

Hydration Resistance and Chemical State Analysis of Zirconia-doped Gadolinia Burnable Absorber

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1. Introduction

Recent developments in Small Modular Reactors (SMRs) focus on low-boron or boron-free system for passive operation concept. Gadolinium-based material can be potential burnable absorber (BA) material which should have large absorption cross-sections. Lumped burnable absorber design integrated into UO₂ pellets have been investigated due to enhanced self-shielding, which supports long-cycle reactor operation. However, pure gadolinia material can be hydrated under high-temperature and high-pressure conditions. This problem can cause swelling, cracking, dissolution of burnable absorber [1]. Earlier work has studied that the addition of oxides such as more than 50 mol.% Al₂O₃ forms stable GdAlO₃ perovskite phases, effectively inhibiting phase transformation and increasing hydration stability [3]. However, this can lead to a reduction in Gd content by at least 50 mol.% compared to a pure gadolinium-based burnable absorber. Consequently, this may cause a loss of neutron absorption cross-section per unit volume.

This study aims to control hydration of Gd-based burnable absorbers by low-level zirconia doping. The chemical state and hydration resistance of zirconia doped gadolinia burnable absorbers were systematically evaluated at high-temperature and pressure environment.

2. Methods and Results

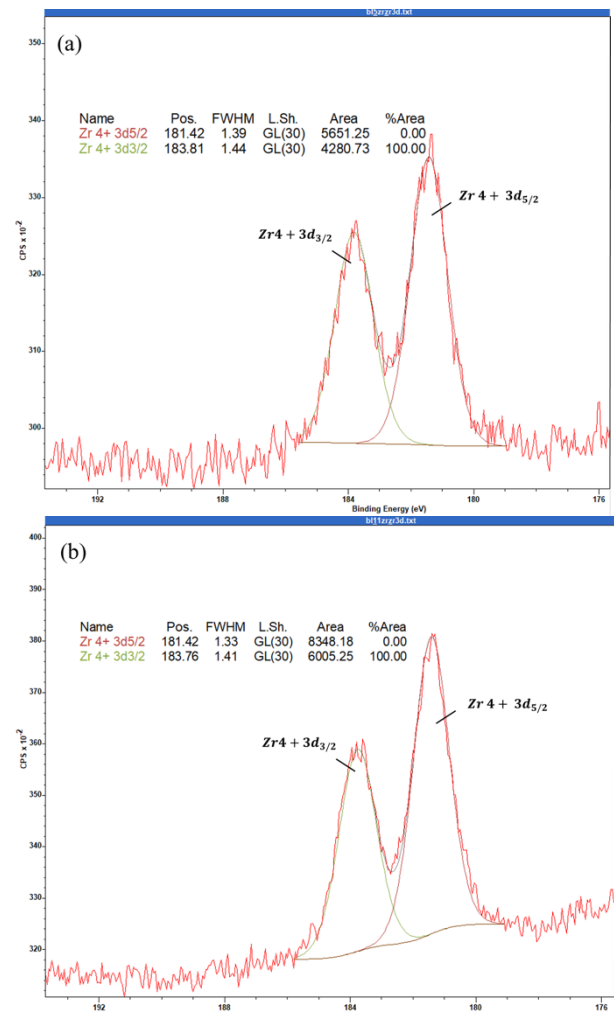
2.1 Fabrication of zirconia doped gadolinia pellet

For chemical state and hydration analysis of the ZrO₂-doped Gd₂O₃ burnable absorbers, pure Gd₂O₃ and ZrO₂ powders were mixed using a planetary ball milling with zirconia balls. The slurry was dried and sieved to fined powder. Cylindrical pellets (Φ11mm) were pressed at 200 MPa and sintered at 1600 °C for 10 hours using a conventional method.

2.2 Chemical state analysis

Chemical state and composition analysis of pellets after ZrO₂ doping and sintering were conducted through an X-ray Photoelectron Spectroscopy (XPS).

Quantitative analysis of Zr³⁺/Zr⁴⁺ state ratio analysis were performed with various ZrO₂-doping contents as shown in **Fig. 1**. These results showed that no Zr³⁺ species were present in the Gd³⁺ lattice of the sintered pellets and that zirconium predominantly existed in the Zr⁴⁺ state. This indicated that the oxygen vacancy concentration within the lattice decreased to maintain charge neutrality through the substitution of Zr⁴⁺ into the Gd³⁺ lattice.



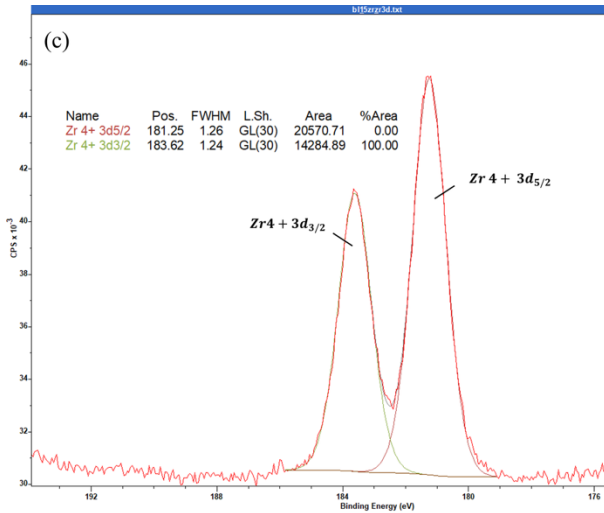


Fig. 1. XPS Zr 3d spectra of (a) 5%, (b) 10%, (c) 15% ZrO₂-doped Gd₂O₃ after sintering.

2.4 Hydration analysis

Hydrothermal tests were performed in a high-pressure reactor at 315 °C and up to 10 MPa for 24 h. Hydration analysis were conducted using XPS, Thermogravimetric Analysis (TGA), and X-ray Diffraction (XRD). The results of hydration resistance were evaluated as doping composition.

3. Conclusions

This study focused on improving the hydration resistance of gadolinium-based burnable absorber by low-content doping of zirconia. XPS results showed that, after sintering, zirconium existed as Zr⁴⁺ oxidation state rather than Zr³⁺. This substitution in Gd³⁺ lattice led to a reduction in the oxygen vacancy concentration within the lattice to maintain charge neutrality. The decreased oxygen vacancy concentration is expected to suppress hydration reactions by limiting proton penetration into the lattice through oxygen vacancy sites under water environments. Hydrothermal tests demonstrated that low-level zirconia-doped gadolinia burnable absorbers exhibit enhanced hydration resistance. This work provided experimental evidence of improved hydration resistance under reactor-simulated environments using boron-free operation system.

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