ScienceDirect

Ceramics International
Volume 46, Issue 9, 15 June 2020, Pages 13170-13179

Structural, thermal, spectral, optical and surface analysis of rare earth metal ion (Gd^{3+}) doped mixed Zn–Mg nano-spinel ferrites

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https://doi.org/10.1016/j.ceramint.2020.02.091
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Abstract

The present work reports the structural, thermal, spectral, optical and surface analysis of rare earth metal ion (Gd^{3+}) doped mixed Zn–Mg nano-spinel ferrites. The samples of Gd^{3+} doped Zn–Mg nano ferrites with equi-amount chemical composition i.e. $Zn_{0.5}Mg_{0.5}Fe_{2-x}Gd_xO_4$ ($0.00 \le x \le 0.10$ in step of 0.02) were prepared by self-ignited sol-gel route. The variance in the thermal behaviour and spinel phase development with weight loss percentage in the prepared samples was investigated by TG-DTA technique. The powder X-ray diffraction (P-XRD) patterns ensured the nanocrystalline mono-phasic formation and spinel-cubic structure of all the samples. The trend of increment in lattice constant (a) and decrement in crystallite (t) size was observed with the doping of Gd^{3+} ions. The appearance of two requisite vibrational stretching modes was affirmed by the FT-IR spectral studies. The UV–Vis optical analysis displayed the augmentation in absorbance and drastic decrement in energy band gap value (1.96 eV–1.83 eV) with Gd^{3+} doping. The photo-luminescent (PL) studies revealed the broad near band-edge emission in visible wavelength range (523 nm–528 nm) for all the samples. The surface parameters investigation was undertaken with the help of BET isotherms recorded by the N_2-physisorption and BJH model. The various surface parameters such as BET surface area, volume and radius of the pores, distribution of the pore sizes etc were construed from the BET data. The enhancement in these surface parameters via Gd^{3+} doping was noted for all the

samples. The outcomes of the present work reflects the influential doping of Gd^{3+} ions in Zn–Mg nano ferrites, which can be implementable for bio-applications as thermal seeds in magnetic hyperthermia or as contrast enhancer in medical MRI imaging.

Introduction

The perception of the materials research has experienced the drastic changes over some decades by the nano-level approach commenced by the Prof. Feynman [1]. The nano scaled materials broadcasts superior properties which are restricted or non accessible in the bulk appearance of the same material. Some imperative features of the nano scaled materials are elevated surface to volume proportion, chemically more stable, dominancy of quantum effects and tuning feasibility [[2], [3], [4]]. In the developed nano scaled materials, the magnetic nanomaterials (MN's) have prominent place due to its significance in numerous technological areas [5,6]. The anisotropy of surface, canting of the spins, superparamagnetic gesture and the multimodal diversity are the key features of the magnetic nanomaterials (MN's) [7,8]. The variety of MN's have shown their usability in the range of applications from magnetic data recorders to micro-wave appliances [9], from heat transport fluids [10,11] to inductive heat production [12,13] and from medical diagnostics [14] to the disease treatments [15,16].

In the class of MN's, nano ferrites (NF's) are the principal materials which consist of the mixed composition of iron oxide and metal ions. Further, depending on the crystal arrangement, NF's can be categorized into three main ways as spinel ferrites (SF's), garnet ferrites (GF's) and hexagonal ferrites (HF's). In these three ferrites, SF's are the most promising and interesting due to its simplistic and stabilized lattice structure, effortless accommodation of guest ions, properties tuning aptitude, preparation easiness and flexible cationic distribution [[17], [18], [19]]. The allocation of cations among two intrinsic lattice sites of SF's is subjected to the balancing of ions. Usually, the site preferential energies of the metal ions define the normal or inverse or mixed behaviour of spinel matrix. Thus, the site preferential allotment of cations over the intrinsic sites and its direct correlation in defining the properties of SF's makes researcher more curious to study the SF's [20]. The physicochemical and other properties of SF's can be altered or enhanced by means of different ways like mode of preparations, stoichiometric proportions, synthesis circumstances, incorporation of guest metal ions (divalent or trivalent) at spinel lattice etc. The modes of preparation and synthesis circumstances mostly affect the size and shape of SF's which subsequently results into a change of structure. However, the incorporation of guest ions either magnetic/nonmagnetic or divalent/trivalent in a spinel lattice consequences the drastic change in structural as well as other properties by means of cationic redistribution. In this light, the appropriate selection of guest ions or dopant is prerequisite to achieve the desired alteration or improvement in the pristine SF's [21,22]. Apart from the conventional dopant, rare earth (RE) metal ion dopant such as Gd^{3+}, La^{3+}, Nd^{3+}, Sm^{3+}, Dy^{3+} etc with nominal concentration give rise to radical changes in SF's [[23], [24], [25], [26]]. Amongst the RE ions, the Gadolinium (Gd^{3+}) with ionic radius of 0.0938 nm is more appreciable for altering the magneto-electric properties of SF's. Also, the Gd^{3+} is most extensively used RE as a contrast enhancer in medical imaging due to its exclusive electronic arrangement. Thus, the inclusion of Gd^{3+} ions in spinel matrix may give rise to noteworthy improvement in contrast enhancing ability.

In the last few decades, researches have made an admirable endeavor to outcome the influence of RE incorporation on the different properties of SF's. Jadhav et al. [27] focused their study on the inductive heating analysis of the Gd^{3+} incorporated Mn–Zn SF's. The elevation in the heating response by Gd^{3+} inclusion was noticed in their work. Kumar et al. [28] investigated the RE (Gd^{3+}) induce influence on impedance and transport phenomenon of Zn–Ni SF's. Mugutkar et al. [29] investigated the doping effect of Gd^{3+} on the magnetic parameters of mixed Co–Zn SF's. Although, investigations on the RE (especially Gd^{3+}) incorporated SF's are extensively rising, still to the best of our information, the systematic investigations related to the influence of Gd^{3+} doping on thermal, spectral, optical and surface parameters of mixed SF's (Zn–Mg SF's) are not reported yet. Also, in accordance with synthesis routes, there are no reports in the literature related to the Gd^{3+} doped Zn–Mg SF's fabricated via self ignited sol-gel synthesis. The self ignited sol-gel route is beneficial to form a pure and mono phase SF's with RE doping which is not easy to obtain by conventional routes.

In view of the above key points, we aimed to prepare Gd^{3+} doped Zn–Mg nano ferrites (ZMGF) with equi-amount chemical composition i.e. $Zn_{0.5}Mg_{0.5}Fe_{2-x}Gd_xO_4$ ($0.00 \le x \le 0.10$ in step of 0.02) by self ignited sol-gel route. The improvement in the structural, thermal, spectral, optical and surface parameters via influential rare earth metal ion (Gd^{3+}) doping in mixed Zn–Mg nano-spinel ferrites was studied by the standard characterization tools.

Section snippets

Synthesis procedure

The high purity precursor materials of the respective metal ions (i.e. Zn^{2+}, Mg^{2+}, Fe^{3+} and Gd^{3+}) in the form of nitrates were procured from *Sigma-Aldrich* and used without subsequent purification for the synthesis ZMGF with equi-amount chemical composition i.e. $Zn_{0.5}Mg_{0.5}Fe_{2-x}Gd_xO_4$ ($0.00 \le x \le 0.10$ in step of 0.02). The auto ignition enabled sol-gel synthesis was employed for the synthesis purpose. For the chelation and combustion of the nitrate solutions, the citric acid was used as a fueling…

Thermal analysis (TG-DTA)

Fig. 2(a and b) illustrates the TG-DTA curves of all the ZMGF samples. The total percentage weight loss of 4.62, 8.52, 13.74, 42.91, 11.74 and 24.17 was noted for x=0.00, 0.02, 0.04, 0.06, 0.08 and 0.10 respectively. It is notable from Fig. 2 (a) that, in the initial stages of loss in weight occurred gradually and at some temperature rapid weight loss began. After the rapid loss in weight, the saturation of loss (no further weight loss) was observed for further temperature region. The…

Conclusions

The nanocrystalline Gd^{3+} doped Zn–Mg ferrites (ZMGF) with pure mono phase spinel structure were successfully prepared by self ignition assisted sol-gel synthesis. TG-DTA results displayed the superior stability over higher temperature range and development of spinel phase for all the ZMGF samples. P-XRD analysis of all the ZMGF samples confirmed the cubic mono phase spinel structure belonging to the *'Fd-3m'* space group. The linear increment in 'lattice constant' and decrement in 'crystallite…

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

Acknowledgement

The author Sandeep B. Somvanshi acknowledges Department of Science and Technology, Government of India for DST-INSPIRE Fellowship (IF170288).…

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