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A new multicomponent material based on carbonyl iron/carbon nanofiber/lanthanum–strontium–manganite as microwave absorbers in the range of 8–12 GHz

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ABSTRACT

We report the design of a new multicomponent microwave absorber in the X band based on carbonyl iron (CI)/carbon nanofiber (CNF)/lanthanum–strontium manganese oxide (LSMO). The citrate precursor method has been used to synthesize LSMO nanopowder. The phase identification has been investigated using XRD patterns. Scanning electron microscope (SEM), vibrating sample magnetometer (VSM) and vector network analyzer (VNA) are used to analyze the morphology of the different components and the magnetic, electromagnetic and microwave absorption properties of the final composite absorbers. To the best of our knowledge, the use of this class of multicomponent microwave absorber, has not been explored before. The results indicate that substituting CI by increasing amounts of CNF and LSMO, not only decreases the weight of the designed absorber, but also has a synergistic effect that promotes its attenuation properties. The absorber of 2 mm thickness and only 30 wt% of loading ratio exhibits an average reflection loss of −8.75 dB over the range 9–12 GHz, while its corresponding absorber without LSMO has only a RL around −5 dB.

1. Introduction

Nowadays, due to the electromagnetic interference caused by rapid development of electronic communication devices, electromagnetic waves absorbers are needed with features like broad absorption bandwidth, strong absorption, simple functionality, low density and thickness. Substantial progress has been achieved in the field of radar absorbent production [1–5]. Generally, microwave absorbing materials are classified into two groups, namely, structural materials and coatings. However, wave-absorbing coatings are more practical because of their especial properties such as low cost, convenient preparation, and the possibility of making thinner absorbers, etc. [6–9].

The fillers of the coatings can be classified into three types according to the wave absorption mechanism: electric loss, magnetic loss or dielectric loss materials [10]. Inherently conductive polymers and carbon materials exhibit high electric loss tangent and considered as electric attenuation absorbers. The electromagnetic energy is mostly attenuated as in ohmic resistor [11–13]. Iron carbonyl and ferrites materials are magnetic loss absorbers and possess high magnetic loss tangent. The electromagnetic energy is attenuated by hysteresis loss and magnetic domain resonance. Whereas many ceramic materials such as barium titanate are considered as dielectric loss absorbers, they attenuate the electromagnetic energy by electronic and ionic polarization. Microwave absorbers based on magnetic materials possess some characteristics such as high complex permeability and therefore high absorption with narrow bandwidth in the low frequency range. But when the dielectric materials are the fillers of the coating, the absorption properties are altered and thick absorbers are needed [13–17]. Dielectric materials have lower density than magnetic materials but they can be used for high range frequency. Therefore in order to achieve an absorber with a suitable absorption in the low frequency range and with minimum thickness and weight, multicomponent absorbents are used such as carbonyl-iron powder and carbon black [9], $La_{0.5}Sr_{0.5}MnO_3/La(OH)_3$ [13], CNF/epoxy composites [18], $NiFe_2O_4$/carbon nanofibers [19] and Fe_3O_4/CNFs [20].

One of the conventional magnetic powders that are used as fillers in microwave absorbers is carbonyl iron (CI) powder with high saturation magnetization and high Snoek's limit. One of the most important characteristics of this magnetic powder is the

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relatively high permeability in the gigahertz frequencies compared to ferrite powders, thus, offering the possibility of making thin absorbents [21–23]. However, tuning the values of magnetic permeability, dielectric constant and their matching, necessitate compositing this powder with other materials, like carbon nano fiber (CNF) and lanthanum strontium manganese oxide (LSMO). In fact, using CNFs with their high electrical conductivity and lanthanum-strontium-manganese oxide as a new class of magnetic nanoparticles at low filler concentration may lead to high electromagnetic losses and offers the possibility of making thin and light absorbents. Moreover, small amounts of CNFs in the matrix may cause substantial variations in the complex permittivity of the composites [15–24].

In this study, we aim to design a relatively lightweight absorber with enhanced microwave absorption properties in the X band. It consists of a gradual substitution of carbonyl iron, a conventional microwave absorber, with the lightweight carbon nanofibers and lanthanum-strontium-manganese oxide. To the best of our knowledge, this multicomponent single layer constitutes a new class of microwave absorber in the X band that has not been explored yet. The electromagnetic characteristics and microwave absorption properties of the samples with various percentages of materials were investigated in details.

2. Materials and test method

Carbonyl iron (CI), carbon nanofiber (CNF) and lanthanum-strontium-manganese oxide (LSMO) is used as magnetic and dielectric fillers in the microwave absorber samples. Carbonyl iron and carbon nanofiber are used as purchased. However lanthanum–strontium–manganese oxide powder was synthesized via citrate precursor method that will be explained in the next section.

2.1. Synthesis of LSMO powder

LSMO powder is synthesized by the citrate precursor method from lanthanum nitrate $La(NO_3)_3$, strontium nitrate $Sr(NO_3)_2$ and manganese nitrate $Mn(NO_3)_2$ with high purity of 99.99%. To achieve $La_{0.6}Sr_{0.4}MnO_4$ composition, stoichiometric amounts of nitrates are dissolved in 200 cc distilled water with constant stirring of 200 rpm and temperature of 80 °C. After stirring for an hour a clear solution is obtained. Subsequently citric acid equivalent to the metal nitrates moles is added to the solution. The homogenized solution slowly evaporated to get a gel, which is decomposed for two hours at 200 °C and annealed at 700 °C for 4 h.

2.2. Preparation of the samples

First, the CI powder is milled by planetary ball mill (Retsch PM100, with 10 hardened steel balls) for 15 h with 300 rpm to decrease grain size. In order to measure the complex permittivity and permeability and also the microwave absorption properties, CI, LSMO and CNF powders are uniformly mixed in paraffin matrix with weight ratio of 30:70. At the beginning, accurate amounts (30 wt.) of CI, LSMO and CNF powders are added into the molten paraffin and homogenized well with ultrasonic vibration for 10 min. Then, single layer microwave absorber samples are prepared using $25 \times 10 \times 2$ mm molds. In this study, four specimens are prepared with different content of CI, LSMO and CNF powders. The weight ratios of them are given in Table 1.

2.3. Evaluation of properties

Phase analysis of the LSMO powder is performed with X-Ray diffraction analysis (XMD 300) in the range of 10–70° with CuK_α radiation. Scanning electron microscope (SEM, MIRA3 TESCAN) is used for determining the morphology and average particle size of the obtained powder. The magnetic properties of the composites are measured with vibrating sample magnetometer (VSM) at room temperature. Microwave absorption characteristics (ε, μ, RL) in the range of X band frequency (8–12 GHz) are measured using vector network analyzer (VNA, Agilent 8510 C) by a waveguide method.

Table 1
The weight percentage of the CI, LSMO and CNF powders.

Sample code	CI (%)	LSMO (%)	CNF (%)
CLC (99-0-1)	99	0	1
CLC (95-3-2)	95	3	2
CLC (91-6-3)	91	6	3
CLC (87-9-4)	87	9	4

3. Results and discussion

3.1. Phase identification analysis and SEM morphology

The result of X-ray diffraction analysis from LSMO powder is shown in Fig. 1. All the characteristic peaks at 22.94°, 32.74°, 40.23°, 46.86°, 52.70°, 58.26° and 68.28° are related to lanthanum-strontium-manganese oxide single phase with the $La_{0.6}Sr_{0.4}MnO_3$ chemical formula that can be indexed to JCPDS no. 50-0308 with rhombohedral crystal structure and R-3c space group. It is clear that there is no impurity formed in the powder due to the selection of appropriate values for annealing time and temperature. In fact, to calculate the crystallite size of the LSMO powder, prominent broad peak is chosen at 32.74° which corresponds to Miller indices (1 0 4). The calculated value of crystallite size according to the Scherrer formula is 32 nm. The inset of Fig. 1 presents the quantitative analysis with MAUD software for estimating the weight percentage of LSMO phase in the diffraction pattern. According to the results, there was 100 wt% of the mentioned phase in the powder which confirms the formation of pure LSMO phase via citrate precursor method.

The SEM images related to magnetic and dielectric components in the microwave absorber samples are shown in Fig. 2. The image of CI powder after milling shows the platy like morphology with an average particle size of 17 μm. Also, in Fig. 2a the presence of agglomerates of particles with different irregular shapes are is evident due to magnetic intrinsic property of CI powders. LSMO

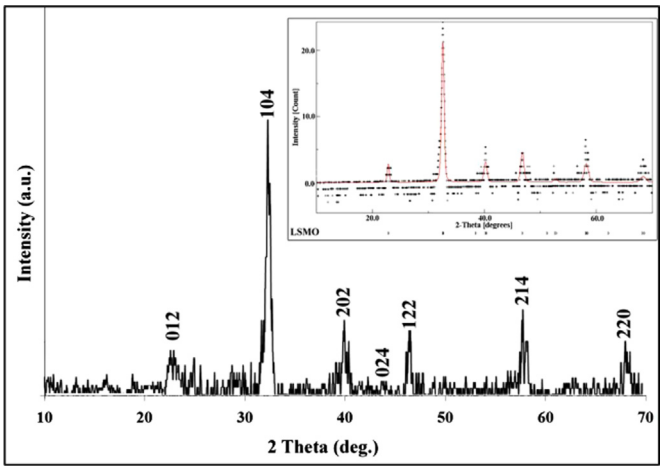

Fig. 1. Diffraction pattern of LSMO powder.