Fabrication and Thermophysical Characterization of High Gd_2O_3 UO_2 Nuclear Fuel

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1. Introduction

In nuclear reactors, electricity is produced through nuclear fission reactions in nuclear fuel, and excess reactivity is high at the beginning of the fuel cycle, making reactivity control essential. Burnable absorbers (BAs) are used within the fuel pellets to effectively control this excess reactivity. Materials such as cadmium (Cd), boron (B), hafnium (Hf), and gadolinium (Gd) are commonly used as burnable absorbers, typically added into UO_2 fuel pellets [1].

Recently, the nuclear industry has focused on developing high-burnup, long-cycle nuclear fuels and advanced reactor concepts. Small modular reactors (SMRs) are gaining attention due to their advantages in flexible deployment and operation compared to conventional large-scale reactors. Additionally, soluble boron-free core designs, currently pursued by various countries, remove soluble boron from the coolant, simplifying reactor systems and improving maintainability. However, the absence of soluble boron makes reactivity control via control rods and burnable absorbers increasingly important. Thus, the development of SMRs and soluble boron-free core designs is closely aligned with advanced reactor designs and high-burnup fuel development trends.

Furthermore, efforts are underway to enhance reactor operation economics by increasing U-235 enrichment beyond the conventional limit to above 5%, thereby enabling extended fuel cycle operation. Higher enrichment levels necessitate increased burnable absorber content within fuel pellets to maintain safe and effective reactivity control.

Determining the fabrication process parameters is a critical step in fabricating mixed-composition sintered fuels containing burnable absorbers. Ceramic sintered pellets, such as UO_2 nuclear fuels, can exhibit significant microstructural changes, including pores and grain structures, depending on fabrication conditions. Therefore, systematic analysis of these processing parameters is essential to ensure the desired pore size and distribution, grain structure uniformity, minimization of microdefects, and homogeneous additive dispersion.

However, incorporating high amounts of burnable absorbers into fuel pellets introduces several technical issues [2]. Gadolinia (Gd_2O_3), commonly added to UO_2 fuel, adversely affects sintering behavior, leading to lower sintered densities and less uniform microstructures. Moreover, increased Gd_2O_3 content significantly reduces the thermal conductivity, melting point, and mechanical integrity of UO_2 pellets, negatively affecting fuel performance [3]. In particular, most thermal conductivity models for UO_2-Gd_2O_3 fuels were developed based on Gd_2O_3 contents below approximately 10 wt%, and their applicability to higher Gd_2O_3 contents has not been sufficiently evaluated.

Considering these issues, this study evaluated the fabrication and thermal conductivity characteristics of UO_2 nuclear fuel pellets containing high-content Gd_2O_3. The measured thermal conductivity was compared with the Hirai model[4] to evaluate its applicability to high-content Gd_2O_3 fuels. Based on the differences between the measured and calculated values, the model parameters were modified using the present experimental data. In addition, the effects of different powder mixing processes on the microstructure, local Gd distribution, resintering behavior, and thermal conductivity of high-content Gd_2O_3 fuel are being evaluated. By providing experimental data not previously reported, this study aims to offer fundamental information useful for improving fabrication processes and enhancing the performance of high-content BA fuels, ultimately contributing to the development of next-generation reactors, including SMRs and soluble boron-free core designs.

2. Experimental & Results

This study analyzed the effects of various fabrication process conditions on the microstructure and thermal properties of UO_2 nuclear fuel containing high-content Gd_2O_3. Fuel pellets with Gd_2O_3 contents ranging from 8 to 20 wt% were fabricated using standard ceramic processing techniques to systematically evaluate their thermal properties.

Green pellets were prepared using different powder mixing processes, including simple mixing, sieve mixing, and planetary milling. The fabricated pellets were sintered under a controlled reducing atmosphere at approximately 1730 °C for 4 hours. Pellet density was evaluated using Archimedes' principle by measuring dry weight, submerged weight, and saturated weight. Fig. 1 shows the variations in the sintered density of pellets fabricated by planetary milling with increasing Gd_2O_3 content. All specimens exhibited relative densities higher than approximately 97% of the theoretical density. Optical microscopy analysis confirmed the absence of significant microdefects in the planetary-milled specimens.

Thermal diffusivity was measured by the laser flash method using disk specimens (~6 mm in diameter and ~1 mm in thickness) over 200–1200 °C, with three measurements performed at each temperature. Specific heat and thermal expansion were measured by DSC and dilatometry, respectively, and thermal conductivity was calculated using $k=\alpha C_p \rho(T)$. The decrease in thermal conductivity with increasing Gd_2O_3 content is attributed to enhanced phonon scattering resulting from lattice distortion and defect formation caused by Gd^{3+} substitution and the associated charge-compensation mechanism. At 1200 °C, the thermal conductivity of the UO_2 pellet containing 20 wt% Gd_2O_3 was approximately 18% lower than that of pure UO_2.

The measured thermal conductivity was normalized to 95% of the theoretical density (K_{95}) and compared with the values calculated using the Hirai model. The original Hirai model showed reasonable agreement with the measured values for the 8 wt% Gd_2O_3 specimen. However, the difference between the measured and calculated values increased with increasing Gd_2O_3 content, particularly for compositions above 12 wt%. This difference may be related to the original model being developed using experimental data for Gd_2O_3 contents ranging from 0 to 10 wt%.

To improve the applicability of the Hirai model to high-content Gd_2O_3 fuels, the temperature-dependent coefficient in the original expression, $D = 3.84\exp(-9.65*10^{-4}T)$, was re-parameterized using the present experimental data. The modified parameters were determined as $D_0 = 3.365$ and $D_1 =$

$-7.856*10^{-4}K^{-1}$ in $D = D_0\exp(D_1T)$. The modified Hirai model reproduced the measured K_{95} values with relative deviations within ±6%, where the deviation represents the difference between the calculated and measured thermal conductivity relative to the measured value.

Additionally, further experiments were conducted on 17 wt% Gd_2O_3 pellets fabricated using different powder mixing processes. Sieve mixing using as-received Gd_2O_3 powder, sieve mixing with pre-milled Gd_2O_3 powder, and planetary milling of UO_2–Gd_2O_3 blended powders were compared. The effects of each mixing route on sintered density, resintering behavior, microstructure, and thermal conductivity were systematically evaluated. In addition, EDS analyses were performed on the bright and dark regions observed in BSE images to identify local variations in Gd content.

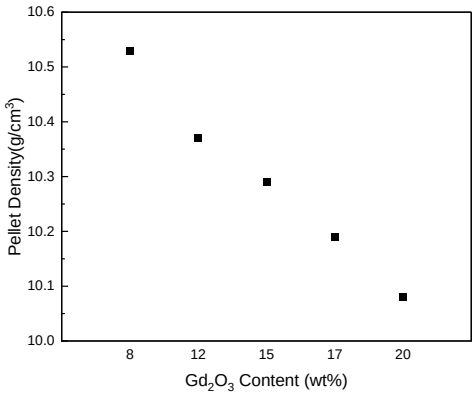


Fig. 1. Sintered density of UO_2 pellets with various Gd_2O_3 contents fabricated by planetary milling method.

3. Conclusions

This study analyzed how fabrication process parameters and Gd_2O_3 content influence the microstructure and thermal properties of nuclear fuels containing high-content burnable absorbers. Adjustments in processing parameters enabled the fabrication of high-density UO_2-Gd_2O_3 fuel pellets with relative densities higher than approximately 97% of the theoretical density.

Thermal conductivity was calculated from the measured thermal diffusivity, specific heat, and temperature-dependent density. Increasing Gd_2O_3 content significantly affected thermal diffusivity and thermal conductivity, demonstrating a direct relationship between the additive content and thermal properties. The thermal conductivity of the specimen containing 20 wt% Gd_2O_3 was approximately 18% lower than that of pure UO_2 at 1200 °C.

The original Hirai model showed increasing differences from the measured values at high Gd_2O_3 contents, particularly above 12 wt%. The temperature-dependent coefficient in the model was re-parameterized using the present experimental data, and the modified Hirai model reproduced the measured K_{95} values within ±6% over the investigated composition and temperature ranges.

The results provide foundational data for evaluating the fabrication and thermal properties of UO_2 nuclear fuels containing 8–20 wt% Gd_2O_3 and can contribute to the development of high-content burnable absorber fuels for long-cycle operation and soluble boron-free core designs.

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