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Spectroscopic reflects of structural disorder in Eu³⁺/Pr³⁺-doped La_{0.4}Gd_{1.6}Zr₂O₇ transparent ceramics

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Abstract

Perfectly transparent ceramics of La_{0.4}Gd_{1.6}Zr₂O₇ doped with either Eu³⁺ or Pr³⁺ ions were prepared by high-temperature vacuum sintering. Results of the Rietveld structure refinement and spectroscopic properties of the ceramics are presented and discussed in details. Photoluminescence characteristics support the non-uniformity in the crystal structure of ceramics. X-ray excited luminescence of La_{0.4}Gd_{1.6}Zr₂O₇:Eu was rather inefficient and the Pr-doped ceramics showed no Pr-related radioluminescence. Instead they produced Eu³⁺ emission characteristic for Gd₂O₃:Eu phase not detected in X-ray diffraction patterns.

1. Introduction

Compounds of the general formula A₂B₂O₇ (A and B stands for 3+ and 4+ ions, respectively) are hosts for materials with a whole variety of actual and potential applications. Examples are thermal barrier coatings [1],[2],[3],[4],[5],[6], nuclear waste forms [7],[8],[9],[10],[11],[12],[13], electrolytes [14],[15], catalysts [16] and optical materials [17],[18],[19],[20],[21],[22],[23]. The A₂B₂O₇ compounds can crystallize in either ordered pyrochlore or disordered fluorite structures. The pyrochlore phase is considered as a superstructure of the fluorite phase with doubled *a* axis. The ratio of ionic radii of cations at A and B sites affects directly the crystal phase they form which in turn influences the compositions spectroscopic properties. What complicates the effect of the structure on the compositions properties is their tendency towards formation of anionic Frenkel defects [24],[25],[26],[27]. Their population quickly increases if processing is performed at high temperatures [28],[29] which is the case of transparent ceramics fabrication.

In the case of rare earth zirconates, RE₂Zr₂O₇, the stable pyrochlore structure is formed when *r*_{RE}/*r*_{Zr} ratio ranges from 1.26 Å (for CN_{Zr} = 8) or 1.48 Å (for CN_{Zr} = 6) up to 1.78 and depends directly on the RE³⁺ ionic radius. Below the correspondingly lower limit values the pyrochlore structure becomes disordered and transfers to a defect-fluorite structure [30],[31]. When the *r*_{RE}/*r*_{Zr} > 1.78, a monoclinic/δ (delta) phase is formed [32]. Furthermore, pyrochlore

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