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Analysis of Fission Gas Release under Post Irradiation Annealing Conditions

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Abstract

Fission gas release under post irradiation annealing condition is analyzed using an empirical fission gas release model for transient conditions. Based on the amount of fission gas retained in the matrix and grain boundary, burst release of fission gas during temperature increase, which is considered to take place via grain boundary separation due to microcracking, is described. Since this mechanism requires certain threshold thermal stress to cause pellet cracking, an experimentally measured threshold temperature is used to activate gas release via microcracking. In addition, diffusional release during holding period at high temperature is considered. The verification of the model has been performed using the data obtained from thermal annealing experiments of BWR fuel pellets base-irradiated to 6~28 MWd/kgU in a commercial reactor. Comparison between the measured and calculated results showed that, to fit the measured data, not only gas atoms retained in the grain boundaries but also some of those in matrices contributed to the burst release during temperature increase. This implies that the discrepancy comes from the difference in diffusion coefficients between the one used in the present calculation and the one in the specimen. The gas inventory in the grain boundary available for release depends on diffusion coefficient. In addition, it was found that even just after the burst release, diffusional release occurs due to the very rapid diffusion of gas atoms from the matrix to the grain boundary.

1. Introduction

Fission gas release in a fuel rod causes an increase of fuel temperature and rod internal pressure. They strongly affect the fuel rod integrity during steady-state and transient operating conditions. Generally fission gas release is higher during transient conditions than steady-state conditions due to higher power and temperature leading to rapid diffusion and rapid change in power and temperature causing grain boundary to separate (pellet microcracking) via thermal stress. Therefore, the accurate prediction of gas release during transient conditions is very important in terms of fuel temperature and rod internal pressure that determines the overall performance of fuel rod. In addition, since the fission gas release during transient conditions depends on the amount of gas retained on the grain boundary, gas release in high burnup fuel whose grain boundaries have large amount of gas inventory would be significant resulting in aggravating thermal performance of fuel. Therefore, the accurate prediction of fission gas on the grain boundary is a prerequisite for the analysis of transient fission gas release.

In this paper an empirical model, which was originally developed to analyze the fission gas release during power change conditions [1], is applied to post irradiation annealing (temperate change) conditions. The model is analyzed using the annealing data obtained from BWR fuel pellets based irradiated to 6~28 MWd/kgU in a commercial reactor [2].

2. Post irradiation annealing experiment

The specimens used in the experiment that is going to be analyzed in this paper were taken from UO_2 fuel pellets irradiated in a commercial BWR reactor for 1 to 4 cycles at maximum linear heat generation rates between 300 and 370 W/cm. The fractional gas releases measured by pin puncturing test after base irradiation were 0.2, 0.8, 21.0 and 21.0 % for each fuel rod. The burnups for each rod were 6, 16, 23 and 28 MWd/kgU, respectively. The basic manufacturing parameters for these fuel rods are given in Table 1.

Table 1. Manufacturing parameters for the base irradiated fuel rods

Manufacturing parameter	Value
Pellet height (mm)	11.0
Pellet diameter (mm)	10.31
Grain size (μm)	9.0
Pellet stack length (mm)	3660.0
Enrichment (%)	1.45
Density	95 % T.D.
Clad outer diameter (mm)	12.27
Clad inner diameter (mm)	10.55
He fill gas pressure (bar)	30.0

Pieces of a fuel pellet with the rim were mounted in resin and sliced into slabs of about 1 mm thickness. Then about 1.5 mm diameter specimens were ultrasonically punched from these slabs. The location of the specimens was roughly between the fuel rim and middle region. Their weights were 30-40 mg. The specimens were contained in a Mo capsule and heated by a high frequency induction coil. Temperature was heated at a rate of 1.5 ^oC/s from 0 to 1800 ^oC and annealed at this temperature for 340 minutes. Using the sweep gas of high purity He/2%H_2 mixed gas at a flow rate of 60 cm^3/min, the released 85Kr was measured by an ionization chamber and a Ge detector [2].

3. Description of the analysis method

As in the case of rapid power increase and/or decrease, pellet microcracking and subsequent fission gas release during the stage of rapid temperature increase is found experimentally in the post irradiation annealing tests. The critical temperature for the onset of burst release at high temperature is found to be a function of fuel burnup. This burnup dependence of burst release is attributed to the increases of fission gas inventory in the grain boundaries and the density of the grain boundary bubbles at higher burnups. Namely higher internal pressures in the bubbles lead to fast growth rates, and the higher densities of the bubbles increase the probability of the bubble interlinkage, which would the cause for the critical temperature for burst release with burnup. In addition, the occurrence of microcracks stemmed from the degradation of mechanical strength of the grain boundary would enhance this trend. In this experiment, the critical temperature for the onset of burst release was 1800^{o}C for the 1 cycle specimen, 1600^{o}C for the 2 cycle specimen and 1500^{o}C for the 3 and 4 cycle specimens [2].

Based on these experimental observations, fission gas release during this post irradiation annealing experiment was simulated as follows:

(1) Fission gas release during base irradiation was calculated using the model for steady-state operation [3].
(2) The gas disposition between the matrix and grain boundary at the end of base irradiation was stored and then used as the initial conditions for post irradiation annealing.
(3) Fission gas release during post irradiation annealing was calculated using the assumption that once the critical temperature was reached all the fission gas retained in the grain boundaries are released immediately to free space. In addition it was assumed that some amount of gas retained in the matrix is released to the free volume. This assumption, which was also used for the analysis of fission gas release during power transient conditions, would be justified if the thermal stress caused by rapid temperature increase is large enough to induce intergranular cracking.
(4) During the annealing period, microcracks in the grain boundaries are assumed to be healed due to rapid diffusion of atoms and then certain amount of gas atoms are needed in the grain face and grain edge for the release path to form. Depending on the time elapsed after the immediate purging of grain boundary inventory, the amount that must be reached in the grain face and grain edge before the release path to form changes.

4. Analysis of experimental data

For the analysis of gas release during base irradiation, the resolution probability of gas atoms in the grain face bubbles being reinjected into the grain by fast neutron b_f, the maximum (saturated) fraction of grain face covered by gas bubbles before gas atoms leak to the grain edge f_b^{max}, the maximum gas bubble swelling at the grain edge at which gas bubbles interlink completely $(\Delta V/V)_e^{max}$, were taken to be 8x10^{-6}, 0.25 and 0.05, respectively [3].

For the specimen of 6 MWd/kgU that were obtained from fuel pellets of 1 cycle base irradiation, the calculated fractional gas release after base irradiation was 0.11%, while the measured one was 0.2%. During the annealing, it was assumed that microcracks open immediately when temperature reaches 1800^{o}C and these microcracks are healed completely after 3.5 hr of annealing at 1800^{o}C. This assumption was simulated as follows in the gas release model: when the microcracks open, both f_b^{max} and $(\Delta V/V)_e^{max}$ is given 0.01, implying that almost all the gas atoms in the grain face and grain edge, that is, in the grain boundary are released to the free space. After 3.5 hr annealing at 1800^{o}C, f_b^{max} and $(\Delta V/V)_e^{max}$ returns to its original value of 0.25 and 0.05, respectively, suggesting that microcracks are healed completely. During this time interval, f_b^{max} and $(\Delta V/V)_e^{max}$ are assumed to increase linearly. Fig.1 shows that the assumption introduced here predicts well the gas release during the annealing experiment.

For the specimen of 16 MWd/kgU that were taken from fuel pellets of 2 cycle base irradiation, the calculated and measured gas release were 0.8% and 0.5%, respectively. The same method was used as for the 6 MWd/kgU specimen except that microcrack opens at 1600^{o}C and crack heals completely at the end of annealing. As can be seen in Fig.2, the model predicts that when the specimen temperature reaches 1600^{o}C almost all the gas atoms in the grain boundary are released and then until gas atoms arrive at the grain boundary by diffusion, no additional release occurs. After around 100 minutes at 1800^{o}C, diffusional release begins to occur. On the other hand, the measured gas release increases abruptly during temperature rise due to cracking and then slowly increases by diffusion from the matrix. This implies that this discrepancy comes from the difference in diffusion coefficients between the one used in the present calculation and the one in the specimen. Depending on the diffusion coefficient, gas inventory in the grain boundary available for release varies and this determines the transient gas release behavior.

For specimens of 23 and 28 MWd/kgU, gas release after base irradiation were both 21.0%, respectively, which are unusually high for this burnup and therefore to fit the calculated gas release with this value, b_f was reduced from 8x10^{-6} to 1x10^{-6}, while using the same valuse for f_b^{max} and $(\Delta V/V)_e^{max}$ as in 6 and 16 MWd/kgU specimens. These model parameters yielded 18.5 and 24.8% release for base irradiation. In analyzing annealing experiment, another assumption that 3% of the gas retained in the matrix would be released due to microcracking was added to get more burst release. Even with this assumption, Fig.3 and 4 still show the same trend that the calculation underpredicts the burst release. However, the diffusional release during 1800^{o}C is well predicted. This fact also supports the idea that the gas inventory retained at the grain boundary were likely to be underpredicted for base irradiation. In addition, it was found that even just after the burst release, diffusional release occurs due to the very rapid diffusion of gas atoms from the matrix to the grain boundary.

5. Conclusion

Fission gas release under post irradiation annealing conditions was analyzed using an empirical fission gas release model for transient conditions. The verification of the model has been performed using the data obtained from thermal annealing experiments of BWR fuel pellets base irradiated to 6~28 MWd/kgU in a commercial reactor. Comparison between the measured and calculated results showed that not only the gas atoms retained in the grain boundaries but also some of those in matrices contributed to the burst release during temperature increase. This implies that the discrepancy originated from the difference in diffusion coefficients between the one used in the present model and the one in the specimen because the gas inventory in the grain boundary available for release, which determines the burst release behavior, depends on the diffusion coefficient. In addition, it was found that even just after the burst release, diffusional release occurred due to the very rapid diffusion of gas atoms from the matrix to the grain boundary.

Acknowledgements

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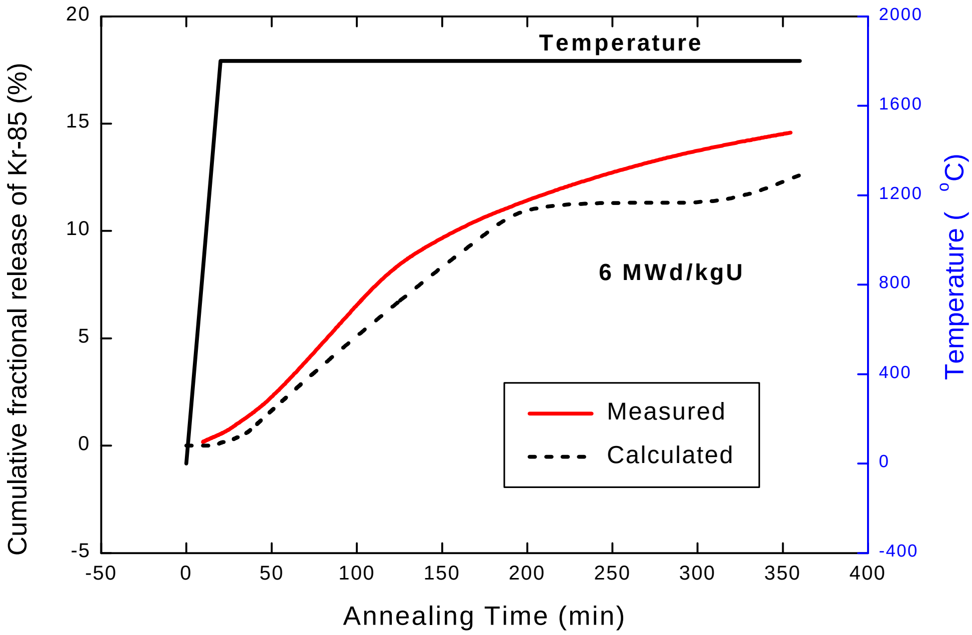

Fig.1. Fission gas release during annealing for 6 MWd/kgU specimen.

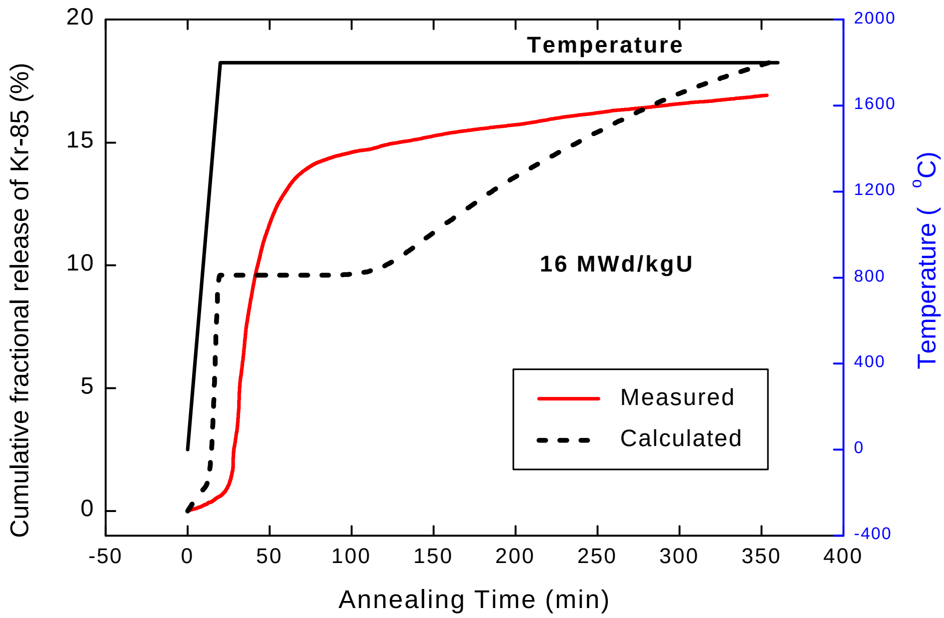

Fig.2. Fission gas release during annealing for 16 MWd/kgU specimen.

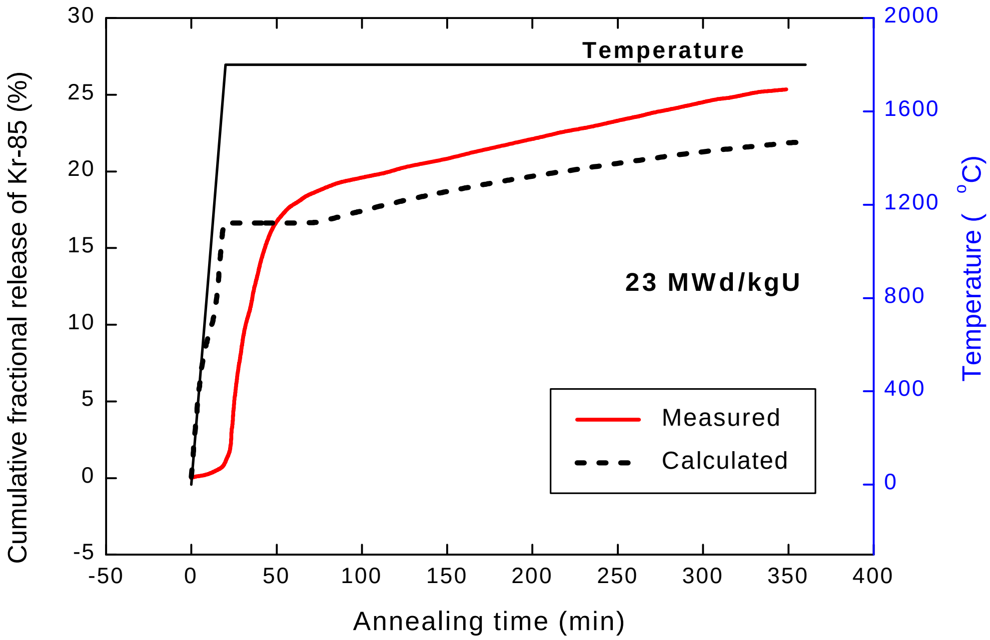

Fig.3. Fission gas release during annealing for 23 MWd/kgU specimen.

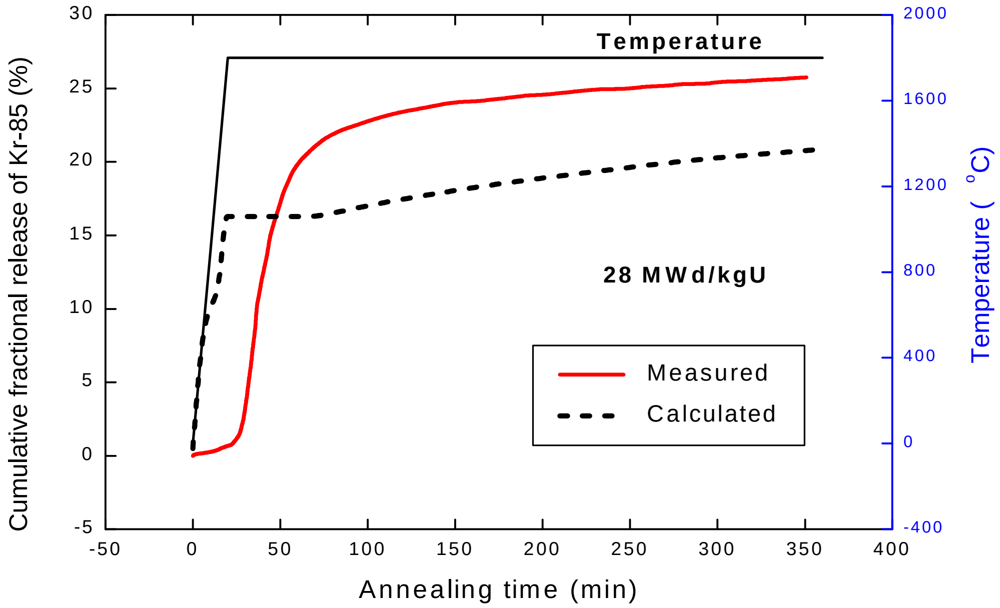

Fig.4. Fission gas release during annealing for 28 MWd/kgU specimen.