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Original Article

Modeling of Pore Coarsening in the Rim Region of High Burn-up UO_2 Fuel

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ABSTRACT

An understanding of the coarsening process of the large fission gas pores in the high burn-up structure (HBS) of irradiated UO_2 fuel is very necessary for analyzing the safety and reliability of fuel rods in a reactor. A numerical model for the description of pore coarsening in the HBS based on the Ostwald ripening mechanism, which has successfully explained the coarsening process of precipitates in solids is developed. In this model, the fission gas atoms are treated as the special precipitates in the irradiated UO_2 fuel matrix. The calculated results indicate that the significant pore coarsening and mean pore density decrease in the HBS occur upon surpassing a local burn-up of 100 GWd/tM. The capability of this model is successfully validated against irradiation experiments of UO_2 fuel, in which the average pore radius, pore density, and porosity are directly measured as functions of local burn-up. Comparisons with experimental data show that, when the local burn-up exceeds 100 GWd/tM, the calculated results agree well with the measured data.

1. Introduction

Sintered UO_2 pellets in rods are the standard nuclear fuel in light water reactors [1–4]. The discharge burn-up of fuel assemblies has been increasing throughout the past few decades in order to improve the economics of power plants. The local burn-up in the rim region of UO_2 pellets will exceed 100 GWd/tU when the average pellet burn-up is about 40 GWd/tU due to the configuration of the neutron energy spectrum in a light water reactor [5,6]. During in-pile irradiation, starting at a local burn-up around 50 GWd/tU and a temperature below 1,100°C, the UO_2 fuel matrix will undergo a restructuring process which results in the appearance of a new fuel structure with the typical characterization of small submicron grains and coarsened fission gas pores. This new structure observed in the rim region of the UO_2 pellets is called high burn-up structure (HBS) or rim structure. Some other researchers [7,8] named this morphology cauliflower structure due to its similar appearance to the biological morphology of the cauliflower.

The occurrence of the HBS results in the fission gas pores coarsening and it will eventually accelerate the fuel swelling; these issues have raised some concern over the reliability and safety of extended fuel operation life in the reactor. Since this

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HBS was first observed in the late 1950s [6], numerous experimental as well as the theoretical studies have been performed in order to investigate this new structure in detail and understand the HBS formation mechanisms. However, a full scientific description of the HBS formation mechanisms is still missing and there are still very few mechanistic models that can be used in the fuel performance analysis code, although several international collaboration programs, such as High Burn-up Effects Program [9] and High Burn-up Rim Project [10], have been established since the early 1980s and several theoretical models have been presented [11–23].

Fortunately, the rim structure of PWR-fuels with local burn-ups of 240 GWd/tM has been studied in recent years and confirms that the coarsening process of the average pore size is close to Ostwald ripening (OR) [24] and coincides with the other studies in the literature [25–27]. Hence, in this paper, a theoretical analysis is presented to investigate the evolution of fission gas pores in the rim region of high burn-up UO_2 fuel pellets based on the OR mechanism, which was originally developed to explain the coarsening of precipitates in solids [28–31]. To describe the evolution of fission gas pores in the HBS of UO_2 fuel pellets, a numerical model of fission gas pore coarsening is developed, which includes the coarsening process of pores, and calculations of the pore radius and density in the fuel matrix during irradiation of UO_2 fuel pellets. The comparisons of theoretical predictions of the mean pore radius, mean pore density, and the porosity with the experimentally measured results is also discussed.

This paper is organized as follows: the next section provides a mechanistic and engineering model for the evolution of fission gas pores in the rim region of high burn-up UO_2 fuel pellets. Then, the calculated results of the mean pore radius, mean pore density, and porosity in the fuel as a function of burn-up are presented and compared to the available literature data. Finally, the main findings of this study are summarized in the conclusions.

2. Physical processes and model

It is well know that the noble gas Xe is originally generated in the UO_2 fuel matrix in atomic form. All atoms formed by fission processes have a strong tendency to diffuse and precipitate into bubbles due to their low solubility. It is assumed that some processes of the pore coarsening in the UO_2 fuel matrix are the same as or similar to the coarsening of precipitates in solids, which are described as follows [28–31]:

(1) The fission gas Xe atoms formed by the fission of uranium isotopes have a strong tendency to diffuse into bubbles due to their very low solubility and this process can decrease the total free energy of the system, which is the same as the second phase particles in solids.
(2) The decreasing of the interfacial energy per unit volume is the driving force for the coarsening of both the fission gas pores in the UO_2 fuel matrix and the second phase particles in solids.
(3) According to the Gibbs–Thomson effect, the growth of the larger second phase particles in solids is at the expense of the smaller ones. Re-solution of fission gas atoms from bubbles and/or pores due to the fission fragment will occur in UO_2 fuel during irradiation, and will induce the shrinkage of the small bubbles but has no obvious effect on the larger bubbles.

The basic premise of the theory presented in this work is that the pore coarsening is driven by the diffusion of fission gas atoms. Based on the similar processes and mechanisms of the pore coarsening and precipitates in solids described above, there is no difficulty in applying OR to develop a mechanistic model for pore coarsening in the rim region of high burn-up UO_2 fuel.

The schematic diagram of the fuel system which contains α phase (Xe pore) and β phase (UO_2 fuel matrix) is shown in Fig. 1. The α phase is composed of Xe atoms (A component). The β phase is composed of Xe atoms and pure UO_2 (B component). The diphase equilibrium condition under a constant temperature and constant pressure requires:

$$dG = 0 \tag{1}$$

More specifically:

$$dG_g + dG_s + dE_{solid} + \sum_i \mu_i dn_i = 0, \tag{2}$$

where the first term on the left hand side of Eq. (2) represents the change of the Gibbs free energy of fission gas in the pore; the second term represents the change of the Gibbs free energy of the fuel system resulting from the change of the interfacial area between the α phase and β phase; and the third term represents the change of the strain energy of the UO_2 fuel matrix during the process of pore coarsening.

The internal pressure in the coarsened rim bubbles of UO_2 pellets is assumed to be much higher than the equilibrium pressure of $P_{eq} = 2\gamma/r$ (γ: surface energy), from the fact that a very high density of dislocations has been observed close to the surface of coarsened rim bubbles with radii between 100 nm and 300 nm in the boiling water reactor pellet peripheral region [32]. These dislocations are generated by a dislocation punching mechanism, and the internal pressure in bubbles is given by $P = \mu b/r + P_{eq}$ (μ is the shear modulus and b

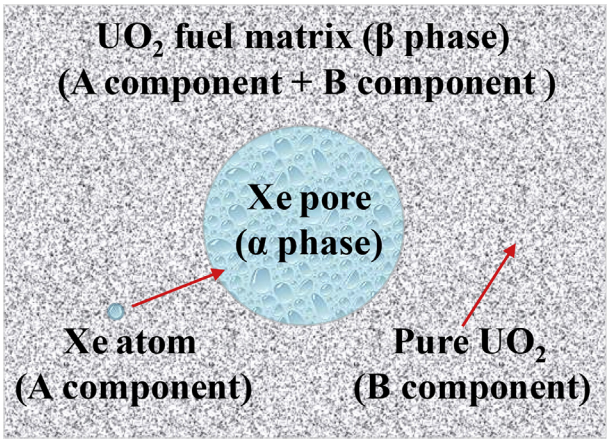

Fig. 1 – Schematic diagram to illustrate the UO_2 fuel system.