

Fission Gas Distribution and Behavior in the High Burn-up Structure

Ch. Hellwig¹, M.I. Horvath¹, P.R. Blair^{1,2}, R. Chawla^{2,3}, D. Günther⁴

¹Laboratory for Materials Behavior, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

²Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

³Laboratory for Reactor Physics and Systems Behavior, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

⁴Laboratory of Inorganic Chemistry, ETH Zurich, CH-8093 Zurich, Switzerland

ABSTRACT

Roughly 20% of all fission products in nuclear fuel pellets are gaseous, the most important are Kr- and mainly Xe-isotopes. They are solved in the fuel matrix or in nm-sized intra-granular bubbles, in μm -sized inter-granular pores or have been released into the plenum of the fuel rod. By reaching a burnup of 60-75 $\text{GWd}/_{\text{THM}}$ a so called High Burnup Structure (HBS) is formed in the rim of the fuel. In this region a depletion of the noble Fission Gases (FG) in the matrix and an enrichment of FG in the pores can be observed. The knowledge on FG distribution in high burn-up fuel is important to understand high burn-up fuel behavior under accident conditions like RIA and LOCA. However, a proper quantification of the FG in the HBS pores is difficult. With analytical methods routinely used for the characterization of solid samples the quantification of FG is impossible, since the FG in the pores close to the surface is already released during sample preparation. Furthermore, mechanistic models that reflect FG transport processes in the HBS and reliably predict the evolution of the remaining FG and its release are largely missing today. At the Paul Scherrer Institut (PSI) we investigated the HBS with respect to FG distribution and behavior experimentally (analytical methods) as well as fundamentally (modelling).

For the modelling approach we developed a gas release model that takes into account the growth of pores via the thermal equilibrium vacancy flux. For typical stereological parameters of the HBS in LWR UO_2 fuel a constant temperature sensitivity analysis has been conducted. We find that there exists a maximum porosity which is dependent on the geometrical characteristics of the system i.e. initial number density, initial pore size and surface-to-volume ratio of the technological open surface. In particular the surface-to-volume ratio significantly affects both the value and time to reach the maximum porosity. Furthermore we see that the absolute value of this maximum porosity is not dependent on parameters affecting the pore growth rate, i.e. temperature and stoichiometry. In spite of this invariance, the release of gas from the porosity is continuous and is affected by the pore growth rate, releasing $\sim 20\%$ of the initial gas inventory once the maximum porosity is achieved.

A first successful validation was done by applying this model to an annealing experiment with high burn-up fuel [1] to assess its predictions in comparison with experimental measurements. We find that a temperature averaged stoichiometry deviation of $\sim 3.5 \times 10^{-3}$ and a low value of the surface-to-volume ratio fits the experimental profile reasonably well. In the case of the vacancy diffusion coefficient we find that within a range of reasonable migration enthalpies the most optimal fit is 2.9 eV/atom. This agrees with the original analysis [1], but suggests that a major component of the release within this experiment is determined by the growth of the porosity rather than volume diffusion of the FG. A qualitative comparison of the out-of-pile and in-pile release with respect to the porosity indicates that for all burn-ups examined to date the maximum porosity has not been reached so far. The behavior prior to reaching the maximum porosity indicates that the release of fission gas from the porosity should be continuous rather than a threshold process. A simplified model derived from this modelling work was then implemented into a fuel performance code.

For the analytical characterization of high burn-up fuel a combination of a laser ablation system (LA) with an inductively coupled plasma mass spectrometer (ICP-MS) was applied. This method offers the advantages of high spatial resolution with laser spot sizes down to 10 μm and very low detection limits. The laser ablation system is custom-build to fit the needs of a shielded ablation environment at PSI. A suitable calibration technique for gases had to be developed in order to allow the quantification of Xe amounts in pores in the HBS. Among different possible calibration strategies, direct injection of a well-defined amount of Xe with a

gas syringe into the carrier gas system of the LA-ICP-MS turned out to be the most suitable and reliable method. The calibrations were performed at the beginning and at the end of a measurement cycle.

Measurements using the LA-ICP-MS method were performed on PWR fuel with a rod average burn-up of $105 \text{ GWd}/\text{t}_{\text{HM}}$. The sample was embedded in a steel sample holder and polished for preceding Scanning Electron Microscope (SEM) and Electron Probe Micro Analysis (EPMA). The SEM measurements were needed to determine the pore size distribution in dependence of the burn-up, resp. pellet radius. EPMA measurements delivered information on the remaining Xe in the matrix. Two ablation modi were applied using i) „single shot” method, where one to several pores were opened and analysed. The estimated pressures in the HBS-pores by the „single shot” method, by assuming 2 – 4 opened pores per LA-pulse (with 2 – 7 μm diameter), are in a range of 2 – 30 MPa, with 30 MPa being the approximate maximum without FG release. By using ii) the „single hole” method, a defined fuel volume was locally drilled out of the matrix. With the porosity determined by SEM in advance the volume of the removed porosity was calculated and the FG pressure in the pores was estimated. The results indicate a significant FG release from the HBS.

Fuel performance modelling with our extended HBS-model was then applied on the origin rod of this well characterized sample. The experimentally determined pressures and pore size distributions are compared with results from the model calculations. The agreement of pore size distribution and FG release was found to be satisfactory. Thus the model seems to adequately depict the main FG diffusion and release mechanism in the HBS. More comparisons will be done to further validate the model.

[1] J.P. Hiernaut, C. Ronchi, J. Nucl. Mater. 294 (2001) 39.