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ARTICLE

Preparation and Microwave Absorption Properties of La-Ho-Fe Alloys

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Abstract: The $La_xHo_{2-x}Fe_{17}$ (x=0.0, 0.2, 0.4, 0.6, 0.8) powders were prepared by arc melting and high energy ball milling. The influence of the La substitution on phase structure, morphology, magnetic properties and electromagnetic parameters were investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), vibrating-sample magnetometry (VSM) and vector network analyzer (VNA), respectively. The results show that the saturation magnetization increases and the average particle size increases with the increase of La content. The minimum absorption peak frequency shifts towards a lower frequency region with the increase of La content. The minimum reflection loss (RL) of $La_{0.2}Ho_{1.8}Fe_{17}$ reaches –28.72 dB at 8.72 GHz, and the frequency bandwidth of RL<–10 dB reaches about 2.32 GHz with the best matching condition d=1.8 mm. The reflection loss with the thickness ranging of 1.2~2.4 mm could reach –10 dB, which indicates that the particles are considered as the promising microwave absorbing materials with better absorption properties.

Key words: $La_xHo_{2-x}Fe_{17}$ alloys; ball milling; electromagnetic parameters; magnetic properties; microwave absorbing properties

With the increasing applications of wireless communications and electronic devices using the electromagnetic wave in the gigahertz (GHz) range, the appearance of electromagnetic interference (EMI) brings serious damages to information security and human health[1-4]. Therefore, the demands for electromagnetic wave absorbing material in the application of electronic devices are rapidly increasing[5]. Due to the increasing demands in electronic devices in industry, commercial and military, wide absorption band-width, light weight and efficient electromagnetic wave absorbing materials have become a significant aim for researchers[6-8].

The Fe-based soft magnetic absorbing materials attract considerable interest on account of their high permeability, Curie temperature and high saturation magnetizations[9,10]. According to the relevant report, Fe-based soft magnetic metals/alloys such as FeCo[11], FeSi[12], NdCeFe[13], and FeCrAl[14] have been researched. Among the raw FeCrAl powders, the effective absorption bandwidth is obtained in the frequency range of 8.3~12.4 GHz in 3.2 mm thickness, and the minimum reflection loss (RL) value is –18.9 dB at 11.6 GHz. The minimum RL of Nd_2Fe_{17} is –32.5 dB at 8.7 GHz with the best matching thickness of 1.8 mm. Thus the Fe-based alloy can be used as one of the ideal microwave absorbing materials in the gigahertz (GHz) range. In addition, rare-earth elements (RE) have typical relaxation characteristics, which may affect the electromagnetic properties[15]. The RE elements may contribute to the change in the magnetic interactions, which improves the magnetic properties to enhance the microwave absorption properties with excellent absorbing bandwidth and optimal matching thickness[16].

In the present work, the main aim is to investigate the influence of La doping on the structure, morphology, magnetic properties, electromagnetic parameters and microwave absorbing properties of Ho_2Fe_{17} alloy.

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1 Experiment

The $La_xHo_{2-x}Fe_{17}$ (x=0.0, 0.2, 0.4, 0.6, 0.8) samples were prepared by arc melting high purity elements La, Ho, Fe (all 99.99% purity) in an arc furnace under high purity argon atmosphere. All samples were heat treated at 1000 °C for 10 d in a quartz tube, and then quenched into ice-water mixture. The samples were polished to the possible oxidized surface and ground into the powders and sieved using a 100 μm pore size metallic sieve. The sieved powders were milled for 15 h on a planet ball mill (QM-1SP) with the mass ratio of the ZrO_2 balls to the powders of 15:1 in absolute ethyl alcohol and the speed of 300 r/min. The phase structure, morphology and magnetic property were characterized by X-ray diffraction (XRD, Empyrean PIXcel3D, Cu-Kα), scanning electron microscopy (SEM, SM-5610LV) and vibrating sample magnetometer (VSM), respectively. Afterward, a toroidal shape composite with an outer diameter of 7.0 mm and an inner diameter of 3.0 mm was fabricated by pressing the mixture in a mold for measuring the absorbing properties. The mass ratio of milled powder to paraffin of composite absorber was 80:20. Then the complex permeability and permittivity were measured by vector network analyzer (VNA, Agilent 8722ES) in the range of 2~18 GHz.

2 Results and Discussion

2.1 Effect of La content on the structure and morphology of La-Ho-Fe

The crystalline structure of $La_xHo_{2-x}Fe_{17}$ (x=0.0, 0.2, 0.4, 0.6, 0.8) alloys was characterized by X-ray diffraction in Fig.1. The results indicate that all the diffraction peaks for the $La_xHo_{2-x}Fe_{17}$ match well with those of Ho_2Fe_{17} structure and a small amount of α-Fe phase is observed. Overall, the α-Fe phase increases with the increase of La content. Besides, the XRD peaks of $La_xHo_{2-x}Fe_{17}$ slightly shift to lower angle as compared to those of the Ho_2Fe_{17} after adding the La. The main reason is that the introduction of La of large atomic radius into Ho_2Fe_{17} lattice would lead to partial substitution of La for Ho, resulting in slight shift of the diffraction angles in agreement with Bragg equation[17].

The SEM morphology in Fig.2 shows that the powders of $La_xHo_{2-x}Fe_{17}$ (x=0.0, 0.2, 0.4, 0.6, 0.8) are flake-like after high-energy ball milling. The flake shape has a larger scattering area, and multiple scattering leads to stronger microwave absorption[18]. It can be clearly found that the particle size of the powders increase with the increase of La content. And the diameters are in the range of 1~10 μm. When

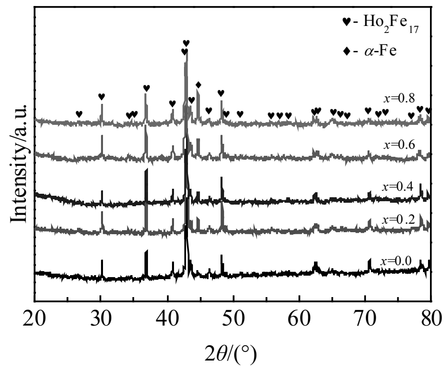

Fig.1 XRD patterns of $La_xHo_{2-x}Fe_{17}$ alloys

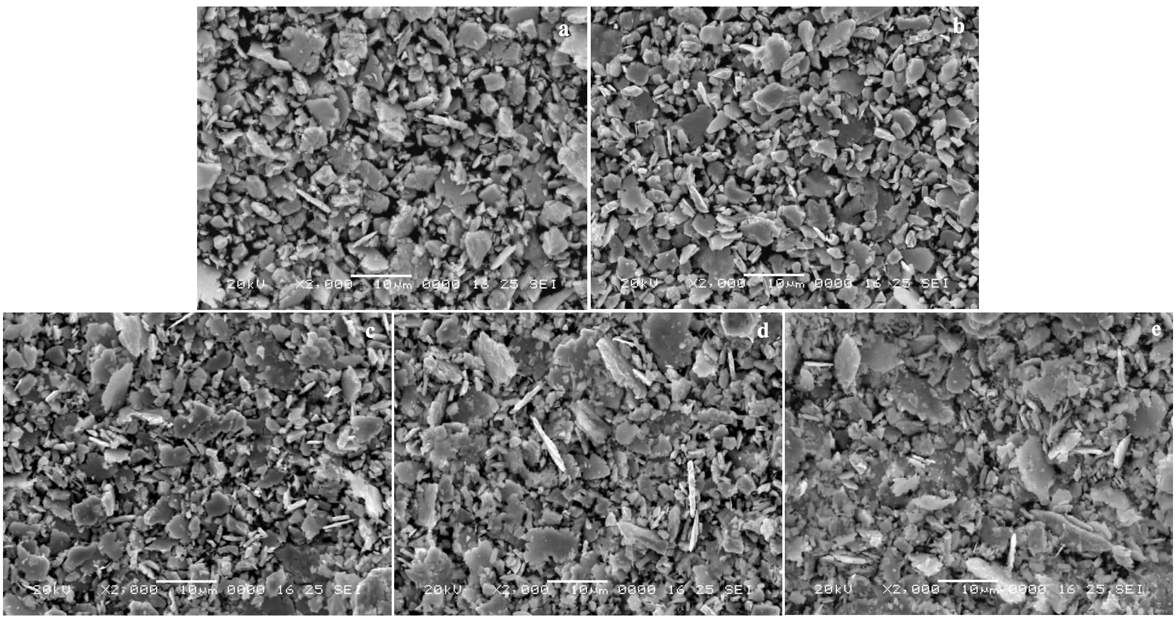

Fig.2 SEM images of the $La_xHo_{2-x}Fe_{17}$ powders: (a) x=0.0, (b) x=0.2, (c) x=0.4, (d) x=0.6, and (e) x=0.8

x=0.2, it can be seen that $La_{0.2}Ho_{1.8}Fe_{17}$ powder is homogeneously distributed.

2.2 Magnetic properties and electromagnetic parameters of La-Ho-Fe powders

The magnetic properties of $La_xHo_{2-x}Fe_{17}$ powders were also studied by measuring the magnetic hysteresis loops, as shown in Fig.3. It can be seen that the saturation magnetization increases with the increase of La content. Furthermore, it shows that the $La_xHo_{2-x}Fe_{17}$ has a low coercive force. A microwave absorbing material with a lower coercive force is suitable to improve the microwave absorption according to Ahmad[19].

The electromagnetic parameters of $La_xHo_{2-x}Fe_{17}$ powders were measured with frequency variation in the range of 2~18 GHz in Fig.4. The result indicates that the real parts (ε') and the imaginary parts (ε'') of relative complex permittivity increase with the increase of La content. This result may be attributed to the variation of electrical resistivity (ρ)[20]. The particle size increases with the increase of La content in Fig.2, which makes the spacing decrease between the particles and the electron hopping barrier. Therefore, it will lead to decrease in the electrical resistivity (ρ)[21]. According to the Debye theory, for the material with a good electrical conductivity, $\varepsilon''\approx 1/2\pi\varepsilon_o\rho f$, where ε_o is the permittivity of a vacuum, ρ is the resistivity, f is the frequency of the electromagnetic wave[22]. It shows that the ε'' is inversely proportional to ρ, and thus the values of ε'' increases with the increase of La content. Furthermore, the resonance frequency of ε'' shifts to the lower frequency region. The real part (μ') of permeability declines with increasing the frequency, which may result from the limited speed of spin and domain-wall motion (displacement/rotation)[23]. The imaginary part (μ'') of permeability also declines as a whole with increasing the frequency, which may be related to the improvement in eddy current effect with the increasing frequency, so it causes larger reverse magnetic field to decrease the permeability[24].

Dielectric loss and magnetic loss are two possible contributions to electromagnetic wave absorption. Fig.5 shows the relationship between the loss tangent of the $La_{0.2}Ho_{1.8}Fe_{17}$ powder and frequency, where the dielectric loss ($\tan\delta_E$) is $\varepsilon''/\varepsilon'$ and magnetic loss ($\tan\delta_M$) is μ''/μ'. It can be discovered in Fig.5 that the $\tan\delta_E$ values are less than $\tan\delta_M$ values from 2 GHz

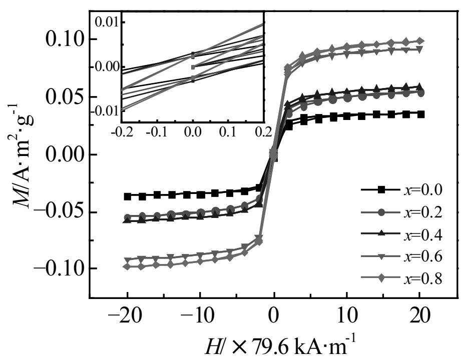

Fig.3 Magnetic hysteresis loops of the $La_xHo_{2-x}Fe_{17}$ powders

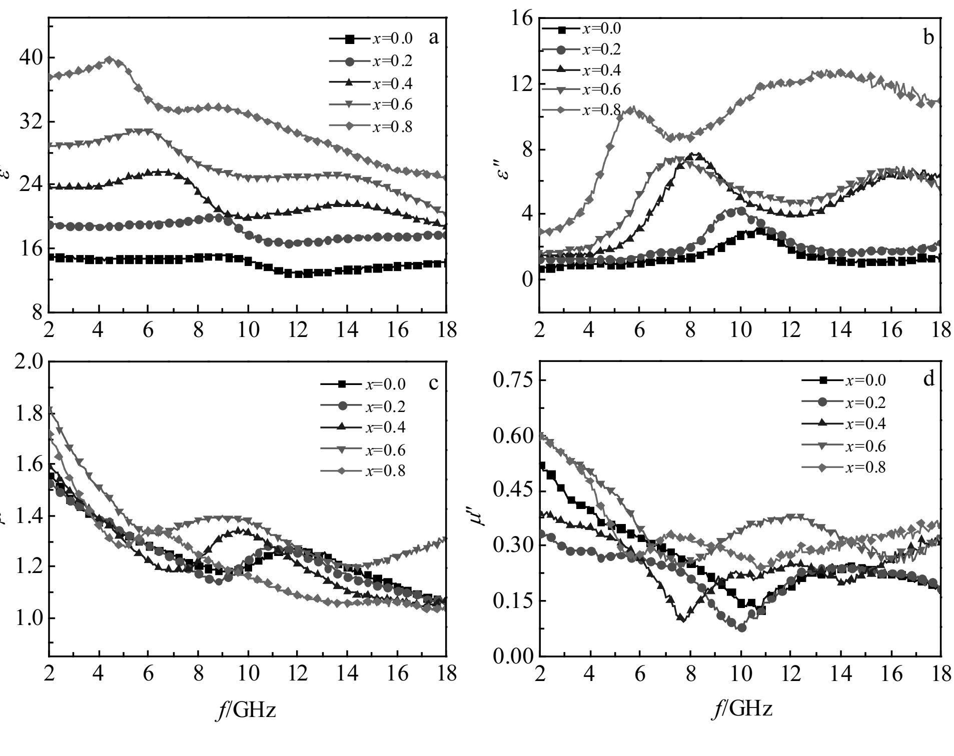

Fig.4 Electromagnetic parameters of $La_xHo_{2-x}Fe_{17}$ powders with the frequency variation: (a) ε', (b) ε'', (c) μ', and (d) μ''

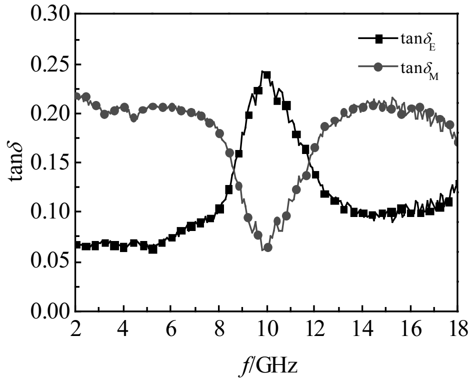

Fig.5 Relationship between loss tangent of $La_{0.2}Ho_{1.8}Fe_{17}$ powders and frequency

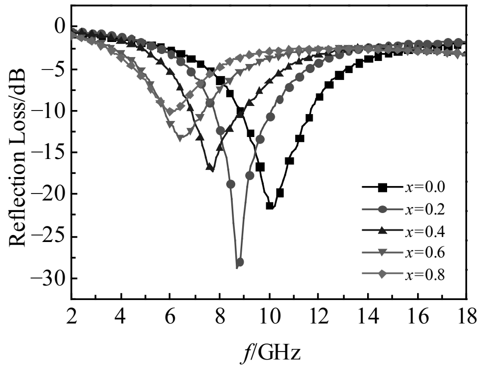

Fig.6 Reflection loss of the $La_xHo_{2-x}Fe_{17}$ powders (d=1.8 mm)

to 8 GHz and 12 GHz to 18 GHz. While the tanδ_E values are much larger than the tanδ_M values from 8 GHz to 12 GHz. Overall, the result shows that the magnetic loss of the $La_{0.2}Ho_{1.8}Fe_{17}$ powder has a greater effect than the dielectric loss in almost all the frequency bands.

2.3 Microwave absorbing properties of La-Ho-Fe

According to the transmission line theory, the reflection loss (RL) of the $La_xHo_{2-x}Fe_{17}$ powders can be calculated with the electromagnetic parameter at a given thickness and frequency according to the following equations[25]:

$$\mathrm{RL} = -20\lg\left|\frac{Z_{\mathrm{in}} - Z_0}{Z_{\mathrm{in}} + Z_0}\right| \tag{1}$$

$$Z_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}} \tag{2}$$

$$Z_{\mathrm{in}} = Z_0\sqrt{\frac{\mu_{\mathrm{r}}}{\varepsilon_{\mathrm{r}}}}\tanh\left[j\frac{2\pi}{c}\sqrt{\mu_{\mathrm{r}}\varepsilon_{\mathrm{r}}}fd\right] \tag{3}$$

where, Z_0 is the characteristic impedance of free space, Z_{in} is the input impedance at free space and absorber interface, ε_0 and μ_0 are the permittivity and permeability of vacuum, respectively. ε_r and μ_r are the relative complex permittivity and permeability of the absorber, respectively. f is the frequency of incident EM wave, d is the thickness of absorbing sample, c is the velocity of light, and j is the imaginary unit.

As shown in Fig.6 and Table 1, the reflection loss of the $La_xHo_{2-x}Fe_{17}$ powder was investigated by Eq.(1), (2) and (3) with the coating thickness of 1.8 mm. The result reveals that the minimum reflection loss of the $La_xHo_{2-x}Fe_{17}$ powders increases first and then decreases with increasing La substitution. Meanwhile, the minimum absorption peak frequency of $La_xHo_{2-x}Fe_{17}$ shifts to a lower frequency range upon the La content. With the increase of La content, the dielectric constant increases and the equivalent thickness increases, and as a result the absorption peak shifts to a low

Table 1 Minimum RL and peak frequencies with the different La contents (d=1.8 mm)

La content	x=0.0	x=0.2	x=0.4	x=0.6	x=0.8
Minimum RL/dB	–21.67	–28.72	–17.24	–13.13	–10.11
f_m/GHz	10.16	8.72	7.68	6.48	6.08
Frequency width of RL<–10 dB/GHz	2.72	2.32	2	1.52	0.16

frequency[18]. In addition, this may be attributed to the increase of the complex permittivity in Fig.4. It is well known that the relationship between matching frequency and thickness can be expressed by the following equation[26]:

$$f_{\mathrm{m}} = \frac{c}{4d_{\mathrm{m}}}\frac{1}{\sqrt{\varepsilon'\mu'}}\left(1+\frac{1}{8}\tan^2\delta_{\mathrm{M}}\right)^{-1} \tag{4}$$

where, f_m and d_m are the matching frequency and thickness, respectively, δ_M is the magnetic loss tangent. It is clear that the matching frequency decreases by $\sqrt{\varepsilon'\mu'}$ times at a given thickness. The values of ε' are obviously increased, and the values of μ' are almost the same. Hence, the reflection loss peaks shift to a lower frequency region for the higher value of ε'. As shown in Table 1, the $La_xHo_{2-x}Fe_{17}$ powders have good microwave properties, especially the powders/paraffin composite absorber which has the best absorbing properties with the La content x=0.2. The minimum RL of $La_{0.2}Ho_{1.8}Fe_{17}$ reaches –28.72 dB at 8.72 GHz, and the frequency bandwidth of RL<–10 dB reaches about 2.32 GHz with the best matching condition d=1.8 mm.

Fig.7 shows the relationship between the reflection loss and frequency for the composites with $La_{0.2}Ho_{1.8}Fe_{17}$ powder at different thicknesses. As shown in Table 2, the minimum reflection loss moves toward the lower frequency region with thickness increasing. It is worth noting that the minimum reflection loss exceeding –10 dB (90% absorption) is obtained in the thickness range from 1.4 mm to 2.4 mm, which shows good microwave absorption properties. Moreover, the absorption bandwidth with reflection loss lower than –10 dB of the composite with the thickness of only 1.2 mm can reach

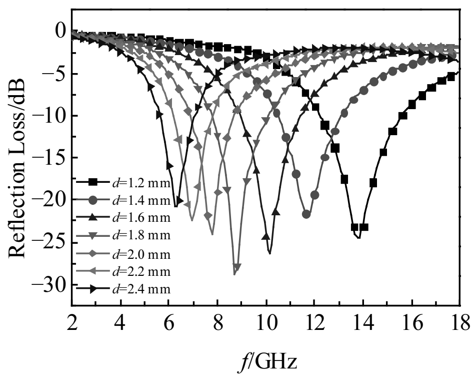

Fig.7 Reflection loss of the $La_{0.2}Ho_{1.8}Fe_{17}$ powder with different thicknesses

Table 2 Minimum RL and peak frequencies of $La_{0.2}Ho_{1.8}Fe_{17}$ with different thicknesses

d/mm	1.2	1.4	1.6	1.8	2.0	2.2	2.4
Minimum RL/dB	–24.47	–22.08	–26.25	–28.72	–24.05	–22.51	–20.85
f_m/GHz	13.84	11.68	10.16	8.72	7.76	6.96	6.32
Frequency width of RL< –10 dB/GHz	3.28	2.56	2.4	2.32	1.76	1.52	1.36

up to 3.28 GHz, and the absorption bandwidth gradually decreases with thickness increasing.

3 Conclusions

1) The $La_xHo_{2-x}Fe_{17}$ (x=0.0, 0.2, 0.4, 0.6, 0.8) alloys prepared by arc melting and high energy ball milling mainly consists of Ho_2Fe_{17} and a small amount of α-Fe phase. The saturation magnetization and the average particle size increases with the increase of La content. The minimum absorption peak frequency shifts towards a lower frequency region with the increase of La content.

2) The minimum RL of $La_{0.2}Ho_{1.8}Fe_{17}$ reaches –28.72 dB at 8.72 GHz, and the frequency bandwidth of RL<–10 dB reaches about 2.32 GHz with the best matching condition d=1.8 mm. The minimum reflection loss exceeding –10 dB is obtained in the thickness range from 1.4 mm to 2.4 mm. The absorption bandwidth with reflection loss lower than –10 dB with the thickness of only 1.2 mm can reach up to 3.28 GHz.

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La-Ho-Fe 合金的制备及其微波吸收特性

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摘　要：采用电弧熔炼及高能球磨工艺制备出 $La_xHo_{2-x}Fe_{17}$ (x=0.0, 0.2, 0.4, 0.6, 0.8) 合金微粉，借助 XRD、SEM、VSM 和网络矢量分析仪等仪器分别研究 La 替换对合金微粉的结构、形貌、磁性能及其微波吸收性能的影响。结果表明, 随着 La 含量的增加，饱和磁化强度和平均颗粒大小都有所增加。$La_xHo_{2-x}Fe_{17}$ 合金的最小反射峰频率向低频方向移动。其中 $La_{0.2}Ho_{1.8}Fe_{17}$ 合金具有最好的吸波效果，在最佳匹配厚度 1.8 mm 下，$La_{0.2}Ho_{1.8}Fe_{17}$ 合金的最小反射损耗在 8.72 GHz 处达到–28.72 dB，反射损耗小于–10 dB 的频带宽度达到 2.32 GHz。当厚度在 1.2~2.4 mm 范围里，$La_{0.2}Ho_{1.8}Fe_{17}$ 合金的反射损耗均小于–10 dB，这表明 $La_xHo_{2-x}Fe_{17}$ 是有前途的微波吸收材料，并具有良好的吸收特性。

关键词：$La_xHo_{2-x}Fe_{17}$ 合金；球磨；电磁参数；磁性能；微波吸收特性

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