Preparation and Microwave Absorption Property of Sr_2FeMoO_6 Powder

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Abstract: Double perovskite oxide Sr_2FeMoO_6 powder is prepared by a solid state reaction method. The microwave absorption properties of Sr_2FeMoO_6 and paraffin wax composites are studied in the frequency range of 2—18 GHz at room temperature. The optimum absorption −36.7 dB is achieved at 17.7 GHz with a matching thickness of 5.0 mm, which indicates that Sr_2FeMoO_6/paraffin composites can be potential microwave absorbers in a relatively high frequency range. The excellent microwave absorption properties are attributed to the good electromagnetic match between dielectric loss and magnetic loss. The dielectric loss is considered to be caused by orientation polarization and interfacial polarization, while the magnetic loss is caused by natural resonance in the low frequency range, eddy current loss as well as exchange resonance in the high frequency range.

Key words: double perovskite; solid state reaction; microwave absorption property

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0 Introduction

With the electromagnetic interference pollution becoming more and more serious, microwave absorption materials have attracted a lot of research interests. Extensive researches have been carried out to improve the microwave absorption properties of traditional absorption materials (such as ferrite[1], carbonyl iron[2], SiC[3]) or develop new types of absorption materials (such as perovskite manganese oxides[4], carbon nanotubes[5], conducting polymers[6], metamaterials[7]). In general, the excellent microwave absorption properties are believed to be resulted from the effective complementarity between the relative complex permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) and permeability ($\mu = \mu' - j\mu''$). Either only the dielectric loss or only the magnetic loss may cause performance degradation due to the imbalance of the electromagnetic match[8]. Thus, magneto-dielectric materials may have a special advantage upon other dielectric materials in microwave absorbing.

Double perovskite Sr_2FeMoO_6 as a magneto-dielectric material has been studied a lot due to the reported substantial low-field magnetoresistance (LFMR) and higher Curie temperature $T_C \approx 400$ K compared with perovskite manganese oxides[9]. Sr_2FeMoO_6 shows ferrimagnetic metallic state with strong magnetism and low resistivity at room temperature[10], which makes it possible for applications as high performance microwave absorption materials. However, as far as we know, few researches have been conducted on the microwave absorption properties of Sr_2FeMoO_6. Most recently, Xi et al. prepared Sr_2FeMoO_6 by sol-gel method and investigated the microwave absorption properties of Sr_2FeMoO_6/epoxy resin composite[11]. Although good microwave absorption properties are obtained, some improvement and

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better understanding of the microwave loss mechanism are still needed.

In this paper, Sr_2FeMoO_6 is synthesized by conventional solid state reaction method since its simpler preparation process. The microwave absorption properties and electromagnetic loss mechanism of Sr_2FeMoO_6/paraffin composite at 2—18 GHz are investigated.

1 Experiment

Sr_2FeMoO_6 is prepared by conventional solid state reaction method. $SrCO_3$ (≥99%), Fe_2O_3 (≥98.6%) and MoO_3 (≥99.5%) are used as starting materials. Stoichiometric amounts of powder are firstly mixed and milled using a high energy ball milling for 3 h. Then the powder are pressed into pellets and sintered at 1 280 ℃ in a H_2/Ar (5%/95%) reduction flow for 6 h. Finally, the sintered sample is grounded to microsized powder.

The crystal structure is examined by powder X-ray diffraction (XRD) technique with Cu Kα radiation. The particle morphology is characterized by scanning electron microscope (SEM). The prepared Sr_2FeMoO_6 is mixed with paraffin wax according to mass ratio of 8 ∶ 3 and then pressed into annular sample with the inner diameter of 3.04 mm, outer diameter of 7 mm and thickness of about 2 mm. The complex relative permittivity and permeability are measured by the co-axial method on an Agilent N5244A microwave vector network analyzer at 2—18 GHz.

2 Results and Discussion

Fig. 1 shows XRD pattern of Sr_2FeMoO_6 powder. It can be seen that single phase of Sr_2FeMoO_6 without detectable secondary phase or impurity is obtained.

SEM image (Fig. 2) of Sr_2FeMoO_6 powder shows that part of the ground powder are flaky like, which may improve the microwave absorption properties[12]. Besides, there exist sharp corners in each powder. It is reported that the sharp corners are also beneficial for electromagnetic energy dissipation because of tip discharge effect[13].

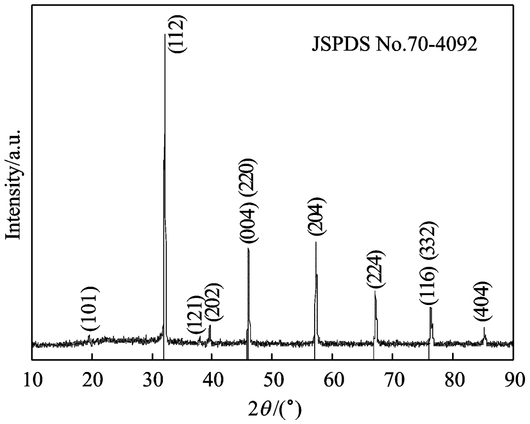


Fig. 1 XRD pattern of prepared Sr_2FeMoO_6 powder

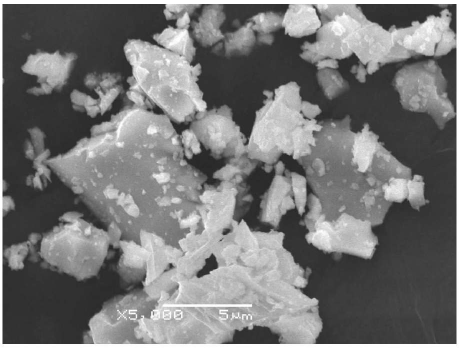
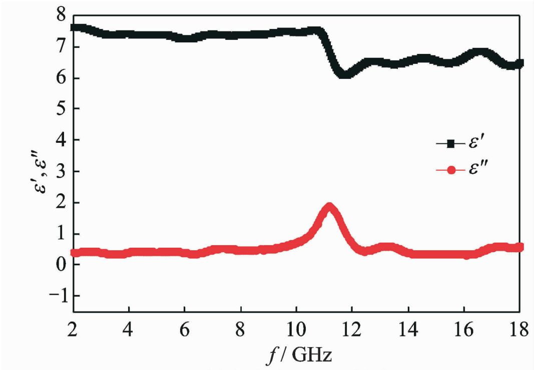
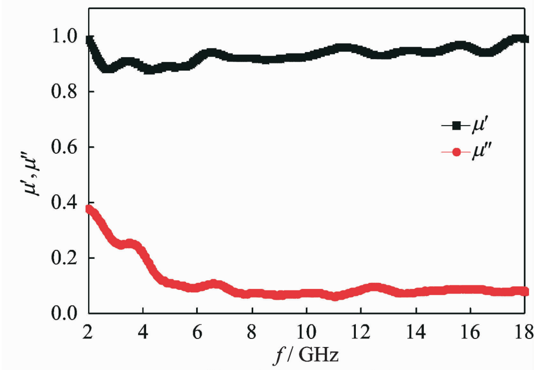


Fig. 2 SEM image of prepared Sr_2FeMoO_6 powder

Fig. 3 shows the frequency dependence of the relative complex permittivity and permeability of Sr_2FeMoO_6/paraffin composite. As shown in Fig. 3 (a), the complex permittivity is typical for nonlinear dielectric resonances and the resonance peak is around 11 GHz. Generally, dielectric loss may be ascribed to electronic polarization, atomic or ionic polarization, orientation polarization, and interfacial polarization[14]. The former two polarization processes mainly occur in the ultraviolet and infrared light ranges, respectively, while the latter two polarization processes occur in the microwave band. This means that the former two polarization processes can be ignored at 2—18 GHz. Orientation polarization originates from the intrinsic dipoles changing polarization direction under the applied electrical field. Interfacial polarization arises from the charge accumulation at the interface of two phases, whose dielectric constant and conductivity are different. Sr_2FeMoO_6 is a magneto-dielectric material and there exists large amount of interface between Sr_2FeMoO_6 particles and paraffin matrix, so it is believed that orientation polarization and interfacial polarization are the main reasons which cause dielectric loss in our experiment. The existing anti-site defects (ASD) and anti-phase domains (APD) in Sr_2FeMoO_6 particles is expected to enhance the dielectric loss[11]. In addition, the 4d electron of Mo^{5+} is itinerant, which is also beneficial for microwave absorbing owing to the electrons hopping between $Fe^{3+}—O—Mo^{5+}—O—Fe^{2+}$ [15].



(a) Relative permitivity



(b) Relative permeability

Fig. 3 Relative permitivity and relative permeability of Sr_2FeMoO_6/paraffin composite as function of frequency

From Fig. 3 (b), it can be seen that the value of μ' changes slightly at 2—18 GHz, however, the value of μ'' decreases in 2—6 GHz range and then fluctuates with the increase of frequency. It is known that there are five major kinds of possible mechanisms which cause microwave magnetic loss as follows: magnetic hysteresis, domain-wall resonance, eddy current loss, natural resonance, and exchange resonance[16, 17]. The magnetic-hysteresis loss can be neglected in weak fields and domain-wall resonance usually occurs in 1—100 MHz range[14]. Therefore, neither the magnetic-hysteresis loss nor the domain-wall resonance is the key contribution to magnetic loss in the present frequency range. If the eddy current loss plays a leading role in magnetic loss, the value of $\mu'' / (\mu'^2 \cdot f)$ should be constant as frequency varies[14, 17]. From Fig. 4, we can see that the eddy current loss is predominant in frequency range higher than 6 GHz while it is negligible in lower frequency range. As mentioned above, the value of μ'' decreases in the low frequency range, from which we can speculate that the natural resonance may occur at a frequency a little bit lower compared with 2 GHz. Therefore, the magnetic loss in the frequency range of 2—6 GHz may be attributed to the natural resonance. We also notice that there exist some small peaks of μ'' in 6—18 GHz range. According to Ref. [18], the natural resonance occurs at a lower frequency compared with the exchange resonance[18]. Thus, the fluctuation of μ'' in the frequency range of 6—18 GHz may be caused by exchange resonance.

The natural resonance frequency f_r is much

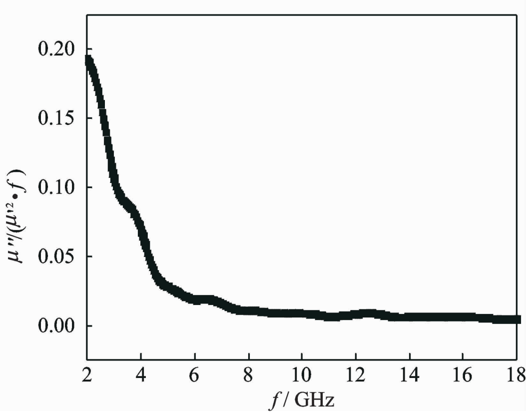


Fig. 4 $\mu''/(\mu'^2 \cdot f)$ spectra of Sr_2FeMoO_6/paraffin composite lower than the result of Ref. [11], which may be caused by different grain sizes. As we know, the anisotropy constant can be significantly increased with the decreasing grain size according to $K_{\mathrm{eff}} = K_{\mathrm{V}} + 6K_{\mathrm{S}}/d$[19], where K_{V} and K_{S} refer to the volume and surface contributions to anisotropy, respectively, d is grain size. The size of grain we obtained is much larger than that in Ref. [11] due to the higher sintering temperature and different preparing method. Therefore, the anisotropy constant may be much smaller. As a consequence, the effective anisotropy field H_{eff} ($H_{\mathrm{eff}} = 4|K_{\mathrm{eff}}| / (3\mu_0 M_{\mathrm{S}})$, μ_0 and M_{S} are permeability of vacuum and saturation magnetization, respectively) will decrease, which will decrease the natural resonance frequency ($2\pi f_{\mathrm{r}} = \gamma H_{\mathrm{eff}}$, where γ is the gyromagnetic ratio)[20].

Fig. 5 shows the reflection loss(RL) spectra of $\mathrm{Sr_2FeMoO_6}$/paraffin composite based on the transmission line theory

$$\mathrm{RL} = 20\log|(Z_{\mathrm{in}} - Z_0)/(Z_{\mathrm{in}} - Z_0)|$$

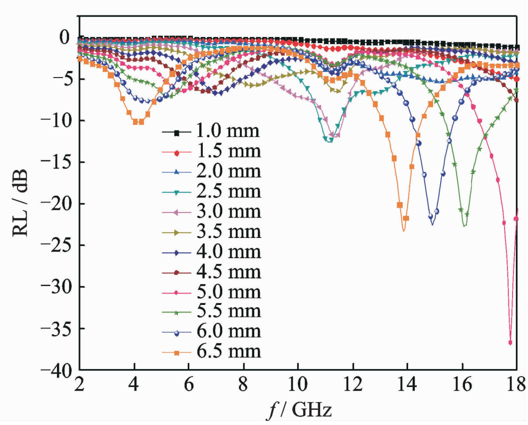


Fig. 5 Frequency dependences of RL for $\mathrm{Sr_2FeMoO_6}$/paraffin composite

where Z_0 represents the characteristic impedance of free space and Z_{in} the input impedance at the absorber/free space interface, which can be expressed as follows

$$Z_{\mathrm{in}} = Z_0\sqrt{\frac{\mu}{\varepsilon}}\tanh\left[\mathrm{j}\,\frac{2\pi ft}{c}\sqrt{\mu\varepsilon}\right]$$

where f is the frequency of incident electromagnetic wave, c the velocity of light, and t the thickness of the composite. ε and μ are the relative complex permittivity and permeability, respectively.

The minimum reflection loss of the $\mathrm{Sr_2FeMoO_6}$/paraffin composite reaches −36.7 dB at 17.7 GHz with a thickness of 5.0 mm, as shown in Fig. 5. This indicates that the $\mathrm{Sr_2FeMoO_6}$/paraffin composites can be used as good microwave absorbers in the higher frequency range. This result is somewhat different from that in Ref. [11], which may be attributed to the different synthesize methods, insulating matrices and $\mathrm{Sr_2FeMoO_6}$ volume concentrations. We can also see that the peak frequency of RL shifts to lower frequency as thickness increases, and there will be more than one peak when the thickness is greater than a critical value (for sample whose thickness is 1 mm, there is no RL peak in the studied frequency). This phenomenon is similar to destructive interference according to quarter-wavelength ($\lambda/4$) matching model[21]

$$t = \frac{nc}{4f_m\sqrt{|\mu\varepsilon|}} \qquad n = 1,3,5,\cdots$$

where f_m is the peak frequency of RL. It can be clearly seen that different values of n can result in multiple peaks, and f_m decreases when t increases.

3 Conclusions

$\mathrm{Sr_2FeMoO_6}$ is prepared by conventional solid state reaction method. The minimum reflection loss of $\mathrm{Sr_2FeMoO_6}$/paraffin composite reaches −36.7 dB at 17.7 GHz with a thickness of 5.0 mm. The excellent microwave absorption properties mainly result from the co-occurrence of the dielectric loss and magnetic loss. As far as we are concerned, the dielectric loss mainly comes from the orientation polarization and interfacial polarization. The magnetic loss origins from the natural resonance in the low frequency range and eddy current loss together with exchange resonance in the high frequency range.

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