Avaliação da liberação de gás de fissão para UO_2 dopado por Cr_2O_3

(Assessment of fission gas release for Cr_2O_3-doped UO_2)

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Abstract

Uranium dioxide has been the most common ceramic fuel used to generate electric power in the last sixty years. The lower addition of chromic oxide (Cr_2O_3) shows the benefits of large grain size. Using (Cr_2O_3-Al_2O_3)-doped UO_2 comprises improved mechanical response and fission gas retention properties. The addition of Cr_2O_3 to UO_2 slightly affected its thermal properties as proposed for pressurized water reactors and boiling water reactor designs. Advanced doped pellet technology (ADOPT) can improve fuel cycle economics and accident tolerance. Today, doped fuel pellets exhibit several favorable characteristics: large grain size, increased density, minor fission gas release (FGR), and reduced pellet cladding interactions. In simulation are used FRAPCON to simulate doped fuel and ferritic alloy as cladding. Most of the material properties of the Cr_2O_3-doped UO_2 were identical to those of the classic UO_2.

Keywords: Cr_2O_3-UO_2, Fission gas release, large grain size, FRAPCON

INTRODUCTION

Over the years, safety margins of operational criteria have faced impact because of the gases generated by fission, such as xenon (Xe) and krypton (Kr), partly released into the pellet-free volume [1]. Thus, Fission Products (FP) promote fuel swelling and decrease pellet thermal conductivity coupled with reduced mechanical stiffness and speed up fuel cracking and fracture. This work shows the fuel rod behavior and computational calculations for large-grained (Cr_2O_3-Al_2O_3)-doped UO_2 fuel based on the FRAPCON fuel performance code [2]. Therefore, it observed discrepancies in the calculated fuel temperature after fuel-cladding gap closure, into the gap fission gases released former mixtures between the noble Xe, Kr, and He.

Sintering additives tested as dopants can promote uranium dioxide grain growth using corundum oxides, such as Al_2O_3, Ti_2O_3, V_2O_3, and Cr_2O_3. Thus, the significant grain size could achieve reduced fission products, reduced fuel swelling, and higher burnup than the standard UO_2 fuel. Table I shows the thermal properties of uranium dioxide and metallic oxides used as dopants.

Table I: Thermal properties of UO_2 fuel and dopants proposed as Cr_2O_3 and Al_2O_3

Thermal Properties	**UO_2**	**Cr_2O_3**	**Al_2O_3**	**SiO_2**	**Ti_2O_3**	**ZrB_2**
Melting point (°C)	2850	2044	2435	1710	2103	3245
Density (g/cm^3)	10.96	4.907	3.950	2.17	4.49	6.08
Thermal conductivity (W/m-)	8.68	5.17	31	1.3	4.8	62
Thermal expansion ($10^{-6}/K$)	9.76	6.20	8.10	0.55	8.4	6.66
Heat capacity (J/kg-K)	235	822	880	680	683	425

Advanced doped pellet technology knowledge as ADOPT® fuel became part of the EnCore® Fuel project sponsored by Westinghouse and developed the PROtect program, covering all safety perspectives for light-water reactors (LWRs) [3]. Other fuel suppliers have developed similar concepts using Chromium-coated fuel rods and Cr_2O_3–doped fuel pellets proposed by Framatome for Enhanced Accidnt Tolerant Fuel (EATF) plan [4].

While Westinghouse employed minor combined additions of Cr_2O_3–Al_2O_3, the Framatome has developed similar concepts using Chromium-coated fuel. In contrast, Westinghouse proposes a Cr–coated ZIRLO, with a thickness of (20–30) µm, whereas Framatome suggests Cr-coated M5, with a thickness of (8–22) µm [5]. Doped fuel shows a lower FGR, especially during transients, and enhanced pellet creep with benefits in operational maneuvers. Doped fuels can operate in PWRs and Boiling Water Reactors (BWRs). There is a noticeable improvement in densification and diffusion through the additives during sintering. Standard UO_2 doped fuels show a higher density of about 0.5%, shorter hold sintering time, and larger grains of around five times that of undoped fuel. Additionally, occurred experiment includes aluminum and manganese oxides. In comparison, the Cr_2O_3-doped UO_2 shows a lower fission gas release of 50% than standard UO_2 pellets at elevated temperatures.

Since 2015, the Global Nuclear Fuel (GNF), lead of BWR technology, submitted a technical license to incorporate aluminum silicate (SiO_2-Al_2O_3) as a dopant into the coating UO_2 grain boundaries, with concentrations of 2500 ppm. GNF plans to test the ARMOR-coated cladding based on iron-chromium-aluminum (known as IronCladding) to improve fuel reliability and operational flexibility.

In the synthesis process employed for UO_2, the routine ceramic path prepares pellets starting with powder compact, producing the green body, and sintering at an elevated temperature of 1500 °C to 1700 °C for a few hours. A large grain needs more hold time, and high temperatures increase the cost of production. The sintering routes using an oxidizing atmosphere are better than those in a reducing atmosphere. In contrast, the wet mixing process has a significant advantage over the dry mixing process, and the low addition of Cr2O3 facilitates sintering. Today, many experiments exist using the Spark Plasma Sintering (SPS) method to activate the powder and realize the densification of high melting point materials in a short hold time.

Fuel performance code

The United States Nuclear Regulatory Commission (US NRC) has widely endorsed the FRAPCON system and updates, computational tools designed to analyze thermal and mechanical behavior under regular operation and accident scenarios [6]. Nowadays, FAST is the current NRC thermal-mechanical fuel performance code that is the next evolution of FRAPCON and FRAPTRAN in one only code. FAST is an audit code adopted by the NRC that allows the simulation of UO_2, $(Pu\text{-}U)O_2$, $Gd_2O_3\text{-}UO_2$, and zirconium-based alloys, also permits adding capacity to simulate $(Cr_2O_3\text{-}Al_2O_3)$-doped fuel and compare fuel performance under any conditions.

MATERIALS AND METHODS

Reports produced from Westinghouse and Framatome for ADOPT and EATF claimed they dismiss the changes in thermal properties for lower concentrations of the additives. In this study, the relevant factors were fuel parameters used in the manufacturing route, such as sintering temperature, hold time, and concentration of oxide dopants. The additions of $(Cr_2O_3\text{-}Al_2O_3)$ doped UO_2 slightly affected its thermal properties. Most of the material properties used are indistinguishable from those of the standard UO_2, impacting only the FGR process for PWRs and BWRs [7].

Physical properties of Cr_2O_3-doped UO_2

The empirical Kopp–Neumann rule (KNR) or the law of mixtures shows broad usage for calculating the heat capacity of chemical combinations and allows for the fitting of the thermal properties of composite ceramics. Vegard's law is valid in multiphase solid solutions with similar crystallography. Equation (A) is the most straightforward linear function of the physical properties of the end content fractions of the composition range. Ceramic fuel has used the mixture rules or KNR to get empirical approximations of its physical properties.

$$a = a_A^0(1-X) + a_B^0(X) \qquad \text{(A)}$$

where $X = X_B$ is the mole fraction of solid component B and a_A^0 and a_B^0 represent the respective lattice parameters of pure components A and B.

Framatome and Westinghouse claimed they rejected the changes in thermal properties for lower concentrations of metallic oxides. The thermal diffusivity of uranium dioxide decreases with increasing temperature, showing the same behavior as UO_2, doped UO_2, and PuO_2 mixed with UO_2. For (Cr_2O_3-Al_2O_3)-doped UO_2 must be identical to those of the undoped fuel, partly because of the smaller amount dissolved in the UO_2 matrix [8].

The physical properties are valid in the temperature range of 300 to 3000 K, rod-average burnup of 0 to 62 GWd/MTU, and as-fabricated density: (92–97)% TD. Equation (B) describes the density given as a function of temperature. Then equation (C) shows the thermal conductivity in W/m-k of standard UO_2. Theoretically, it should contain slight modifications concerning (Cr-Al) doped UO_2 but not significantly change the thermal fuel properties.

$$\rho_{UO2}(T) = -2.966 \times 10^{-11}T^3 + 5.04 \times 10^{-8}T^2 - 0.0003495T + 11.05 \quad \text{(B)}$$

where ρ is density of UO_2 in g/cm^3, T is the temperature in K

$$K_{UO2}(T) = -5.81 \times 10^{-10}\, T^3 + 4.68 \times 10^{-6}\, T^2 + 0.01T + 11.44 \qquad \text{(C)}$$

where K is the thermal conductivity in W/m-K, and T is the temperature in K, valid below melting point 2850 °C to pure uranium dioxide.

Equation (D) expresses the linear expansion of UO_2. A small reduction in volumetric expansion delays the gap closures of the space between the pellet and the cladding filled with noble gas. Equation (E) represents the specific heat capacity of pure UO_2 given as a function of temperature in K.

$$\frac{\Delta L}{L_0}(UO2) = 9.8 \times 10^{-6}T - 2.61 \times 10^{-3} + 0.316\left(-\frac{E_D}{kT}\right) \quad \text{(D)}$$

where T is the temperature in K, E_D is the power to form defect given in Joules, and k represents the Boltzmann constant (1.38×10^{-23} J/K).

$$Cp_{UO2}(T) = 5.69 \times 10^{-8}T^3 - 9.96 \times 10^{-5}T^2 + 0.39T + 183.91 \text{ (E)}$$

where Cp is the heat capacity in (J/kg-K) and T is the temperature in K.

Fission Gas Release and Swelling Model

Today, it used fuel codes at least for different to estimate gas release, which calculates the gas released in part stored in the plenum. FRAPCON model divide effect in the function of the temperature ranges defining diffusion coefficients changed for doped fuels. Then, the equations (F) and (G) express the total diffusion coefficients and show diffusion under temperatures below 1381 K, respectively. Equation (H) represents diffusion for the intermediate temperature range of 1381 K to 1650 K. Equation (I) defines the diffusion rate for temperatures above 1650 K.

$$D = D_1 + D_{2+} D_3 \qquad \text{(F)}$$

$$D_1 = 1.51 \times 10^{-17}. \exp(-9808/T\,) \qquad (\,T < 1381\,K) \qquad \text{(G)}$$

$$D_2 = 2.14 \times 10^{-13}. \exp\,(-22884/T) \qquad (1381\,K \leq T \leq 1650\,K) \qquad \text{(H)}$$

$$D_3 = 1.09\ \times 10^{-17}. \exp(\,-6614/min(T, 1850)) \qquad (T > 1650\,K) \qquad \text{(I)}$$

The empirical fuel swelling model calculates the volumetric deformation of UO_2 pellets caused by the buildup of stable solid and gaseous fission products during irradiation, given a function of burnup.

This model combines creep-induced elongation with densification due to pressured sintering and irradiation to determine the overall dimensional changes in fuel. Besides, the UO_2 shows pores precipitated at the grain boundary, while the (Cr-Al) doped pellet contains pores inside the grain. Also, additives improve the pore shape, reducing irregularities. The swelling models have suffered many updates and consider at least two effects produced by intragranular and intergranular fission gas bubble behavior as a function of irradiation time, temperature, fission rate, and burnup. Equation (J) represents an empirical correlation used in the swelling model for solid fission products. Equation (K) shows the effects of gaseous fission products.

$$S_{solid} = 2.5 \times 10^{-29} Bs \qquad \text{(J)}$$

where S_{solid} is the fractional volume change due to solid fission products (m^3 volume change/m^3 fuel) Bs is burnup during a time step (fissions/m^3).

$$S_{gas} = 8.8 \times 10^{-56} (2800 - T)^{11.73} e^{[-0.0162(2800-T)]} e^{[-8.0\times10^{-27}B]} B_S \qquad \text{(K)}$$

where S_{gas} is the fractional volume change due to gas fission products (m^3 volume change/m^3 fuel), T is temperature (K), B is total burnup of fuel (fissions/m^3), and) Bs is burnup during a time step (fissions/m^3).

Measurements of creep models are the decisive for integrity of the fuel response, avoiding fragmentation started from crystallographic defects. In a reactor core with over oxygen, a stoichiometric deviation occurs in the core environment, forming many UO2 unbalanced formulations, which distribute many point defects and clusters. Thus, we can conclude that the increased size of the grains should influence the creep mechanism.

RESULTS AND DISCUSSION

IFA-677, formed by standard fuel and doped fuel with additives, was irradiated for six cycles over 500 days from 2005 to 2007. The fuel rod Halden IFA-677.1 test used six rods produced by Westinghouse, Framatome, and GNF, cases chosen are IFA-677.1 rods 1 and 5 with a slight variation in dopant concentrations. Westinghouse made IFA-677 Rod 1 with Cr_2O_3 and Al_2O_3 doped UO_2. Framatone manufactures IFA-677 Rod 2. In contrast, IFA-677 Rod 3fabricaated by GNF with standard UO_2 pellets.

The first rod 1 contains (Cr-Al) doped UO_2 pellets with 900 ppm of Cr_2O_3 and 200 ppm of Al_2O_3. In rod 5 used, the (Cr-Al) doped UO_2 comprised 500 ppm Cr_2O_3 and 200ppm Al_2O_3. Using diffusion coefficient change, FGR models found diffusion results using pure UO_2 and the effects of Cr_2O_3 and Al_2O_3 dopants. Figure 1 displays the diffusion coefficient versus reciprocal temperature, which expresses an intersection point of 1650 °C.

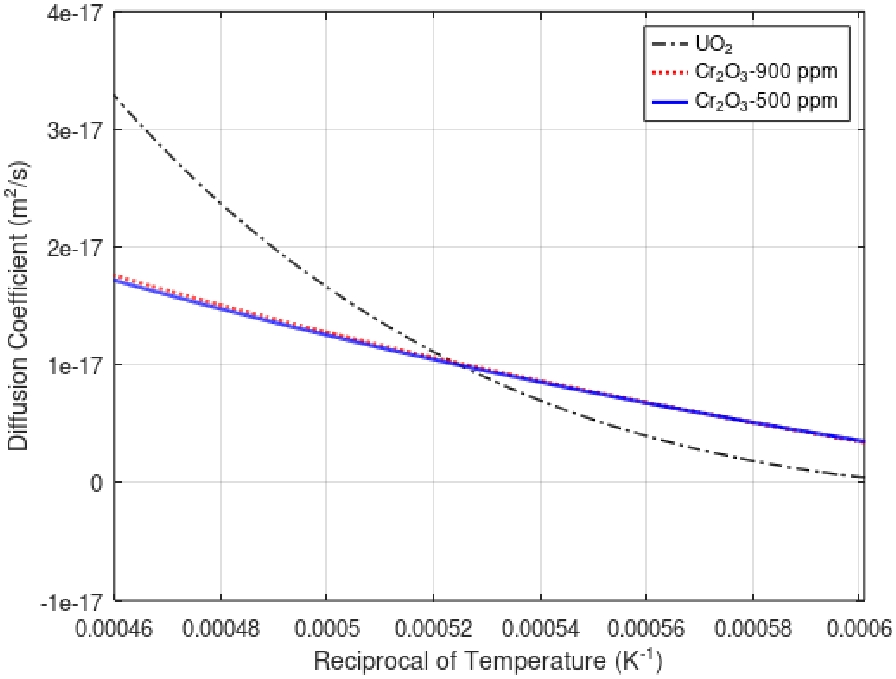

Figure 1. Diffusion coefficient versus reciprocal temperature.

The IFA-677 experiment investigated a higher initial rate using doped fuel produced by fuel suppliers. The fuel rod had an active length of 400 mm using (Cr-Al) doped UO_2 with large grains sizes. Figure 2 shows a comparative between fission gas release under burn cycle

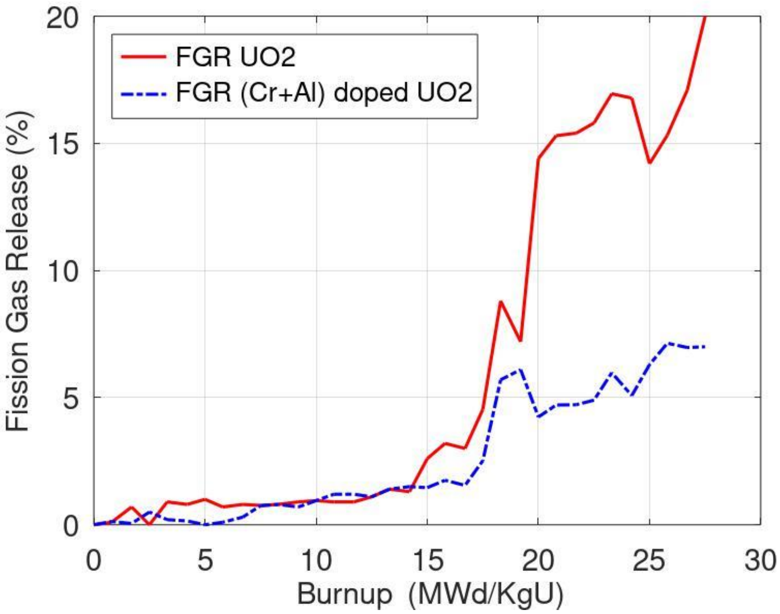

Figure 2: Fission gas release of the fuels UO_2 and UN-U_3Si_2.

A linear heating rate ranged from 42 to 46 kW/m applied during the cycle, reaching the centerline temperature assessed from 1500 °C to 1750 °C. The other rods used standard UO_2 and served as a comparative response. Figure 4 illustrates the FGRs of UO_2 and doped UO_2.

CONCLUSION

The large grain is one of the most promising technological advances. Doped fuel show advantages resulting in more safety, economy, and compatibility. Most of the material properties of the (Cr_2O_3-Al_2O_3)-doped and (SiO_2-Al_2O_3)-doped fuels were identical to those of undoped UO_2. The solubility limit at sintering temperatures restricts the concentration of additives to around ∼ 1000 ppm. Following Vegard's law, formulations use a lower concentration of metallic oxides, Cr_2O_3 varying from 500 ppm to 900 ppm, and Al_2O_3 at ppm.

Commercial fuels fabricated by Westinghouse, Framatone, and GNF showed several favorable characteristics: a large-grain size (> 20 µm), increased density (to 10.67 g/cm3), and lesser fission products enhanced the creep rate, also reducing PCI problem. Adding (Cr-Al) doped UO2 showed slight effects on all the thermal properties. The addition of Chromia and Alumina improved the oxidation resistance in air and water. The cracking pattern was homogeneous and could relieve the cladding stress. Thus, the dopant concept will probably be perfectly acceptable for more tolerant nuclear fuels. Using FRAPCON has got satisfactory simulation results. However, few of the above literature reported the influence of the preparation process on the grain size of UO_2.

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