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ARTICLE

Effect of Dy Content on Microwave Absorption Properties of Pr_2Fe_{17} Alloy

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Abstract: The $Dy_xPr_{2-x}Fe_{17}$ (x =0.0, 0.1, 0.2, 0.3, 0.4) powder was prepared by arc-smelting and high energy ball milling. The phase structure, morphology, magnetic properties and electromagnetic parameters of the powder were investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), vibrating sample magnetometer (VSM) and vector network analyzer (VNA), respectively. The results reveal that the saturation magnetization of the $Dy_xPr_{2-x}Fe_{17}$ alloys decreases with the increase of Dy content. The minimum absorption peak frequency shifts towards higher frequency region upon the Dy substitution. And the minimum reflection loss of the $Dy_xPr_{2-x}Fe_{17}$ alloys increases firstly and then decreases upon the Dy content. The $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder exhibits the best microwave absorbing properties. The minimum reflection loss of $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder is about –42.38 dB at 5.04 GHz, and the frequency bandwidth of R_L <–10 dB reaches about 1.20 GHz with the best matching thickness of 2.5 mm.

Key words: Dy-Pr-Fe alloys; high energy ball milling; magnetic properties; electromagnetic parameters; reflection loss

Microwave absorbing materials play an important role in preventing the electromagnetic radiation and interference and extend the applications of civilian wireless communications technology, satellite broadcast communications, and military technology[1-7]. They can dissipate the electromagnetic wave energy into heat energy, reduce the electromagnetic radiation and interference, and have attracted strong interests [8-12].

At present, the soft magnetic absorbing materials mainly include Fe, Co, Ni with the corresponding metallic alloys in the frequency range of 2~18 GHz [13]. The Fe-based soft magnetic absorbing materials have attracted immense attention on account of their excellent saturation magnetization, high permeability, and lower price [14]. The high saturation magnetization is favorable to obtain desirable microwave absorbing properties. And Fe-rich Pr_2Fe_{17} alloys are crystallized in the Th_2Zn_{17}-type rhombohedral crystal structure with R3-MH space group, and exhibit high saturation magnetization, which is favorable to improve the microwave absorbing properties [15,16]. Therefore the Fe-rich Pr_2Fe_{17} alloy possesses the potential to be the promising microwave absorbing materials. In our previous work[17, 18], it was found that the Pr_2Fe_{17}-based alloys possessed higher complex permittivities and permeabilities and good microwave absorbing properties under the small thickness. In the present paper, the main aim is to investigate the influence of heavy rare earth Dy doping on the structure, morphology, magnetic properties and electromagnetic parameters and microwave absorbing properties of the Pr_2Fe_{17} alloy.

1 Experiment

The samples of $Dy_xPr_{2-x}Fe_{17}$ (x=0.0, 0.1, 0.2, 0.3, 0.4) were prepared by arc-melting pure metal elements Dy, Pr and Fe (all 99.99% purity) under high purity argon atmosphere. The samples were melted several times in order to achieve good homogenization. All ingots were encapsulated

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in a quartz tube, heat-treated at 1000 ℃ for 10 d, and then quenched into ice-water mixture. Samples for measuring microwave absorption properties were mechanically milled for 20 h under the protection of absolute ethanol by using a planet ball mill (QM-1SP) with the speed of 320 r/min. The mass ratio of the ZrO_2 balls to the powders was 20:1.

The ground powders were investigated by X-ray diffraction (XRD, Empyrean PIXcel 3D, Co Kα). The morphology of the ball milled powder was characterized by scanning electron microscopy (SEM, SM-5610LV). The saturation magnetization of the Dy-Pr-Fe powder was measured by vibrating sample magnetometer (VSM). The composite samples for microwave absorption study were prepared by uniformly dispersing the milled powder in paraffin with a mass fraction of 80%. The mixture was made into a toroidal specimen with an inner diameter of 3.0 mm, outer diameter of 7.0 mm and controlled a thickness of about 3.5 mm. Then the complex permeability and permittivity of the composite absorbers in the frequency range of 2~18 GHz were measured by vector network analyzer (VNA, Agilent 8722ES).

2 Results and Discussion

2.1 Effect of Dy substitution on the phase composition and morphology of Pr_2Fe_{17} alloy

Fig.1 shows XRD patterns of $Dy_xPr_{2-x}Fe_{17}$ (x = 0.0, 0.1, 0.2, 0.3, 0.4). The results indicate that all the samples maintain the Pr_2Fe_{17} phase. Compared to Pr_2Fe_{17}, the peaks in XRD patterns are found to shift towards higher angle with the increasing amount of Dy substitution. It is mainly because that the introduction of the Dy atoms into Pr_2Fe_{17} lattice would lead to Dy partial substitution for Pr, resulting in the decrease of lattice parameters[19].

Fig.2 shows the SEM images of the $Dy_xPr_{2-x}Fe_{17}$ powder. It can be observed that most of $Dy_xPr_{2-x}Fe_{17}$ powder becomes flaky after ball milling. The particle size becomes much finer with the increase of Dy content. The flakiness or needle form is the best shape of microwave absorbing materials on the basis of the reference[20]. It is obvious that the large size and amount of flake particles decrease significantly with the increasing amount of Dy substitution. The morphology of samples implies that $Dy_xPr_{2-x}Fe_{17}$ alloys possess the potential to be a promising microwave absorbing agent.

2.2 Effect of Dy substitution on the magnetic properties and electromagnetic parameters of Pr_2Fe_{17} alloy

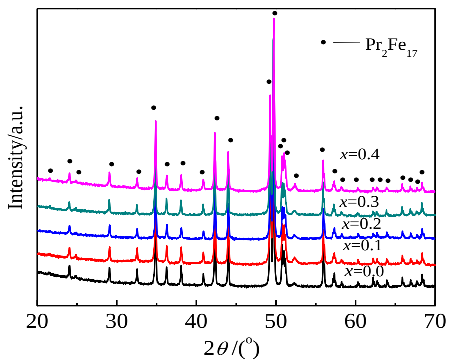

Fig.1 XRD patterns of $Dy_xPr_{2-x}Fe_{17}$

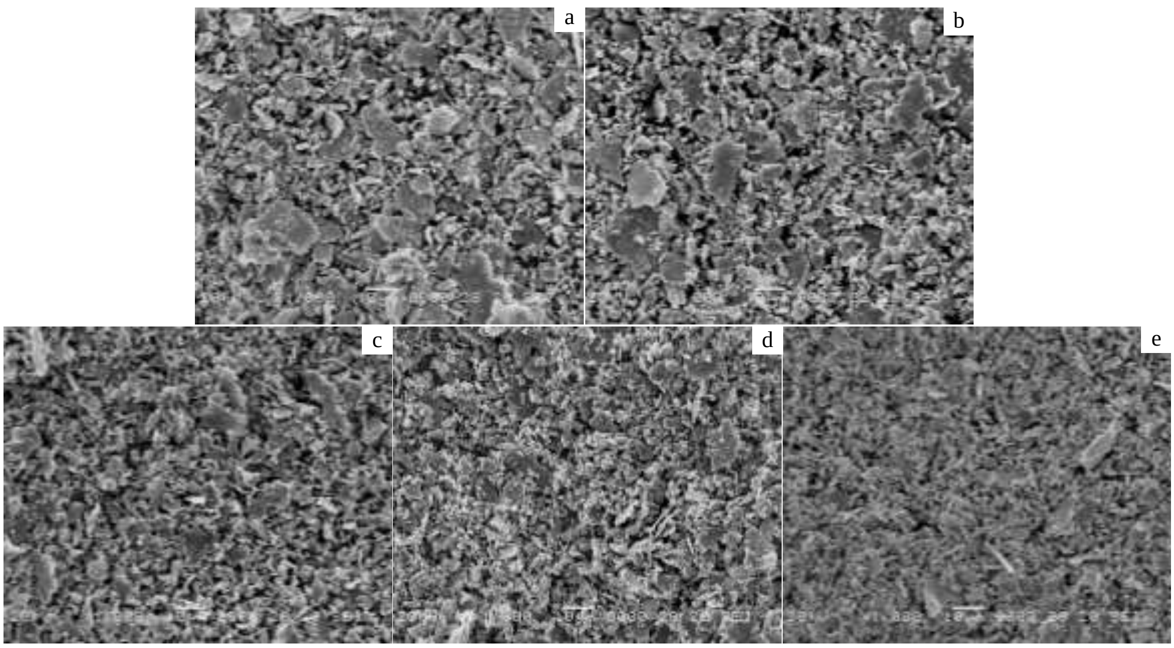

Fig.2 SEM images of the $Dy_xPr_{2-x}Fe_{17}$ powder: (a) x =0.0, (b) x =0.1, (c) x =0.2, (d) x =0.3, and (e) x =0.4

The magnetic properties of $Dy_xPr_{2-x}Fe_{17}$ powder were also studied by measuring the magnetic hysteresis loops, and the results are depicted in Fig.3. It can be seen that the saturation magnetization decreases with the increase of Dy content.

Fig.4 shows the electromagnetic parameters of $Dy_xPr_{2-x}Fe_{17}$(x =0.0~0.4) powder in the frequency range of 2~18 GHz. The results indicate that the real parts (ε') and the imaginary parts (ε'') of relative complex permittivity of the composite decrease with the increasing amount of Dy substitution. The interfacial polarization is dominant in the composite on account of the large particle size and surface area of flake particles [21]. The grain size shrinkage induced by the difference of radius between Dy and Pr could change the surface state and grain surface energy level change obviously. The enhanced interface polarization and repetitious reflection is favorable to improve the microwave absorbing properties [22]. And the flaky particles possess high electrical conductivity and enhance the space charge polarization between adjacent conductive particles, which is conducive to obtain higher ε' and ε'' [23-25]. And the size and amount of the large flake particles decreases significantly with the increasing amount of Dy substitution; therefore the values of ε' and ε'' decrease with the increasing amount of Dy content.

The resonance frequency of ε'' shifts towards lower frequency region with the increase of Dy content. It is mainly because that the particle size becomes fine, which increases the crystal defects and decreases conductivity, finally causing the resonant frequency of ε'' to move to lower frequency region.

The real parts (μ') of the complex permeability increase with the increase of Dy content. This feature can be attributed to the fact that μ' is in direct proportional to $(1/M_s^2)^2$ [26], while the saturation magnetization (M_s) of (Pr, Dy)$_2$Fe$_{17}$ increases with increasing Dy content. In addition, the imaginary parts (μ'') of complex permeability also increase with the increase of Dy content. It is because that the μ'' is in direct proportional to $1/M_s^2$ [26].

2.3 Effect of Dy substitution on the microwave absorbing properties of Pr_2Fe_{17} alloy

According to the generalized transmission line theory, the reflection loss of the powder can be calculated by Eq. (1) [27]:

$$R = -20\lg\left[\left|\frac{Z\tanh(jkd)-Z_0}{Z\tanh(jkd)+Z_0}\right|\right] \tag{1}$$

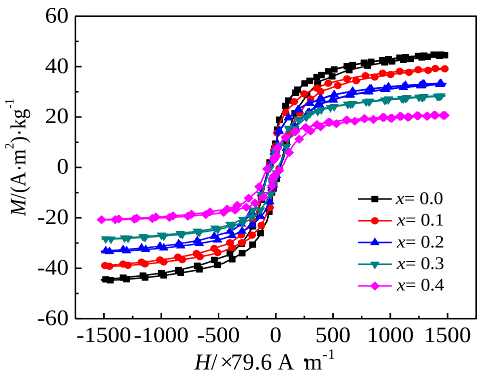

Fig.3 Magnetic hysteresis loops of the $Dy_xPr_{2-x}Fe_{17}$

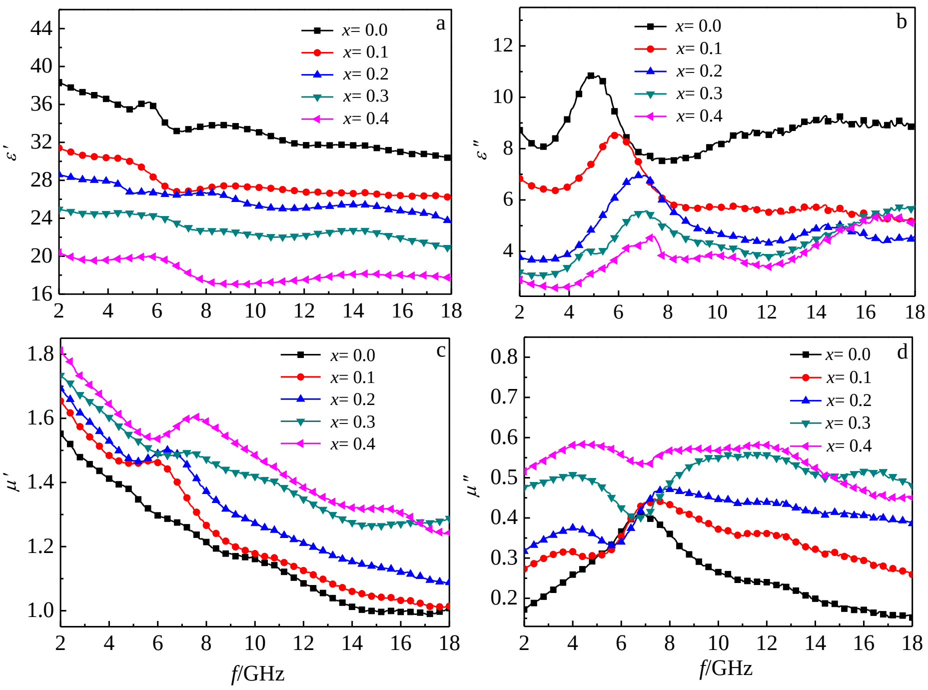

Fig.4 Electromagnetic parameters of $Dy_xPr_{2-x}Fe_{17}$ powder: (a) ε', (b) ε'', (c) μ', and (d) μ''

Where, the propagation constant $k=\sqrt{\varepsilon_0\mu_0(\varepsilon'-\mathrm{j}\varepsilon'')(\mu'-\mathrm{j}\mu'')}$, the wave impedance $Z=\sqrt{(\mu'-\mathrm{j}\mu'')\mu_0/(\varepsilon'-\mathrm{j}\varepsilon'')\varepsilon_0}$, μ_0 and ε_0 are the permeability and permittivity in vacuum respectively, d is the matching thickness of the absorbers, Z_0 is the vacuum wave impedance, and j is the imaginary unit.

The reflection loss of the $Dy_xPr_{2-x}Fe_{17}$ (x =0.0~0.4) powder with the matching thickness of 1.8 mm were investigated through Eq.(1), which are shown in Fig.5 and Table 1. It can be seen that the minimum absorption peak frequency shifts towards higher frequency region upon the Dy content. The resonance frequency is in direct proportional to $1/M_s$, so the absorption peak frequency will shift towards higher frequency region as the M_s decreases [26].

The powders possess good microwave absorbing properties in the frequency range of 2~18 GHz with the matching thickness of 1.8 mm. It can be seen that the minimum reflection loss (R_L) of the $Dy_xPr_{2-x}Fe_{17}$ alloys increase firstly and then decrease with the increase of Dy content. The absorption bandwidths (R_L< –10 dB) become wider. And the absorption bandwidths is in direct proportional to $1/M_s^2$, and the M_s decreases with the increase of Dy content, which results in the absorption bandwidths increase [26]. The $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder shows the best microwave absorbing properties, the minimum reflection loss of $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder is about –28.91 dB at 7.36 GHz, and the bandwidth of R_L < –10 dB reaches about 1.92 GHz with the matching thickness of 1.8 mm.

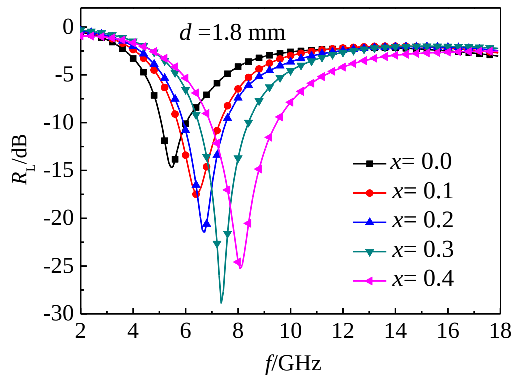

Fig.5 Reflection loss of the $Dy_xPr_{2-x}Fe_{17}$ powder (d = 1.8 mm)

Table 1 Minimum R_L and peak frequencies with different Dy contents (d =1.8 mm)

x	0.0	0.1	0.2	0.3	0.4
Minimum R_L/dB	-14.62	-17.49	-21.49	-28.91	-25.28
f_m/GHz	5.52	6.40	6.72	7.36	8.08
Frequency width of (R_L<–10 dB)/GHz	0.88	1.50	1.60	1.92	2.48

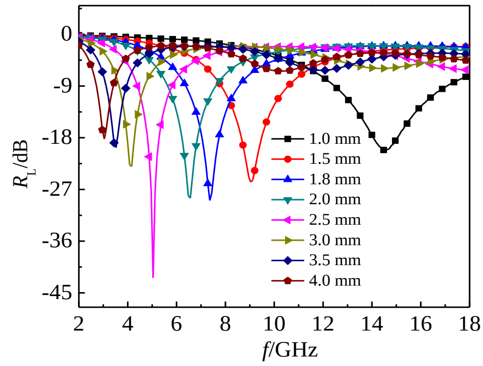

Fig.6 Reflection loss of the $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder with different thicknesses

Fig.6 shows the reflection loss of $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder with different thicknesses in the frequency range of 2~18 GHz. Apparently, it can be observed that the absorption peak shifts towards lower frequency region with the increase of absorbing coating thickness. The phenomenon can be explained by Eq.(2) [28]:

$$f_m=\frac{c}{2\pi\mu''d} \tag{2}$$

where, f_m, c and d are the matching frequency of the minimum reflection loss, the velocity of light, and the coating thickness of the composite absorbers, respectively. Eq.(2) indicates that the f_m shifts towards lower frequency region upon the microwave absorbing coating thickness. The $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder has the best microwave absorbing properties with the matching thickness of 2.5 mm. And the minimum reflection loss of $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder is –42.38 dB at 5.04 GHz, and the frequency bandwidth of R_L < –10 dB reaches about 1.20 GHz with the best matching condition d = 2.5 mm. In addition，the reflection loss with the thickness ranging of 1.0~4.0 mm all could reach –15.0 dB, which show the perfect microwave absorption properties.

3 Conclusions

1) This work demonstrates that the $Dy_xPr_{2-x}Fe_{17}$ (x = 0.0, 0.1, 0.2, 0.3, 0.4) alloys with tunable Dy content can be synthesized on the single Pr_2Fe_{17} phase. The saturation magnetization decreases with the increasing amount of Dy substitution.

2) The minimum absorption peak frequency shifts towards higher frequency region with the increase of Dy substitution. The minimum reflection loss of the $Dy_xPr_{2-x}Fe_{17}$ powder increases firstly and then decreases with the increase of Dy content.

3) The $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder exhibits the best microwave absorbing effect with the matching thickness of 2.5 mm. The minimum reflection loss of $Dy_{0.3}Pr_{1.7}Fe_{17}$ powder is –42.38

dB at 5.04 GHz, and the frequency bandwidth of *RL* < −10 dB reaches about 1.20 GHz with the best matching condition *d* =2.5 mm. Overall, the Dy-Pr-Fe alloys possess the immense potential to be a promising microwave absorbing agent in the frequency range of 2~18 GHz.

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Dy 对 Pr_2Fe_{17} 合金微波吸收特性的影响

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摘　要：采用电弧熔炼及高能球磨工艺制备出 $Dy_xPr_{2-x}Fe_{17}$ (x =0.0, 0.1, 0.2, 0.3, 0.4) 合金微粉，借助X 射线衍射(XRD)、扫描电镜(SEM)、振动样品磁强计(VSM)和网络矢量分析仪等仪器分别对合金微粉的结构、形貌、磁性能及其微波吸收性能进行了研究。结果表明，随着Dy含量的增加，$Dy_xPr_{2-x}Fe_{17}$微粉的饱和磁化强度降低。$Dy_xPr_{2-x}Fe_{17}$合金的最小反射峰频率随Dy含量的增加往高频方向移动，最小反射损耗呈先增大后减小的变化趋势；其中$Dy_{0.3}Pr_{1.7}Fe_{17}$合金具有最好的吸波效果，在最佳匹配厚度2.5 mm下，$Dy_{0.3}Pr_{1.7}Fe_{17}$合金的最小反射损耗在5.04 GHz处达到−42.38 dB左右，反射损耗小于−10 dB的频带宽度达到了1.20 GHz。

关键词：Dy-Pr-Fe 合金；高能球磨；磁性能；电磁参数；反射损耗

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