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Original Research

Preparation and characterization of doubly substituted microwave absorbing material by sol-gel technique for super high frequency applications

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ABSTRACT

Rare earth Dy^{3+} and divalent Mn^{2+} elements substituting W-type hexagonal ferrites $Ba_{1-x}Dy_xZn_2Fe_{16-y}Mn_yO_{27}$ (x = 0, 0.02, 0.06, 0.1 and y = 0, 0.1, 0.3, 0.5) were prepared by sol-gel method. The thermo- gravimetric analysis (TGA) and differential scanning calorimetry (DSC) was carried out to find the temperature at which single phase can be obtained. XRD patterns indicate the presence of the single phase for all the synthesized samples with the absence of any extra peak due to unreacted material and secondary phases. The occurrence of absorption bands at low wave numbers (563 and 446 cm^{-1}), can be assigned to the stretching vibration of metal and oxygen ions in FTIR spectra, which also confirms the single hexagonal phase for prepared material. The grains are of platelet like structure, which enhances the microwave absorption properties of hexagonal ferrites. The synthesized material exhibits the minimum reflection loss of −20.9 dB at 11.8 GHz frequency, which reflects the applications of this material in super high frequency devices. The microwave conductivity of the material increases with frequency.

1. Introduction

Owing to the expanding usage of wireless and super high frequency operating devices, the electromagnetic interference (EMI) and electromagnetic radiations (EMR) are becoming an emerging problem. The public concerns the serious health hazards due to this electromagnetic pollution [1–4]. In medical, some devices like ultrasound and Magnetic resonance imaging can be disturbed by this electromagnetic pollution. So, there is a need to make an enhancement in microwave absorbing fabrication technology to cope up with these problems. Most recently, the researchers are focusing on carbon nano tube, hexagonal ferrites, ZnO composites, Graphene composites etc. as microwave absorbing material [2,5,6]. Due to low cost, high efficiency, high saturation magnetization and high anisotropic field, the hexagonal ferrites could serve well as microwave absorber than the other material. It is well established that the substitution of rare earth elements in pure ferrites alters the structural and microwave absorption properties of hexagonal ferrites [7]. Many researchers have reported the microwave absorption properties of hexagonal ferrites in bulk and thin/thick film form. The structural, magnetic and microwave properties of screen printed thick films of $Ba_xSr_{1-x}Fe_{12}O_{19}$ (BSF) (x = 0.0–1.0) has been reported by Velhal et al. [8]. The maximum crystallite size for 60% Ba^{2+} substituted thick film was 55.44 nm and it also shows the magnetic moment of 76.62 emu/g. The microwave absorption properties were measured in the range of 8–18 GHz. The author found this material suitable for using as microwave absorbing material (MAMs). Stergiou et al. [9] has prepared the $BaSrCo_{2-x}Ni_xFe_{12}O_{22}$ Y-type hexagonal ferrites. It was seen that the complex permeability and permittivity increases with the substitution of Ni^{2+} contents. Furthermore, the samples exhibit the refractive index of 1 GHz, which makes this material an important candidate for UHF antenna design with smaller dimensions.

The main aim of this work was to synthesize microwave absorbing material and understand the substitutional effect of rare earth element Dy^{3+} and divalent Mn^{2+} on structural and microwave absorption properties of hexagonal ferrites.

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2. Experimental procedure

2.1. Sample preparation

The polycrystalline samples of composition $Ba_{1-x}Dy_xZn_2Fe_{16-y}Mn_yO_{27}$ (x = 0, 0.02, 0.06, 0.1 and y = 0, 0.1, 0.3, 0.5) were prepared by Sol-Gel method. The stoichiometric ratios of raw materials ($Sr(No_3)_2$, $Mn(No_3)_2{\cdot}4H_2o$, $Zn(No_3)_2$, $Dy(No_3)_3{\cdot}H_2o$, Iron nitrate, Citric Acid) were mixed in ultrapure deionized water. The PH value was maintained 7–8 by using ammonia solution. The gel was obtained by stirring the solution at 90 °C. The gel was burnt at 130 °C for 24 h in an oven and converted into ash. The ash was grinded in mortar and pestle for 20 min to obtain fine powder. The powder was sintered at 1000 °C for 3 h to get the required W-type hexagonal ferrite phase.

2.2. Sample characterizations

The thermal analysis (of precursor) to confirm the temperature at which required phase can be obtained was carried out by using Mettler Toledo TGA/DSC 1 STAR^e system equipped with Nitrogen gas. The crystalline phase after heat treatment was identified by Schimadzu X-Ray diffractometer equipped with Cu-K_a radiations (λ = 1.5406 Å). The FTIR Spectra was recorded by C91020 Perkinelmer FTIR Spectrometer in the near infrared region over the range of 4000–450 cm^{-1}. The surface morphology was examined by JEOL JSM- 6500 F field emission scanning electron microscopy.

2.3. Microwave absorption measurements

Complex permittivity (ε′, ε″) and permeability (μ′, μ″) measurements were carried out through Agilent E8361A PNA Network Analyzer by using cavity Perturbation Method [10]. The sample was prepared by properly mixing of 20% Paraffin wax and 80% of the ferrite material to insert into the slot of the waveguide. The reflected and transmitted parameter (S_{11}, S_{21}) were measured through Network Analyzer. The complex permittivity and permeability were calculated by using following relations

$$\varepsilon' - 1 = \left[\frac{V_c}{V_s}\right]\left[\frac{f_c - f_s}{2f_s}\right] \quad (1)$$

$$\varepsilon'' = \left[\frac{V_c}{4V_s}\right]\left[\frac{1}{Q_s} - \frac{1}{Q_c}\right] \quad (2)$$

$$\mu' - 1 = \left[\frac{V_c}{kV_s}\right]\left[\frac{f_c - f_s}{f_s}\right] \quad (3)$$

$$\mu'' = \left[\frac{V_c}{2kV_s}\right]\left[\frac{1}{Q_s} - \frac{1}{Q_c}\right] \quad (4)$$

Where V_c and V_s are the volumes of the empty cavity and the sample, f_c and f_s are the resonance frequencies of the empty cavity and the sample, Q_c and Q_s are the quality factor for the empty cavity and perturbed case. $K = 2a^2/(a^2+l^2)$ is the geometrical parameter, where 'l' is the length and 'a' is the breadth of the cavity.

3. Results and discussion

3.1. Thermal analysis

The thermo-gravimetric analysis and differential scanning calorimetry curves of as prepared material are presented in Fig. 1. The TGA curve shows the two endothermic peaks at 230 °C and 450 °C with the weight loss of 53.2% and 34.9% respectively. The weight loss at 230 °C is mainly due to the removal of water molecules from the material and

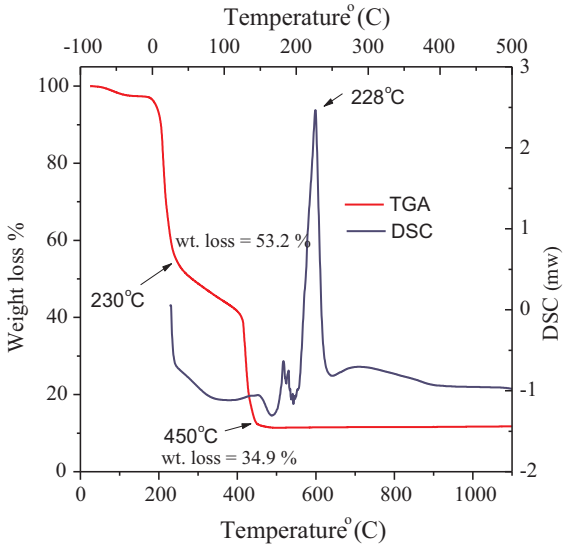

Fig. 1. TGA and DSC analysis of as prepared material.

the weight loss at 450 °C indicates the liberation of gasses e.g CO_2, NO_2 etc. and oxides from the material. Above 450 °C, no endothermic or exothermic peak is observed in TGA curve which indicates the formation of hexagonal phase. The huge exothermic peak at 228 °C in DSC curve reveals the evaporation of adsorbed water molecules from the material [11].

3.2. Phase composition of material

The XRD patterns for all the prepared samples sintered at 1000 °C are presented in Fig. 2. XRD patterns exhibit the single phase for all the samples. There is no extra peak due to unreacted material and secondary phase as confirmed by JCPDS File (52–1868). It is worth noting that the Dy-Mn substitution in pure ferrites presents the single W-type hexagonal phase for all the samples. Huang et al. [12] prepared same single phase unsubstituted material by using adsorbent combustion method by sintering at 1200 °C for 5 h but in present case, the single phase W-type hexagonal ferrites has been prepared by sintering at 1000 °C for 3 h by using sol gel method which is less expensive.

The lattice parameters a (Å) and c (Å) were calculated by using the formula;

$$\frac{1}{d_{hkl}^2} = \frac{4(h^2 + hk + k^2)}{3a^2} + \frac{l^2}{c^2} \quad (5)$$

where d_{hkl} is the interplanar crystal spacing of the corresponding crystallographic plane in the XRD pattern and h, k and l are the

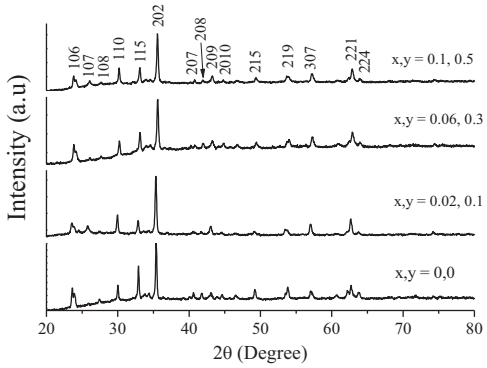

Fig. 2. XRD patterns for $Ba_{1-x}Dy_xZn_2Fe_{16-y}Mn_yO_{27}$ (x = 0, 0.02, 0.06, 0.1 and y = 0, 0.1, 0.3, 0.5) W-type hexagonal ferrites sintered at 1000 °C.