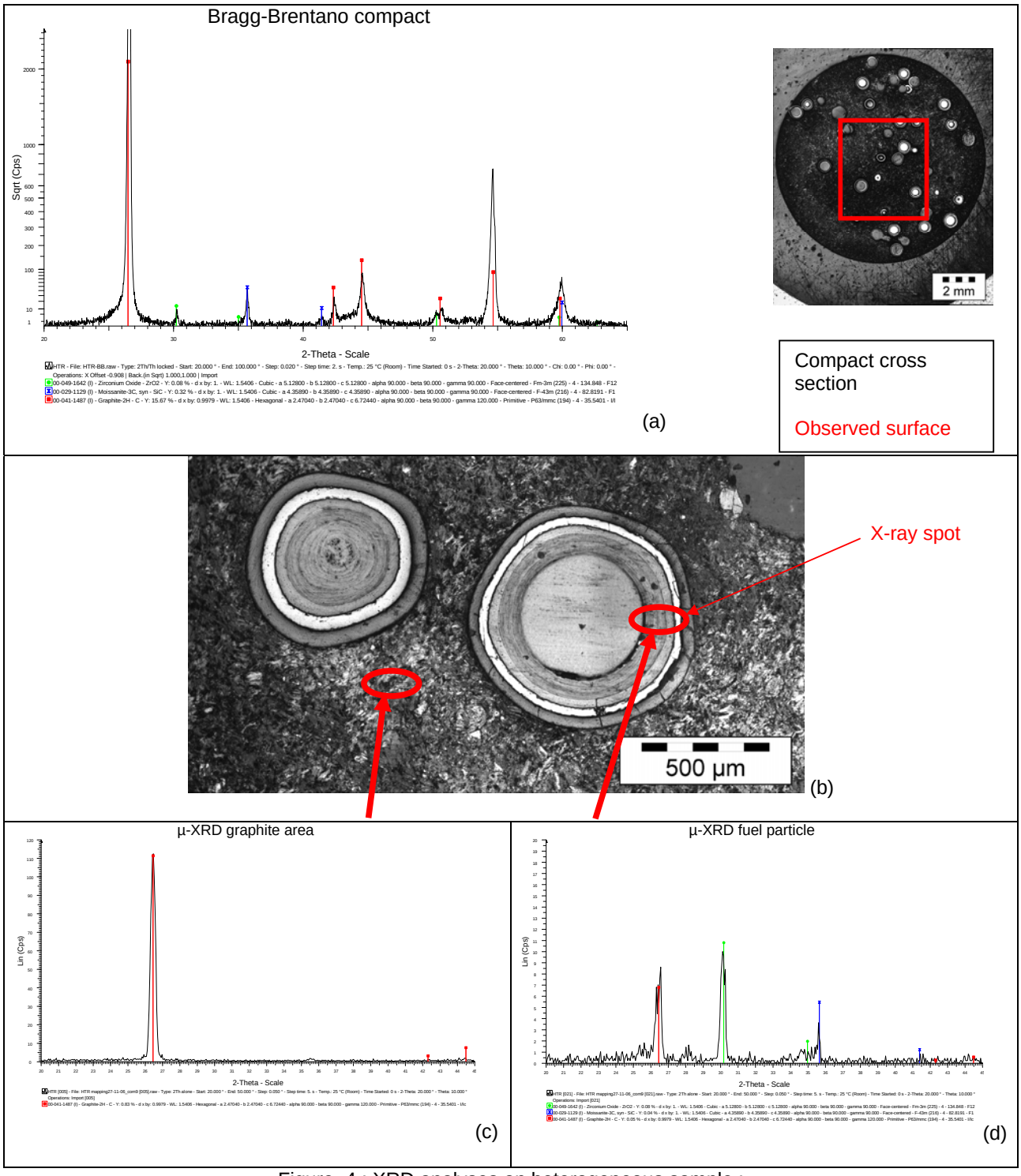


Figure. 4 : XRD analyses on heterogeneous sample ;
(a) : Bragg-Brentano measurement
(b) : sample cross section
(c) : µ-XRD on graphite matrix ; (d) : µ-XRD on particle

3.2. local strain-stresses analyses

The behavior of gases produced by fission is of great importance for nuclear fuel operation. It controls the release in the free volume and hence internal presure of the fuel rod together with fuel swelling and the rod beahavior in case of transient (especially in fast transients). The ultimate goal of fission gas behavior analyses is to determine the following quantities as a function of position in the fuel pellet and irradiation time:

- The concentration of gas atomically dissolved in the fuel matrix
- The concentration of gas in bubbles located in fuel grains and/or grain boundaries
- The amount of gas released from the fuel

The sum of previous concentrations is equal to fission gas production.
The size of gas bubbles is a function of the gas quantity, the surface tension and the stress state of the surrounding fuel matrix. Consequently, the determination of stress-strain field on irradiated fuel will be helpful to better-characterized bubbles behavior.
Mechanical Strain measurements on a fuel material may be carried out using X-ray diffraction methods [1].
Actually, The d-spacing (d_{hkl}) of a crystalline material acts as a strain sensor. The strain measurement $\varepsilon_{(hkl)} = \frac{\Delta d_{(hkl)}}{d_{0(hkl)}}$ is a function of line shift $\Delta 2\theta$ of a given (hkl) plane (Fig. 5).

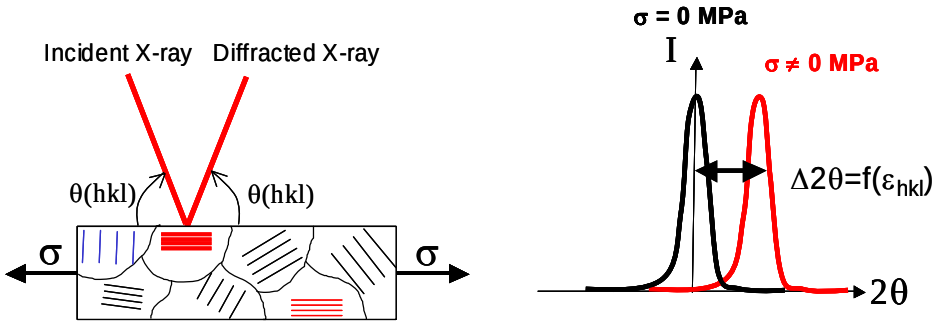


Figure. 5: X-ray Stress Analysis (XSA) method

The sensitivity of strain measurements is illustrated on a model material, UO_2 pellet sintered with high pressure Argon atmosphere.
SIMS analyses [2] exhibit a high density of precipitated Argon bubbles at the pellet center (several peaks detected on the SIMS depth profiles). At the pellet periphery, a decrease in Argon bubbles density is evidenced (Fig. 6a and 6b).
Strain analyses have been performed on the same area using XRD. It is demonstrated that stress level may be related to gas behavior: at equilibrium in the bubbles-UO_2 system, the gas pressure acting to expand the bubbles is balanced by the stress field in the surrounding solid. The more bubble density is increasing, the higher stress level is

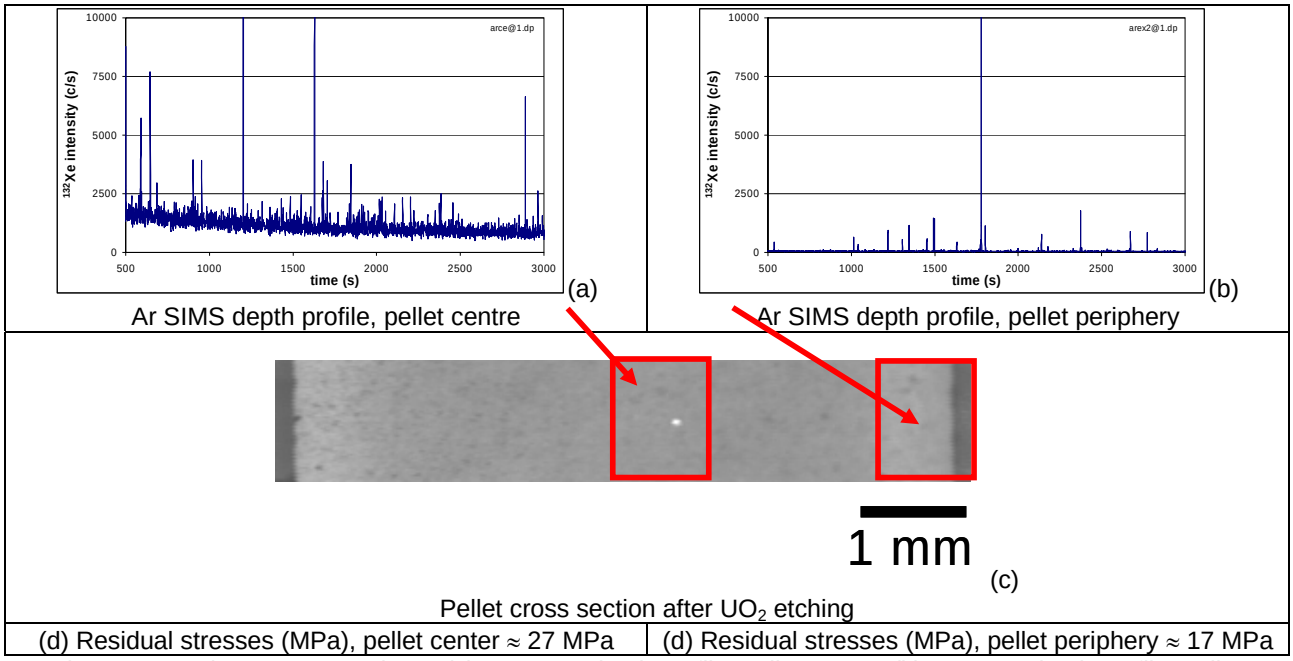


Figure. 6: Strain-stresses analyses (a): SIMS Ar depth profile, pellet center; (b) SIMS Ar depth profile, pellet periphery; (c): pellet cross section, SIMS and XRD measurement area are indicated in red; (d): Residual stress determined at the centre and pellet periphery

4. Conclusions

The modification of a standard XRD device to be installed in a hot cell has been detailed. The two main modification targets have been reached:

- to avoid contamination of X-Ray goniometer and to make easier the servicing of the goniometer.
- to reduce gamma radiation level on detection device in order to improve the quality of diffraction patterns.

New XRD capabilities have been demonstrated on model materials:

- local measurements on heterogeneous sample will allow to determine structure modification after irradiation (i.e phase transformation, amorphous materials, irradiation defects)
- Further informations on fission gases behavior may be obtained by using X-ray Stress analyses (XSA) methods

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