

Magnetic properties of samarium and gadolinium co-doping Mn-Zn ferrites obtained by sol-gel auto-combustion method

JI Bifa (季必发)^{1,2}, TIAN Changan (田长安)^{1,*}, ZHANG Quanzheng (张全争)^{1,3}, JI Dongdong (吉冬冬)¹, YANG Jie (阳杰)¹, XIE Jinsong (谢劲松)¹, SI Jingyu (司靖宇)^{1,*}

(1. Department of Chemistry and Materials Engineering, Hefei University, Hefei 230601, China; 2. Institute of Nano Science and Technology, University of Science and Technology of China, Suzhou 215123, China; 3. State Key Laboratory of Structural Chemistry, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou 350002, China)

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Abstract: Mn-Zn ferrites doped with different contents of Sm³⁺ and Gd³⁺ ions were prepared by sol-gel auto-combustion method and characterized by Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TG), X-ray diffraction (XRD), scanning electron microscopy (SEM) and vibrating sample magnetometer (VSM). When samples were calcined in a relatively low temperature below 1100 °C, secondary phases (α -Fe₂O₃) could be identified. Therefore, in order to acquire pure and better crystallinity, the suitable calcining temperature of powders was selected at 1200 °C. It was also found that all the samples consisting of ferrite phases of typical spinel cubic structure and average crystallite sizes between 31.5 and 38.2 nm were obtained after calcining at 1200 °C for 4 h. The lattice parameters increased almost linearly with increasing Sm content. A dense microstructure was obtained after sintering at 1250 °C for 4 h. Through the analysis of magnetic properties, hysteresis loops for all the samples were narrow with low values of coercivity and retentivity, indicating the paramagnetic nature of these samples. And saturation magnetization M_s strongly depended on the type of additive to reach a maximum of 47.99 emu/g for $x=0.015$, which showed a great promise for hyperthermia applications.

Keywords: Mn-Zn ferrites; co-doping; sol-gel auto-combustion method; microstructure; magnetic properties; rare earths

Spinel ferrites have attracted considerable attention in recent years due to their interesting properties as well as scientific and technological significance^[1–3]. As an important soft magnetic ceramic, manganese zinc (Mn-Zn) ferrites with excellent performance is one of the well-known spinel-type ferrites having high saturation magnetization, high initial permeability, high electrical resistivity, relatively high Curie temperature, and low hysteresis loss. These ferrites are extensively used in inductance components, noise filters, microwave devices, broad band impulse transformers, and magnetic recording media^[4–6].

The applications of ferrites are influenced by the physical and chemical properties of the materials themselves, meanwhile, the magnetic and electric properties of ferrite materials strongly depend on their chemical compositions, additives and substituents. In this study, small amounts of cationic substitutions were used in Mn-Zn ferrites because foreign ions in the ferrites can dramatically change their properties. Mn-Zn ferrites have a spinel structure, Fe ions occupy both tetrahedral site (A-site) and octahedral site (B-site); however, Mn²⁺ and

Zn²⁺ ions occupy tetrahedral site (A-site) in the spinel unit cell. The spinel structure allows introduction of other suitable ions into the lattice, which replace the metallic cations in regular A- or B-sites, and thus alter the microstructure and properties of the Mn-Zn ferrites^[7]. According to the literature^[8–11], rare-earth (RE) ions including Co²⁺^[12], Cu²⁺^[13], Mo⁶⁺^[14], Ti⁴⁺^[14], Al³⁺^[15], and Sn⁴⁺^[16] have been used to improve the microstructure and electric and magnetic properties of the Mn-Zn ferrites.

RE ions-doped ferrites have significant importance and they appear to be a promising candidate for numerous industrial applications; therefore, RE ions play an increasingly important role as dopants. Iqbal et al.^[8] studied the electrical transport properties of Eu³⁺ substituted Mn-Zn ferrites. They found that the dielectric constant, complex dielectric constant, and loss tangent of these materials decreased with an increase in Eu concentration. Islam et al.^[9] investigated the structural, magnetic, and electrical properties of Gd-substituted Mn-Zn mixed ferrites. They demonstrated that the saturation magnetization decreases with Gd-content and the substantial en-

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* **Corresponding authors:** TIAN Changan, SI Jingyu (E-mail: tca2204@126.com, sijyo@hfu.edu.cn; Tel.: +86-551-62158440, +86-551-62158047)

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hancement of resistivity due to Gd-substitution. Venkataraju et al.^[10] studied the effect of Cd on the structural, magnetic, and electrical properties of nanostructured Mn-Zn ferrites. They found that the maximum magnetization of all the samples decreases with increase in Cd content. Sun et al.^[11] studied the effect of added ZrO₂ on the magnetic properties of Mn-Zn ferrites. They demonstrated that the initial permeability, saturation induction, and power loss were significantly improved when the ZrO₂ concentration was 0.02 wt.%.

Moreover, the performance of a material is closely related to the synthesis technique. Several methods have been developed for synthesizing Mn-Zn ferrite materials. Currently, the most commonly applied synthesis techniques include hydrothermal synthesis, co-precipitation reactions, solid-state reactions, ball milling, and sol-gel auto-combustion methods^[17]. Among these methods, sol-gel auto-combustion method has been recently studied in synthesizing different materials. It combines the virtues of the sol-gel process and low-temperature combustion process^[18]. The products prepared by sol-gel auto-combustion, in general, exhibit better performance due to their smaller particle size, improved purity, surface area, homogeneity in composition, and higher reactivity^[19–21].

Mn-Zn ferrites, such as Mn_{0.5}Zn_{0.5}Fe₂O₄, show good magnetic properties and the effect of doping rare earth element will improve the magnetic properties^[8–11]; however, the effect of co-doping of samarium (Sm) and gadolinium (Gd) on magnetic properties of Mn-Zn ferrites has rarely been studied so far. Therefore, in this study, Sm and Gd co-doping Mn_{0.5}Zn_{0.5}Fe₂O₄ ferrites (Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄, where $x=0.00, 0.005, 0.01, 0.015$ and 0.02) were synthesized via the sol-gel auto-combustion method. The synthetic process and characterization and the magnetic properties of the prepared Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄ were comprehensively investigated.

1 Experimental

1.1 Sample preparation

Nanocrystalline Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄ powders were prepared by the sol-gel auto-combustion method. In this process, all starting materials and reagents employed, including citric acid (C₆H₈O₇·H₂O), manganese sulfate monohydrate (MnSO₄·H₂O), zinc nitrate hexahydrate (Zn(NO₃)₂·6H₂O), iron(III) nitrate nonahydrate (Fe(NO₃)₃·9H₂O), samarium(III) nitrate hexahydrate (Sm(NO₃)₃·6H₂O), gadolinium(III) nitrate hexahydrate (Gd(NO₃)₃·6H₂O), and ethylene glycol (C₂H₆O₂), were of analytical purity.

In this method, MnSO₄·H₂O, Zn(NO₃)₂·6H₂O, Fe(NO₃)₃·9H₂O, Sm(NO₃)₃·6H₂O, and Gd(NO₃)₃·6H₂O were dissolved in distilled water, followed by the addi-

tion of C₆H₈O₇·H₂O and C₂H₆O₂, as complexing agents and fuel, to form a homogeneous solution. The above solution was prepared with a C₆H₈O₇·H₂O to metal ion (Mn, Zn, Fe, Sm, Gd) molar ratio of 1.5:1 and a C₂H₆O₂ to C₆H₈O₇·H₂O molar ratio of 1.2:1. The pH of the solution was adjusted to 7 using ammonia solution. After this stage, the solution was continuously stirred with magnetic agitator at 80 °C. After several minutes, the mixed solution formed a homogeneous solution. Further, the temperature was increased to 90 °C, which led to the formation of a viscous gel. The obtained gel was dried in an oven at 120 °C for 24 h to remove the solvents; the dried gel was then calcined at 300 °C for 4 h in a muffle furnace, where the auto-combustion reaction occurred to produce the precursors. These powders were then calcined at 1200 °C for 4 h to obtain the pure phase Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄ powders. The synthesized Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄ powders were mixed with an appropriate amount of 5 wt.% polyvinyl alcohol as the binder and granulated by using a 180-mesh sieve. Subsequently, the granulated powders were uniaxially pressed at 100 MPa to form green specimens. These materials were then sintered at 1250 °C for 4 h to study the magnetic properties. The experimental flowchart of Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄ preparation by the sol-gel auto-combustion method is shown in Fig. 1.

2.2 Property measurements

Fourier transform infrared (FTIR, Model 8400S, Japan) spectroscopic analysis was performed in the wavenumber range of 400–4000 cm^{–1} on samples pelletized using dry KBr. Xerogels were studied by simultaneous thermogravimetry (TG, Model Netzsch STA 409PC, Germany) under a flow of air at a rate of 10 °C/min. The phase of the calcined powders was analyzed by X-ray diffraction (XRD, Model Rigaku, Japan). Scans were taken with a 2 θ step of 0.02° (over the range of 20°–70°, 2 θ) using Cu K α radiation (18 kV, $\lambda=0.154056$ nm). The microstructure was analyzed by using a scanning electron microscope (SEM, Model S4800, Japan). In order to study the magnetic properties, a vibrating sample magnetometer (VSM, Model MrcroSense EV7, USA) was used at room temperature with a maximum field of 10 kOe.

2 Results and discussion

2.1 FTIR investigation

The FTIR spectra of the as-prepared Mn_{0.5}Zn_{0.5}Sm_xGd_{0.02–x}Fe_{1.98}O₄ xerogels in the range 400–4000 cm^{–1} are shown in Fig. 2. Characteristic absorption peaks are clearly observed at around 3000–3500, 1610, 1380, 1120, 1050, 830, 610, and 400 cm^{–1}. A broad absorption band appearing at around 3000–3500 cm^{–1} in FTIR spectra of the xerogels is characteristic of absorbed water or the

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