Phase and Dissolution Analysis of ZrO_2, CaO_2 Doped Gd_2O_3

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1. Introduction

Recent developments in innovative Small Modular Reactors (i-SMRs) aim for passive operation systems through boron-free or low-boron conditions. Consequently, pure gadolinia which has high neutron absorption cross-sections could be effectively adopted as a burnable absorber (BA). However, pure gadolinia burnable absorbers in pressurized water reactor (PWR) environments would have the problem of hydration reaction. Previous studies have shown that under PWR conditions of high temperature and pressure, gadolinia burnable absorber forms hydroxide when reacting with water. This can cause various problems, including swelling and subsequent cracking, decreased heat conduction due to pore formation, increased core temperature, and increased reactivity as the gadolinia loses its function, resulting in a reactivity-initiated accident [1]. Previous attempts have been made to solve this problem by adding other elements such as Al and Ti. For instance, doping gadolinia with Al_2O_3 resulted in the formation of a stable perovskite phase, such as $GdAlO_3$, specifically at Al_2O_3 concentrations exceeding approximately 50 mol.%. This perovskite structure effectively suppressed phase transformation even at high temperatures, significantly improving material stability [2]. However, doping with more than 50 mol.% of Al_2O_3 introduces a problem where the amount of gadolinia within the burnable absorber is reduced significantly. This resulted in a loss of neutron absorption cross-section per unit volume and a subsequent decrease in reactivity control ability.

This study aims to prevent hydration and subsequent dissolution by doping small amounts of specific elements into burnable absorbers. Specifically, the phase and dissolution behaviors of gadolinia burnable absorbers doped with two types of oxides (ZrO_2, CaO_2) were analyzed.

2. Methods and Results

2.1 Fabrication of ZrO_2, CaO_2 doped gadolinia burnable absorber

For dissolution analysis of the doped gadolinia burnable absorbers, sample pellets were fabricated. Mixed powders for each element and composition were produced, along with pure gadolinia and $GdAlO_3$ powders for comparison of leaching resistance. The fabrication process from mixed powders to pellets was conducted through conventional sintering.

2.2 Phase analysis

Phase analysis was conducted using X-ray diffraction (XRD). A stable crystalline phase was formed when gadolinia burnable absorbers were doped with certain concentrations of specific oxide elements. In the case of ZrO_2 doping, as the ZrO_2 content increased, the crystal structure of Gd_2O_3 transitioned toward the cubic phase, accompanied by a corresponding decrease in the monoclinic phase. In contrast, for pure gadolinia, during the fuel sintering process, sintering temperatures typically reach approximately 1600 °C, resulting in an irreversible cubic-to-monoclinic phase transition at around 1200 °C. Thus, pure gadolinia forms a monoclinic structure.

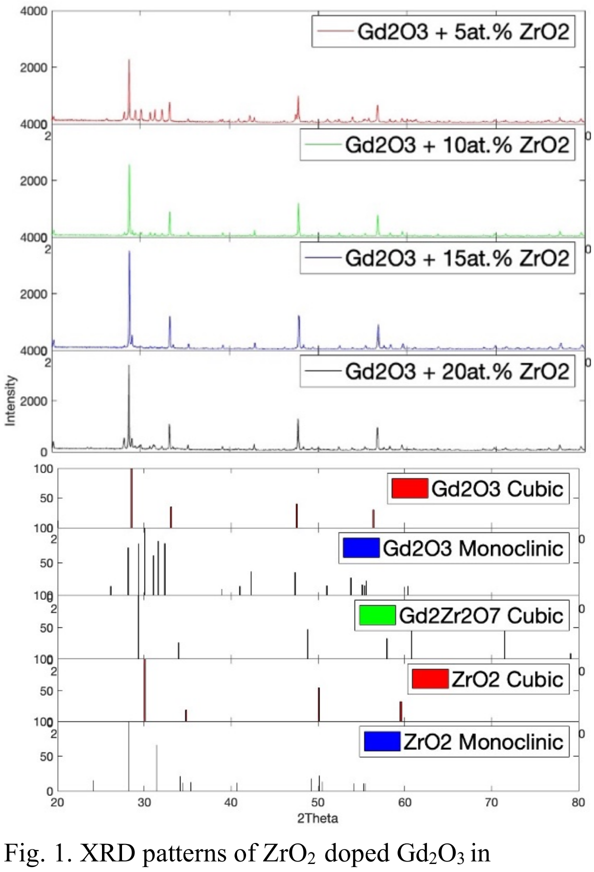

Fig. 1. XRD patterns of ZrO_2 doped Gd_2O_3 in composition ranges.

2.3 Dissolution analysis

Leaching tests were conducted in accordance with the American National Standard ANSI/ANS-16.1-2019. The experimental conditions were set at 25°C temperature and atmospheric pressure for 5 days. The sample data was then measured by inductively coupled plasma mass spectrometry (ICP-MS). The results confirm the dissolution behavior of each element at each concentration.

3. Conclusions

The aim of this study is to control the hydration behavior of gadolinia burnable absorber by doping low amounts of ZrO_2, and CaO_2. The low ZrO_2, CaO_2 doped gadolinia burnable absorber samples are fabricated by sintering, and the phase analysis is carried out using XRD. The XRD results show that the gadolinia burnable absorber doped with a low amount of specific elements forms a stable phase. In addition, the leaching test was carried out by the American National Standard ANSI/ANS-16.1-2019 method. The results show that gadolinia burnable absorber doped with a low percentage of specific elements can control hydration more effectively. This work contributes to innovative neutron absorber elements for SMRs targeting boron-free or low-boron coolants as a screening experiment.

REFERENCES

[1] Balestrieri, D. "A study of the UO_2/Gd_2O_3 composite fuel." (1998).
[2] Kim, J. S., Jung, T. S., & Yoon, J. (2024). Reactor core design with practical gadolinia burnable absorbers for soluble boron-free operation in the innovative SMR. Nuclear Engineering and Technology, 56(8), 3144-3154.

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