

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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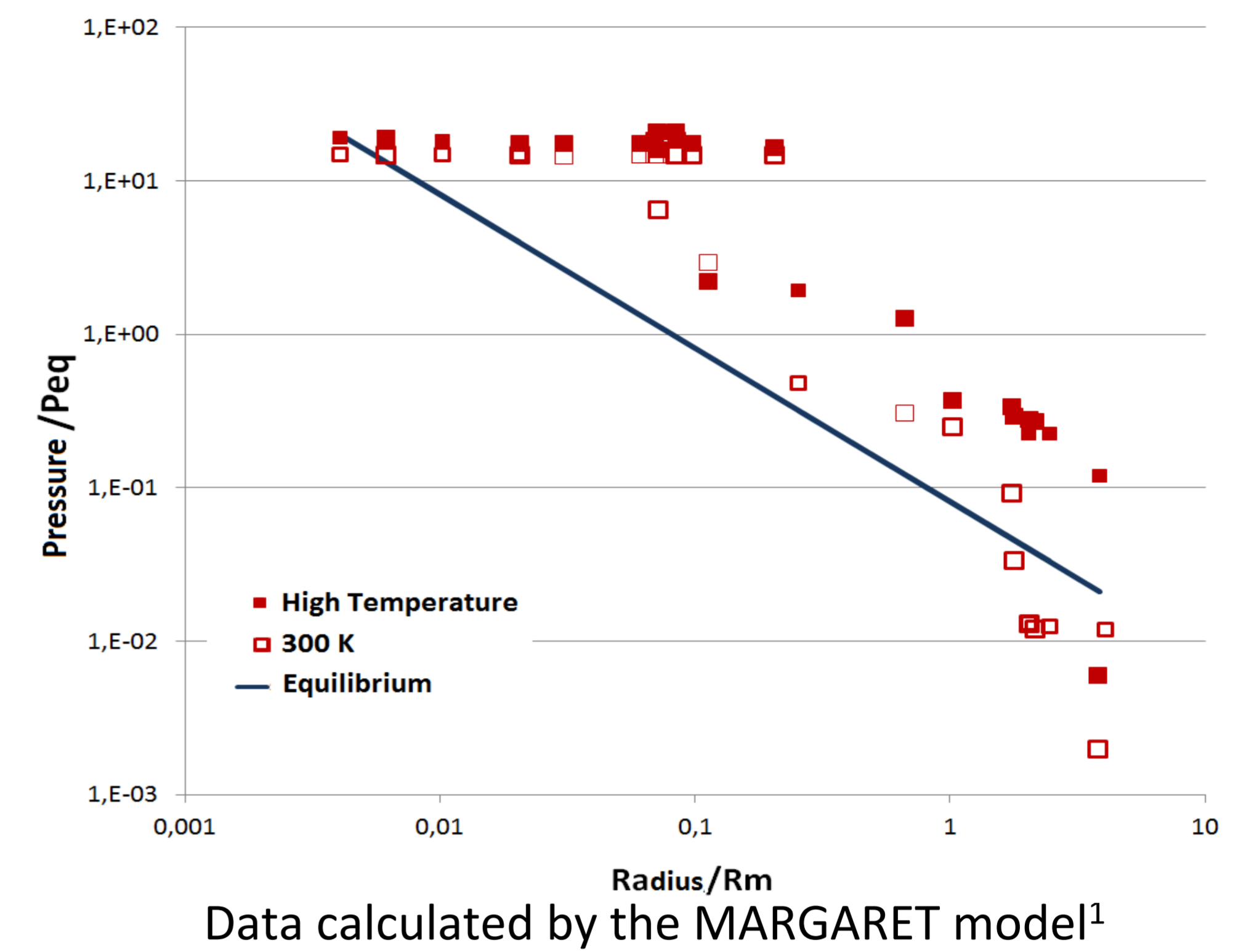
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During the operation of a Pressurized Water Reactor (PWR), the fuel is subjected to different loads - mainly mechanical and thermal - that modify its behaviour when combined with the effects of irradiation. The irradiation generates fission gases (Xe and Kr) in the fuel pellets which are generally found in bubble form. The internal pressure of these bubbles can be high and thus generate internal stress in the fuel.

This study* focuses on developing methods capable of measuring the deformation fields around the bubbles and the pressure causing this deformation.

* Thesis in collaboration with the CEA Cadarache center and the ENSAM engineering school in Aix-en-Provence

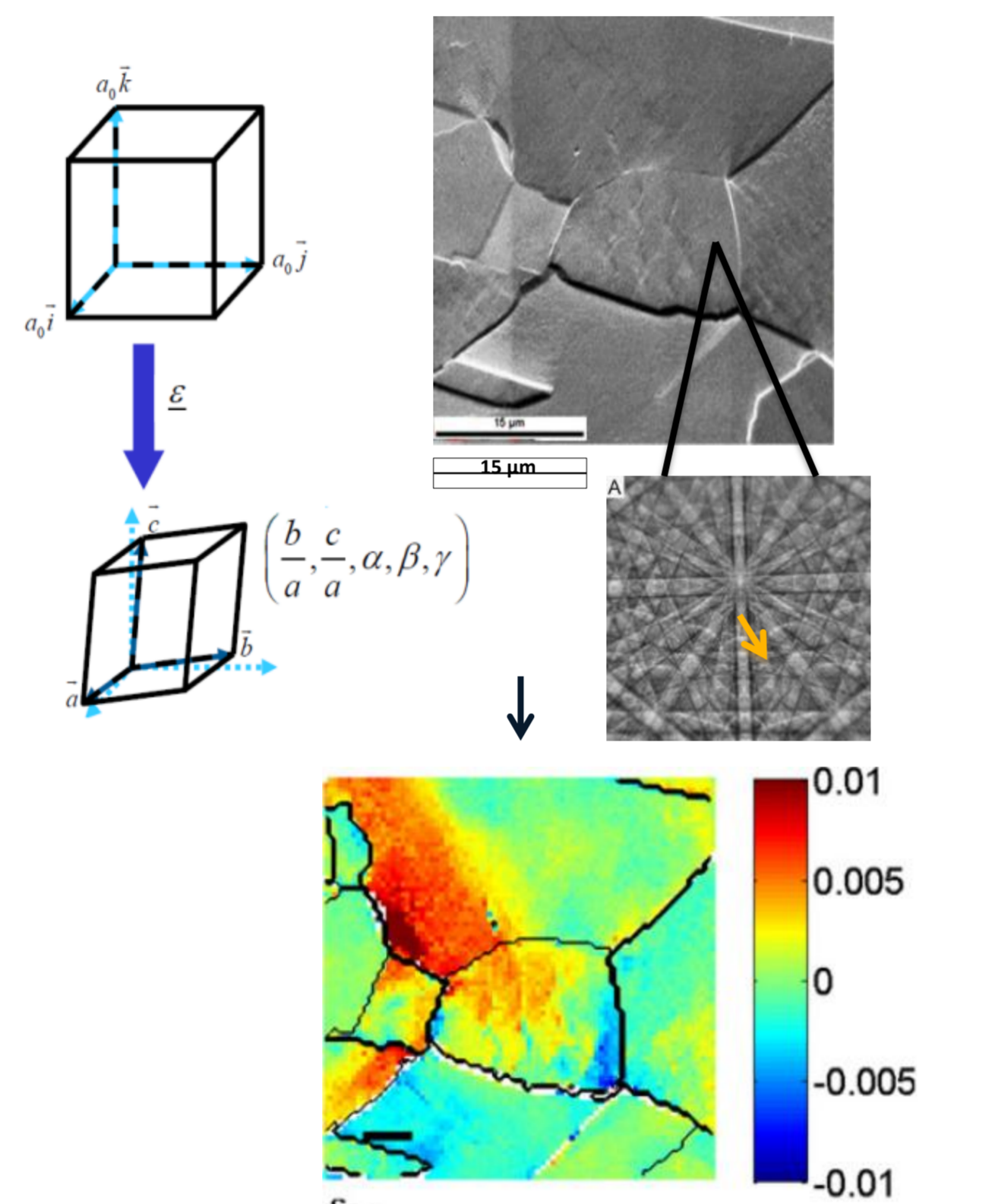


Methodology

Measuring deformation around pressurized bubbles using EBSD

EBSD and Cross-Correlation - Principle

When stress is applied to a material, it will deform the crystal lattice which leads to the displacement of the Kikuchi bands as shown in the EBSD diagram.



It is possible to determine the elastic **strain tensor**, **lattice rotation tensor** and **stress tensor** at any point in the sample. The pressure can thus be determined by using an inverse method and a finite-element model related to the sample.

Deformation sensibility : $2 \cdot 10^{-4}$

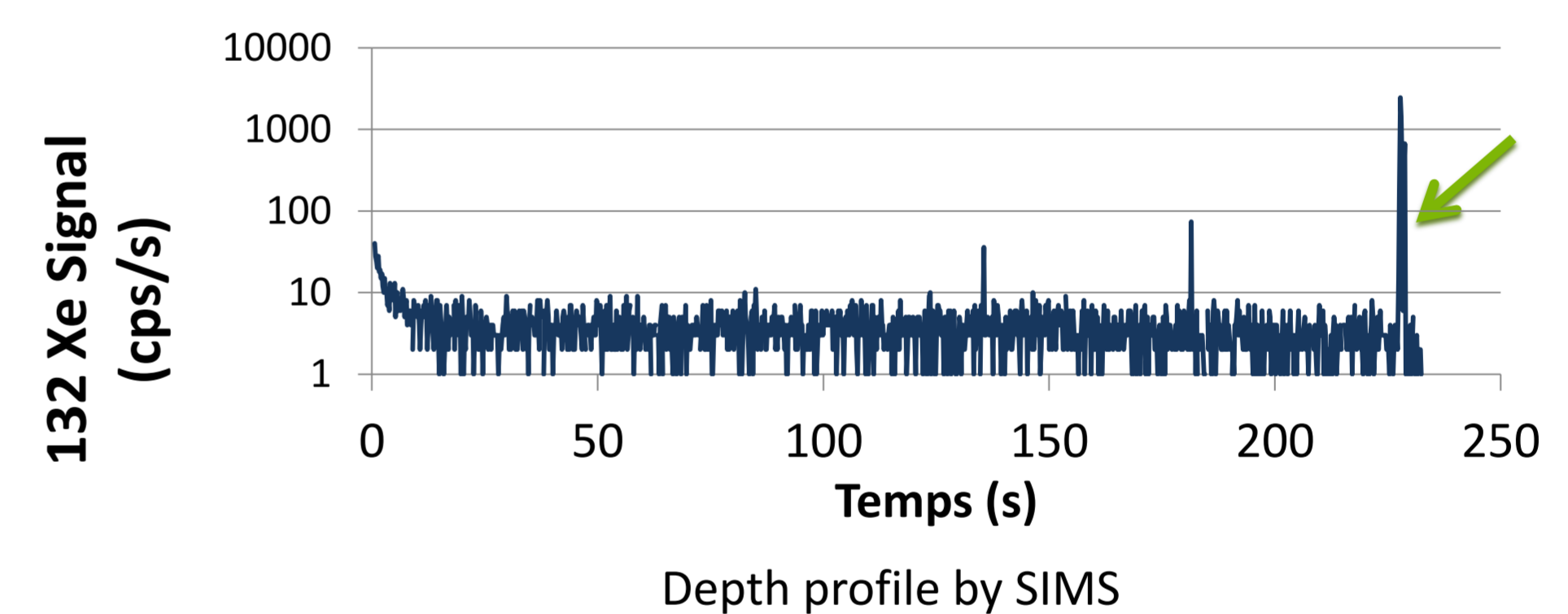
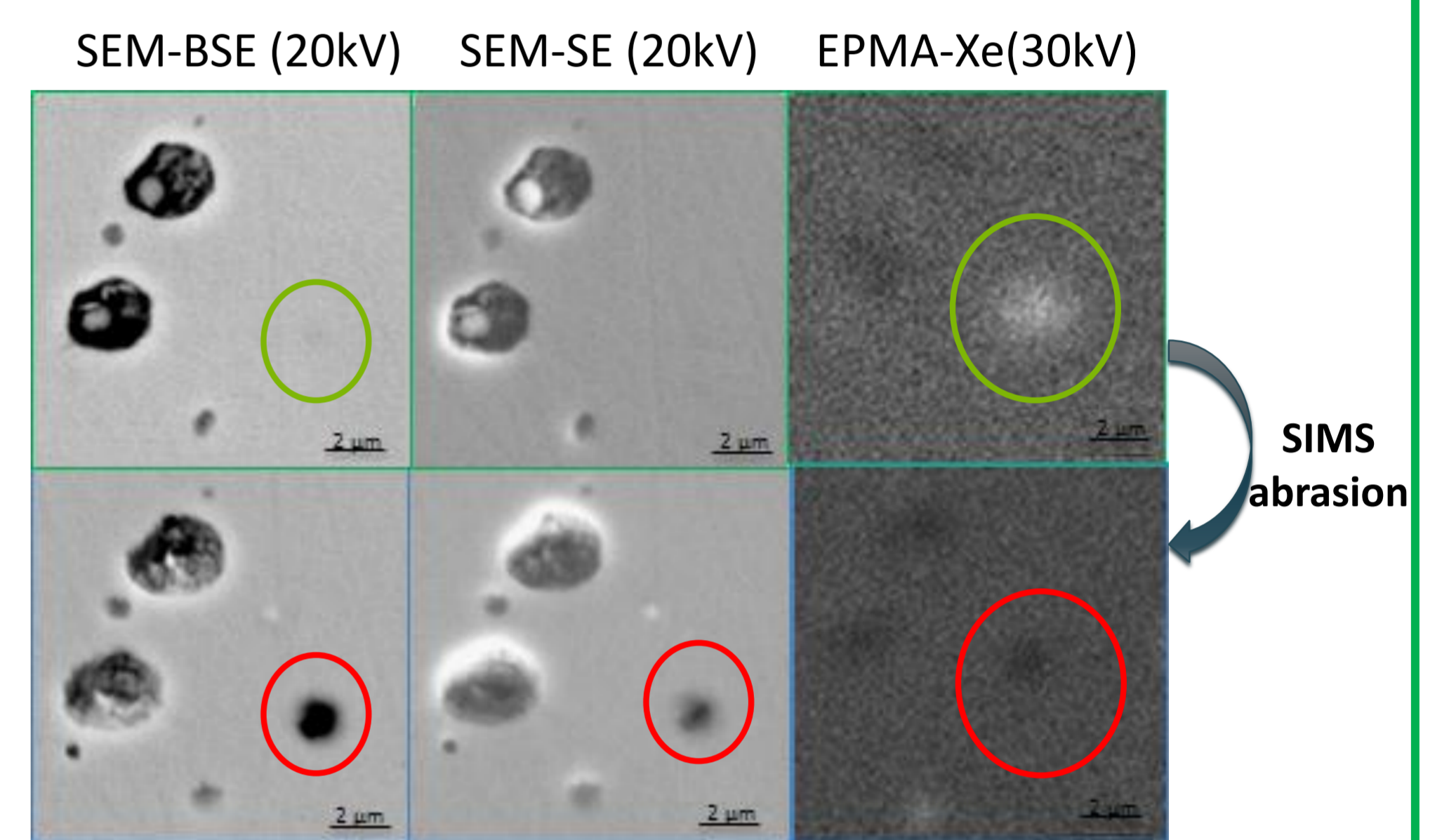
Measuring deformation by cross-correlation²

Measuring bubble pressure by SEM-SIMS-EPMA on irradiated fuel

- An area with isolated bubbles just below the surface is identified with SEM and EPMA.

- Opened bubbles are identified by a second sweep of the SEM and EPMA.

- The depth profile by SIMS reveals a number of peak. Assumption: one peak corresponds to the opening of one bubble.



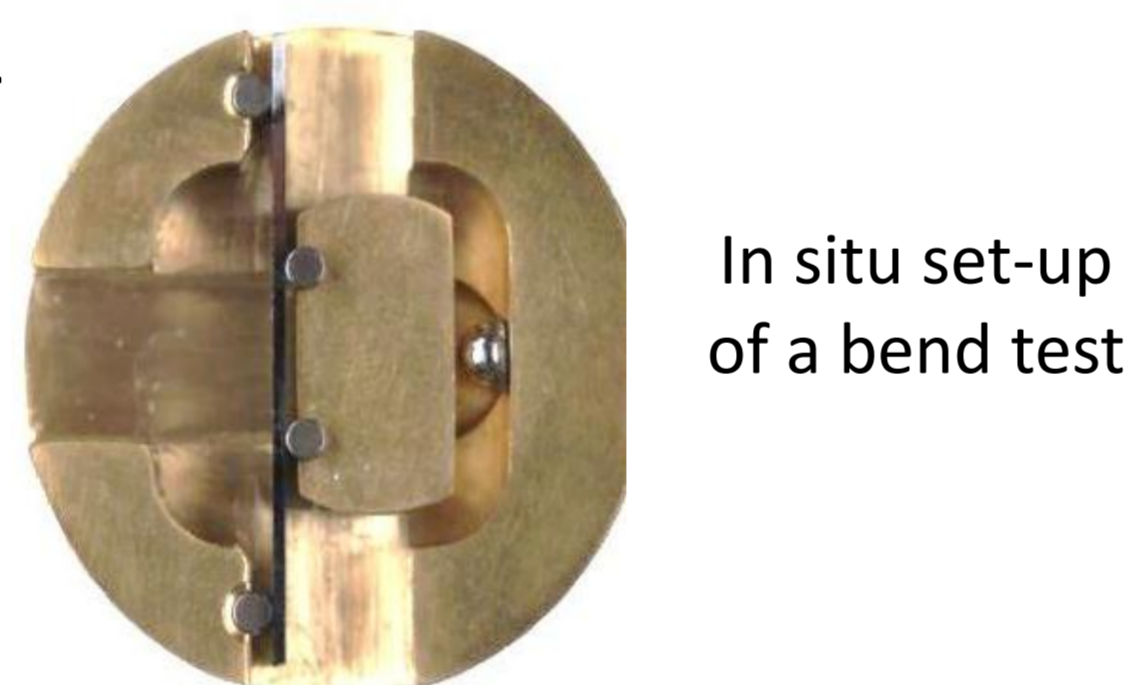
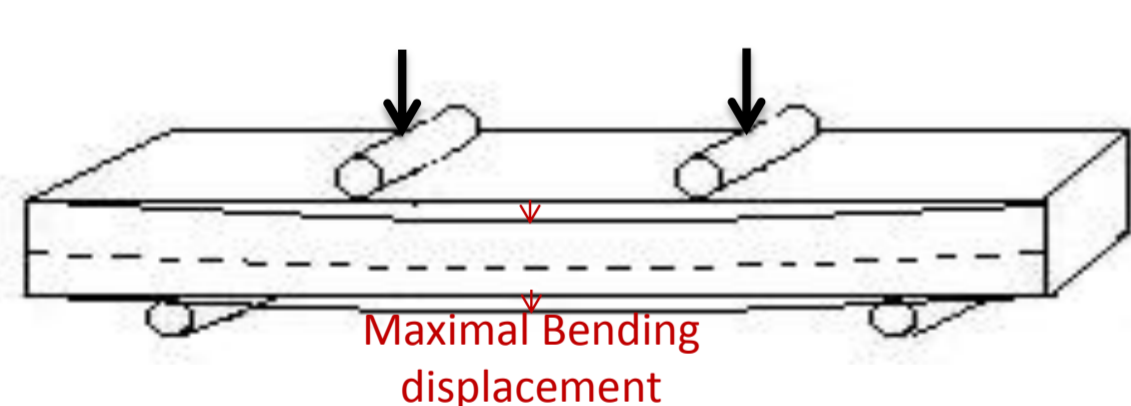
- The **SIMS** measurements are quantified by readjusting the Xe-132 intensities with quantitative **EPMA** measurements in areas where the total xenon inventory can be measured by both techniques, i.e. where Xe is present in dissolved form.

- Assessing bubble volumes by **SEM**
The **atomic volume of xenon** is determined and **appropriate equations of states** are used to calculate the pressure in the bubbles.

Validation of a simple case

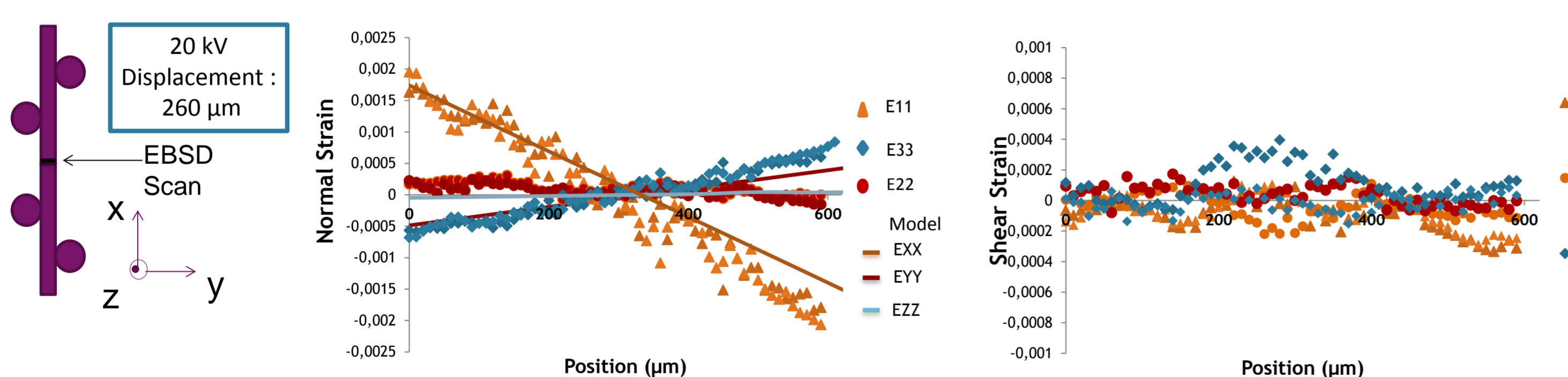
4-point bending test on a single-crystal silicon bar
Order of magnitude of expected deformation:

$10^{-4} - 10^{-3}$



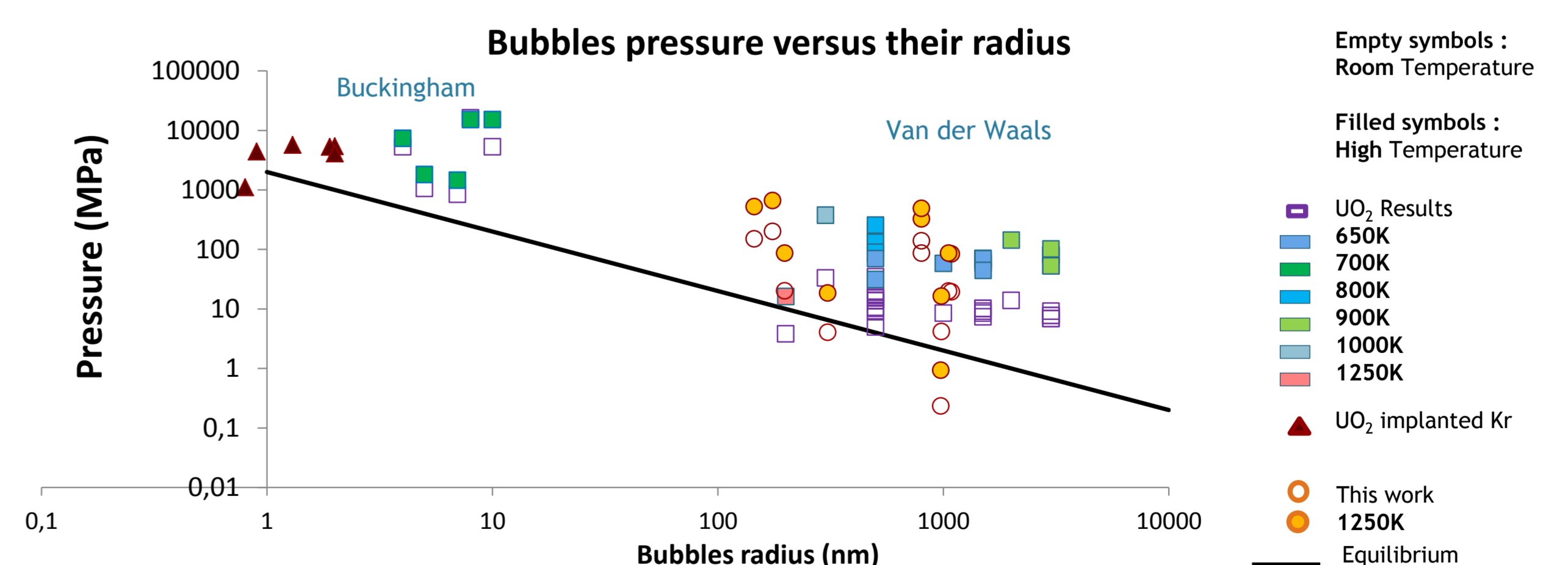
The study has reached the stage where the method is being validated with a simple case. The first deformation measurements by EBSD on an unstressed silicon bar revealed the importance of the surface state, the flatness and the position of the sample for cross-correlation.

Results



The center point of the line is considered as an unstrained reference. Every other EBSD pattern is compared to this reference. In order to validate the results, the four-point bending experiment was simulated by a FE model assuming anisotropic elastic 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 $2 \cdot 10^{-4}$. Special care is required to avoid any projection parameters variations, with a flat and parallel sample surface to the stage surface.

The pressure can be calculated on the basis of the Van der Waals equation of state at 300K. The resulting pressures can be compared with other experimental data (EDX and SIMS)⁴⁻⁵. These first results are in good agreement with the results from the literature.

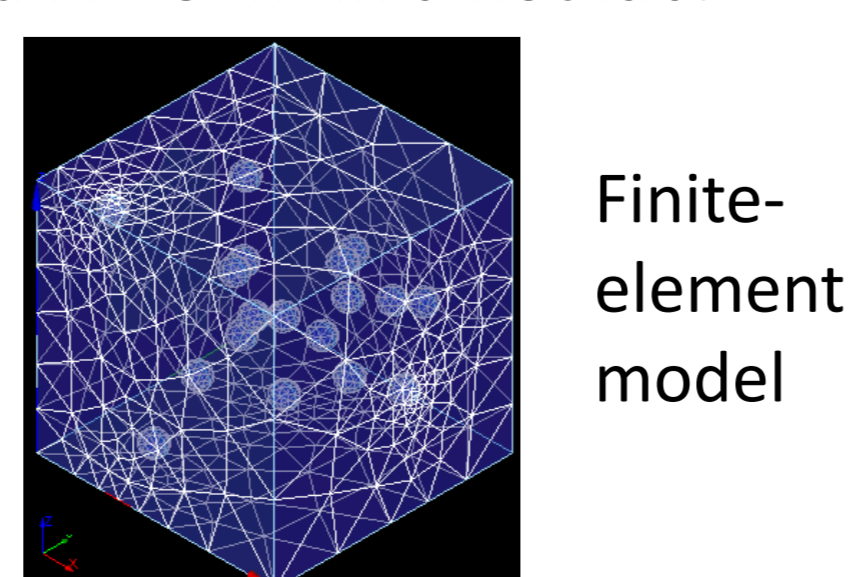


Future prospects

EBSD and Cross-Correlation method

- Application of the method on irradiated fuels thanks to a FIB-SEM in a hot lab.

- Application of a finite-element model to determine deformation caused by fission gas bubbles in the irradiated fuel and therefore their pressure.



SEM/SIMS/EPMA method

- This study will be continued with the aim of reducing the data acquisition time 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, peak intensity and the equation of state.
- The results will be compared to a more global method³.

1. L. Noirot, *Nuclear Engineering and Design*, 241(6), pp. 2099-2118, 2011 2. T.B. Britton, *Ultramicroscopy*, 111, pp. 1395-1404, 2011 3. J. Lamontagne, *Microchim Acta*, 155, pp. 183-187, 2006 4. K. Nogita, *Nuclear Instruments and Methods in Physics Research Section B*, 141(11), pp. 481-486, 1998 5. J. Noirot, *Journal of Nuclear Materials*, 372, pp. 318-339, 2008.

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