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Fission product release and microstructure changes of irradiated MOX fuel at high temperatures

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ABSTRACT

Samples of irradiated MOX fuel of 44.5 GWd/tHM mean burn-up were prepared by core drilling at three different radial positions of a fuel pellet. They were subsequently heated in a Knudsen effusion mass spectrometer up to complete vaporisation of the sample (~2600 K) and the release of fission gas (krypton and xenon) as well as helium was measured. Scanning electron microscopy was used in parallel to investigate the evolution of the microstructure of a sample heated under the same condition up to given key temperatures as determined from the gas release profiles. A clear initial difference for fission gas release and microstructure was observed as a function of the radial position of the samples and therefore of irradiation temperature. A good correlation between the microstructure evolution and the gas release peaks could be established as a function of the temperature of irradiation and (laboratory) heating.

1. Introduction

Plutonium recycling in mixed oxide fuel (MOX) occurs in around 45 PWR reactors worldwide and 20 other reactors are licenced for use of MOX (one up to 100%). Today most of these reactors accept from 25% to 50% of MOX fuel elements in the reactor core and advanced reactors like the EPR or ABWR are capable to operate with a complete loading of MOX. Initially the burn-up of the MOX fuel was lower than that of UO_2 fuel, i.e. three annual cycles versus four, but for economic reasons some utilities strive for parity between the two fuel types [1]. Because of the differences in composition and fabrication processes, the behaviour of MOX and UO_2 fuel differs from the point of view of physical properties and microstructure, and as a consequence also the irradiation behaviour is not identical. Extensive studies have already been performed to understand this, considering the influence of microstructure, burn-up, radial position and Pu-content [1–9].

The fission gas release from MOX fuel is a critical issue. As discussed by Cazalis and Georgenthum [10] the fission gas production in MOX fuel is slightly lower than in UO_2, but for similar burnup the concentration of fission gas (FG) at grain boundaries in MOX fuel produced with the most current fabrication technology (MIMAS) is significantly higher. This is due to the fact that fuel from this process is inhomogeneous and the local burnup in the Pu-rich agglomerates is relatively high. A higher fission gas release for MOX fuels from commercial reactor irradiation has been observed [10], but this has been attributed to the differences in power rating. In contrast, the helium production in MOX fuel is significantly higher than in UO_2 as a result of the alpha-decay of transuranium nuclides, both during irradiation and after discharge from the reactor.

In this paper we report the results of a study of the release of fission gas and helium from irradiated MOX samples that were taken from different radial positions in a fuel pellet, thus representing different irradiation temperatures in the range of 800 K (contact with cladding) to 1573 K (fuel core) and burnups. We also studied the microstructural changes in the samples as a function of annealing temperature. The results allow us to understand the fission gas release during accidental transients in reactor and better understand the observed behaviour of MIMAS MOX pellet during irradiation.

2. Fuel sample and experimental method

The experiments were performed on commercial PWR MOX fuel with an average burn-up of 44.5 GWd/tHM. The fuel investigated was manufactured by Siemens KWU using the OCOM process (similar to MIMAS) and was irradiated at a low power rating in the KWO reactor in Obrigheim in Germany during the period July 1986 till May 1990. The fuel has a microstructure characterized by plutonium rich agglomerates (PA) up to 200 μm in size randomly dispersed in a matrix of natural UO_2 (UM), and was extensively described in Walker et al. [11] who designated it as OCOM30. They also reported that a puncturing test showed that about 2.5% of the fission gas was release to the plenum.

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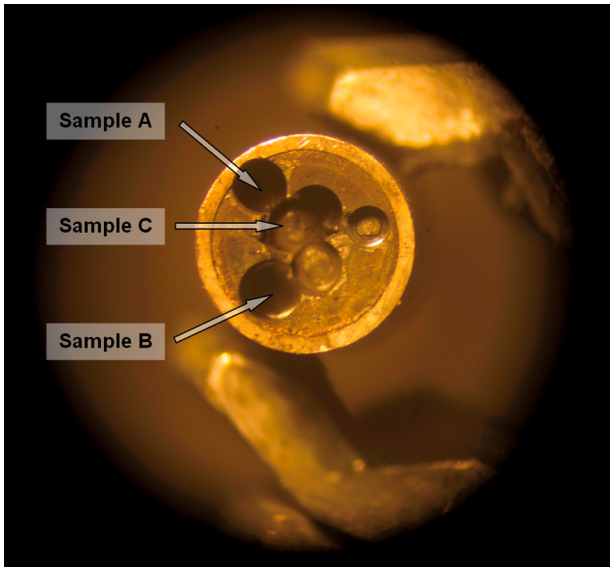

Fig. 1. Optical view of the fuel rod section showing the regions drilled to obtain the fuel samples.

Three samples were examined in this work; they were single fragment taken from parts extracted by core drilling from a fuel pellet (see Fig. 1):

- The region closest to the cladding ($0.58 < r/r_0 < 0.96$), designated as sample type A in Fig. 1. It represents the "cooler" part of the fuel pellet. The irradiation temperatures (T_{irrad}) in this range are from 854 to 1312 K (ΔT: 458 K).
- The intermediate radial zone of the pellet ($0.42 < r/r_0 < 0.81$), designated sample type B in Fig. 1, has a T_{irrad} ranging from 1068 to 1434 K (ΔT: 365 K).
- The central zone of the pellet ($0.003 < r/r_0 < 0.41$), designated sample type C in Fig. 1, which was close to the hottest part of the pellet, has a T_{irrad} ranging from 1442 to 1572 K (ΔT: 131 K).

The sample irradiation temperatures were determined from the calculated temperature profile (exponential function) knowing the core temperature of the fuel (1573 K) [11], the standard temperature for this type of fuel at the inner side of the cladding (800 K). The average burnup was calculated with TRANSURANUS code [12] and the PA burnup is the average burnup multiplied by the ratio of the fissile Pu concentration in PA over average fissile Pu concentration in fuel [11]. Calculated burnups correspond reasonably well with measurement of Walker et al. [11]. All those data are shown Fig. 2.

Fragments of 2–8 mg were chosen for the experiments. Since these specimens are small compared to the drilled sample size and were taken randomly, the precise radial position could not be determined, in particular the specimens of sample type, A and B could be from close radial locations.

Specimens from each drilled sample type were annealed up to complete vaporisation (~2600 K) at a speed of about 10 K min^{-1} in a Knudsen effusion mass spectrometer (KEMS) described previously [13,14]. In addition to helium and to the FGs all the species present in the vapour between 83 and 300 a.m.u. were measured during the heating. Additionally, the ^{85}Kr isotope was analysed in a cold trap by β and γ counting. The long-lived fission gas isotopes correspond to masses 131, 132, 134 and 136 for Xe and 83, 84, 85 and 86 for Kr. The absolute quantities of gas released from specimens of sample types A and B were also determined using the in-house built Q-GAMES (Quantitative gas measurement system), described in detail in [15].

For each of the samples, fragments were also annealed and measured in the KEMS up to specific temperatures corresponding to different stages of the FGs or He release. These fragments were subsequently analysed by Scanning Electron Microscopy (SEM, Philips XL40) [16] in order to investigate the relationship between structural changes, burn-up, irradiation temperature and fission products release. SEM observations were also done on the samples before the KEMS experiments and the fracture surface appearance of the samples is shown in Fig. 3, revealing the presence of the high burnup structure (HBS) in the Pu-rich agglomerates.

A summary of the 12 samples analysed by KEMS, SEM and Q-GAMES is given in Table 1.

3. Fission gas and helium release

3.1. Sample type A: closest to the pellet edge

The evolution of the Xe release is shown in Fig. 4. It exhibits parallel curves for all the isotopes, except for ^{131}Xe at low

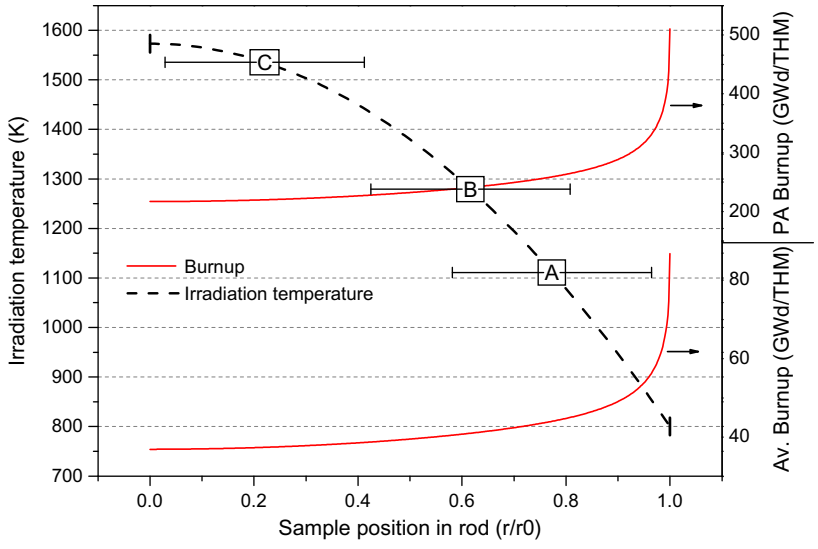

Fig. 2. Radial position of the samples with the corresponding calculated irradiation temperature (left axis), average burnup (lower right axis) and PA burnup (upper right axis).