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Magnetic and microwave absorption properties of $SrZnCoFe_{16}O_{27}$ powders synthesized by solution combustion method

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ABSTRACT

ZnCo-substituted strontium W-type hexaferrite ($SrZnCoFe_{16}O_{27}$) has been prepared by solution combustion synthesis method. The effects of fuel content ($\phi = 0.75$, 1 and 1.5) on phase evolution, microstructure and magnetic properties were characterized by X-ray diffractometry, infrared spectroscopy, electron microscopy and vibrating sample magnetometry techniques. Fourier transform infrared analysis and theoretical calculations were conducted to determine and control the concentration of metal citrates in solution precursors. Single phase $SrZnCoFe_{16}O_{27}$ powders with the large platelet-like particles were achieved at $\phi = 1.5$ following calcination at 1200 °C, while the M-type hexaferrite and spinel ferrite phases were formed at $\phi < 1.5$. With the increase of fuel content, the saturation magnetization increased from 64.8 to 76.8 emu/g and coercivity firstly increased from 355 to 585 Oe and then decreased up to 34.1 Oe. The microwave absorption measurements in X-band (8–12 GHz) exhibited the maximum absorption of −33.6 dB at 10.4 GHz with an absorption bandwidth of more than 2.5 GHz at −20 dB for the powders synthesized at $\phi = 1.5$.

1. Introduction

Microwave radars have been used to detect distant aircrafts which affect not only their survival in the hostile territory but the mission success rate, resulting in a growing and widespread interest in stealth technology [1,2]. Therefore, it is necessary to explore methods to minimize the radar signal reflected from a vehicle. There are only four basic techniques including of shape design of the vehicle, electromagnetic absorbing materials, passive and active cancellation for reducing electromagnetic wave energy [2]. The platform is firstly designed for reducing radar reflection in the primary threat sectors. However, microwave absorbing materials are required to treat areas whose shape could not be optimized. Therefore, knowledge of the design and application of microwave absorbing materials is vital to minimize the radar reflection signals [3].

The microwave absorbing materials usually consist of dielectric or magnetic materials as filler and a polymer like paraffin, epoxy, etc. [4,5]. The lossy dielectric materials such as carbon, graphite and metal flakes, etc. with the low density and perfect temperature stability have been used for absorbing electromagnetic wave. However, the dielectric materials are usually too bulky for convenient operation in comparison with the magnetic absorbing materials [2,6]. Therefore, the magnetic materials including spinel ferrites and hexaferrites have attracted considerable attention for their utilization as microwave absorbing materials [7–9].

Among all types of hexagonal ferrites, W-type hexaferrite ($Ba(Sr)M_2Fe_{16}O_{27}$, $M = Fe^{2+}$, Co^{2+}, Ni^{2+}, Zn^{2+}, etc.) possesses the highest saturation magnetization (Ms) of 78–80 emu/g and anisotropy field (H_a) of 19 kOe and high ferromagnetic resonance frequencies of 30–60 GHz which make it as a strong candidate for the microwave absorber applications [10–12]. The W-type hexaferrite powders have been synthesized by various techniques such as solid state reaction, coprecipitation, glass crystallization and solution combustion [9,12–14]. The facile operation, low cost, energy-efficient and a short reaction time are benefits of the solution combustion method. The solution combustion method exploits an exothermic reaction between the oxidants (metal salts like nitrates, chlorides, etc.) and a suitable organic fuel, such as citric acid, glycine, urea, etc. [15–17]. The fuels can also act as chelating agents for achieving homogeneous cation distributions in the gels, guaranteeing the direct formation of complicated structures or following calcination at low temperature. The formation of the coordination complexes between cations and citrates strongly

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depends on the pH of the starting solution and fuel contents [18].

In this work, the cation distributions in the dried gels have been evaluated based on the calculation of the citrate complexes and verified by infrared spectroscopy method. Furthermore, the influence of fuel content on the phase, morphology, magnetic properties and microwave absorption of the $SrZnCoFe_{16}O_{27}$ powders have been characterized. The more fuel content led to the homogeneous cation distributions in precursor solutions, promoting the formation of single phase $SrZnCoFe_{16}O_{27}$ with platelet-like particles following calcination at 1200 °C.

2. Experimental procedures

The required amounts of ferric nitrate ($Fe(NO_3)_3\cdot 9H_2O$), strontium nitrate ($Sr(NO_3)_2\cdot 6H_2O$), cobalt nitrate ($Co(NO_3)_2\cdot 6H_2O$), zinc nitrate ($Zn(NO_3)_2\cdot 6H_2O$) and citric acid ($C_6H_8O_7$) were dissolved in the distilled water in which the fuel to oxidant molar ratios (ϕ) were 0.75, 1 and 1.5. After homogenization, the pH value of the solution was adjusted to 7 using 25 wt% ammonia (NH_4OH) solution [19]. The dark brown homogeneous solution was poured into a dish and heated till to transform into a gel while by further heating up to a certain temperature, ignition reaction started from a point which was the most ready for this ignition and then the combustion front propagated spontaneously towards the walls of the dish due to the exothermic reaction. The as-combusted powders were calcined at 1200 °C for 2 h in air atmosphere.

IR spectra in the range of 400–4000 cm^{-1} were measured by Fourier transform infrared (FTIR) spectrometer (8500S SHIMADZU).

Phase evolution was analyzed by PANalytical X-ray diffractometer (XRD) using monochromatic CuKα radiation. The XRD patterns were also submitted to a quantitative analysis by Rietveld method using MAUD program.

The morphology and microstructure of the particles were observed by TESCAN Vega II field emission scanning electron microscopy.

A vibrating sample magnetometer (Meghnatis Daghigh Kavir Co., Iran) was also employed to measure the magnetic properties of the powders at room temperature.

The as-calcined $SrZnCoFe_{16}O_{27}$ powders were mixed with epoxy to prepare X-band rectangular standard samples (0.4×0.9 in^2) in 3:1 weight ratio. The complex permeability μ_r and permittivity ε_r were simultaneously obtained by a vector network analyzer (VNA) which measure the scattering parameters. The scattering parameters show the amount of reflected and transmitted energy. Once the *S*-parameters have been found, they can be converted into the complex electromagnetic properties of the material [20]. Electromagnetic parameters were measured in frequency range of 8–12 GHz at room temperature by a PNA, Agilent E8364B instrument using Agilent 85071D materials measurement software.

3. Results and discussion

Solution combustion synthesis is based on the exothermic reaction between oxidants like metal nitrates and fuels such as glycine, urea, citric acid, etc. [21]. Considering CO_2, N_2 and H_2O as byproducts, the redox processes that are taking place during combustion reactions can be written as follows:

$$Sr(NO_3)_2\cdot 6H_2O + Zn(NO_3)_2\cdot 6H_2O + Co(NO_3)_2\cdot 6H_2O + 16Fe(NO_3)_3\cdot 9H_2O + 15\varphi C_6H_8O_7 + \frac{135}{2}(\varphi-1)O_2 \rightarrow SrZnCoFe_{16}O_{27} + 90\varphi CO_2 + (162+60\varphi)H_2O + 27N_2 \quad (1)$$

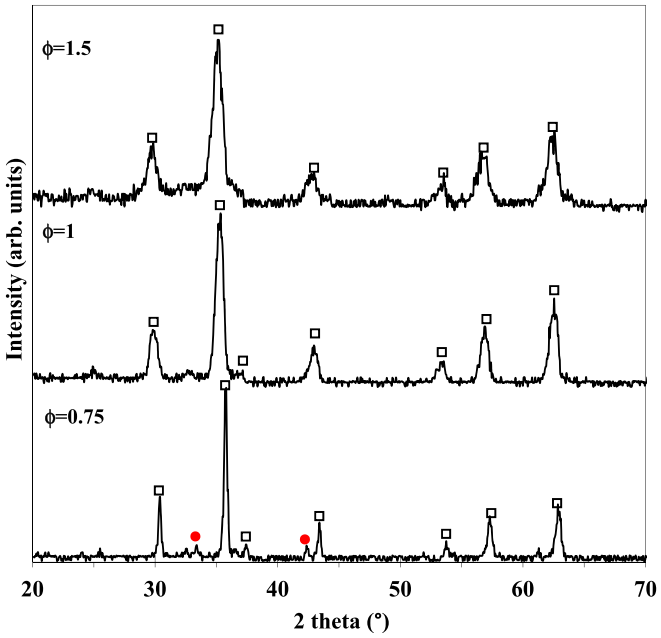

Fig. 1. XRD patterns of the as-combusted powders at the various ϕ values. (: α-Fe_2O_3,: Spinel ferrite).

The stoichiometric mixture ($\phi = 1$) does not require atmospheric oxygen for complete fuel oxidation, while $\phi > 1$ (or <1) implies fuel-rich (or lean) conditions. The fuel to oxidant ratio (ϕ) mainly effects on the combustion behavior, phase, morphology and magnetic properties of the combusted products [22,23]. It is well established that the stoichiometric mixture ($\phi = 1$) shows the maximum adiabatic combustion temperature. The adiabatic temperature decreases steadily for either the leaner or richer mixture due to the need to heat up either the excess reactants or products [24].

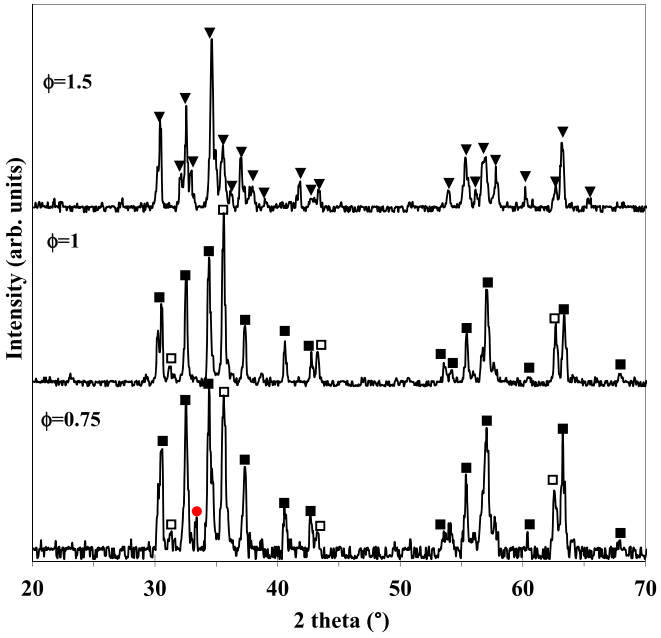

Fig. 2. XRD patterns of the as-combusted powders following calcination at 1200 °C prepared at the various ϕ values (: α-Fe_2O_3,: Spinel ferrite,: M-type hexaferrite and: $SrZnCoFe_{16}O_{27}$).