Double-layer microwave absorber of nanocrystalline strontium ferrite and iron microfibers*

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Microwave absorption properties of the nanocrystalline strontium ferrite ($SrFe_{12}O_{19}$) and iron (α–Fe) microfibers for single-layer and double-layer structures are investigated in a frequency range of 2 GHz–18 GHz. For the single-layer absorbers, the nanocrystalline $SrFe_{12}O_{19}$ microfibers show some microwave absorptions at 6 GHz–18 GHz, with a minimum reflection loss (RL) value of −11.9 dB at 14.1 GHz for a specimen thickness of 3.0 mm, while for the nanocrystalline α–Fe microfibers, their absorptions largely take place at 15 GHz–18 GHz with the RL value exceeding −10 dB, with a minimum RL value of about −24 dB at 17.5 GHz for a specimen thickness of 0.7 mm. For the double-layer absorber with an absorbing layer of α–Fe microfibers with a thickness of 0.7 mm and matching layer of $SrFe_{12}O_{19}$ microfibers with a thickness of 1.3 mm, the minimum RL value is about −63 dB at 16.4 GHz and the absorption band width is about 6.7 GHz ranging from 11.3 GHz to 18 GHz with the RL value exceeding −10 dB which covers the whole K_u-band (12.4 GHz–18 GHz) and 27% of X-band (8.2 GHz–12.4 GHz).

Keywords: nanocrystalline microfibers, microwave absorber, double-layer structure

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1. Introduction

The electromagnetic wave absorbing materials for both commercial and military applications have attracted numerous studies.[1−4] The absorbing materials can usually be classified as single-layer absorber and multi-layer absorber. Lu *et al.*[5] reported on the single-layer absorber of the amorphous Ni–P nanotubes. Sun *et al.*[6] investigated the single-layer absorber containing hierarchical dendrite-like magnetic materials of Fe_3O_4, γ–Fe_2O_3 and Fe. As one layer absorbing material is hard to simultaneously meet the demands for broad frequency (f) range, light in weight and strong absorption, multi-layer microwave absorbing materials therefore are required technologically. Recently, researchers were interested in double layer and multi-layer microwave absorbers.[7−9] Qing *et al.*[10] investigated the single-layer and the double-layer microwave absorbers of $BaTiO_3$ and carbonyl iron powders with various constituents. The results showed that the double-layer absorber with a thickness of 1.4 mm, which consisted of the matching layer with 50 wt% $BaTiO_3$ and the absorption layer with 60 wt% $BaTiO_3$ and 20 wt% carbon iron, had a much better microwave absorbing characteristic than the single-layer absorber, with a band width of 10.8 GHz–14.8 GHz where the reflection loss (RL) value was below −10 dB and the minimum RL value was −59 dB at 12.5 GHz.

Hexagonal ferrites are common microwave absorbers in the gigahertz (GHz) range because of their high saturation magnetization, large anisotropy field, excellent chemical stability and high microwave magnetic loss.[11] The strontium ferrite is a typical hexagonal ferrite and has been investigated extensively.[11−13] Ghasemi *et al.*[14] reported on the electromagnetic properties and the microwave absorbing characteristics of the doped barium hexaferrite. Meanwhile, as ferromagnetic metals each have a high saturation magnetization and complex permeability, they can be good microwave absorbers.[15−18] However, the ferromagnetic metals usually each have a high electric conductivity and their permeability will drastically decrease at high frequencies due to the eddy current effect induced by electromagnetic waves.[19] As the hexagonal ferrite fibers and ferromagnetic metallic fibers generally feature shape anisotropy, they can be used as structural function materials. According to our previous preparation of the nanocrystalline M-type ferrites and ferromagnetic metals microfibers,[20,21] in the present

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work we investigate the electromagnetic characteristics and microwave absorption of the nanocrystalline $SrFe_{12}O_{19}$ and α–Fe microfibers.

2. Experiment

The nanocrystalline $SrFe_{12}O_{19}$ microfibers and α–Fe microfibers were prepared by the citrate–gel process at a calcination temperature of 950 °C for 4 h and by the citrate–gel precursor reduction process under the reduction atmosphere (V (H_2): V (N_2) = 1:4) at 700 °C for 2 h, which was described in detail in our previous investigations.[20,21] The microfiber morphology was investigated with field emission scanning electron microscope (FESEM, JSM-7001F) and the X-ray diffraction (XRD) patterns were collected on a Rigaku D/Mmax2500PC diffractometer with Cu–Kα radiation. The magnetic properties of the microfibers were investigated at room temperature using a vibrating sample magnetometer (VSM).

The microfibers-wax composites for microwave absorption estimation were prepared by mixing 67 wt% nanocrystalline $SrFe_{12}O_{19}$ microfibers and 33 wt% wax (Specimen 1) and 50 wt% nanocrystalline α–Fe microfibers and 50 wt% wax (Specimen 2), respectively. Then, the microfibers-wax composite was cast into toroidal-shape specimens each with an inner diameter of 3.04 mm and an outer diameter of 7.00 mm. The scattering parameters of the specimens were measured using a Hewlett Packard 8722ES network analyzer. The relative permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) and permeability ($\mu_r = \mu' - j\mu''$) values for these specimens were determined from the scattering parameters measured at a frequency range of 2 GHz–18 GHz.

For a single layer microwave absorber, the RL value was calculated from the complex relative permeability and permittivity at a given frequency and absorber thickness by the following equations:[22,23]

$$Z_{in} = Z_0(\mu_r/\varepsilon_r)^{1/2} \tanh[\, j(2\pi f d/c)(\mu_r \cdot \varepsilon_r)^{1/2}], \quad (1)$$

$$RL = 20 \log |(Z_{in} - Z_0)/(Z_{in} + Z_0)|\,, \quad (2)$$

where f is the frequency, d the absorber thickness, c the light velocity, Z the air impedance and Z_{in} the absorber impedance.

For a multi-layer microwave absorber, it is assumed that the relative complex permeability, the relative complex permittivity and the thickness for each layer are: μ_{ri}, ε_{ri} and d_i (i = 1, 2, ..., n), respectively. According to the transmission line equivalent theory,[22,23] the input impedance of every layer is

$$Z_i = \eta_i \frac{Z_{i-1} + \eta_i \mathrm{th}(k_i d_i)}{\eta_i + Z_{i-1} \mathrm{th}(k_i d_i)}. \quad (3)$$

Here, η_i is the characteristic impedance of the i-th layer and expressed as

$$\eta_i = \sqrt{\mu_{ri}/\varepsilon_{ri}} = \sqrt{(\mu_i' - j\mu_i'')/(\varepsilon_i' - \varepsilon_i'')}, \quad (4)$$

k_i is the magnitude of the i-th wave vector and the electromagnetic wave incidence is vertical, so it is a constant and given as

$$k_i = j\omega\sqrt{(\mu_{ri}\varepsilon_{ri})/c}, \quad (5)$$

therefore the RL value for a multi-layer microwave absorber can be calculated by Eq. (2).

3. Results and discussion

3.1. Characteristics of nanocrystalline $SrFe_{12}O_{19}$ and α–Fe microfibers

Figure 1 shows the SEM morphologies and XRD patterns of the nanocrystalline $SrFe_{12}O_{19}$ microfibers and α–Fe microfibers. From Figs. 1(a) and 1(b), it can be observed that the microfibers are characterized by diameters about 1 μm–8 μm and high aspect ratios about 20–150. Their XRD reflections (Figs. 1(c) and 1(d)) belong correspondingly to the magnetoplumbite structure (JCPDS 33-1340) and the α–Fe structure (JCPDS 06-0696). The values of average crystallite size ($\boldsymbol{D}$) of $SrFe_{12}O_{19}$ and α–Fe can be estimated from the prominent reflections (114) and (110) using the Scherrer formula ($D = 089\lambda/(\beta \cos\theta)$, where λ is the wavelength of the X-ray radiation, β is the full width at half maximum (FWHM) of relevant reflection peak, and θ is the diffraction angle. And the calculated $\boldsymbol{D}$ values are 67 nm for $SrFe_{12}O_{19}$ microfibers and 44 nm for α–Fe microfibers respectively.

The magnetic hysteresis loops of nanocrystalline $SrFe_{12}O_{19}$ and α–Fe microfibers are shown in Fig. 2. From Fig. 2 it follows that the nanocrystalline $SrFe_{12}O_{19}$ microfibers have an average specific saturation magnetization ($\boldsymbol{M}_s$) about 64.2 Am^2/kg and an average high coercivity (H_c) of 319.5 kA/m while the specific saturation magnetization (M_s) and coercivity ($\boldsymbol{H}_c$) of the nanocrystalline α–Fe microfibers are on average about 142.5 Am^2/kg and 7.6 kA/m respectively. In the present work, these hard magnetic nanocrystalline $SrFe_{12}O_{19}$ microfibers are used as the matching layers and the soft magnetic nanocrystalline α–Fe microfibers are the absorbing layers for the design of double-layer microwave absorbers.

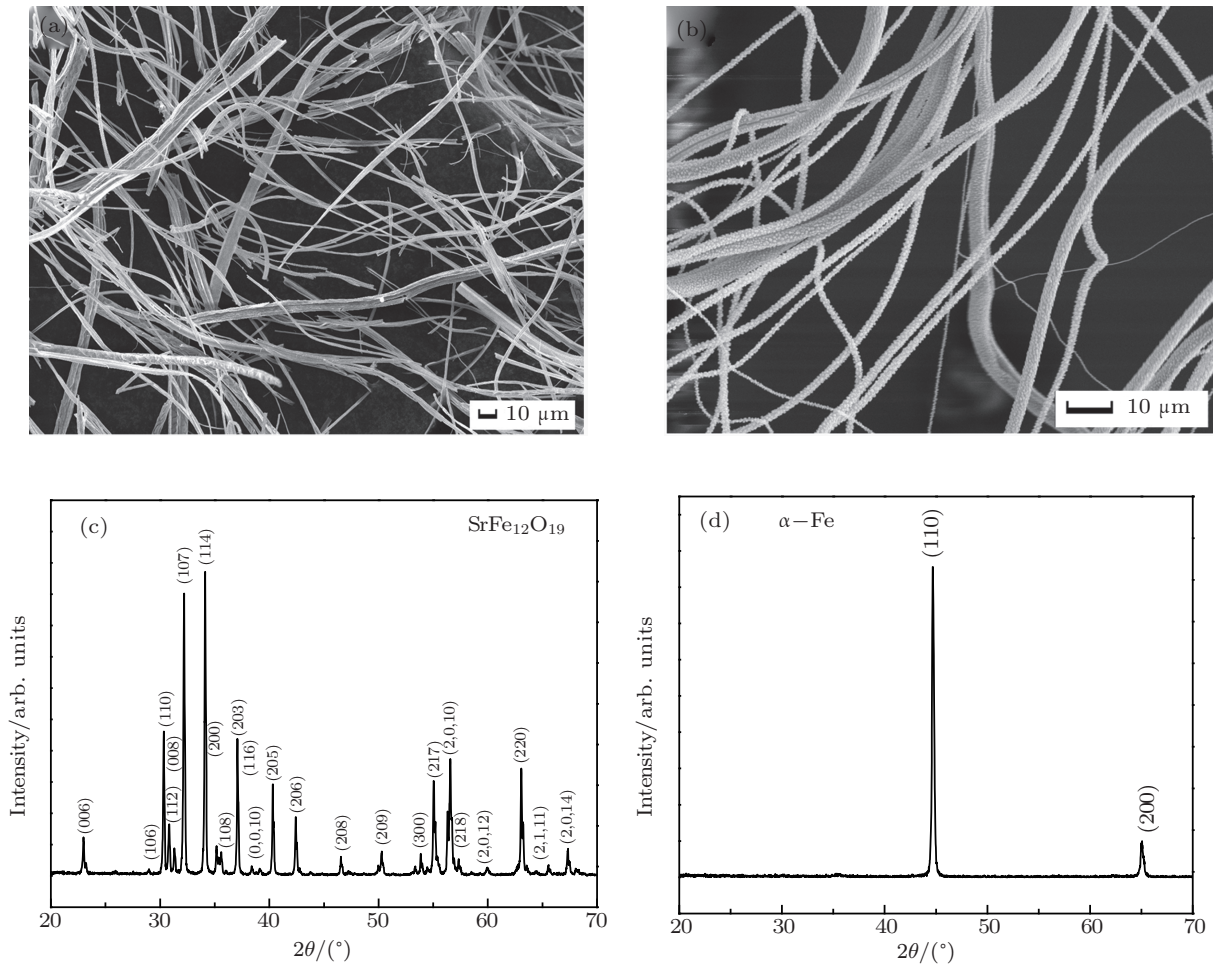


Fig. 1. SEM morphologies of nanocrystalline $SrFe_{12}O_{19}$ (a) and α–Fe (b) microfibers, XRD patterns of nanocrystalline $SrFe_{12}O_{19}$ (c), and α–Fe (d) microfibers.

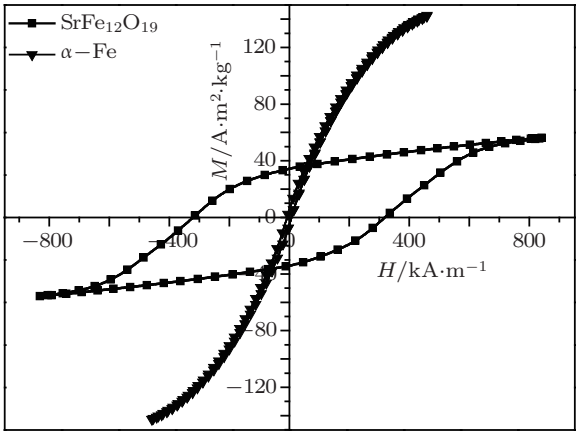


Fig. 2. Magnetic hysteresis loops of nanocrystalline $SrFe_{12}O_{19}$ and α–Fe microfibers.

3.2. Electromagnetic properties

Figure 3 shows the relative permeability (μ_r) and permittivity (ε_r) values of specimen 1 and specimen 2 in a frequency range of 2 GHz–18 GHz. From Fig. 3(a) for the nanocrystalline $SrFe_{12}O_{19}$ microfibers, it can be seen that the real part ε' exhibits a decrease tendency with frequency increasing, except for a small peak around 8.5 GHz, which can be due to the damping of the electrical dipole vibrations and interface polarizability.[24,25] For the imaginary part $4\varepsilon''$, it is almost constant with the frequency increasing from 2 GHz to 12 GHz, while in the frequency range 12 GHz–18 GHz, it has a broad peak around 14 GHz and an increasing tendency from about 16 GHz to 18 GHz, implying that the nanocrystalline ferrite microfibers can have some dielectric losses at a frequency of about 14 GHz and in a range from about 16 GHz to 18 GHz due to intrinsic electric dipoles and interfacial polarization. The frequency dependences of the real part μ' and the imaginary part μ'' of μ_r are shown in Fig. 3(b) for the nanocrystalline $SrFe_{12}O_{19}$ microfibers. The μ' value shows a reduction tendency in a frequency range of 2 GHz–10.5 GHz, with a broad peak around 8 GHz and then increases in the remaining range. For the imaginary part μ'', it is basically located at zero in a frequency range of 2 GHz–7 GHz,

while it shows an increase with a value 0.1 in a frequency range from about 10.5 GHz to 15.5 GHz and then decreases. Thus, it can be reasoned that the magnetic loss should be little in a low frequency range (2.0 GHz–7.0 GHz). The microwave adsorption in a frequency range from about 10.5 GHz to 15.5 GHz can be attributed to the magnetic hysteresis and domain wall displacement[22,26]

Figures 3(c) and 3(d) show the values of relative permittivity and permeability for the nanocrystalline α–Fe microfibers. From Fig. 3(c), the real part ε' exhibits a decrease tendency in a frequency range of 2 GHz–18 GHz, while the imaginary part ε'' has an increasing tendency from 2 GHz to 5 GHz and then decreases in a frequency range of 5 GHz–18 GHz. The observed phenomena can be generally attributed to the damping of the electrical dipole vibration and interface polarizability.[24,27] The frequency dependences of real part μ' and imaginary part μ'' of $\mu_{\rm r}$ for the nanocrystalline α–Fe microfibers are shown in Fig. 3(d). The μ'' value shows a reduction tendency in a frequency range of 2 GHz–14 GHz, with a broad peak around 14 GHz and then increases in the remaining range. For the imaginary part μ'', it exhibits a decrease tendency in a frequency range of 2 GHz–18 GHz except for some small peaks. These peaks can be attributed to the domain wall displacement and the eddy current loss arising from the intrinsic damping, interface effect, small size effect and spin wave excitations.[26,28]

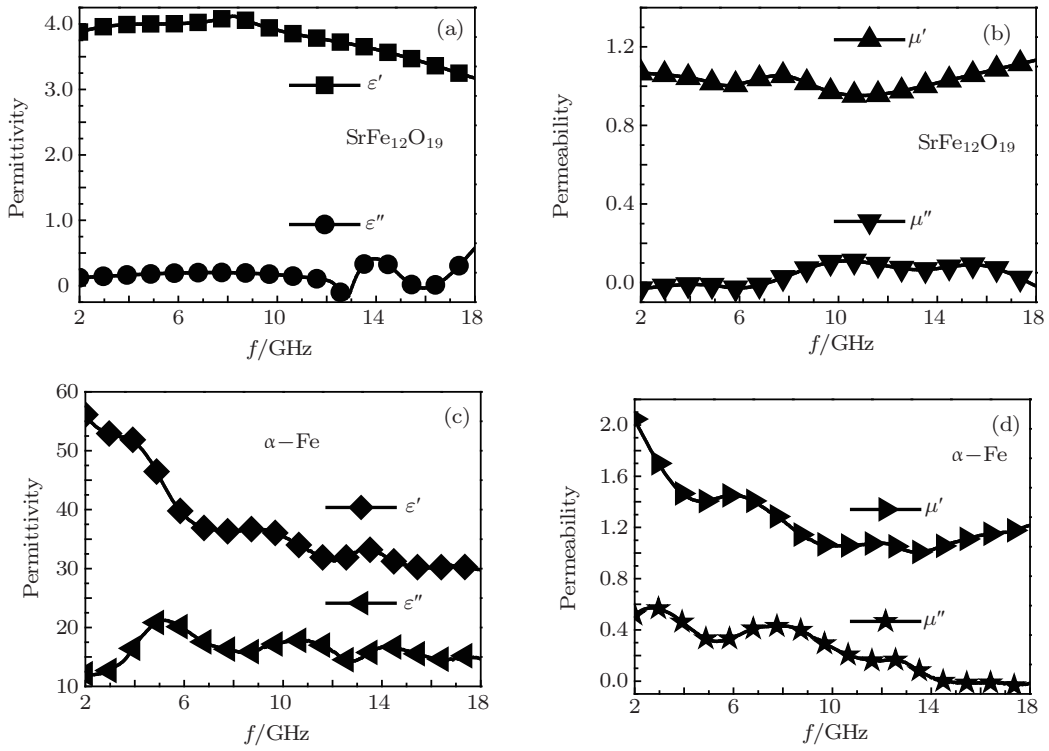


Fig. 3. Values of relative permeability and permittivity of specimen 1 (panels (a) and (b)) and specimen 2 (panels (c) and (d)) in a frequency range of 2 GHz–18 GHz.

3.3. Microwave absorption of single-layer and double-layer microwave absorbers

Figure 4 shows the RL values and frequencies for single-layer microwave absorber of specimen 1 and specimen 2 with different thicknesses in a frequency range of 2 GHz–18 GHz. From Fig. 4(a), for the absorber of nanocrystalline $SrFe_{12}O_{19}$ microfibers the effective microwave absorptions take place in a frequency range of 6 GHz–18 GHz, with a minimum reflection loss of −11.9 dB at 14.1 GHz for the specimen thickness of 3.0 mm. Referring to the relative complex permittivity and permeability as showed in Figs. 3(a) and 3(b), the absorption peaks for $SrFe_{12}O_{19}$ microfibers (Fig. 4(a)) are largely due to the magnetic losses arising from the magnetic hysteresis and domain wall displacement, as the natural resonance frequency for $SrFe_{12}O_{19}$ ferrites generally is around 50 GHz.[29] While for the absorber of the nanocrystalline α–Fe microfibers (Fig. 4(b)), it has microwave absorptions in a frequency range of 2 GHz–18 GHz. For the specimen

with a thickness of 0.7 mm, the minimum RL value is about −24 dB at 17.5 GHz and its absorption band width reaches about 3 GHz ranging from 15 GHz to 18 GHz with the RL value exceeding −10 dB, which can be mainly attributed to the dielectric loss resulting from the eddy current dissipation. With the α–Fe microfibers absorber thickness increasing to 2.0 mm, the absorption peak is shifted to a lower frequency region where the absorption is caused by the combination effect of dielectric loss and magnetic loss according to Figs. 3(c) and 3(d).

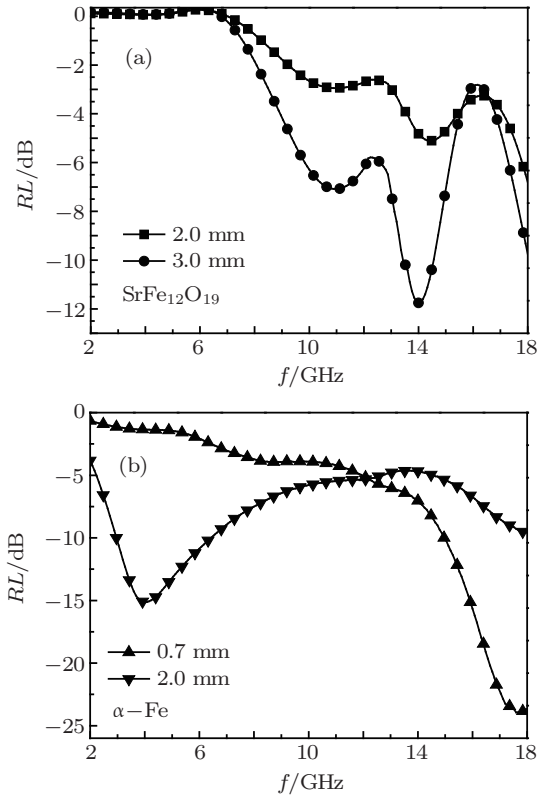


Fig. 4. Reflection losses of single-layer absorbers for specimen 1 and specimen 2 with different thicknesses in a frequency range of 2 GHz–18 GHz.

For the double-layer of the nanocrystalline $SrFe_{12}O_{19}$ and α–Fe microfibers, as the electromagnetic energy can be produced by the motion of charges and induced into a dissipative current,[15,23] the absorption peaks are the complementary results based on the Maxwell equations for permittivity and permeability. The reflection losses of double-layer absorber with a total thickness of 2.0 mm, which is composed of the matching layer filled with specimen 1 and the absorption layer filled with specimen 2 with various thicknesses, are shown in Fig. 5. It can be observed that with the absorbing layer thickness increasing from 0.5 mm to 1.8 mm, the minimum microwave absorption peak is shifted to a lower frequency region. When the absorption layer thickness is 0.7 mm and the matching layer thickness is 1.3 mm, the RL value achieves a minimum value of about −63 dB at 16.4 GHz, which means that two layers are matching well. With this coupling thickness, the absorption band width is about 6.7 GHz ranging from 11.3 GHz to 18 GHz with the RL value exceeding −10 dB, which covers the whole K_u-band (12.4 GHz–18 GHz) and 27% X-band (8.2 GHz–12.4 GHz). By comparison with the single-layer absorbers as showed in Fig. 4, the double-layer absorbers are efficient in a frequency range of 2 GHz–18 GHz.

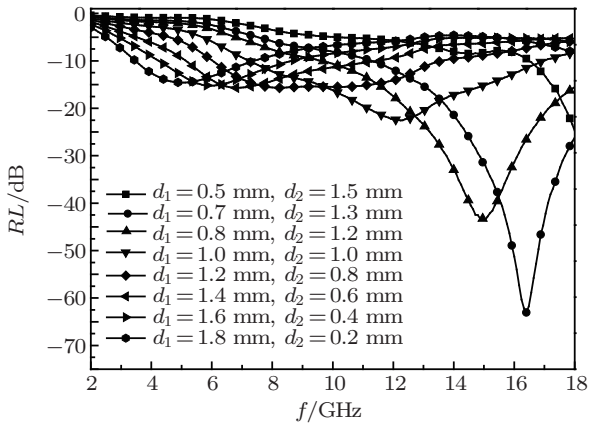


Fig. 5. Reflection losses of double-layer absorbers consisting of matching layer (d_1) filled with specimen 1 and absorption layer (d_2) filled with specimen 2 in a frequency range of 2 GHz–18 GHz.

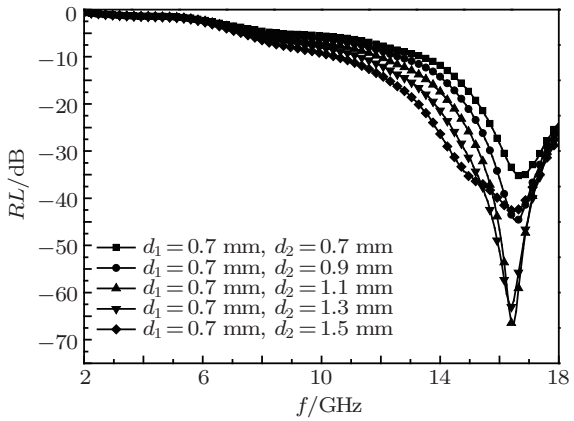


Fig. 6. Reflection losses of double-layer absorber with constant absorption layer thickness (d_1) and values of matching layer thickness (d_2) ranging from 0.7 mm to 1.5 mm in a frequency range of 2 GHz–18 GHz.

For an optimized absorbing layer thickness of 0.7 mm for the nanocrystalline α–Fe microfiber (specimen 2), the variations of RL values for the double-layer absorbers with matching layer thicknesses ranging from 0.7 mm to 1.5 mm of the nanocrystalline $SrFe_{12}O_{19}$ microfibers (specimen 1) are shown in Fig. 6. With the increase of matching layer thickness, the minimum RL value first increases, reaches about

−67 dB at 16.5 GHz when the matching layer thickness is 1.1 mm and then decreases. This coupled layer thickness is basically consistent with the above calculated results as shown in Fig. 5. With this coupled double-layer thickness, the absorption band width has about 6.1 GHz ranging from 11.9 GHz to 18 GHz for the RL value exceeding −10 dB.

Table 1 shows the microwave absorption properties of some recently reported double-layer absorbers. Compared with the double-layer absorbers reported in Refs. [4], [16] and [19], the double-layer absorber in the present work is thin and strong and in particular, its absorption covers the whole K_u-band (12.4 GHz–18 GHz). The enhancement in microwave absorption for the nanocrystalline $SrFe_{12}O_{19}$ and α–Fe microfibers each with a large surface-to-volume ratio can be ascribed to their shape anisotropy, small size effect and interface polarization.[13,25,30] As the permittivity and the permeability are coupled on the basis of the Maxwell equations, an obvious coupling frequency band occurs around 16.4 GHz (Fig. 6) for the 0.7-mm thick absorbing layer of α–Fe microfibers and the 1.3-mm thick matching layer of $SrFe_{12}O_{19}$ microfibers.

Table 1. Comparison of microwave absorption properties of some recently reported double-layer absorbers.

Absorbing layer	Matching layer	Absorbing layer thickness/mm	Total thickness /mm	Minimum RL value/dB	Band width/GHz ($RL < -10$ dB)	Ref.
50 wt% $Ba(Zn_{0.5}Co_{0.5})_2Fe_{16}O_{27}$ powder	50 wt% carbonyl iron powder	2.7	3.7	–55 (at 7.5 GHz)	5.2–9.3	[4]
30 wt% carbon black powder	10 wt% α–MnO_2 powder	3	4.5	–28 (at 15.7 GHz)	13–18	[16]
80 wt% carbonyl iron powder	50 wt% $BaTiO_3$ powder	1.9	2.3	–64 (at 3.5 GHz)	2.8–4.3	[19]
50 wt% α–Fe microfibers	67 wt% $SrFe_{12}O_{19}$ microfibers	0.7	2.0	–63 (at 16.4 GHz)	11.3–18	This Work

4. Conclusions

(i) For the single-layer absorbers, the nanocrystalline $SrFe_{12}O_{19}$ microfibers show some microwave absorptions in a frequency range of 6 GHz–18 GHz, with a minimum RL value of −11.9 dB at 14.1 GHz for the specimen thickness of 3.0 mm, while for the nanocrystalline α–Fe microfibers, their absorptions largely take place in a frequency range of 15 GHz to 18 GHz with the RL value exceeding −10 dB, with a minimum RL values about −24 dB at 17.5 GHz for the specimen thickness of 0.7 mm.

(ii) With the double-layer structure consisting of the nanocrystalline α–Fe microfibers and nanocrystalline $SrFe_{12}O_{19}$ microfibers, the microwave absorption performance is clearly enhanced by comparison with the single-layer absorbers.

(iii) For the total thickness of 2.0 mm, the microwave absorption peaks for the double-layer absorber of the matching layer filled with 67 wt% $SrFe_{12}O_{19}$ microfibers and the absorption layer filled with 50 wt% α–Fe microfibers shift toward a lower frequency region with the increase of absorption layer thickness. With a coupled absorbing layer thickness of 0.7 mm and a matching layer thickness of 1.3 mm, the RL value is about −63 dB at 16.4 GHz, the absorption band width is about 6.7 GHz ranging from 11.3 GHz to 18 GHz with the RL value exceeding −10 dB, which covers the whole K_u-band (12.4 GHz–18 GHz) and 27% X-band (8.2 GHz–12.4 GHz).

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