Effect of 15% Er_2O_3 and Dy_2O_3 Doping on the Gd_2O_3 Crystal Structure at Elevated Temperature

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1. Introduction

High-performance burnable absorbers (BAs) are required to maintain small excess reactivity in the concept of soluble-boron-free (SBF) pressurized water reactors (PWRs). Advanced lumped-gadolinia designs (e.g., CSBA and CIMBA) have been proposed and shown good performance in minimizing excess reactivity [1], [2]. However, gadolinia (Gd_2O_3) can undergo hydration under high-temperature and high-pressure conditions, leading to mechanical degradation of the absorber. To overcome this problem, previous studies have explored the addition of small amounts of rare-earth oxides (REOs); in particular, CeO_2 has been reported to improve hydration resistance under hydrothermal conditions [3]. Based on this approach, this study examines alternative REO additions by investigating the effect of 5-15 at% Er_2O_3 and Dy_2O_3 on the phase of Gd_2O_3 at elevated temperatures. X-ray diffraction is used to identify phase evolution and establish a baseline for subsequent evaluation of hydration resistance.

2. Methods and Results

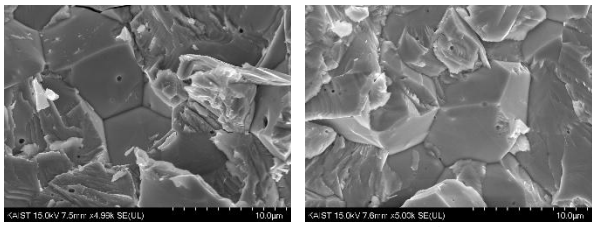
2.1. Fabrication of REO-doped Gadolinia

REO-doped gadolinia compositions were prepared in 15 at.% dopant, alongside an undoped Gd_2O_3 reference. Starting powders of Gd_2O_3, Er_2O_3, and Dy_2O_3 (purity ≥ 99.9%) were weighed according to the target compositions and homogenized by planetary ball milling in ethanol for 20 hours. The mixed powders were dried and pressed into pellets at 180 MPa. The undoped Gd_2O_3 and the REO-doped Gd_2O_3 pellets were sintered at 1600^0C for 10 hours in air.

Both doped materials exhibit good densification during sintering, with relative densities of around 95%, indicating minimal porosity. This is also confirmed by SEM images (Fig. 1), which shows interconnected grains with few porosities.

Table 1. Density of doped-Gd_2O_3 pellets after sintering

Samples	Theoretical Density (g/cm^3)	Relative Density (%TD)
15at%Er_2O_3-Gd_2O_3	8.407	95.40
15at%Dy_2O_3-Gd_2O_3	8.397	95.59



a b

Fig. 1. (a) 15at%Er_2O_3-Gd_2O_3 and (b) 15at%Dy_2O_3-Gd_2O_3 after sintering

2.2. XRD characterization

Sintered pellets were ground into powder for X-ray Diffraction (XRD) characterization. The samples were analyzed in the 2θ range from 20°-80° using Cu Kα radiation.

The XRD results demonstrate that both undoped Gd_2O_3 and REO-doped Gd_2O_3 exhibited a monoclinic phase after sintering (Fig. 2). This phase is a common crystal structure for Gd_2O_3 above ~1250^0C, and it remained intact even after doping with Er and Dy. However, slight peak shifts towards larger 2θ can be observed in the doped Gd_2O_3 samples. The peak shifts indicate lattice distortion due to the doping process. Er and Dy have smaller ionic radii compared to Gd. This difference causes a shrinkage of the unit cell, as the smaller dopant ions replace the larger ions within the crystal lattice.

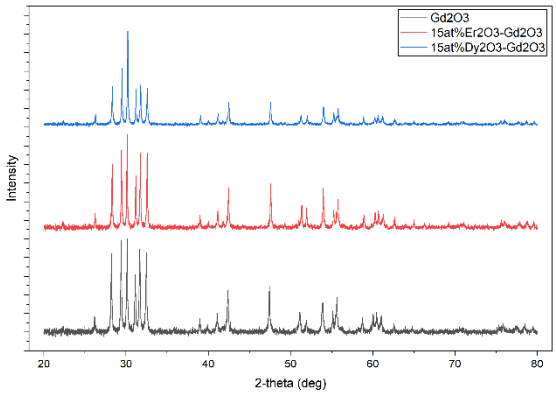


Fig. 2. XRD patterns of undoped and REO-doped Gadolinia after sintering

The lattice distortion is confirmed by the difference in lattice parameters and unit cell volume. As shown in Table 2, the addition of REO (Er_2O_3, Dy_2O_3) leads to a reduction in the lattice parameter and unit cell volume.

Table 2: Lattice parameters and unit cell volume of undoped and REO-doped Gd_2O_3 after sintering

Samples	Phase(s)[a]	a (Å)	b (Å)	c (Å)	β (deg)	Volume ($Å^3$)
Gd_2O_3	B	14.083	3.570	8.756	100.02	433.6
15at% Er_2O_3-Gd_2O_3	B	14.063	3.557	8.735	100.14	429.6
15at% Dy_2O_3-Gd_2O_3	B	14.046	3.559	8.733	100.16	430.11

[a] B: monoclinic

3. Conclusion

This study aimed to establish a baseline before conducting the hydration testing. The sintered pellets introduced high relative density values close to 95% of the theoretical density. This indicates that the fabrication method and parameters was produce well-densified pellets.

Despite the changes in lattice parameters and unit cell volume due to doping, the monoclinic phase is remained unchanged after doping with 15at%Er_2O_3 and 15at%Dy_2O_3. This suggests that the doping concentrations of 15at% for both Er and Dy do not disrupt the overall crystal structure of Gd_2O_3 at elevated temperature.

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