New methods to estimate the internal pressure of xenon in fission bubbles using SEM-EPMA-SIMS and EBSD in irradiated fuels

C. Cagna, I. Zacharie-Aubrun, P. Bienvenu, B. Michel, L. Barrallier

FROM RESEARCH TO INDUSTRY

New methods to estimate the internal pressure of xenon in fission bubbles using SEM-EPMA-SIMS and EBSD in irradiated fuels

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C. CAGNA, I. ZACHARIE-AUBRUN, P. BIENVENU, B. MICHEL –
CEA Cadarache, DEC
L. BARRALLIER – Arts et Métiers ParisTech, Laboratory MSMP

EMAS2015 |

5 MAY 2015

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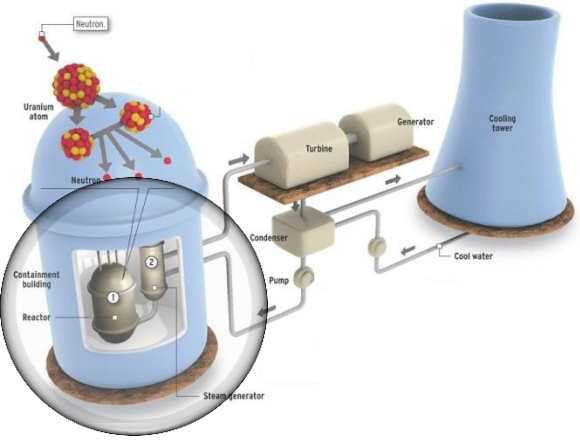
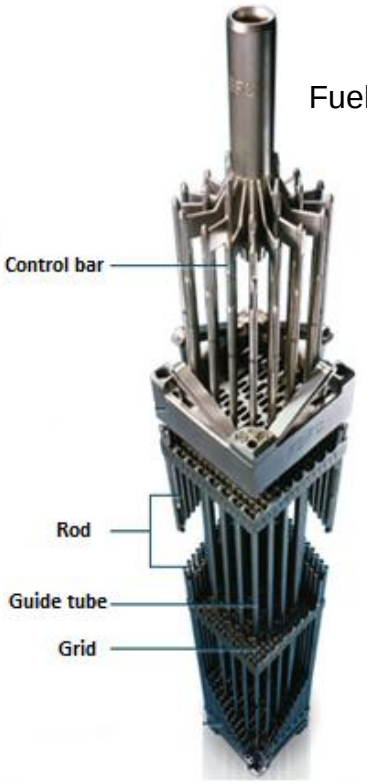
I- CONTEXT



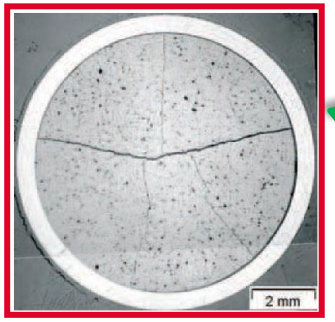
PW Reactor



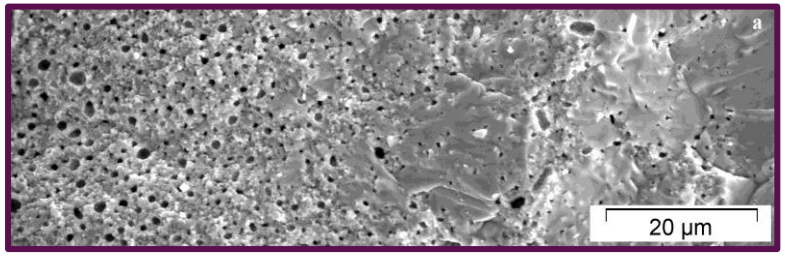
Fuel assembly

Rods : fuel + cladding

Fuel : UO_2 pellets



Radial section of a pellet



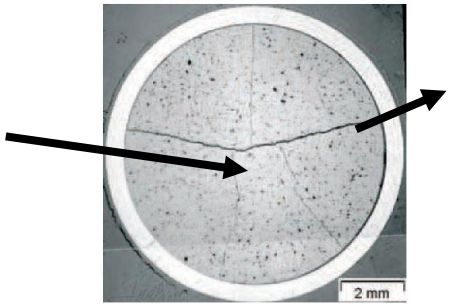
Fuel's microstructure

I- CONTEXT

- Modeling is an essential tool that is now seeking to translate all phenomena and calculate all physical quantities (temperature, stress, swelling, release of fission gas…) that modify the behavior of PWR fuels.

- Providing new experimental data, such as **fission gas bubbles pressure**, is a way to validate and improve the models' predictions of the fuel behaviour during :

  - Nominal Irradiation

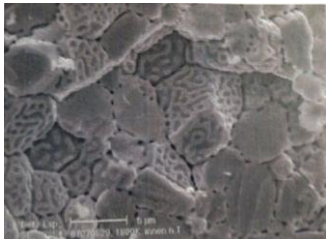


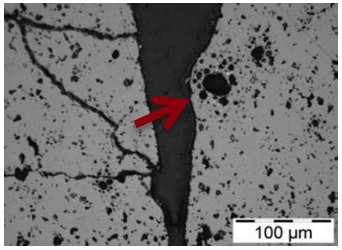
Irradiated 1 cycle

  - Power ramps

  - Accidental situation – RIA (Reactivity Insertion Accident)

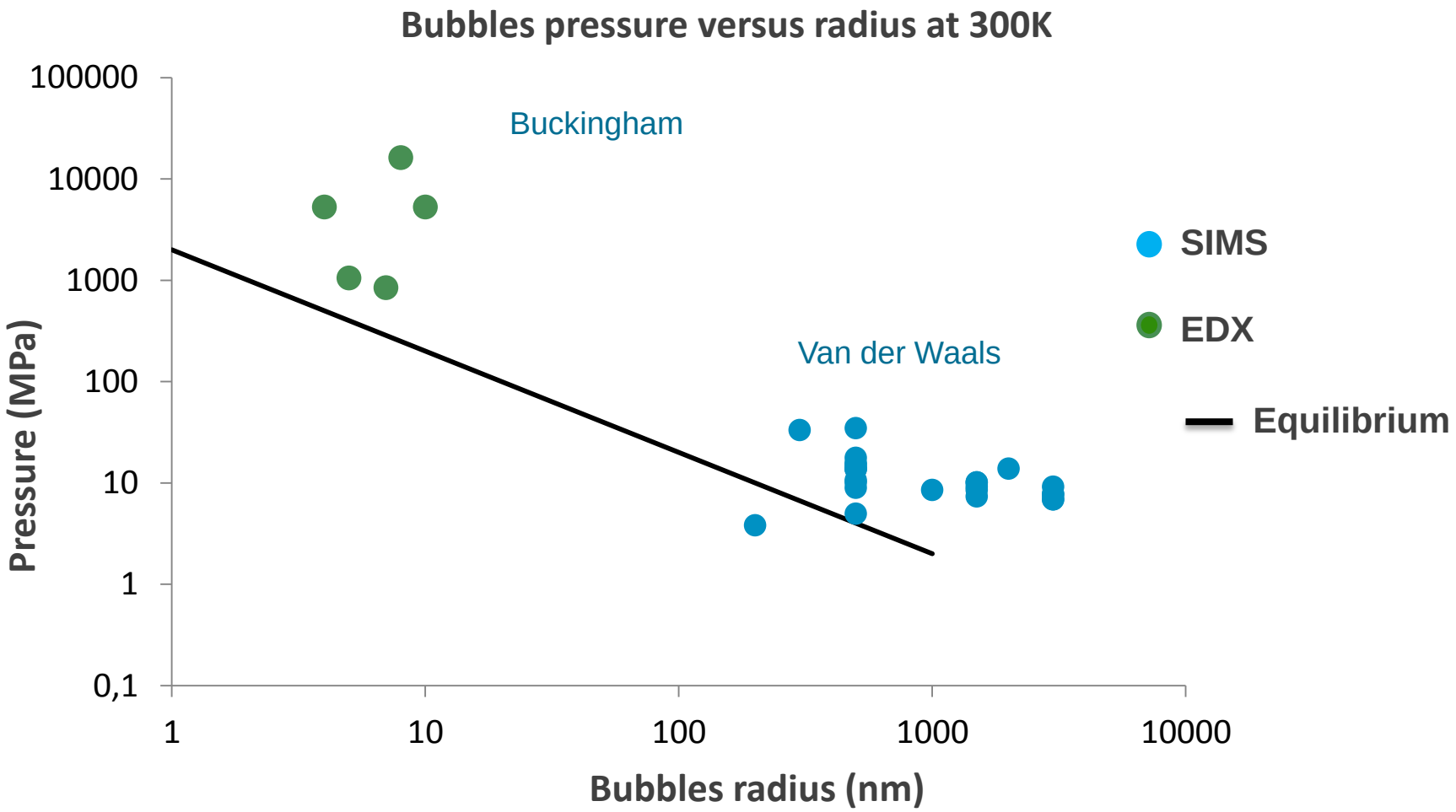
- **Localisation of fission gases**
  - In solution inside the fuel
  - Intra- and/or intergranular bubbles


Coalescence of bubbles after a ramp



HBS in MOX

Experimental data from the literature



J.Noirot, NFIR report, 2012
J.Noirot, JNM 372, 2008,
K.Nogita, Nuclear Instruments and Methods in Physics Research B, 141, 481-486,1998 |

III- ESTIMATING THE PRESSURE IN BUBBLES

I. CONTEXT

II. ESTIMATING THE PRESSURE IN BUBBLES

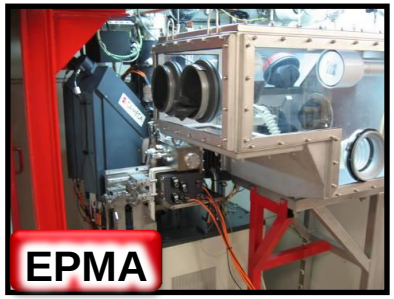
- **Direct method : Measurement by SEM/EPMA/SIMS**

- Indirect method :
  - Strain measurement by EBSD/cross-correlation,
  - Finite element modeling to calculate the pressure

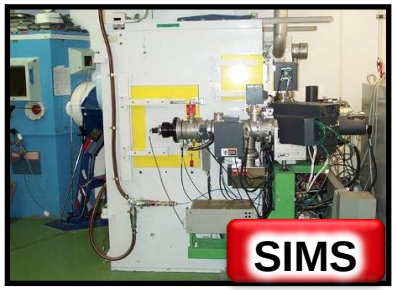
II. CONCLUSION AND FUTURE PROSPECTS

III- ESTIMATING THE PRESSURE IN BUBBLES – GLOBAL METHODOLOGY

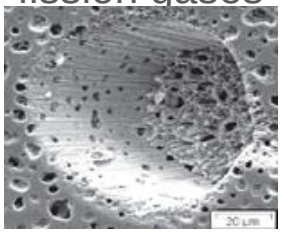
Measuring the amount of xenon in bubbles



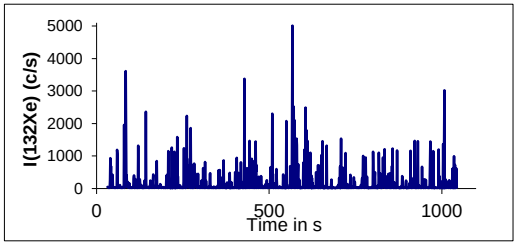
Quantitative analysis



Total inventory of fission gases



SIMS crater

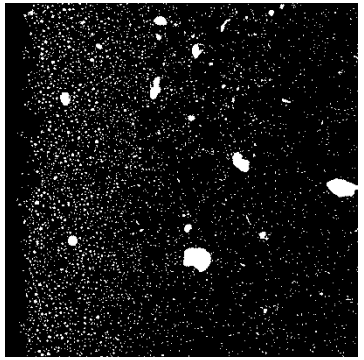


SIMS depth profile

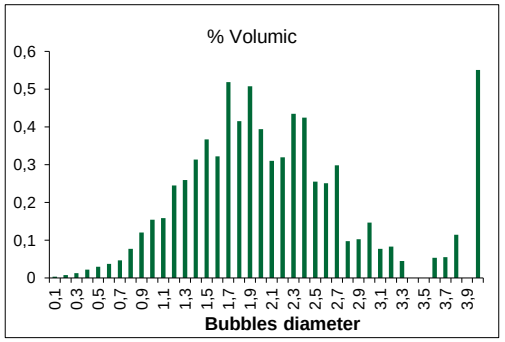
- Quantity of xenon inside the bubbles

Measuring the volume fraction of bubbles

Morphology Image analysis



Binary image



- 2D -> 3D Volume fraction

Atomic Volume

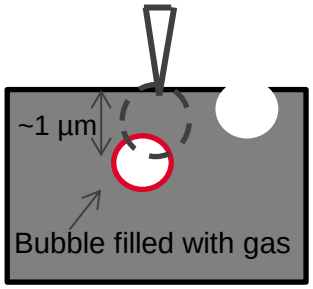
Mean pressure

) Equation of state

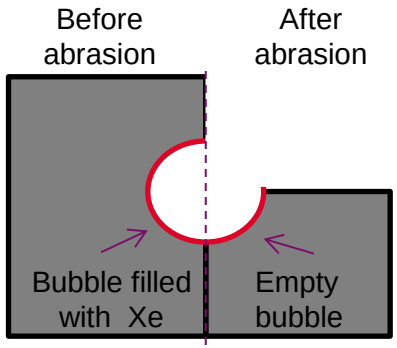
III- ESTIMATING THE PRESSURE IN BUBBLES – SINGLE BUBBLE METHODOLOGY

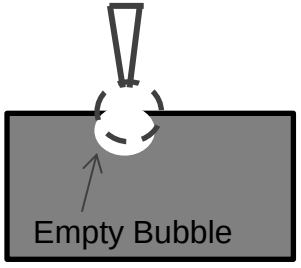
① SEM
Identification and **localization of one bubble under the surface**

② EPMA
Checking that the bubble is filled with xenon – X Carto



③ SIMS
Sample abrasion



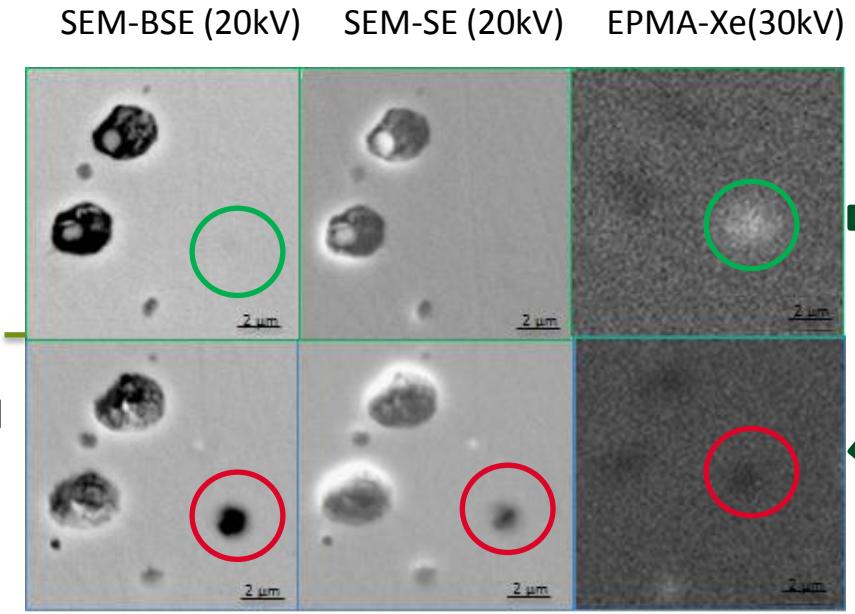


④ EPMA
Checking that the bubble has been opened– X Carto

⑤ SEM
Measurement of the bubble diameter

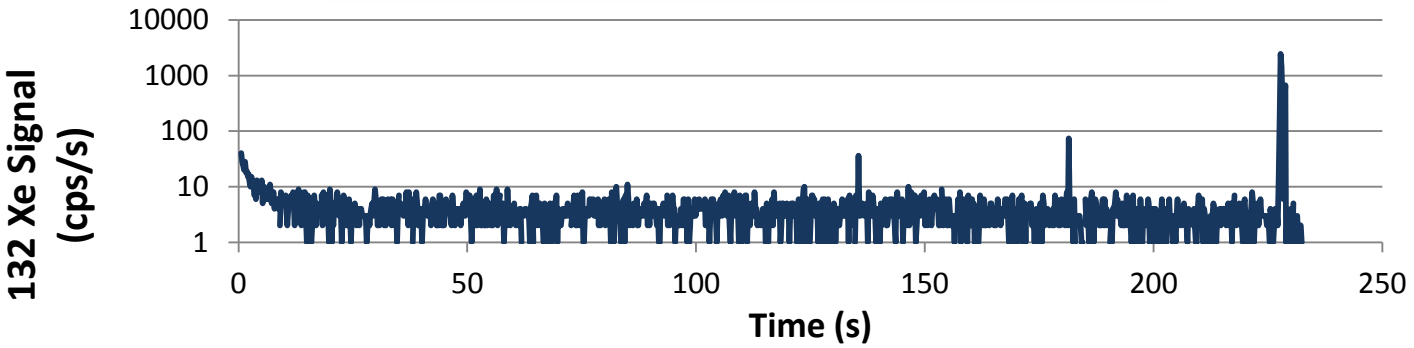
III- ESTIMATING THE PRESSURE IN BUBBLES – SINGLE BUBBLE METHODOLOGY

- An area with isolated bubbles just below the surface is identified by SEM and EPMA.
- Opened bubbles are identified by a second sweep by SEM and EPMA.



SIMS abrasion

Associating peaks and bubbles

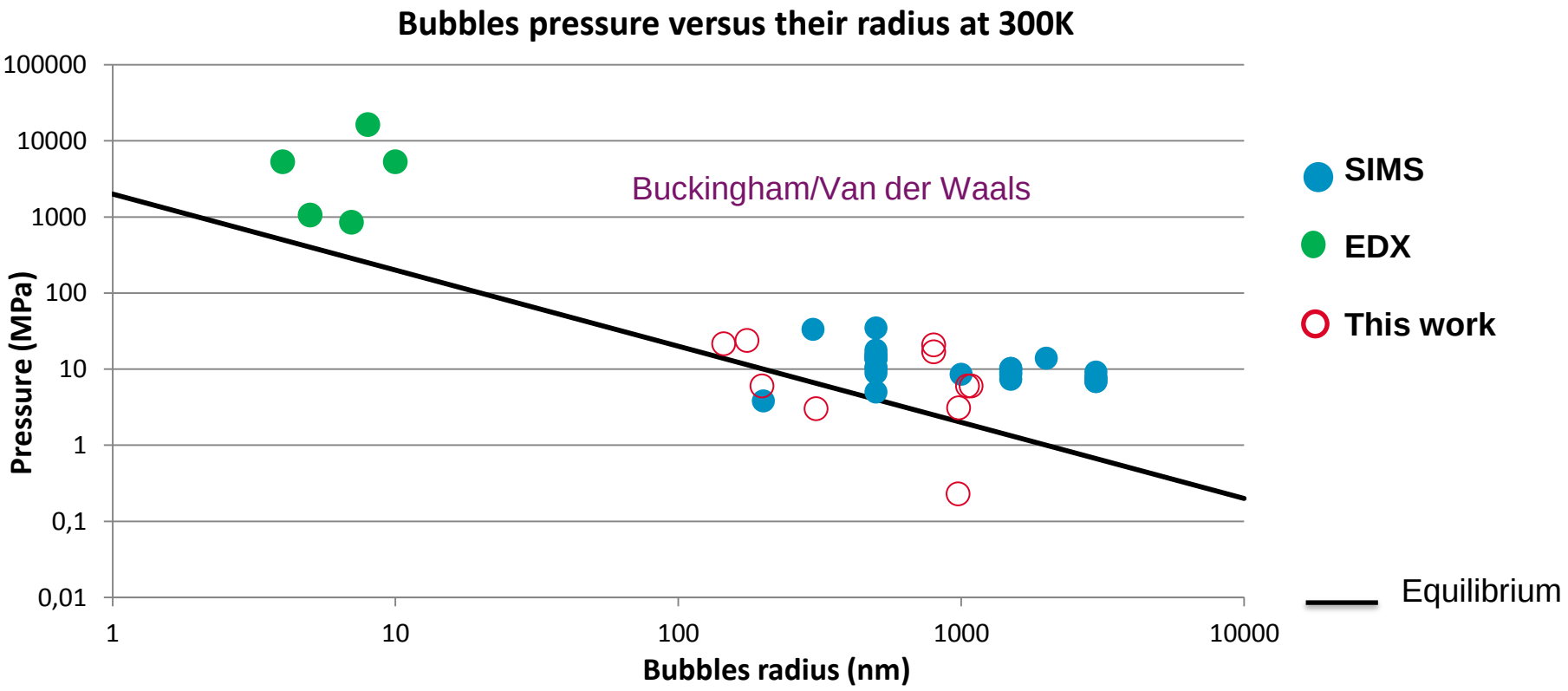


Depth profile by SIMS

III- ESTIMATING THE PRESSURE IN BUBBLES – SINGLE BUBBLE METHODOLOGY

Results

Equation of state : Molar volume < 90 $m^{-3}.mol^{-1}$ Buckingham
Molar volume > 90 $m^{-3}.mol^{-1}$ Van der Waals



At room temperature, the pressure tends to be close to the equilibrium

III- ESTIMATING THE PRESSURE IN BUBBLES

I. CONTEXT

II. ESTIMATING THE PRESSURE IN BUBBLES

- Direct method : Measurement by SEM/EPMA/SIMS

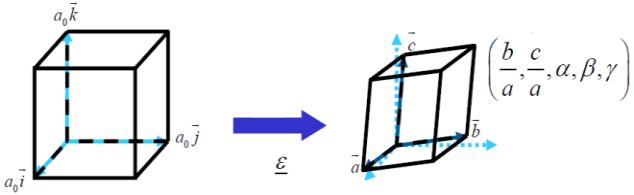
- **Indirect method :**
  - **Strain measurement by EBSD/cross-correlation,**
  - **Finite element modeling to calculate the pressure**

II. CONCLUSION AND FUTURE PROSPECTS

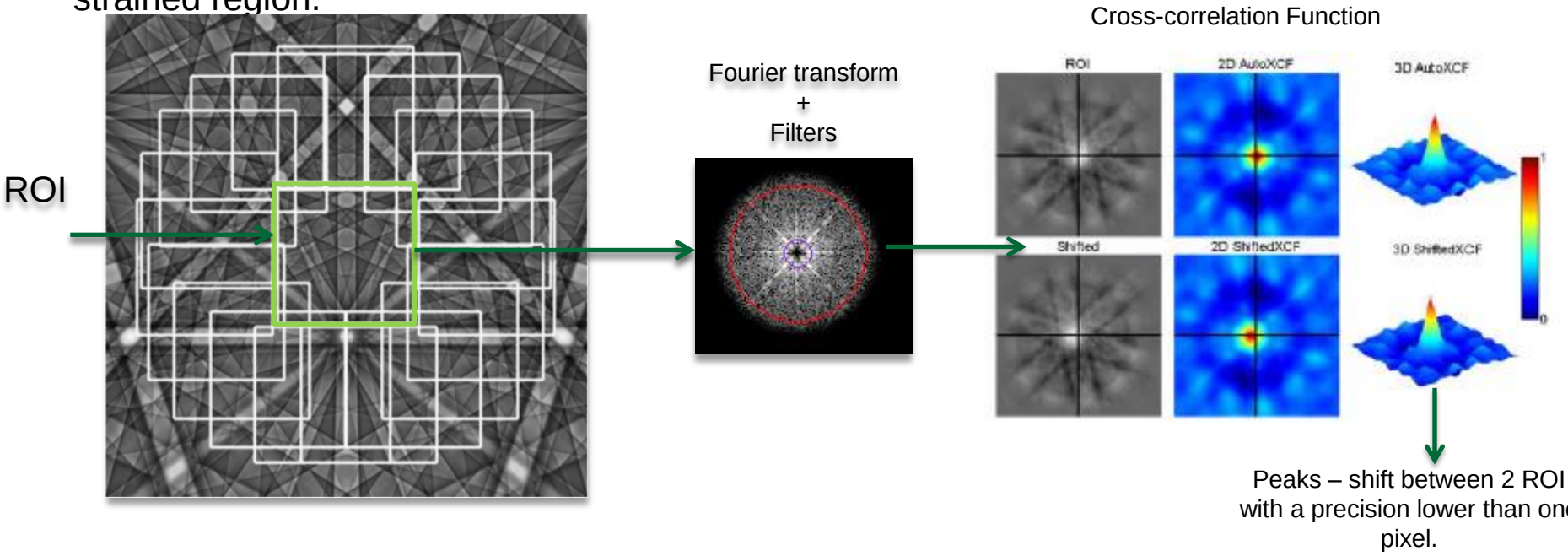
III- ESTIMATING THE PRESSURE IN BUBBLES BY EBSD

- The stress applied to a material will lead to a **distortion** of its crystal lattice and thus a movement of bands on the EBSD Kikuchi diagram.

Strain precision estimated at ~**2.10^{-4}**



- Two diagrams are taken from a grain, one in a unstrained region (reference) and another in a strained region.

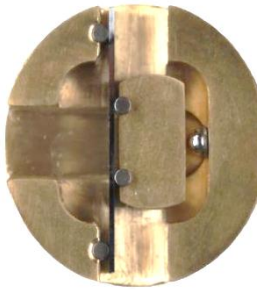


- It is then possible to calculate at any point of the sample **the elastic strain and rotation tensors of the crystal lattice.**

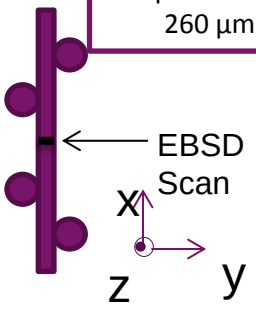
Réf : A.J. Wilkinson *et al*., Ultramicroscopy, 106, 307-313, 2006
T. Britton, A.J. Wilkinson, Ultramicroscopy, 111, 1395-1404, 2011

III- ESTIMATING THE PRESSURE IN BUBBLES BY EBSD

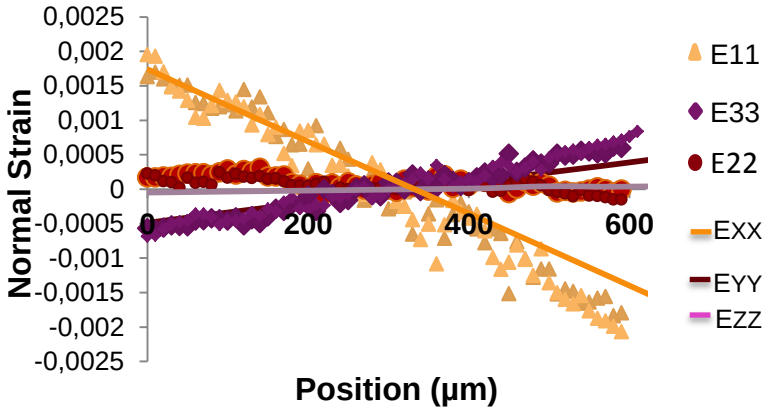
First step : calibration

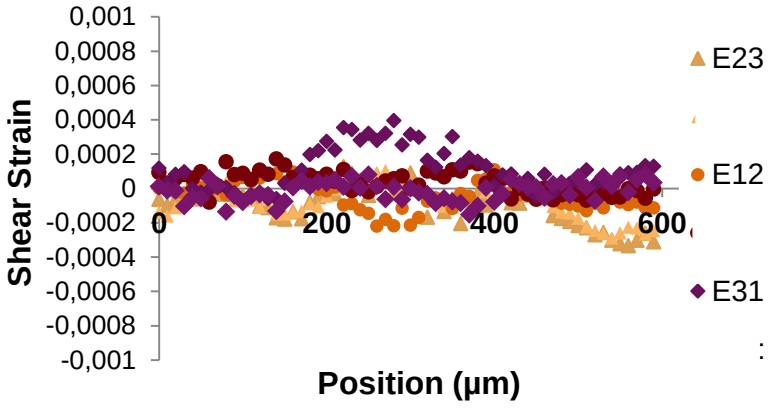


20 kV
Displacement :
260 µm



Four point bending test on a silicon single crystal



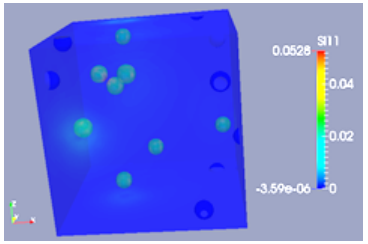


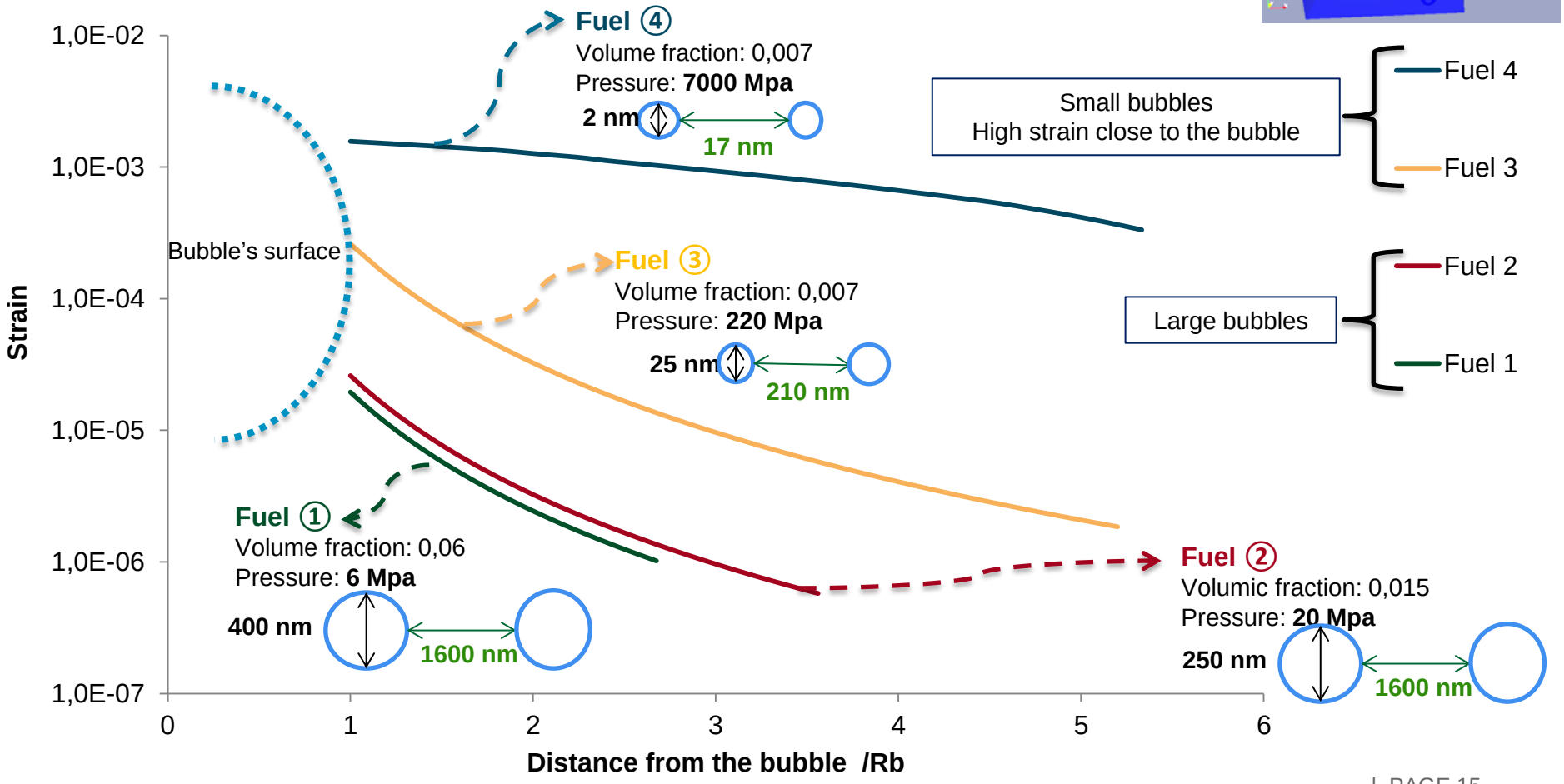
- Center point of the line = unstrained reference. EBSD patterns are compared to this reference.
- In order to validate the results, the four-point bending experiment was simulated by a FE model assuming anisotropic elasticity of cubic materials (C_{11} = 396 Gpa, C_{12}= 121 Gpa, C_{44} = 64 Gpa).
- According to the calculations, the shear strain should be null over the area. It appears that some points are over 2.10^{-4}. Special care is required to avoid any projection parameters variations, with a flat and parallel sample surface to the stage surface.

III- ESTIMATING THE PRESSURE IN BUBBLES BY EBSD

From calibration to irradiated fuels

With a finite element model it is possible to estimate the strain reached around the bubbles in irradiated fuels for different microstructures





Rb : Bubble radius

III- CONCLUSION AND FUTURE PROSPECTS

I. CONTEXT

II. ESTIMATING THE PRESSURE IN BUBBLES

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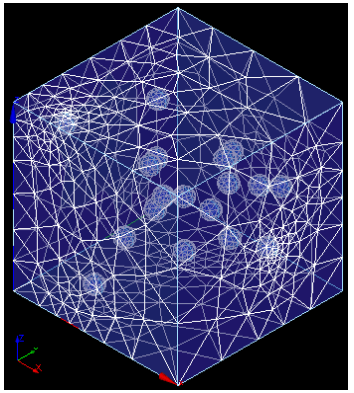
III- CONCLUSION AND FUTURE PROSPECTS

SEM/EPMA/SIMS method

- This study will be continued with the aim of adjusting acquisition confitins and adapting the SIMS beam to detect a smaller number of bubbles.
- Work is performed to improve the precision and accuracy of the results by working on the measurement of the bubble diameter (FIB-SEM), peak intensity and the equation of state.
- The results will be compared to a more global method.

EBSD and Cross-Correlation method

- Application of the method on material models such as ceramics implanted in xenon.
- Application on irradiated fuels thanks to a FIB-SEM in a hot lab.
- A finite-element model is developed to determine strain caused by bubbles in the irradiated fuel and therefore fission gas pressure.



Finite-element model

Thank you for your attention

Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Cadarache | 13108 Saint Paul Lez Durance
T. +33 (0)4 42 25 53 10
celine.cagna@cea.fr

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