Fission gases pressure evaluation in irradiated PWR fuels : complementarities of microanalyses techniques, SEM, EPMA and SIMS

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Abstract

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 pressure of the fuel rod, the fuel swelling and also part of the behavior of the fuel during off-normal or accidental events. An experimental method was developed in CEA Cadarache hot laboratory, to perform quantitative microanalyses of xenon on irradiated fuel.
Local xenon inventory along a pellet radius was determined from SIMS measurement calibrated to EPMA data.
From SIMS data collection, local xenon depth profiles were converted into distribution chart to separately analyze, dissolved and gas precipitated xenon gas.
The number of bubbles containing xenon was sorted as a function of the cube root of SIMS intensity of each bubble.
From SEM analyses, porosities were sorted as a function of the ECD (Equivalent Circular Diameter) of each bubble.
Both SEM and SIMS distribution charts were then compared. Assuming that both maxima of each distribution correspond to the largest bubbles population. Xenon molar volume of this population was determined, and the mean pressure was obtained using a Van der Waals equation of state.
An illustration of the method is detailed on a UO_2 fuel rod irradiated 5 annual cycles in an EDF French Pressurized Water Reactor, and submitted to a power ramp in OSIRIS reactor.

***Keywords:* SIMS, EPMA, SEM, Xenon pressure, irradiated nuclear fuel.**

1. Introduction

The behavior of fission gases (FG) produced during irradiation in Pressurized Water Reactor (PWR) has a prominent impact on the fuel itself. They are released in the free volume and may have a major impact on both internal pressure of the fuel rod, fuel swelling and fuel behavior during incidental and accidental events. Integral measurement of released gas is obtained from rod puncturing. In that case local behavior of FG is not accessible. Up to now, Electron Probe Micro Analysis (EPMA) and Scanning Electron Microscope (SEM) techniques are dedicated to investigate FG local behavior. In order to improve the detection of xenon in irradiated fuels some experiments have been conducted with the CEA Cadarache shielded Secondary Ions Mass Spectrometer (SIMS) [1, 2]. In previous papers it was shown that SIMS was a useful technique to characterize total inventory of xenon in addition to EPMA [3, 4]. In the present paper, the quantitative analysis of xenon behavior is detailed using the micro-analysis devices available in Cadarache Hot laboratory.

2. Experimental

2.1 Sample

A 5 mm thick slice was taken out of a rollet, which underwent a power ramp in OSIRIS reactor at 400 W/cm during 12 hours, and which was refabricated from a UO_2 nuclear fuel rod already irradiated five cycles in an EDF French Pressurized Water Reactor. The local burn-up at the sampling position was calculated to be 63 GWd/tM. The sample was embedded in a low melting point metallic alloy and polished along a cross section perpendicular to the rod axis.

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2.2 Experimental devices

2.2.1 Electron Probe Micro Analysis

EPMA was performed using a shielded CAMEBAX device (CAMECA). Xenon radial distribution and mapping were collected with a primary electron beam (acceleration voltage : 25 kV, primary beam current 250 nA). Quantitative radial xenon analysis was obtained thanks to an UO_2 low Burn-Up fuel (25 GWd/tM) used as standard sample for xenon because it contained only dissolved xenon [5].

2.2.2 Scanning Electron Microscope

SEM was performed using a shielded XL 30 model (PHILIPS) equipped with a Centaurus KE developments BSE detector and SIS ADDA acquisition software to collect high resolution images.

2.2.3 Secondary Ions Mass Spectroscopy

SIMS experiments were carried out using a shielded IMS 6f (CAMECA). ^{132}Xe isotope depth profile was collected using a 30 nA oxygen primary beam defocused on a 30 µm diameter area. The ion sputtering time was 2000 s and the xenon signal was collected every 1s.

3. Results and discussion

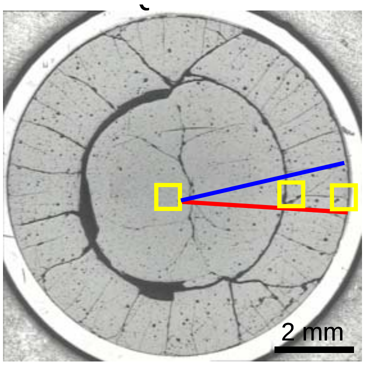

figure 1: photograph of the sample. EPMA, SEM and SIMS measurements location are indicated

A view of the embedded sample is given in figure 1. Location of EPMA , radial xenon profile in red, and xenon mapping in yellow (together with SEM imaging). SIMS radial ^{132}Xe profile is indicated in blue. Moreover 12 additional SEM images were obtained at various positions on the pellet.

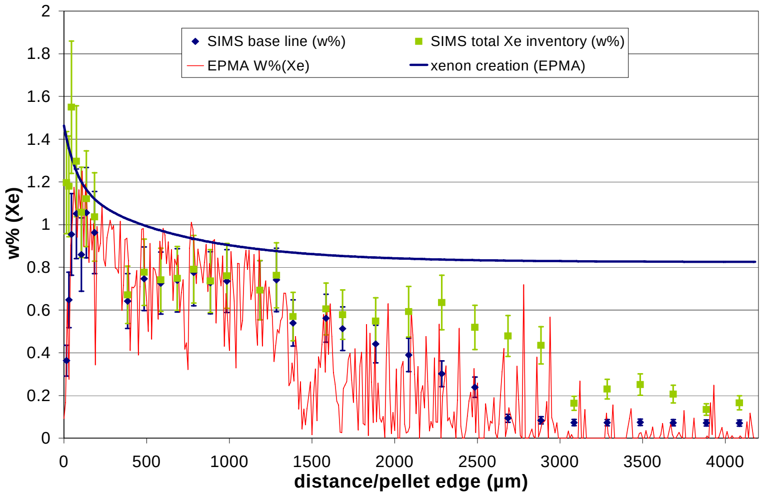

figure 2: EPMA and SIMS radial measurement compared to creation

3.1 Local xenon inventory

From both EPMA and SIMS measurements xenon concentration was determined using an analysis protocol detailed in [**Fehler! Textmarke nicht definiert.**] and [6]. Local inventory was then compared to local xenon creation obtained from local neodymium measurement (considered as directly connected to local burn-up) using either EPMA or SIMS [7] (figure 2).

EPMA measurements exhibit two areas where local xenon concentration is lower than what expected by creation values.

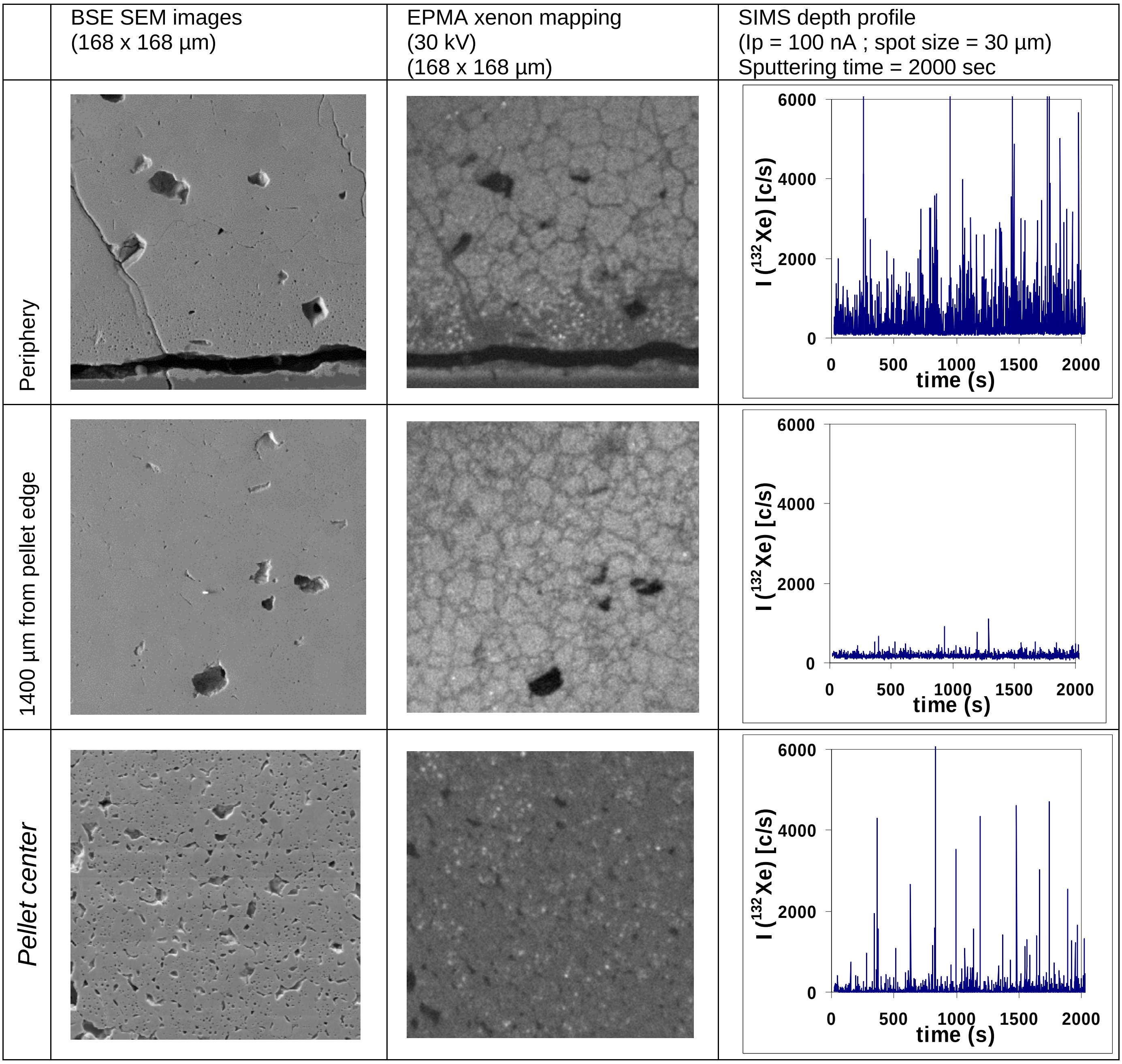

figure 3: SEM images, EPMA xenon mapping and SIMS depth profiles, collected in the same area

The first one is located at the periphery of the pellet (0-100 µm) where the rim effect occurred (High Burn-up Structure HBS [8]).

The second one, located from the mid-radius1300µm from the pellet edge to the pellet center, is connected to the high temperature value reached during power reactor or ramp irradiation : as a consequence part of fission xenon is precipitated into porosities whose sizes are determined from SEM results.

In the last area (from 100 to 1300 µm), xenon concentrations measured with EPMA are closed from creation values. In this area both SIMS and EPMA measurement should be similar (i.e. no significant xenon release and xenon considered in solid solution). Thus SIMS measurement were correlated to EPMA one. The SIMS depth profile evidenced no significant gas inventory in bubbles. It is then reasonable to consider that all the gas is dissolved or in small bubbles. In such conditions, the gas quantity measured by SIMS ans EPMA is equal, that is the reason why the SIMS data are normalized to the EPMA ones in this area.

From SIMS measurements a gas release was evidenced (SIMS total xenon inventory in figure 2) in the mid-radiusfrom 1300 µm from the pellet edge to center pellet area. In the same time bubbles growth (still containing xenon) was detected in this area(SIMS depth profile in the pellet center area on figure 3).

No significant release was noticed in the rim area, nevertheless a gas precipitation was clearly evidenced from both SEM images (increasing porosities observed on figure 3 in the pellet periphery), EPMA mapping results (xenon bubbles detected under the sample surface as presented on figure 3 in the pellet periphery) and SIMS depth profiles (large number of peaks connected to xenon bubbles punching by the primary ion beam)

3.2 SEM and SIMS porosities analyses

The porosities size distribution was determined by SEM analyses in three areas at different radial position from the edge pellet to its center. For each position the volume fraction of porosities is presented (figure 4 C).
Bubbles distribution was extracted from SIMS depth profiles (figure 3) collected in the same area. Each data points of the depth profile were sorted as a function of its intensity. These charts, as presented in figure 4 A allow the separate analysis of depth profile baseline and peaks:
The peak always observed on the distribution chart was fitted using a gauss function, its maximum gives the mean value of the baseline. The baseline corresponds to the straight line on which peaks are superimposed in the depth profiles.
After removal of the baseline (continuous line on figure 4 A distribution chart), residue was sorted as a function of cube root of ^{132}Xe intensities of each bubble: SIMS distribution chart of the number of porosities containing xenon was extracted. SIMS number distribution chart were compared to SEM number distribution diagram. From SEM diagram the whole porosities population was observed irrespective of the xenon content of each observed bubbles. Moreover smallest bubbles observed with SEM (i.e. 0,1-0,2 µm ECD) were not detected as peaksbut included in the baseline instead of peaks in the SIMS depth profiles [**Fehler! Textmarke nicht definiert.**]. SIMS distribution chart of the number of porosities did not contain 0,1-0,2 µm size class. The main bubbles population observed in the rim area did contain xenon as evidenced by similar values of SIMS and SEM bubbles number. From 1400 µm from pellet edge to its center, most of porosities were xenon emptied, this is consistent with the fraction gas released evidenced on the radial profile (figure 2)
The SIMS quantity distribution chart (figure 4) was finally calculated : for each class, the number of bubbles was multiplied by the molar contain of each bubble.

	Periphery	1400 µm from pellet edge	Center
Bubbles number/µm^3 SIMS	0,13	0,07	0,08
Bubbles number/µm^3 SEM 0,3 < ECD < 4 µm	0,18	0,3	0,2

Table 1: Comparison of bubbles densities from SIMS data (xenon contained bubbles) and SEM images analyses.

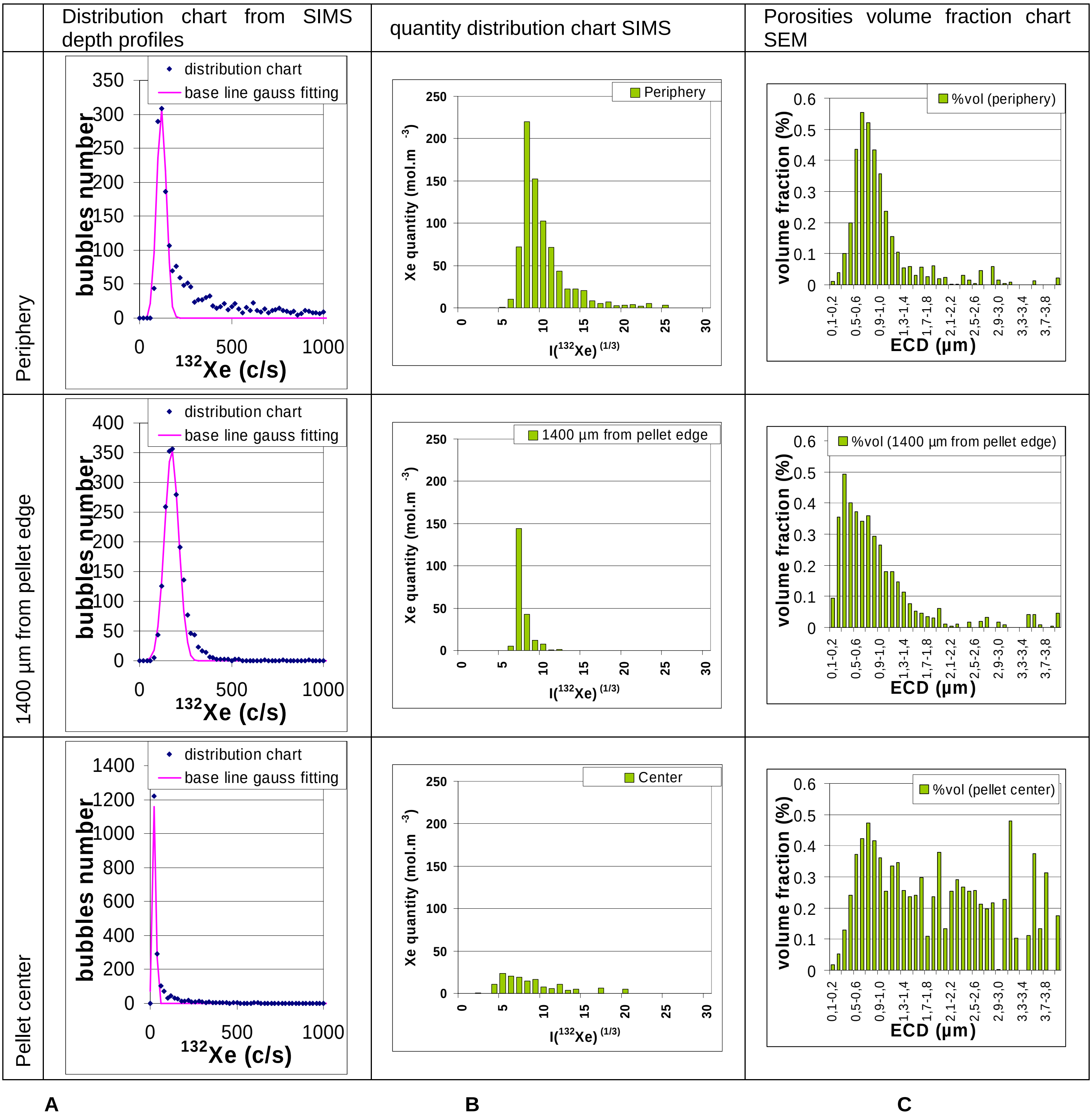

figure 4 : A. SIMS xenon intensities distribution charts computed from Xe depth profile
B. quantity of xenon contained in bubbles charts deduced from previous chart
C. Porosities volume fraction diagram, SEM

3.3 Atomic volume and Xe pressure determination

Assuming that both SIMS quantity distribution and SEM volume fraction distribution are representative of the same bubbles population, xenon pressure of the most important bubbles population was determined.
The molar volume of xenon contained in this population was computed by identification of both maximum of SIMS and SEM distribution. Using a modified Van der Waals equation of state, the mean pressure was estimated. The pressure estimation is the most consistent in the rim area where no significant number of emptied porosities is evidenced.

	atomic volume (Å/at)	Mean pressure (MPa) at 293 °K	Mean pressure (MPa) at ramp temperature °K
Periphery	155	17	135 (900 °K)
Pellet center	234	6	170 (2000°K)

Table 2: mean pressure estimation from identification of SIMS and SEM most important bubbles population

The pressure estimation in the rim area is consistent with previous results [**Fehler! Textmarke nicht definiert.**, 9] presented on fuels irradiated in power reactor up to a similar burn-up.
In the pellet center, results have to be carefully analyzed: a significant xenon fraction has been released during power and ramp irradiation, at ambient temperature the mean pressure in remaining xenon bubbles is three time lower compared to rim area, at ramp temperature center xenon bubbles are the most pressurized.

4. Conclusions

We have shown that local behavior of xenon can be quantitatively analyzed using complementarities of micro-analyses tools SEM, EPMA and SIMS :
The radial location of fission gas release is determined, in our sample we've shown that no significant FGR from the rim area was resulting from irradiation in both power and test reactor.
The xenon contained bubble population is analyzed. It's demonstrated that pellet center bubbles are highly pressurized at ramp temperature compared to rim area bubbles.

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