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Effect of fission gas diffusivity of dopant in Cr_2O_3 doped UO_2 pellet

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1. Introduction

Currently, nuclear fuel development aimed at improving the safety of reactors is being carried out worldwide. Reducing the fission gas release and pellet-cladding-interaction can strengthen the performance and the safety of nuclear fuel. This can be achieved by using UO_2 pellets with large grain size, as the enlargement of grain size is expected to not only improve the ability to retain fission gases but also enhance fuel plasticity at high temperatures during transient operation.

The method of mixing additives is one of the ways to improve grain size. Framatome (formerly known as AREVA) developed advanced fuel with enlarged grain size by doping UO_2 with Cr_2O_3[1]. In the fission gas release model, grain size is a key model parameter that increases intra-granular diffusion length, thereby reducing the rate of arriving at the grain boundaries. In addition, the diffusivity of gas atoms is another key parameter that affects the rate of gas diffusion to grain boundaries.

The research results of Killeen[2], Kashibe, and Une[3] indicated that Cr-doped UO_2 can exhibit higher fission gas diffusion than undoped UO_2. Additionally, Cooper demonstrated that additives can improve fission gas diffusion using an atomistic model.

This paper investigates the effect of additives on the thermal diffusivity of fission gases in Cr2O3-doped UO2 pellets. In addition, the fission gas release behavior was evaluated considering the effect of thermal diffusivity due to additives.

2. Methods and Results

2.1 Effect of fission gas diffusion

Fission gas diffusion in irradiated UO_2 fuels is described by the three different terms that characterize distinct process steps that apply to different temperature ranges[4], which in general form can be written as :

$$D = D1 + D2 + D3 \quad (1)$$

The term D1 represent the intrinsic diffusion at high temperature(T > 1650 K). The term D2 represent irradiation-enhanced diffusion at moderate high temperature (1300 K < T < 1650 K). the fission gas diffusion is enhanced by the presence of vacancies concentration. The term D3 represent athermal contribution at low temperature (1300 K < T).

Figure 1 shows the effect of the Cr dopant on the thermal diffusivity of UO_2 pellet. Killeen[2] provides a one each of the fission gas diffusion results of doped and undoped UO_2. Kashibe and Une[3] measured fission gas diffusivity for doped and undoped UO_2 between 1472 K and 1878 K. The diffusivity above 1525 K was enhanced in doped UO_2 compared to undoped UO_2, while diffusivity was suppressed below 1525 K. These results indicate that there is significant variability for D1 diffusivity. This is likely caused by different oxygen potentials and deviations from stoichiometry in $UO_{2\pm x}$, which strongly influences the concentration of the defects responsible for diffusion.

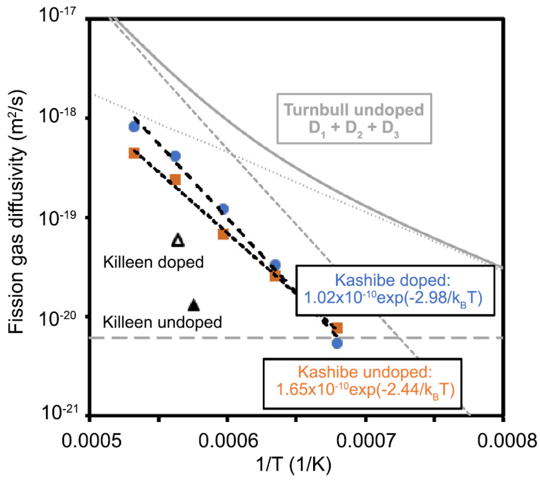

Fig. 1. The out-of.-pile fission gas diffusivity data from Killeen and Kashibe for Cr doped (blue) and undoped UO_2(orange). The gray lines is fission gas diffusivity model for undoped UO_2 from turnbull model[5].

Cooper[5] suggested that the addition of Cr_2O_3 to UO_2 could change the oxygen potential and therefore the fission gas diffusivity of doped UO_2 compared to undoped UO_2. More specifically, the possibility of defining the oxygen potential of doped UO_2 with Cr-Cr_2O_3 two-phase equilibrium under a wide range of reaction conditions was tested through thermodynamic investigation. To provide an accurate prediction of diffusion(D1) through the U vacancy assisted transport of fission gas, empirical potential entropy calculation was performed in conjunction with density functional theory(DFT) calculations of the density function of enthalpy. To examine the influence of these on the fission gas diffusivity, two cases are tested: one where minimal alterations are made to the underlying DFT data (Case A), and another where the possibility that

DFT overestimates the data is tested (Case B). Additionally, a cluster dynamics model was formulated in terms of the system's free energy using DFT data to consider the role of defect production in diffusion(D2). These results showed that intra-granular diffusion was enhanced through doping in the Cr-Cr_2O_3 two-phase oxygen potential.

Cooper[5] developed an enhanced diffusivity model by the addition of Cr_2O_3 to UO_2 as shown in equation (2) for application to the fuel performance code. The parameters of the model for Cr_2O_3 doped UO_2 are summarized in Table 1. Equation (2) is given by:

$$D^{doped} = \exp\left(-\frac{\Delta H_1}{k_B}\left[\frac{1}{T}-\frac{1}{T_1}\right]\right)D_1^{undoped} + \exp\left(-\frac{\Delta H_2}{k_B}\left[\frac{1}{T}-\frac{1}{T_2}\right]\right)D_2^{undoped} + D_3^{undoped}$$

-- (2)

Where T_1, T_2, $\triangle H_1$ and $\triangle H_1$ are reported in Table 1 for Case A and Case B

Table I. Parameters for enhanced diffusivity models derived for Cr doped UO_2

Parameter	Case A	Case B
T1 = T1 (K)	1773	1773
$\triangle H_1$ (eV)	0.3198	0.3282
$\triangle H_2$ (eV)	-0.3345	-0.6998

Figure 2 shows the effect of dopant on the fission gas diffusivity of undoped and doped UO_2 by applying equation (2). The diffusivity of doped UO_2 was improved by dopant in the temperature ranges corresponding to D1 and D2. Additionally, Case B showed greater improvement in the D2 range compared to Case A.

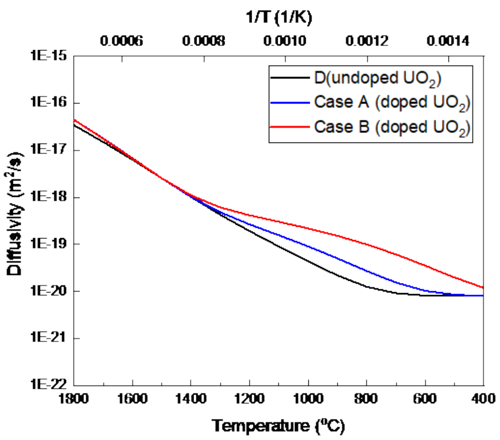

Fig. 2. Fission gas diffusivity in Cr_2O_3 doped UO_2 calculated by applying equation (2) to fission gas diffusion model

2.2 Effect of fission gas release

The Steady-State UO_2 FGR assessment case used in FRAPCON-4.0[6] was used to examine that the FGR effect of dopant prior to application to the in-pile test result (IFA-716) of Cr_2O_3 doped UO_2. The enhanced diffusivity model of equation (2) was applied to the fission gas model of FRAPCON-4.0 in three models (undoped, Case A, Case B).

Figure 3 shows the FGR behavior as function of burnup according to the burnup of the 332 rod irradiated in the BR-3 reactor. Grain size was set to 5.5 μm(standard) and 35 μm to observe the effect of FGR by grain size. As the grain size increased from 5.5 μm to 35 μm, the amount of FGR decreased because the intra-granular diffusion length increased and the speed at which it reached the grain boundary decreased. When the grain size was 35 μm, the amount of FGR increased in Case A and Case B considering the effect of dopant. Consequently, it is demonstrated that total amount of FGR in doped pellet during irradiation can be increased because the fission gas diffusivity owing to the dopant is affected.

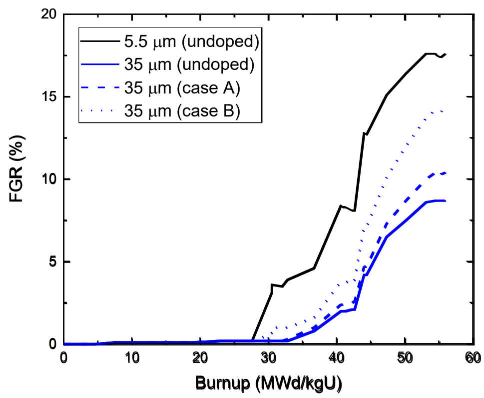

Fig. 3. FGR as a function of burnup for 332 test

3. Summary

The doping of UO_2 with Cr_2O_3 is designed to increase grain size during fabrication. One expected benefit of large grain is to reduce fission gas release. Adding Cr_2O_3 to UO_2 may also alter the oxygen potential, which could result in a change in diffusivity for doped UO_2 compared to undoped UO_2. Therefore, in order to predict FGR of Cr_2O_3 doped UO_2, it should be considered with the effect of the large grain and the enhanced diffusivity model, which act to suppress and enhance FGR, respectively. For further study, FGR behavior of experiment (IFA-716) will be studied with the proposed model. In the experiment (IFA-716), Cr_2O_3 doped pellet was irradiated and the amount of FGR was measured as the higher amount of FGR than normal UO_2.

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