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Novel lead – free relaxor ferroelectric based on $Mg_{1-x}Sr_xTiO_3$ (MTO:Sr) nanostructured for radar absorbing applications

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ABSTRACT

Novel relaxor ferroelectric based electromagnetic wave shielding at microwave frequency materials, containing Sr doped $MgTiO_3$, were developed and investigated. The structural and morphological characteristics of the products were analyzed using scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS). The samples were found to sensitive to Sr doping level and the presence of Sr was confirmed. The optical band gap of pure $MgTiO_3$, 1% Sr doped, 3% Sr doped and 5% Sr doped $MgTiO_3$ was estimated to be 3.75, 4.02, 4.06 and 4.10 eV, respectively. The microwave dielectric properties of Sr-doped $MgTiO_3$ as a function of microwave frequency in the range of 1–20 GHz were examined. With Sr - doping, the microwave dielectric properties of the $MgTiO_3$ is improved considerably. The applicability of as fabricated Sr doped $MgTiO_3$ for electromagnetic wave shielding effectiveness at microwave frequency was studied. The dependence of the polarization of $MgTiO_3$ as a function of Sr content was examined in details. This makes the Sr doped $MgTiO_3$ good candidates for uses as high permittivity materials and it will be versatile for various electromagnetic wave shielding applications at microwave frequency.

1. Introduction

Lead – free ceramic materials have been extensively investigated for technological, manufacturing and fabrication applications such as solar cells [1–3]. Researchers have devoted resources to the fabrication and manufacturing processes of the electronic devices [4]. Lead – free ceramic materials have been used in fabrication of perovskite solar cells for low-cost energy generation. Facile technologies that have been developed to break the paradigm of fabrication of perovskite solar cells abound; notable among them is a recent work by Bag et al. [5]. By using a desktop inkjet printer, they demonstrated an efficient method of fabricating perovskite solar cells. This technology is particularly promising method for perovskite solar cell fabrication. The solid state hole transport mechanism enhances the solar cell stability. The efficiency of these solar cells is about 20% [6] and it presents a promising material worthy of the current considerations [7]. Perovskite have been utilized for the fabrication of light emitting diodes (LEDs), electroluminescent devices (ELDs), scintillators, sensors, fuel cells, superconductors and more [4]. Perovskite are characterized by ABX3 formula, where A and B represent cations, and X is an anion. The A-site cation is normally electropositive and larger than the B-site cation, while the X-site is commonly occupied by oxide/halide ions [8]. The basic structure of the perovskites is cubic; however, structural distortions can occur giving way to a variety of structures such as tetragonal, orthorhombic, trigonal and monoclinic. Perovskites such as magnesium titanate $MgTiO_3$ (MTO) has been extensively investigated due to its interesting properties and wide range of applications in optoelectronic materials including photovoltaic (PV), photosensors and light emitting diodes (LED), [9,10]. Bhalla et al. [11] have reported that the ferroelectric, piezoelectric and superconductive properties of perovskite oxides can be tuned by an appropriate chemical composition. Ferroelectric properties have been reported to affect the photovoltaics properties of perovskite oxides [12,13]. The perovskite oxide crystal structure lends itself to band gap manipulation which can affect its visible light absorption properties. Reports in literature have confirmed visible light absorption for perovskite materials [14–18] It was demonstrated clearly by appropriate modification such as doping or manipulation of composition, sample showed significant response to solar light. Cai et al. [19] have studied the photoactivity of Cr doped $SrTiO_3$. They reported improved photoactivity for Cr/Sr wt % of 0.5%–2%, while Cr/Sr wt % of 3%–10% experienced decrease in activity. Yu and

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his research group have used one-step hydrothermal method to fabricate Cr-doped $SrTiO_3$ samples [20]. They calculated a band gap of 2.45 eV and 3.21 eV for Cr-doped $SrTiO_3$ and pure $SrTiO_3$ respectively. Ferri et al. [21] have prepared $MgTiO_3$ powders using complex polymerization method. They observed optical band gap values ranging from 3.07 eV to 4.05 eV for heat treated sample from 450 °C to 700 °C respectively. Tseng et al. [22] reported the growth of $Mg(Zr_{0.05}Ti_{0.95})O_3$ thin films by the sol–gel process. In a more recent work, Zr doped $MgTiO_3$ materials were prepared using a sputtering method [23]. The optical band gap values have been changed with thermal annealing. With this connection, the aim of the present study is to synthesize $Mg_{1-x}Sr_xTiO_3$ (MTO:Sr) nanoparticles through solution method. The effect of Sr - doping on the microstructure and optical properties of the ceramic products were mainly investigated. The applicability of $Mg_{1-x}Sr_xTiO_3$ for electromagnetic wave shielding effectiveness at microwave frequency was studied in details. Furthermore, the effect of Sr - content on ferroelectric properties of as fabricated products was examined, too.

2. Experimental details

2.1. Materials and synthesis

The used precursors are magnesium acetate $(CH_3CO_2)_2Mg$ (98%, Aldrich), titanium butoxide $Ti[O(CH_2)_3CH_3]_4$ (99%, Aldrich), strontium nitrate ($Sr(NO_3)_2$) (99.93%, Aldrich), ethylene glycol, $C_2H_6O_2$ (99.5%, Synth), and citric acid $C_6H_8O_7$ (99.5%, Synth). Firstly, titanium butoxide was dissolved in an aqueous solution of citric acid and it was stirred was stirred at 90 °C. The nominal value of $(CH_3CO_2)_2Mg$ was put into Ti citrate solution and then, $C_2H_6O_2$ was added to the solution. The prepared solutions were ultrasonically stirred using a FYRONIX sonicator. Finally, the various molar ratios of $Sr(NO_3)_2$ were added to the solution to obtain MTO:Sr powder samples. The powders was obtained at 350 °C for 2 and they were thermally treated at 700 °C in a furnace.

2.2. Characterization techniques

The surface morphologies of MTO:Sr powder samples were investigated using a JEOL 7001 scanning electron microscopy (SEM) Optical properties of MTO:Sr powder samples were examined using Shimadzu 3600 UV/VIS/NIR spectrophotometer at room temperature. The electromagnetic shielding properties such as reflection loss (SER), absorption loss (SEA) and return loss (RL) were determined by a ANRITSU network analyzer. The polarization-electric field measurements of the samples were measured at room temperature with a Radiant Precision ferroelectric testing system.

3. Results and discussion

3.1. Morphological and elemental properties of Sr doped Mg-TiO_3

The surface morphologies of Sr doped Mg-TiO_3 powder materials were examined by a SEM. Fig. 1(a–d) shows the SEM images of un-doped MTO samples and with 1% Sr doped, 3% Sr doped and 5% Sr doped, respectively. From the obtained SEM micrographs, it is clear that the synthesized products are consisting of particles uniformly distributed throughout the surface. The particles were irregular in shape for all samples but highly agglomerated for the Sr doped samples. The images also show appreciable densification of the Sr doped samples. This behavior confirms the similar observation already reported in the literature [24,25]. The composition of the fabricated nanoparticles were examined by energy dispersive spectroscopy (EDX) and demonstrated in Fig. 2. As can be seen from the EDX spectrum, the synthesized nanoparticles are made by Mg,

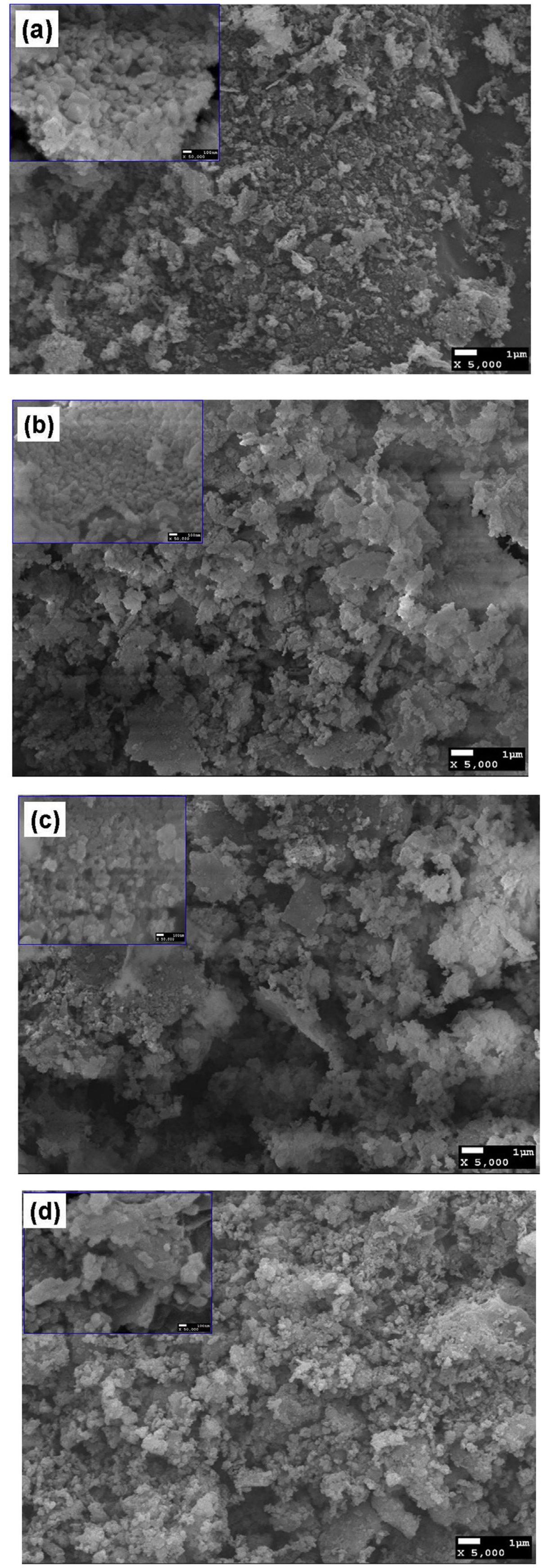

Fig. 1. SEM micrographs of samples (a) undoped MTO (b) 1% Sr doped (c) 3% Sr doped, and (d) 5% Sr doped MTO.