Hydration Resistance of Zr-doped Gadolinia for Burnable Absorber Applications

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Keywords : Gadolinia burnable absorber, Hydration Resistance

1. Introduction

Advanced pressurized water reactors (PWRs) and small modular reactors (SMRs) focus on reducing soluble boron in the coolant, requiring burnable absorbers (BAs) with high thermal neutron absorption cross-sections. Gadolinium oxide (Gd_2O_3) is a potential material due to the large absorption cross-sections of ^{155}Gd and ^{157}Gd. However, under the high-temperature, high-pressure conditions of PWR operation, gadolinia is easily hydrated to form gadolinium hydroxide, leading to volumetric swelling, microstructural cracking, dissolution, and eventual degradation of thermal and neutronic performance [1]. Previous studies have used oxides such as Al_2O_3 to form stable phases such as $GdAlO_3$ perovskites that suppress phase transformation and enhance stability [2]. However, this method requires Al_2O_3 content of more than 50 mol%, which greatly reduces gadolinium content and neutron absorption capacity.

This research focuses on enhancing hydration resistance and preserving high gadolinium content through zirconia doping. Zirconia has a high melting point, chemical stability, and compatibility with oxide fuel matrix, and can form a solid solution or gadolinium zirconate ($Gd2Zr_2O_7$) pyrochlore. The microstructure, chemical state and composition of zirconia doped gadolinia ceramics were systematically evaluated before and after hydration.

2. Methods and Results

2.1 Processing

Pure Gadolinia (Gd_2O_3, 99.9%) and Zirconia (ZrO_2, 99.9%) powders were used for fabrication. They were then combined with ethanol and mixed using a planetary ball milling with zirconia balls. The resulting slurry was filtered, dried, and sieved to obtain fine powder. Cylindrical green compacts (Φ11mm) were formed by uniaxial pressing, followed by sintering in a furnace.

2.2 Microstructural Grain Analysis

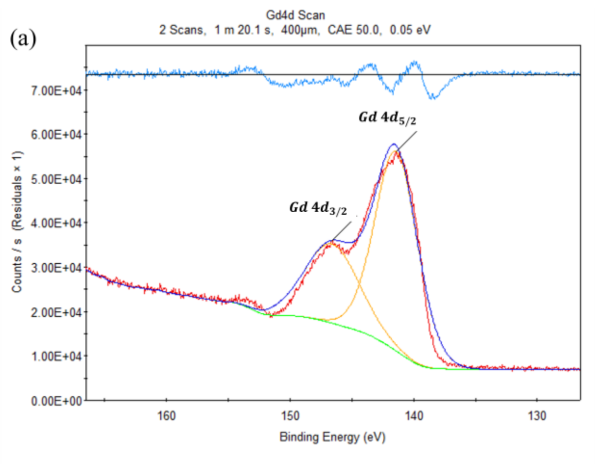
Microstructure Grain analysis was performed through a scanning electron microscope (SEM). Pure gadolinia had a monoclinic phase after sintering, with an average grain size in the order of tens of micrometers [3,4]. In contrast, it was confirmed that as the zirconia doping content increased, a mixed monoclinic–cubic phase was observed and the microstructure grain size decreased rapidly by several micrometers.

2.3 Chemical State and Composition Analysis Before and After Leaching Test

Chemical state and composition analysis according to doping content and before/after hydration were conducted through an X-ray Photograph Spectrometer Spectral (XPS). Quantitative analysis of hydroxyl group bond production, Gd-O ligand bond reduction, and Gd bond reduction analysis were performed through XPS measurement before hydration, respectively as shown in **Fig. 1**. These measurements provided insight into how zirconia doping content influences the hydration resistance of gadolinia through comparative analysis of XPS results after hydration.

2.4 Hydration Behavior and Resistance Evaluation Analysis

Hydration behavior and resistance evaluation analysis were performed in an autoclave under conditions simulating a PWR environment. The experimental conditions were conducted at 300 °C and up to 15 MPa for 24 h. The sample data were analyzed using XPS and inductively coupled plasma mass spectrometry (ICP-MS). The results were used to evaluate the effect of zirconia doping content on the hydration behavior of gadolinia.



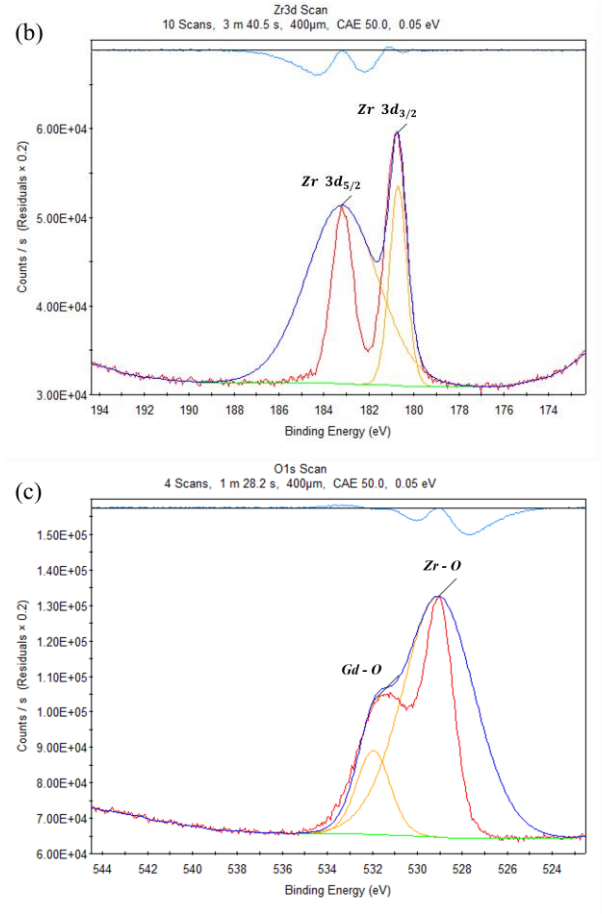


Fig. 1. XPS spectra of (a) Gd 4d, (b) Zr 3d, (c) O 1s of ZrO_2 doped Gd_2O_3 before leaching experiment

3. Conclusions

This study aims to improve the hydration resistance of Gadolinia burnable absorber by low-level doping of zirconia. Low-zirconia-doped gadolinia burnable absorber samples were fabricated via planetary ball milling, followed by compaction and sintering. Microstructural grain analysis is performed through scanning electron microscopy (SEM), and chemical state and composition analysis according to doping is performed through X-ray photoelectron spectroscopy (XPS). SEM results show that even a small amount of zirconia-doped Gadolinia burnable absorber significantly reduced the grain size from tens of micrometers to only a few micrometers. Leaching tests were carried out under autoclave conditions simulating the PWR environment. The results show that a low percentage of zirconia-doped Gadolinia burnable absorbers can more effectively suppress hydration. This work provides experimental evidence of hydration resistance in reactor-simulated environments, contributing to the development of advanced nuclear fuels for PWRs or SMRs targeting low- or boron-free operation.

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Acknowledgement

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Ministry of Science and ICT (MSIT) of the Republic of Korea [No. RS-2024-00420956] and Korea Institute of Energy Technology Evaluation and Planning (KETEP) and the Ministry of Trade, Industry & Energy (MOTIE) of the Republic of Korea [No. RS-2025-02633904].